UNIVERSITY OF NOTTINGHAM

DOCTORAL THESIS

Empirical and theoretical investigations of fine particle emission from cooking

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Abstract

Exposure to fine particulate matter with diameter $\leq$ 2.5 µm ($PM_{2.5}$) is associated with elevated risks of acute and chronic health effects. These health effects are established for ambient $PM_{2.5}$ concentrations; however, the average UK resident spends 70% of their time indoors at home. $PM_{2.5}$ is also a criteria indoor air pollutant, so indoor $PM_{2.5}$ concentrations could contribute more to total personal exposure. Cooking is a primary $PM_{2.5}$ source in non-smoking dwellings. Crucially, with cooking, complete source removal is not feasible; instead ventilation strategies are used to dilute $PM_{2.5}$ concentrations or to remove emitted $PM_{2.5}$ at source. In response, this thesis used empirical and theoretical methods to measure and predict $PM_{2.5}$ concentrations in dwellings of the English housing stock, and to test the suitability of the current ventilation requirements of the English building regulations to mitigate against $PM_{2.5}$ emitted during cooking. This investigation was composed of three stages.

First, concentrations were measured in the kitchens of five multiple occupancy student dwellings situated in Nottingham, UK. Measured $PM_{2.5}$ concentrations were elevated above background levels during cooking. A behavioural intervention to encourage ventilation use during cooking resulted in a statistically significant, small decrease in concentrations in two of the five houses. Changes in cooking activity between the measurement weeks was a confounding factor.

The second stage was to develop a method to measure emission rates, using toasting bread as a simple repeatable source. Key factors for a repeatable test were controlled mixing and ventilation conditions, sufficiently small concentration logging interval, and determining custom calibration factors using gravimetric sampling to ensure accurate mass measurements. A total of 40 tests were used to produce an empirical cumulative distribution function of emission rates from toasting bread. Emission rates from cooking full meals composed of multiple stovetop processes were measured following an adapted method. Concentration

profile, average emission rate, source strength and calibration factor all varied between meals. Therefore, concurrent gravimetric sampling is imperative when using optical devices to monitor cooking source $PM_{2.5}$ concentrations. Frying in a non-stick pan and using a cooker hood during cooking reduced room emissions, and so are recommended to minimise occupant exposures.

In the final stage, probabilistic modelling techniques were used to predict 24-hour average concentrations in kitchens of the English housing stock. Over 98% houses were predicted to be too airtight to sufficiently dilute concentrations by infiltration alone. Current ventilation requirements from the building regulations were predicted to be adequate in less than 11% houses, which improved to 46% when intermittent ventilation was continued for 10 minutes after cooking. Therefore, householders are recommended to use externally exhausting mechanical ventilation throughout cooking and for a further 10 minutes. Recommended ventilation rates for concentrations to remain below guidelines in 75% kitchens are 61 L/s ($11\,h^{-1}$) constant or 79 L/s ($13\,h^{-1}$) intermittent ventilation during cooking and for a further 10 minutes. The relationship between cooker hood airflow rate and capture efficiency was defined in the form of a design tool. A cooker hood with high capture efficiency is recommended to achieve the same air quality at lower ventilation rate, and consequently lower noise and energy impacts.

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For Nana O'Leary and Grandma Lloyd

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List of Abbreviations

AC	**A**ir **C**onditioning
ACH	**A**ir **C**hanges per **H**our
ADF	**A**pproved **D**ocument **F**; guidelines to meet the UK building regulations
APS	**A**erodynamic **P**article **S**izer
AQSR	**A**ir **Q**uality **S**tandards **R**egulations
ASHRAE	The **A**merican **S**ociety of **H**eating, **R**efrigerating and **A**ir-Conditioning **E**ngineers
CDF	**C**umulative **D**istribution **F**unction
eCDF	**e**mpirical **C**umulative **D**istribution **F**unction
CE	**C**apture **E**fficiency
CF	**C**alitbration **F**actor
COPD	**C**hronic **O**bstructive **P**ulmonary **D**isease
CPC	**C**ondensation **P**article **C**ounter
DALY	**D**isability **A**djusted **L**ife **Y**ear
DNFCS	**D**utch **N**ational **F**ood **C**onsumption **S**urvey
EC	**E**lemental **C**arbon
EHS	English **H**ousing **S**urvey
ETS	**E**nvironmental **T**obacco **S**moke
GBD	The **G**lobal **B**urden of **D**isease study
GM	**G**eometric **M**ean
GSD	**G**eometric **S**tandard **D**eviation
HEPA	**H**igh **E**fficiency **P**articulate **A**ir (Filter)
I/O	**I**ndoor/**O**utdoor Ratio
IAQ	**I**ndoor **A**ir **Q**uality
IEQ	**I**ndoor **E**nvironmental **Q**uality
IUGR	**I**ntra**u**terine **G**rowth **R**estriction
IWBI	The **I**nternational **W**ell **B**uilding **I**nstitute
LH(S)	**L**atin **H**ypercube (**S**ampling)
LQ	**L**ower **Q**uartile or 25th percentile
MVHR	**M**echanical **V**entilation with **H**eat **R**ecovery
MW	**M**ann-**W**hitney U Test or Wilcoxon's Rank-Sum Test
NAAQS	**N**ational **A**mbient **A**ir **Q**uality **S**tandards (USA)
NDNS	**N**ational **D**iet and **N**utrition **S**urvey
OC	**O**rganic **C**arbon
OPC	**O**ptical **P**article **C**ounter

PAH	**P**olycyclic **A**romatic **H**ydrocarbons
c-PAH	**c**arcinogenic fraction of **PAH**
PDF	**P**robability **D**ensity **F**unction
PM$_{2.5}$	Fine **P**articulate **M**atter with diameter $\leq 2.5\,\mu m$
PM$_{10}$	**P**articulate **M**atter with diameter $\leq 10\,\mu m$
PM$_x$	**P**articulate **M**atter with diameter $\leq x\,\mu m$
PM$_{x-y}$	**P**articulate **M**atter with diameter in range $x - y\,\mu m$
PNC	**P**article **N**umber **C**oncentration
PPV	**P**urpose **P**rovided **V**entilation
PSL	**P**oly**s**tyrene **L**atex
QALY	**Q**uality **A**djusted **L**ife **Y**ear
RH	**R**elative **H**umidity
SD	**S**tandard **D**eviation
SHS	**S**econd **H**and **S**moke
SVOC	**S**emi-**V**olatile **O**rganic **C**ompound(s)
TEOM	**T**apered **E**lement **O**scillating **M**icrobalance
UFP	**U**tra**f**ine **P**articles with diameter $\leq 0.1\,\mu m$
UQ	**U**pper **Q**uartile or 75th percentile
US EPA	**U**nited **S**tates **E**nvironmental **P**rotection **A**gency
VOC	**V**olatile **O**rganic **C**ompounds
WHO	**W**orld **H**ealth **O**rganisation

List of Symbols

$C(t)$	$PM_{2.5}$ concentration at time t	$\mu g/m^3$
$\overline{C}$	Average $PM_{2.5}$ concentration	$\mu g/m^3$
$\overline{C(t)}$ or $\overline{C(T)}$	Average $PM_{2.5}$ concentration over time t or T	$\mu g/m^3$
C_b	background $PM_{2.5}$ concentration	$\mu g/m^3$
C_a	ambient $PM_{2.5}$ concentration	$\mu g/m^3$
C_{max}	maximum $PM_{2.5}$ concentration	$\mu g/m^3$
$\overline{C_{opt}}$	Average concentration from optical device	$\mu g/m^3$
$\overline{C_{ref}}$	Average concentration from reference device	$\mu g/m^3$
C_p	peak $PM_{2.5}$ concentration	$\mu g/m^3$
C_ss	steady state $PM_{2.5}$ concentration	$\mu g/m^3$
C_{Tp}	theoretical peak $PM_{2.5}$ concentration	$\mu g/m^3$
d_{AE}	aerodynamic diameter	µm
d_{opt}	optical diameter	µm
d_{VE}	volume equivalent diameter	µm
g	emission rate, constant	µg/s, mg/min
$\underline{g(t)}$	emission rate, as a function of time	µg/s, mg/min
$\overline{g(t)}$	emission rate, average	µg/s, mg/min
G	source strength	µg, mg
k	decay due to factors other than ventilation	h^{-1}, s^{-1}
m	mass	µg, mg
n, N	number of samples	-
P	penetration factor or coefficient	-
p	p-value, statistical significance test threshold	
t_{ext}	external temperature	°C
t_i	external temperature	°C
t, T	time	h, min, s
q, Q	volume flow rate	L/s, m^3/s, m^3/h
$\overline{q}$	average volume flow rate	L/s, m^3/s, m^3/h
r	effect size	
V	volume	m^3
z	statistical z-score	
α	standard error	
λ	ventilation rate	h^{-1}, s^{-1}
μ	arithmetic mean	
η	capture efficiency (CE)	%

ϕ	total decay rate	h^{-1}, s^{-1}
σ	standard deviation	

Chapter 1

Introduction

Around 40 000 deaths in the UK are attributable to exposure to outdoor air pollution each year, with more linked to indoor pollutants [1]. Possible links have been found between air pollution and cancer, asthma, stroke and heart disease, diabetes, obesity, and changes linked to dementia [1]. Detrimental effects include both increased mortality and increased morbidity, for example, there are observed links between $PM_{2.5}$ and respiratory mortality [2], but also increased hospital admissions due to respiratory symptoms [3] and exacerbation of asthma [4]. Both of these latter outcomes do not necessarily cause death. In response, targets to improve outdoor air quality have been included in the UN Sustainable Development Goals [5] and the UK Air Quality Standards Regulations set targets for reducing external concentrations of key pollutants to comply with EU legislation [6].

However, people in the UK spend an average of 70% of their time in their own houses [7]. In addition, more than a quarter of national annual energy use and carbon dioxide (CO_2) emissions can be attributed to energy use in housing [8]. Improvements to dwelling airtightness, part of the move to improve the thermal performance of buildings, without compensatory purpose provided ventilation has led to lower ventilation rates [9] and a reduction in contaminant dilution. Total

exposure can be considered as a function of the time spent and concentrations found in different micro-environments [10], and so indoor concentrations, especially in dwellings, may have a greater influence on personal exposures, and so health.

Buildings contain airborne pollutants from indoor sources, such as emissions from building materials and furnishings, and occupant activities, and from external sources. Of these pollutants, airborne fine particles of diameter $\leq 2.5\,\mu m$, known as $PM_{2.5}$ [11], are shown to pose the greatest risk [12]. $PM_{2.5}$ is respirable, so particles bypass the body's primary defences [13]. Exposure to elevated concentrations over extended periods has been linked to chronic respiratory and cardiovascular diseases, and cancer [13]. Indoor sources of $PM_{2.5}$ include cooking, smoking, spray aerosols, and various forms of combustion, including burning candles and incense [14]. Furthermore, indoor sources have been found to have a greater effect on indoor $PM_{2.5}$ concentrations than ambient sources [15]. Cooking is a source of particular interest as an activity conducted in the majority of homes.

It is impossible to stop people cooking in their own homes, therefore complete source removal is not feasible. There is a direct conflict between the need to reduce ventilation to a minimum to save energy, and the need to provide enough ventilation to ensure adequate indoor air quality, and thus reduce the risk of $PM_{2.5}$ exposure. If additional purpose provided ventilation (PPV) is not provided when retro-fitted measures are undertaken to minimise infiltration, the logical consequence is a reduction in the total building ventilation rate and an increase in the risk of $PM_{2.5}$ exposure. However, localised kitchen ventilation is not always available or used by cooks. Furthermore, current UK building regulations [16] only require the maintenance of existing ventilation sources in domestic kitchens during renovations. There are potential consequences for population health and well-being, but also financial penalties as the tax payer pays for both the energy efficiency measures and the treatment for the health damage they cause, where

social health care is available.

1.1 Background to Air Quality Research

Research into the health impacts of air pollution began in the early to mid 20th Century, triggered by sudden increases in sickness and death following several extreme pollution episodes [4], such as the Meuse Valley fog of 1930 or the London Smog of 1952. The observed increased mortality offered a quantitative measure of adverse health effects [4].

By the 1970s, there were well established links between respiratory disease and particulate and sulfur oxide pollution [4]. However, there remained disagreement on concentrations responsible for significant health impacts [4]. Epidemiologic studies provided more quantification of particulate air pollution levels found in urban areas [4], and the US Clean Air act required pollutants which 'may reasonably be anticipated to endanger public health and welfare' to be identified [17]. This move triggered further research into air pollution and health.

In the 1980s, links between increased morbidity and mortality due to cardiopulmonary disease, and very high concentrations of PM air pollution became more generally accepted [17]. It was suggested smaller particles played a larger part in the health threat [17]. Additionally, several epidemiological studies in the US demonstrated apparent health effects at lower ambient concentrations than previously expected [11]. This was likely due to a change in the size distribution and composition of airborne particles leading to an altered toxicity, and rising concentrations of nitrogen oxides, from the increased use of motorised vehicles [18]. Subsequent research, up to the end of the 20th century, found $PM_{2.5}$ able to penetrate deep into the lungs, to the alveolar gas exchange region [17].

The 2015 Global Burden of Disease Study (GBD) estimated the burden of disease attributed to 79 risk factors in 195 countries and territories [19]. Overall, long-term

exposure to ambient $PM_{2.5}$ was the fifth-ranked risk factor for global mortality, 4.2 million deaths, and sixth-ranked for global disability adjusted life years (DALYs), 103.1 million DALYs. However, the associated risks were higher in low-income and middle-income countries. In addition, the contribution of ambient air to the global burden of disease was found to have increased over the previous 25 years, with recommendations for major reductions in pollution levels to avoid increases in disease burden. Population-weighted average concentrations, used to compare the proportion of populations living in areas with different levels of pollution, show ambient $PM_{2.5}$ concentrations and household air pollution from using solid fuels vary regionally and globally [20]. In 2016, the population-weighted annual mean ambient $PM_{2.5}$ concentration for the UK was 11 $\mu g/m^3$ [20]. By comparison, for China it was 53 $\mu g/m^3$, 98 $\mu g/m^3$ in Nepal and 6 $\mu g/m^3$ in New Zealand [20].

Research over the past century has shown human health may be affected by exposure to airborne pollutants in all stages of life [1, 21]. Risks associated with exposure start at the earliest stages of life: pregnancy, infancy and early childhood, with some pollutants able to cross the placenta [1]. Childhood exposures have been found to suppress natural lung function development, and exposure later in life can lead to increased rates of lung function decline [1]. Additionally, effects of exposure are amplified for the vulnerable [1].

Overall, the most studied health endpoint in association with air pollution is mortality [21]. Additionally, associations have been made between $PM_{2.5}$ exposure and (i) pulmonary effects [21] including suppressed lung function, accelerated lung function decline in adults, and chronic exposure causing new-onset asthma in children and adults [1], and (ii) cardiovascular effects [21] such as exacerbated cardiovascular disease (CVD) including myocardial infarction, heart failure and stroke, and even deaths caused my CVD [1]. More recent studies have also investigated possible associations between air pollution and (i) reproductive and

prenatal outcomes [21], (ii) neurotoxic effects [21] and neurodevelopment [1], and (iii) onset of type II diabetes [1]. Whilst epidemiology provides some evidence of health impacts, ongoing research into the mechanistic pathways to explain the epidemiological associations is also necessary [21].

1.2 The Indoor Environment

An interest in the links between outdoor or indoor air quality (IAQ) and health date can be traced back through much of history, including Roman and Biblical references [22]. From the 1700s to the early 20th century, the focus remained on the indoor environment over outdoors. However, in the 1930s, the focus shifted to outdoors due to a lack of evidence that explained the relationship between IAQ and health [22]. IAQ was then considered a comfort parameter rather than one relating to health [22]. Both providing indoor air that should not have negative health impacts, and air that is perceived by the occupants to be acceptable are considerations when achieving good IAQ [23]. The resurging interest in the health impacts of IAQ began towards the end of the 20th century, with studies citing increasing prevalence of asthma and allergies in developed countries as their motivation [22, 23]. Globally, the burning of solid fuels indoors for both heating and cooking has a significant impact on IAQ and health [24]. This is not applicable in developed nations, such as the UK, where gas and electricity are the primary energy sources in housing [22]. Therefore, the the effect of burning solid fuels at home is not considered within this work.

IAQ research in developed countries has largely been conducted in northern Europe and North America [22]. Investigations have included the effect of damp housing and mould growth [25], links between asthma and domestic air quality [26], and the effects of household smoking behaviour [27]. The strategies to improve IAQ include source control, ventilation, air cleaning and air conditioning [23]. Some indoor pollutant sources, such as smoking or building materials, can be

dealt with using source control. However, the most appropriate mitigation strategy depends on the source and the pollutant type.

Multiple studies have found associations between cooking behaviour and health impacts. Elevated risks of lung cancer have been associated with cooking emissions and poor kitchen ventilation [28] and with cooking with traditional woks in kitchens without a cooker hood, for Taiwanese women [29]. Cooking, using both gas and electric appliances, has been found to emit multiple pollutants, including $PM_{2.5}$ [30], ultrafine particles (UFPs) [30, 31], nitrous oxides (specifically gas cookers)[31], polycyclic aromatic hydrocarbons (PAHs) [32] and volatile organic compounds (VOCs) [33]. Particulate matter from cooking sources is complex, and has been found to contain several known carcinogens [34], and as it is a priority indoor pollutant, $PM_{2.5}$ emitted from cooking sources is of interest.

There are two main methods to investigate IAQ, particularly in housing. The first is to measure concentrations in dwellings, either using static or portable personal measurement devices; for example [35, 36]. Alternatively, a variety of simulation tools can be used to model the indoor environment and predict concentrations [37–39]. Both methods have their advantages, however, large scale *in situ* monitoring is often invasive, and cost and time prohibitive. Modelling stocks of dwellings can not only estimate exposures at the population scale, but can also be used to predict the impacts of interventions. Model inputs include: weather variables, dwelling physical characteristics, indoor temperatures, ambient pollutant concentrations, dwelling ventilation rate, indoor deposition rates, and indoor emissions, based on the list compiled by Das *et al.* [37]. In the case of cooking source $PM_{2.5}$, indoor emissions are a function of emission rates from cooking and occupant cooking behaviour. Model predictions are highly sensitive to the emission rates used [37], and reported emission rates for cooking are highly variable, even for the same cooking methods and ingredients; for example, reported emission rates for the toasting of bread vary between 0.11 mg/min [35] and 9.5 mg/min [40].

1.3 Aims and Objectives

This project aims to investigate $PM_{2.5}$ emissions from cooking meals typical to a European or western diet, and to investigate associated risks within the UK housing stock. A further aim is to assess the suitability of the current ventilation requirements as set out within Approved Document F of the UK Building Regulations (ADF) [16], although similar methods could be used for international housing stocks to assess the relevant building regulations. These aims require a multifaceted approach, and are achieved through the following objectives:

(i) Measure $PM_{2.5}$ concentrations in domestic kitchens, to investigate the concentrations that might be found in housing, and to examine the effect of cooking on these concentrations. Furthermore, investigate the potential to reduce cooking related indoor $PM_{2.5}$ concentrations using existing ventilation mechanisms, using a simple intervention study.

(ii) Develop a method to measure $PM_{2.5}$ emission rates from a variety of cooking sources. The resulting emission rates should be suitable for use in statistical modelling.

(iii) Measure the $PM_{2.5}$ emission rate from cooking complete meals, to add to the existing cooking emission rate data.

(iv) Measure the effect of equipment and ingredients on $PM_{2.5}$ emissions from cooking, including the potential to reduce emissions using a cooker hood

(v) Synthesise probability density functions (PDFs) which predict $PM_{2.5}$ concentrations due to cooking in English domestic kitchens for a range of scenarios. And through this statistical modelling approach, investigate the current ventilation requirements within Approved Document F.

1.4 Thesis Outline

The aims and objectives of this thesis, identified through a literature review, were achieved by conducting multiple empirical and theoretical investigations. The findings from earlier investigations were used to inform the methodologies for later work, and as such, the methods developed organically. Therefore, Chapters $3-6$ of this thesis, see below, each contain their own methods, results and discussion, with the overall findings drawn together in Chapter 7.

The thesis is divided into the following chapters:

1. Introduction.
2. Literature Review. Including a critical literature review of existing measurement techniques and calculation methods for measuring $PM_{2.5}$ emission rates, an analysis of previous measurement and modelling studies, and current knowledge of the health impacts of exposure to $PM_{2.5}$.
3. Concentrations and Reductions Measured in Domestic Kitchens. An investigation using in situ measurements in student housing to study the effect of cooking on $PM_{2.5}$ concentrations
4. A Method to Measure $PM_{2.5}$ Emission Rates. The development of a method to measure emission rates, using toasting bread as a simple source type.
5. Emission rates from cooking full meals and reduction potential of using a cooker hood.
6. Predicted $PM_{2.5}$ Concentrations in the English Housing Stock. The development and findings of a statistical modelling approach to predict $PM_{2.5}$ in the English Housing stock, and test the ventilation requirements from ADF.
7. Discussion and Recommendations for Future Work.
8. Conclusions.

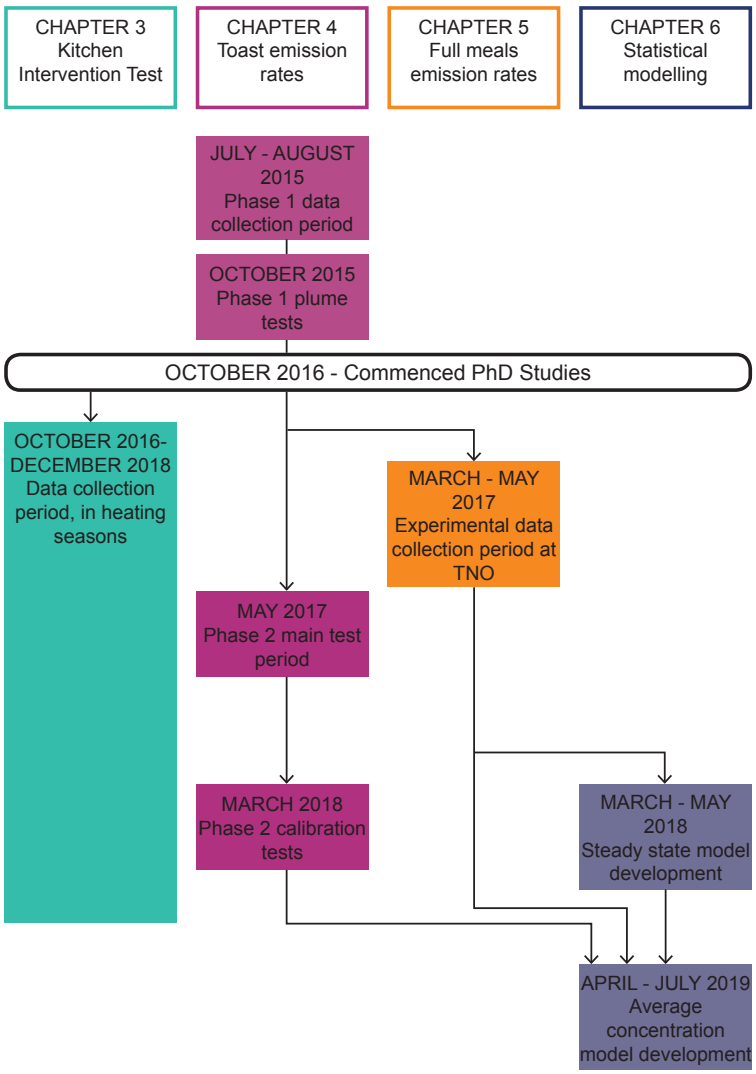


FIGURE 1.1: Flow chart of primary activities

Much of the empirical work included in Chapters $3-5$ was conducted concurrently, and so they are not presented chronologically. Figure 1.1 shows a flow chart of main activities; subsequent data analysis periods are not included.

The intervention test presented in Chapter 3 ran alongside other investigations, as data was collected in conjunction with undergraduate dissertation students. Intervention studies might normally be designed to incorporate findings from more fundamental work, such as those in Chapters $4-6$. In this case, the intervention was a baseline assessment of existing kitchen ventilation and the degree to which it could be used to reduce $PM_{2.5}$ concentrations.

Chapter 4 is split into two distinct methodologies, *Phase 1* and *Phase 2*. As shown in Figure 1.1, the Phase 1 measurements were conducted prior to commencing this PhD. This phase is included as the problems identified in the methodology informed subsequent decisions in the experimental design for Chapter 4 Phase 2 and Chapter 5.

In turn, explorations of mixing and calibration factor results from Chapter 5 informed methodological decisions in Chapter 4 Phase 2.

Chapter 2

Literature Review

Some work from this chapter has previously been published in conference and journal papers. Specifically, Section 2.5.2 was published in:

O'Leary, C. *et al.* Investigating measurements of fine particle (PM2.5) emissions from the cooking of meals and mitigating exposure using a cooker hood. *Indoor Air* **29,** 423–438. ISSN: 0905-6947 (2019)

Section 2.5.3 was presented in a conference paper and poster at the 2017 AIVC conference:

O'Leary, C. & Jones, B. *A Method to Measure Emission Rates of $PM_{2.5}$s from Cooking* in *38th Air Infiltration and Ventilation Centre Conference* (Nottingham, UK, 2017), 463–472

Some content, especially parts of Section 2.4.2, was included in:

O'Leary, C. *et al.* Setting the standard: The acceptability of kitchen ventilation for the English housing stock. *Building and Environment*, 106417. ISSN: 0360-1323 (2019)

2.1 Health Impacts of $PM_{2.5}$

There is historic scientific evidence linking $PM_{2.5}$ concentrations and exposure to a variety of health effects, as outlined in Chapter 1. However, before investigating $PM_{2.5}$ in the indoor environment, these health effects should be further investigated

to establish the limits to existing knowledge, and to inform decisions about threshold levels.

The key principles and terms involved in exposure analysis are important to help understand the complex issues at play. Within the body of this work, the main *pollutant* or *agent* considered is $PM_{2.5}$, and the primary *target* is humans, although other pollutants or targets may be mentioned. As $PM_{2.5}$ is an airborne pollutant, the *exposure pathway*, or the course the pollutant takes to reach the target from the source [44], mainly involves the movement of air. The *exposure route* differs from the exposure pathway as it is how the pollutant enters the body after contact [44]. For $PM_{2.5}$ this primarily involves the respiratory system. The distinction between exposure pathways and exposure routes is similar to distinction between *exposure* and *dose*. These two terms are often used interchangeably in the literature, but this is not wholly accurate. Exposure is the contact between a pollutant and target[44, 45], in air quality work this is usually expressed as the pollutant concentration in the environment in which the person is located. In contrast, *dose* refers to the amount of the pollutant that enters the the target [44, 45]. At the most basic level, dose is a function of the concentration in a specific environment and the amount of time a person is exposed to said concentration [44], although other factors including activity levels and respiratory rate may also impact dose [44, 46].

Two main approaches are frequently used in literature to investigate the health impacts of airborne pollutants such as $PM_{2.5}$. *Epidemiology* identifies the risk of specific diseases or health outcomes among populations, and identifies risk factors [45]. It is a powerful process that can help to identify possible causes of a disease or illness before the mechanisms that cause the disease are understood, by using statistical methods to find associations between potential causes and incidence rates of diseases and injuries in a specific population [45].

Once epidemiological methods have established associations between potential causes and effects, the next stage is frequently to conduct controlled tests to

investigate the toxicology and mechanisms by which the observed health effects might occur. These might take the form of *in-vitro* tests, which involve exposing cultured cells to the pollutant of choice, or *in-vivo* tests where a whole animal is exposed and the effects observed to simulate the response in a human [45]. Other tests also include controlled exposure studies, where human participants are exposed to controlled concentrations of a specific pollutant and specific health effects are monitored [47]. Such studies are less common due to the ethical implications of such a study, and the fact that they are limited to only observing short term effects.

With a pollutant such as $PM_{2.5}$, the potential exposures and health effects are split into two categories: *acute* (short term) effects and *chronic* (long term) effects [45]. Acute exposure periods can be anything from a few hours to a few days, and chronic health effects usually include the effects of exposures to lower concentrations for extended periods, from years to lifetimes [45]. Acute and chronic exposures are typically linked to different health effects, and both are important when considering the health impacts of a pollutant on a population. The health endpoints considered can also be split into two key categories: *mortality* (death rates), and *morbidity* (rates of persons falling ill) [48]. Both of these can be further broken down to relate to specific diseases or injuries, or considered in their entirety.

2.1.1 Epidemiology

Numerous existing studies investigate associations between $PM_{2.5}$ concentrations and health outcomes. These exist for chronic and acute effects, considering morbidity and mortality endpoints, however they most frequently consider ambient or outdoor $PM_{2.5}$ concentrations as a proxy for exposure. This limitation will be discussed later. Additionally, some earlier studies investigate PM_{10}, a larger size fraction which includes particles with diameter less than 10 µm. A select few of these studies are included here, as $PM_{2.5}$ is a subset of the PM_{10} size fraction.

However, it is not possible to attribute the effects observed in these studies specifically to either size fraction.

2.1.1.1 General Health Effects

Mortality, both all-cause and cause-specific, is the most studied health endpoint in association with air pollution, for both chronic and acute effects [21]. Multiple studies have found small but constant positive associations between PM_{10} concentrations and mortality rates [21]. For $PM_{2.5}$, there is a clear link between ambient concentrations and all-cause mortality; however this is especially true for cardiovascular or cardiopulmonary and respiratory mortality [21]. Hospitalisation rates are frequently used morbidity health outcome, usually reported as cause-specific, for example hospitalisation for respiratory symptoms. Some evidence relating to mortality and hospitalisation endpoints will now be further explored.

Ambient air pollution, in particular PM_{10}, is well established as a risk factor for early mortality, with an estimated increased daily mortality rate between 0.7% and 1.6% for each $10\,\mu g/m^3$ increase in PM_{10} reported in 1995 [49]. Similar associations have been found for $PM_{2.5}$ in multiple locations. In Canada, Burnett *et al.* [50] reported positive, statistically significant associations between daily variations in gas and particulate pollution and mortality rates, where they found particulate matter explained 28% of the total health effect, although the effect of $PM_{2.5}$ varied between cities. And in a global review of the health effects of air pollution, Brunekreef & Holgate [18] found associations between both acute and chronic exposures to air pollution, and cardiac death and hospitalisations for respiratory symptoms.

In studies specifically relating to ambient $PM_{2.5}$, effects have been found for both acute and chronic exposure. Associations with short term concentrations include increased daily all-cause mortality [51–54], respiratory mortality and

cardiovascular mortality [51, 52, 54], and hospital admissions [51]. These findings have been reproduced in meta-analyses [51, 52, 54] and for different locations. Atkinson *et al.* [51] reported global associations between $PM_{2.5}$ and mortality and hospitalisations. However there was substantial variation in the size of the effect between different global regions. Additionally, they found larger associations for respiratory mortality than cardiovascular. In terms of long term $PM_{2.5}$ concentrations, associations have been found with all-cause, cardiopulmonary and lung cancer mortality [2, 55] cardiovascular, and elderly mortality [2], and non-accidental mortality [56]. Thurston *et al.* [56] also found an association with respiratory mortality, but this effect was restricted to never-smokers. Again, these effects have been observed in a global meta-analysis [2], where a larger effect was observed for all-cause mortality when average concentrations remained below $10\,\mu g/m^3$. Additionally, a larger effect was observed for cardiopulmonary, cardiovascular and elderly mortality. There is some evidence that individuals with diabetes, myocardial infarction (MI) survivors, the elderly, and those with lower socio-economic status may be more susceptible to these health effects [21].

To summarise, there is substantial evidence of associations between both short and long term ambient $PM_{2.5}$ concentrations and increased hospitalisations and death rates for multiple causes. Studies have observed some level of heterogeneity in these findings, the reasons for which are unclear. Additionally, the relative effect on different cause mortality does also vary between studies, however respiratory and cardiovascular mortality and morbidity appear to have the strongest associations with ambient $PM_{2.5}$.

2.1.1.2 Pulmonary and Respiratory Effects

The effect of $PM_{2.5}$ on the lungs and respiratory system is perhaps the most obvious potential health effect, as this is the system through which airborne pollutants enter the body. Within this area there has been some emphasis on the effect on children,

who are seen as particularly vulnerable as their lungs are still immature and their respiratory defence mechanisms are still developing [21]. Higher acute PM_{10} concentrations have been associated with increased reports of asthmatic attacks [4], and ambient particulate exposure has been associated with increased risk of bronchitis, reduced lung function and asthma rates in children [57]. Greater acute effects on Peak expiratory flow (PEF) and lower respiratory symptoms or cough in children have been observed for $PM_{2.5}$ than PM_{10}, although there is some evidence of heterogeneity and study bias in this finding [21]. Observed chronic effects of PM in children include reduced lung function and development and respiratory symptoms, and in adults include lung function deficits, increased symptoms of obstructive airway disease, and increased risk of lung cancer mortality [21].

In a review, Xing *et al.* [58] identify multiple epidemiological studies that identify associations between both chronic and acute ambient PM and respiratory health effects. Elsewhere, meta-analyses of acute $PM_{2.5}$ exposure found exposure to elevated ambient $PM_{2.5}$ was associated with increased chronic obstructive pulmonary disease (COPD) hospitalisations and mortality [3]. This meta-analysis included 12 studies examining the effect on hospital admissions and 6 examining mortality. There was some observed heterogeneity, with higher effects observed in single city studies than multi-city, however the authors concluded that there is evidence to suggest that ambient $PM_{2.5}$ concentrations are a risk factor in COPD onset and progression.

Two meta-analyses consider the possible association between $PM_{2.5}$, among other air pollutants, and pneumonia in children [59, 60] . MacIntyre *et al.* [60] investigated associations between air pollution and early childhood respiratory infections, namely pneumonia, croup and otitis media. In the ten European birth cohorts which formed the basis of the study, $PM_{2.5}$ was not significantly associated with any of the respiratory infections considered. However, Nhung *et al.* [59] did find a significant positive association between ambient $PM_{2.5}$ concentrations and hospital

admissions for children with pneumonia, especially in lower income countries.

To summarise, there is evidence that ambient $PM_{2.5}$ concentrations are associated with pulmonary and respiratory health effects in children and adults. Observed health effects include reduced lung function and development, asthmatic symptoms, increased risk of COPD development and progression, and lung cancer. There is also some evidence that there may be an association between $PM_{2.5}$ and pneumonia in children, however the evidence for this is inconsistent. Studies also observe some heterogeneity, the reasons for which are as yet unclear.

2.1.1.3 Cardiovascular Health

After respiratory or pulmonary effects, cardiovascular health effects associated with air pollution are next most widely studied. There is evidence to suggest cardiovascular health impacts are have the highest attributable risk associated with $PM_{2.5}$ [21].

In a statement from the *American Heart Association*, exposure to $PM_{2.5}$ was deemed to contribute to cardiovascular morbidity and mortality [61]. The statement cited evidence that both chronic and acute $PM_{2.5}$ exposures are linked to increased risk of cardiovascular disease related mortality and non-fatal events. This conclusion is supported elsewhere in the literature, with studies including meta-analyses finding significant associations between short term $PM_{2.5}$ and increases in markers for cardiovascular disease [62], increased blood pressure [63], cardiac arrhythmia hospitalisations and mortality [64], and MI risk [65]. Studies investigating long term $PM_{2.5}$ concentrations have found associations with increases in markers for cardiovascular disease [62], and cardiovascular disease mortality, especially among never smokers [66]. However, Polichetti *et al.* [62] question whether it is appropriate to always separate acute and chronic concentration effects as acute exposure may be superimposed on effects of chronic exposure.

There is also some evidence of associations between particulate air pollution and cerebrovascular effects, including a small association between ultrafine particle (UFP) concentrations and stroke observed in Helsinki [21]. UFPs are themselves the smaller size fraction of $PM_{2.5}$, consisting of particles with diameter $\leq 0.1\,\mu m$. In particular, there is evidence to suggest an acute PM exposure is a risk factor for ischemic stroke [67]. Long term ambient $PM_{2.5}$ exposure was associated with increased risk of incident stroke in European cohorts of never-smoking individuals aged over 60 years [68]. Significant associations were found for incidence of stroke and stroke mortality for global, European and North American populations [69], although here there was no significant association for Asia. Yuan *et al.* [69] also found the significant association among never smokers, whereas there was no significant association among current smokers.

In summary, there is evidence to suggest ambient $PM_{2.5}$ is a risk factor for multiple cardiovascular morbidities and mortalities, including MI, cardiac arrhythmia and stroke. Again, there is some evidence of regional heterogeneity and smoking behaviour is a confounding variable.

2.1.1.4 Other Physical Health Effects

The physical health effects investigated so far are the most comprehensively researched. However, there is some limited evidence of other potential physical health effects of $PM_{2.5}$ exposure.

There is some, limited evidence of potential neurotoxic effects. This includes associations with inflammatory processes in the central nervous system crucial in neurodegenerative diseases such as Alzheimer's and Parkinson's disease, and some evidence of increased risk of developing Alzheimer's disease associated with PM exposure in smokers [21]. These links are less widely researched and warrant more investigation.

Two studies also suggest an association between chronic $PM_{2.5}$ exposure and onset of diabetes mellitus (commonly known as diabetes), particularly Type 2 [70, 71]. This includes an association between increased annual $PM_{2.5}$ concentration in one to five years preceding diagnosis and increased risk of Type 2 diabetes mellitus found in a meta-analysis of studies from Europe and North America.

Some studies have investigated prenatal outcomes in association with $PM_{2.5}$, although this does remain a comparatively new field [21]. The main outcomes considered include premature birth, birth weight, intrauterine growth restriction (IUGR) and small for gestational age, and birth defects [21]. Associations were observed between ambient $PM_{2.5}$ concentrations during pregnancy and low term birth weight [72, 73]. Additionally, one study found some association with small for gestational age newborns [73]. Like many effects described previously, there was some regional heterogeneity. Few studies investigated birth defects, such as congenital heart defects, and those that did generally found no association with $PM_{2.5}$ [21].

In summary, there is limited yet growing evidence of some physical health effects of ambient $PM_{2.5}$ other than the most widely reported cardiovascular and respiratory morbidities and mortalities. These potential health effects include low birth weight, small for gestational age, diabetes mellitus and neurodegenerative disease such as Alzheimer's disease. However, the evidence for these health endpoints is highly limited, and warrants further research.

2.1.1.5 Mental Health Effects

Another new and less widely researched field is the potential impacts of air pollution, including $PM_{2.5}$, on mental health. This is a growing area for research, and therefore is briefly considered here. In a recent review, Hoisington *et al.* [74] posed ten questions concerning the built environment and mental health. Of these the most relevant is "*What is the impact of indoor air pollution, or outdoor air pollution*

that can access the indoor environment, on mental health?". The review itself is focused on the indoor environment, however, due to the lack of available data on indoor air, considered studies relating to ambient air. The authors cite studies with associations between acute ambient PM_{10} exposure and increased hospital admissions for mental disorders, between ambient $PM_{2.5}$ and increased risk of suicide and depressive disorders, and between maternal exposure to ambient air pollution in late second and early third trimester of pregnancy and increased risk of autism spectrum disorder in the child.

In Europe and North America, a longitudinal study found $PM_{2.5}$ significantly associated with increased psychological distress in the USA [75], $PM_{2.5}$ exposure at age 12 was significantly associated with increased occurrence of depressive disorder at age 18 among London based children [76], chronic exposure was associated with increased depressive symptoms in older Americans [77], and older Americans exposed to increased chronic $PM_{2.5}$ exposures were at risk of decreased cognitive function equivalent to ageing 1.6 years [78]. Studies of acute $PM_{2.5}$ concentration have found an association with increased suicide rates after a 2 day lag in Salt Lake County, Utah [79], and increased anxiety symptoms in older Americans [77].

More investigations have been conducted into mental health impacts in Asia, particularly in South Korea and China. Air pollution including PM_{10} was associated with reduced hedonic happiness and increased rate of depressive symptoms in China [80], and in Guangzhou, China acute PM_{10} was associated with a significant increase in suicide risk, especially in males and people educated to a higher level [81]. Some associations were observed between acute $PM_{2.5}$ and hospitalisations for mental disorders in Beijing, China [82], and a similar observation was made for PM_{10} concentrations in Shanghai, China [83]. In South Korea, long term PM_{10} concentrations may be a risk factor for a range of mental health outcomes including suicide mortality [84], subjective stress and suicidal

ideation [85]. Additionally, a meta-analysis found weekly suicide rates significantly associated with PM_{10} level in South Korea [86].

In summary, there is limited research which finds associations between ambient particulate matter and mental health outcomes, and much of the existing research focuses on PM_{10} rather than the smaller size fraction $PM_{2.5}$, the focus of this thesis. Additionally, multiple studies exist for China and South Korea, where equivalent work has not been conducted in Europe. Given the regional heterogeneity found in many studies of physical health effects, it would be appropriate to conduct repeat studies in Europe and other parts of the world before assuming the same effects can be found globally. Therefore, there is some limited evidence that $PM_{2.5}$ may be a risk factor for mental ill-health, but more research is needed to confirm these associations.

2.1.1.6 Concentration-, Exposure- and Dose-Response

In establishing the health effects of any pollutant, it is beneficial to define the relationship between dose or exposure and a specific health endpoint. Within the literature, these curves can take the form of dose-, exposure- or concentration-response curves, depending on the method by which they were created. Whilst the different response curves do exist in the literature, concentration-response curves are most frequently available. Additionally, epidemiological studies frequently report the increased risk of a specific health outcome associated with a set increase in the pollutant of interest. This second approach usually assumes a constant effect. Existing findings from both approaches will be explored here.

Studies that report a constant effect are considered first. Tables 2.1 to 2.4 compile reported associations between short and long term $PM_{2.5}$ concentrations and mortality and morbidity effects from the literature. Both all-cause and cause-specific associations are included, and grouped by cause. Preference was

given to results from meta-analyses, and studies were excluded if they did not report an effect per increase in $PM_{2.5}$ concentrations in $\mu g/m^3$, and if 95% confidence intervals were not provided. Reported effects in the literature were mostly presented as percentage increase, hazard ratio (HR), odds ratio (OR) or relative risk (RR). Additionally, two studies reported effects on health outcomes where an absolute rather than relative outcome is the most appropriate measure. One reported an association with term birth weight in grams [73], and the second reported an association with raised blood pressure in mmHg [63]. Both are included in the appropriate tables, but mainly as a point of interest, as they are not directly comparable with other findings. All the included studies are for ambient $PM_{2.5}$ concentrations.

The observed increased risk of mortality associated with a $10\,\mu g/m^3$ increase in long term $PM_{2.5}$ concentration, is between 3 and 27%, see Table 2.1. Additionally, Vodonos *et al.* [2] report a larger increase associated risk of all-cause mortality for $1\,\mu g/m^3$ at lower average concentrations than higher. This suggests a non-linear exposure-response relationship. Most chronic morbidity health endpoints included in Table 2.2 are hazard ratios for incidence. These range between 1.09 and 1.74 per $5\,\mu g/m^3$ increase in $PM_{2.5}$, and indicate increased risk for people over 65 years of age and never smokers.

The short term risk of death associated with a $10\,\mu g/m^3$ increase in $PM_{2.5}$, at $0.4 - 3.36\%$, is lower than the long term risk, see Table 2.3. The increased risk of all-cause mortality associated with a $10\,\mu g/m^3$ increase in daily $PM_{2.5}$ is $0.4 - 1.5\%$. The highest observed mortality risk is for ischaemic heart disease, at 3.36%, although the 95% confidence intervals for this finding are wide [51]. Most acute morbidity findings are for hospital admissions, see Table 2.4 which includes both physical and mental health associations. An $0.96 - 3.94\%$ increased risk of hospitalisation due to respiratory or pulmonary conditions is associated with a $10\,\mu g/m^3$ increase in $PM_{2.5}$. Similar risk is observed for cardiovascular hospitalisation, with the exception

TABLE 2.1: Associations between long term $PM_{2.5}$ concentrations and chronic mortality health endpoints

Health effect	$\Delta PM_{2.5}$ ($\mu g/m^3$)	Time Period	Associated Risk	95% CI	Source
All-cause	10	annual	HR 1.03	1.00, 1.05	Thurston *et al.* [56]
All-cause	10	long term	RR 1.06	1.02, 1.11	Pope *et al.* [55]
All-cause	10	long term	RR 1.06	1.04, 1.08	COMEAP [87]
All-cause	10	annual	+7.3%	+7.1, 7.5%	Di *et al.* [88]
All-cause, person-years $< 12\,\mu g/m^3$	10	annual	+13.6%	+13.1, 14.1%	Di *et al.* [88]
All-cause, $\mu = 10\,\mu g/m^3$	1	long term	+1.29%	+1.09, 1.50%	Vodonos *et al.* [2]*
All-cause, $\mu = 15.7\,\mu g/m^3$	1	long term	+1.03%	+0.97, 1.11%	Vodonos *et al.* [2]*
All-cause, $\mu = 30\,\mu g/m^3$	1	long term	+0.82%	+0.52, 1.12%	Vodonos *et al.* [2]*
Respiratory,never smokers	10	annual	HR 1.27	1.03, 1.56	Thurston *et al.* [56]
Respiratory, $\mu = 10\,\mu g/m^3$	1	long term	+1.13%	+0.85, 1.41%	Vodonos *et al.* [2]*
Lung cancer, $\mu = 10\,\mu g/m^3$	1	long term	+1.22%	+0.87, 1.39%	Vodonos *et al.* [2]*
Lung cancer	10	long term	RR 1.14	1.04, 1.23	Pope *et al.* [55]
Cardiopulmonary	10	long term	RR 1.09	1.03, 1.16	Pope *et al.* [55]
Cardiopulmonary, $\mu = 10\,\mu g/m^3$	1	long term	+1.92%	+1.59, 2.25%	Vodonos *et al.* [2]*
Cardiovascular	10	annual	HR 1.10	1.05, 1.15	Thurston *et al.* [56]
Cardiovascular disease	10	annual	+12%	+8, 16%	Liu *et al.* [66]*
Cardiovascular disease, never smokers	10	annual	+18%	+6, 32%	Liu *et al.* [66]*
Cardiovascular, $\mu = 10\,\mu g/m^3$	1	long term	+1.46%	+1.25, 1.67%	Vodonos *et al.* [2]*
Stroke	5	long term	HR 1.11	1.05, 1.17	Yuan *et al.* [69]*
Elderly, $\mu = 10\,\mu g/m^3$	1	long term	+1.61%	+1.35, 1.85%	Vodonos *et al.* [2]*

*indicates results from a meta-analysis

TABLE 2.2: Associations between long term $PM_{2.5}$ concentrations and chronic morbidity health endpoints

Health effect	$\Delta PM_{2.5}$ ($\mu g/m^3$)	Time Period	Associated Risk	95% CI	Source
Stroke: incidence	5	annual	HR 1.19	0.99, 1.62	Stafoggia *et al.* [68]
Stroke: incidence	5	long term	HR 1.11	1.05, 1.17	Yuan *et al.* [69]*
Stroke: incidence, North America	5	long term	HR 1.09	1.05, 1.14	Yuan *et al.* [69]*
Stroke: incidence, Europe	5	long term	HR 1.07	1.05, 1.10	Yuan *et al.* [69]*
Stroke: incidence, $\geq$60 years	5	annual	HR 1.40	1.05, 1.87	Stafoggia *et al.* [68]
Stroke: incidence, never smokers	5	annual	HR 1.74	1.06, 2.88	Stafoggia *et al.* [68]
Stroke: incidence, $< 25\,\mu g/m^3$	5	annual	HR 1.33	1.01, 1.77	Stafoggia *et al.* [68]
Type 2 Diabetes, development	10	annual	+10%	+2, 18%	Eze *et al.* [71]
Small for gestational age (SGA)	10	pregnancy	OR 1.04	1.01, 1.07	Stieb *et al.* [73]
Reduced term birth weight	10	pregnancy	$-20.5\,g$	$-24.7\,g$, $-16.4\,g$	Stieb *et al.* [73]
Moderate-to-severe anxiety symptoms	5	180 days	OR 1.55	1.31, 1.85	Pun *et al.* [77]

*indicates results from a meta-analysis

of one finding from Atkinson *et al.* [51] who found a possible decreased risk of stroke hospitalisation in those over 65 years of age. There are relatively few studies which report observed acute mental health risks in a suitable form, although for those that do, the increased risk is comparable to that for acute physical health. All of the studies included in Table 2.4 assume a constant exposure-response relationship.

These findings report relatively consistent health risks associated with exposure to increased $PM_{2.5}$ concentrations. Additionally, there is some suggestion that the assumption of a constant effect of $PM_{2.5}$ may not be justified, as demonstrated by Vodonos *et al.* [2] who observed larger chronic effects at lower concentrations. Concentration-response curves can further illuminate this relationship.

TABLE 2.3: Associations between short term $PM_{2.5}$ concentrations and acute mortality health endpoints

Health effect	$\Delta PM_{2.5}$ ($\mu g/m^3$)	Time Period	Associated Risk	95% CI	Source
All-cause	10	daily	+1.04%	+0.52, 1.56%	Atkinson *et al.* [51]*
All-cause	10	daily	+1.5%	+1.3, 1.7%	Karimi *et al.* [54]*
All-cause	10	daily	+1.2%	+0.5, 1.9%	Levy *et al.* [89]*
All-cause, older adults	10	daily	+1.05%	+0.95, 1.15%	Di *et al.* [53]
Total non-accidental	10	daily	+0.4%	+0.22, 0.59%	Lu *et al.* [52]*
Respiratory	10	daily	+1.51%	+1.01, 2.01%	Atkinson *et al.* [51]*
Respiratory	10	daily	+0.8%	+0.4, 1.0%	Karimi *et al.* [54]*
Respiratory disease	10	daily	+0.75%	+0.39, 1.11%	Lu *et al.* [52]*
COPD	10	daily	+2.5%	+1.5, 3.5%	Li *et al.* [3]*
COPD, excluding asthma	10	daily	+2.86%	-0.12, +5.93%	Atkinson *et al.* [51]*
Cardiovascular	10	daily	+0.84%	+0.41, 1.28%	Atkinson *et al.* [51]*
Cardiovascular	10	daily	+0.5%	+0.1, 2.0%	Karimi *et al.* [54]*
Cardiovascular disease	10	daily	+0.63%	+0.35, 0.91%	Lu *et al.* [52]*
Ischaemic heart disease	10	daily	+3.36%	+0.68, 6.10%	Atkinson *et al.* [51]*
Stroke	10	daily	+1.85%	+0.74, 2.97%	Atkinson *et al.* [51]*

*indicates results from a meta-analysis

TABLE 2.4: Associations between short term $PM_{2.5}$ concentrations and acute morbidity health endpoints

Health effect	$\Delta PM_{2.5}$ ($\mu g/m^3$)	Time Period	Associated Risk	95% CI	Source
Hospitalisation: respiratory diseases	10	daily	+0.96%	-0.63, +2.58%	Atkinson *et al.* [51]*
Hospitalisation: COPD	10	daily	+3.1%	+1.6, 4.6%	Li *et al.* [3]*
Hospitalisation: COPD incl asthma, 65+	10	daily	+1.85%	-2.07, +5.93%	Atkinson *et al.* [51]*
Hospitalisation: COPD excl asthma, 65+	10	daily	+2.36%	+1.0, 3.73%	Atkinson *et al.* [51]*
Hospitalisation: Lower respiratory infection, 65+	10	daily	+3.94%	+2.11, 5.80%	Atkinson *et al.* [51]*
Hospitalisation: asthma, 0-14	10	daily	+2.29%	-0.09, +4.73%	Atkinson *et al.* [51]*
Hospitalisation: respiratory, 0-14	10	daily	+2.45%	+0.12, 4.85%	Atkinson *et al.* [51]*
Hospitalisation: pneumonia, children	10	daily	+1.8%	+0.5, 3.1%	Nhung *et al.* [59]*
Hospitalisation: cardiovascular disease	10	daily	+0.90%	+0.26, 1.53%	Atkinson *et al.* [51]*
Hospitalisation: cardiac, 65+	10	daily	+3.05%	+1.64, 4.48%	Atkinson *et al.* [51]*
Hospitalisation: ischaemic heart disease, 65+	10	daily	+2.52%	+0.53, 4.55%	Atkinson *et al.* [51]*
Hospitalisation: stroke, 65+	10	daily	-0.45%	-2.24, +1.33%	Atkinson *et al.* [51]*
Hospitalisation: heart failure, 65+	10	daily	+4.39%	+1.35, 7.53%	Atkinson *et al.* [51]*
Hospitalisation: dysrhythmias, 65+	10	daily	+0.60%	+0.03, 1.17%	Atkinson *et al.* [51]*
Hospitalisation or mortality: cardiac arrhythmia	10	$\leq$ 7 days	RR 1.015	1.006, 1.024	Song *et al.* [64]*
Systolic blood pressure (SBP)	10	daily	1.393 mmHg	0.874, 1.912 mmHg	Liang *et al.* [63]*
Diastolic blood pressure (DBP)	10	daily	0.895 mmHg	0.49,1.299 mmHg	Liang *et al.* [63]*
Hospitalisation: schizophrenia, lag day 3	10	daily	+0.49%	+0.04, 0.95%	Gao *et al.* [82]
Hospitalisation: schizophrenia, lag day 0-6	10	daily	+1.01%	+0.02, 2.00%	Gao *et al.* [82]
Moderate-to-severe depressive symptoms	5	7 days	OR 1.09	1.01, 1.17	Pun *et al.* [77]
Moderate-to-sever depressive symptoms	5	30 days	OR 1.20	1.08, 1.33	Pun *et al.* [77]

*indicates results from a meta-analysis

Pope *et al.* [55] estimated concentration-response curves for long term ambient $PM_{2.5}$ in the American Cancer Society study. They found the concentration responses for all-cause mortality, cardiopulmonary mortality and lung cancer mortality to appear monotonic and are not significantly different from literature, although elsewhere a log-linear fit where the slope decreases at higher concentrations was shown to fit the data [11]. Additionally, the shape of the concentration-response function at concentrations above those measured in the study is unknown [11]. Pope & Dockery [11] compared these curves to other data found in literature, which they also found could be represented reasonably by a linear model. These included two studies on mortality rates in the US, and one for children's lung growth.

Thurston *et al.* [56] also considered the relationship between annual $PM_{2.5}$ concentrations and all-cause and cardiovascular mortality rates. Here, the predicted effect of $PM_{2.5}$ on all-cause mortality was lower than previous estimates. The concentration responses were also found to be monotonic in shape, especially for cardiovascular mortality, for annual $PM_{2.5}$ concentrations $2.9 - 28.0\,\mu g/m^3$. This concentration response curve for all-cause mortality also has some suggestion that at concentrations below $10\,\mu g/m^3$ the estimated mortality rate may not continue to decrease, however there is much larger uncertainty in the predictions in this range, and fewer observed concentrations at lower levels.

In contrast, there is some evidence from that at overall average concentrations below $10\,\mu g/m^3$, there is a larger increase in mortality rate per unit increase $PM_{2.5}$ [2]. In a global meta-analysis, Vodonos *et al.* [2] produced curves to describe how the concentration-response relationship changed with mean exposure level. The authors reported the effect of an increase in $PM_{2.5}$ at three different concentration levels: $10\,\mu g/m^3$, $15.7\,\mu g/m^3$ (the overall mean concentration across all studies), and $30\,\mu g/m^3$. They found a $1\,\mu g/m^3$ increase in $PM_{2.5}$ was associated with respective increases of 1.29%, 1.03% and 0.82% in all-age, all-cause mortality. This suggests that rather than a lower concentration threshold for observed effects, there may be greater benefits of further decreasing in already low concentrations. Similar to the models produced by Thurston *et al.* [56] there was also less certainty in the curves a the extreme high and low $PM_{2.5}$ concentrations, at which there were fewer published studies. The GBD study included exposure-response functions for relative risk of ischaemic heart disease, cerebrovascular disease, lower respiratory infection, lung cancer and COPD associated with long term ambient $PM_{2.5}$ concentrations [19]. These were also non-linear and showed a similar shape to those discussed previously, with the highest associated relative risk for ischaemic heart disease at age 25.

Similar relationships have also been reported for acute $PM_{2.5}$ exposure. Di *et al.* [53]

estimated exposure-response curves for short term $PM_{2.5}$ concentrations. At concentrations below $15\,\mu g/m^3$, they found a linear relationship, with relatively small 95% confidence intervals. Above this concentration, the gradient dropped, and the 95% confidence intervals grew, indicating less certainty in the predictions.

Generally these few studies indicate that there is no lower threshold of effect for ambient $PM_{2.5}$, and there is some evidence to suggest there may be greater benefits to further reducing low concentrations. The three key caveats should also be remembered: (i) these studies are primarily for outdoor concentrations of outdoor source $PM_{2.5}$; (ii) the rationalisation is based on associations observed in epidemiological evidence, which does not prove causation; and (iii) there is greater uncertainty in the estimated concentration-response curves at lower concentrations, largely due to there being less available data.

To this point, individual morbidities and mortalities associated with $PM_{2.5}$ concentrations have mostly been considered separately. However, methods by which these effects can be combined into a single metric are useful for both predicting the total burden of disease across a population and for comparing with other pollutants or unrelated diseases. Two such metrics exist and are commonly used: *Quality Adjusted Life Years* (QALYs) and *Disability Adjusted Life Years* (DALYs). These two measures are close to inverse [90], where one QALY is equivalent to 1 year of life in perfect health [91], and one DALY is equivalent to 1 year lost of life in perfect health [92]. Both work by assigning weights between 0 and 1 to conditions, where 0 is death and 1 is full health for QALYs, and 1 = death and 0 = full health for DALYs [90]. Different agencies show different preferences, however both metrics can be used to for similar purposes, and are useful in cost effective analysis, to test how cost effective different interventions might be [90].

One example study by Logue *et al.* [12] estimated overall DALY losses attributable to chronic indoor exposures to multiple nonbiological air pollutants. Here the authors used a linear intake-incidence-DALY (IND) approach for $PM_{2.5}$ to first

estimate the disease incidence rate and then the combined DALY losses for estimated indoor exposures. The disease incidence rates were estimated using concentration-response functions from epidemiological literature. For $PM_{2.5}$ the health endpoints included were total mortality, chronic bronchitis and non-fatal stroke. Other pollutants were also considered using either the IND method or an intake-DALY (ID) approach based on animal toxicity literature. Indoor exposures were estimated using the results of a previous review which estimated mid-range and upper-bound concentrations relevant to chronic exposures for 267 pollutants [93]. This review included studies which measured indoor pollutant concentration, primarily from the USA but also other industrialised countries. These indoor concentrations were adjusted by factor 0.7 to estimate indoor exposures, as the average American spends 70% of their time indoors [12]. Logue *et al.* [12] predicted that DALYs lost due to $PM_{2.5}$ contributed substantially to annual health impacts. The authors identified $PM_{2.5}$, acrolein, formaldehyde, radon and secondhand smoke (SHS) as the most harmful nonbiological air pollutants in residences. The risks associated with $PM_{2.5}$, acrolein and formaldehyde apply to the entire population, whereas the radon and SHS are only found in a fraction of dwellings.

The Logue *et al.* [12] study highlighted the potential effect of indoor $PM_{2.5}$ on health, however there are a few limitations to this study that are worth considering. Firstly, the authors only included $PM_{2.5}$ concentration-response functions for three health endpoints. At the time of writing in 2011, these were the health effects with the most established epidemiological evidence, and yet the authors state that their approach likely underestimated the total impact of $PM_{2.5}$ on heart disease. Additionally, more associated health effects of $PM_{2.5}$ have since been identified, as previously explored, therefore the estimations of this paper are likely to underestimate the total DALYs lost due to chronic $PM_{2.5}$ exposure. For example, the total effect of ambient PM in the UK was estimated to be 360700 DALYS lost, where 61% were attributed to ischaemic heart disease, 20% to cerebrovascular disease, 11% to trachea, bronchus and lung cancers, 8% to COPD, and $<$1% to

lower respiratory infections [1]. Secondly, Logue *et al.* [12] assumed a linear concentration-response relationship; this may not be true, although it does appear to be at low concentrations. Thirdly, whilst these estimates are based on measured indoor concentrations for $PM_{2.5}$ and other pollutants, the $PM_{2.5}$ concentration-response functions used to estimate disease incidence (from Pope *et al.* [55], Brook *et al.* [61], and Abbey *et al.* [94]) are all based on ambient $PM_{2.5}$. This assumption that $PM_{2.5}$ from all sources and across all environments has the same toxicity is consistent across the epidemiological literature. The basis and implications of this assumption are explored in Section 2.1.3.

This process can be taken one step further, by assigning an economic value to a human life [95]. Governments or health care systems use this to estimate the impact on public finances, and to estimate whether the cost of implementing an intervention would be reflected in resulting savings for the health care system.

2.1.2 Toxicology and Mechanisms

Epidemiological evidence may have identified associations between air pollution, especially ambient $PM_{2.5}$, and physical and mental outcomes. However, it is key to remember that correlation does not equate to causation. Investigations into $PM_{2.5}$ toxicology and mechanisms may explain the observed endpoints.

2.1.2.1 Physical Health

The first key mechanism explored in multiple in-vivo and in-vitro studies is inflammation. In an in-vitro study, Yan *et al.* [96] tested the effect of exposing cultured cells to sampled ambient $PM_{2.5}$. Soluble and insoluble particles were separated and then cell cultures were exposed to a range of concentrations, with a diesel exhaust sample was also included as a positive control. Cells exposed to $PM_{2.5}$ showed increased expression of inflammatory mediator IL-8 [96]. Two

animal models have tested for system inflammation [97, 98], vasoconstriction of arteries in the heart and lung [97], and changes to cardiac function [98] in rats. These studies collected ambient $PM_{2.5}$ samples on filters, in Sao Paulo, Brazil [97] and Shanghai, China [98]. In both studies, the collected particles were extracted from the filters and installed in the rats' tracheas. Rivero *et al.* [97] found environmental $PM_{2.5}$ promoted pulmonary and cardiac alterations, and significant vasoconstriction (the constriction of blood vessels) in rats. However, the study did not include a comprehensive chemical analysis of the sampled particles. Wang *et al.* [98], also studied the effect of ozone on rats, which did not trigger changes alone, but ozone and $PM_{2.5}$ together had a larger effect than $PM_{2.5}$ alone [98].

Donaldson *et al.* [99] and C. L. Tran [100] investigated the inflammatory response to low toxicity, low solubility particles using both in-vivo and in-vitro techniques. They were interested in the proximal alveolar region of the lung, where there is thought to be a high rate of particle deposition and low clearance levels. Rather than mass based concentrations, particle surface area was used to standardise concentrations across particle types. The results of the in-vivo study in rats and in-vitro study conducted on epithelial cells show good agreement, with a threshold for effect observed at $1\,cm^2/cm^2$ for both approaches.

Zwozdziak *et al.* [101] took an alternative approach, using a dosimetry model to predict where in the respiratory system $PM_{2.5}$ is deposited. Whilst this did not directly consider the means by which $PM_{2.5}$ might trigger responses, knowing where most particles are deposited could help to identify where to focus studies. The model was based on ambient $PM_{2.5}$ and included seasonal variation [101]. The largest predicted deposits were in the upper respiratory tract, where 37.5% of the total particle mass was predicted to deposit in winter and 48.3% in summer. In contrast, only 3.4% $PM_{2.5}$ mass was predicted to deposit in the trachea and bronchiolar regions, and 7.5% in the pulmonary alveoli, the deepest region considered. However, some of this is to be expected, as different processes affect

the way particles progress through the different regions of the respiratory system. Four key processes have previously been identified to stop particulates as they are inhaled: *interception*, *impaction*, *gravitational settling* and *Brownian motion*. Ott *et al.* [44] identified that larger $PM_{2.5}$ is mainly deposited in the nose and upper throat due to impaction, while smaller $PM_{2.5}$ is mainly deposited in the pulmonary region due to Brownian diffusion. Therefore, it is unsurprising that the largest total mass deposits were predicted in the upper regions where larger, heavier particles are likely to deposit. However, mass deposits do not give much resolution on how many particles progressed deeper, and it does not answer whether mass or particle counts are more important to health.

In relation to this, Churg & Brauer [102] used analytical electron microscopy to count, size and identify particles in autopsy upper lung tissue. Tissue samples were obtained for 10 never-smoking, long term residents of Vancouver. The authors identified that 96% particles were in the $PM_{2.5}$ size range, and included metals and silica. UFPs accounted for less than 5% of the total particle count, although the authors highlight that this observation does not mean that UFPs are not deposited in the lungs, suggesting that they do not accumulate in large numbers possibly due to rapid removal by macrophages or by dissolving in lung tissue.

Elsewhere, multiple mechanisms have been proposed and investigated. Peixoto *et al.* [103] examined potential mechanisms by which $PM_{2.5}$ exposure might trigger cell death. These mechanisms included inflammatory related cascade, oxidative stress, and DNA damage. Inflammation has been identified as a short term response to $PM_{2.5}$ exposure at the cellular level, a response which is associated with acute responses in asthma and emphysema [103], and long term some DNA damage may remain. However, the authors do emphasise that there are still major gaps in the current knowledge.

Xing *et al.* [58] also cite a range of studies that investigate the impact of $PM_{2.5}$. Alveolar macrophages, a type of cell in the respiratory system that form part of the

immune system, have been found to have decreased viability with $PM_{2.5}$ exposure, and other mechanisms mentioned in this review include DNA damage possibly caused by free radical peroxidation, inbalanced intracellular calcium homeostasis and inflammatory injury. These studies offer some evidence that some particles can cause some damage to the body when inhaled. Xing *et al.* [58] did make an important conclusion, similar to that of Peixoto *et al.* [103], that few studies have investigated the means by which $PM_{2.5}$ might cause disease as a whole, and so suggest an integrated approach for future research.

These studies identify some means by which inhaled $PM_{2.5}$ might trigger responses in the body to explain observed health effects. The exact details are less important from an engineering perspective. If, as Peixoto *et al.* [103] suggest, particle composition and not just size affects particle toxicity, then $PM_{2.5}$ composition and source are important. All the studies mentioned in this section focus on ambient $PM_{2.5}$, and it is not clear whether $PM_{2.5}$ from indoor sources would have the same composition or effects.

2.1.2.2 Mental Health

With regards mental health, little research has been conducted into toxicology or mechanisms, and possible mechanisms remain unclear [74]. In the review conducted by Hoisington *et al.* [74], some potential mechanisms were proposed; for example chronic low grade inflammation is thought to be a risk factor for stress-related psychiatric and trauma-related disorders. Studies discussed previously found inflammatory effects of $PM_{2.5}$ therefore this is a possible mechanism. Additionally, Hoisington *et al.* [74] cited animal studies which found air pollution to impair cognitive function and increase depressive behaviours in association with neuroinflammation in the hippocampus. These potential mechanisms warrant further investigation.

In contrast, Sass *et al.* [75] proposed mechanisms that would pertain specifically to outdoor air pollution, and which relate to human behavioural changes in reaction to poor outdoor air quality. For example, the authors suggested that in areas with high ambient pollutant concentrations, people would be likely to spend less time outdoors. This is then linked to reduced physical activity, vitamin D deficiency, reduced contact with green space and social isolation, which are themselves risk factors for psychological distress.

Some plausible mechanisms relating to inflammation have been proposed, but further research is needed for them to become established. Additionally, some behavioural mechanisms were proposed, but these would not apply to poor indoor air quality, which may even have the opposite effect. Further understanding of physiological changes related to the development of adverse mental health conditions could help to direct future research in this area, and it would be beneficial to consider both ambient and indoor $PM_{2.5}$ within future investigations.

2.1.3 Is all $PM_{2.5}$ the same?

Most existing literature considers $PM_{2.5}$ as something of a single, homogeneous pollutant. In 2010, the World Health Organisation (WHO) published guidelines for indoor air quality, in which it concluded that "there is no convincing evidence of a difference in the hazardous nature of particulate matter from indoor sources as compared to those from outdoor sources" [104]. Conversely, $PM_{2.5}$ is defined as airborne solid and liquid particles with a diameter less than or equal to 2.5 µm [105], so by its very definition, all $PM_{2.5}$ is not the same. Limited research has directly investigated the health effects of indoor source $PM_{2.5}$. However studies investigating ambient $PM_{2.5}$ have observed heterogeneity of effects between locations [3, 21, 73]. Pope & Dockery [11] noted a need for healthy scepticism as they observed inadequate consistency in the toxicological and epidemiological evidence. Additionally, air composition in indoor environments, where people

spend the majority of their time [7], is distinctly different from local ambient air [106]. Therefore it is worth considering how the physical and chemical composition vary between sources and environments, and possible implications on health. Multiple chemical formulae are introduced in this section, therefore a periodic table is included in Appendix A for ease.

2.1.3.1 $PM_{2.5}$ composition and associated health effects

Epidemioligical studies have often observed geographical and seasonal heterogeneity of effects of $PM_{2.5}$. Previous studies hypothesised that the observed seasonal and spatial variation in health response could be related to seasonal and geographic variation in $PM_{2.5}$ composition [107]. Such variation in composition has previously been observed across the USA [107]. Particle solubility, metal content, surface area, reactivity [11] and solubility [4] are all possible factors which may influence overall particle toxicity.

Burnett *et al.* [50] considered the effect of elemental and sulfate components of $PM_{2.5}$ on daily mortality in eight Canadian cities. Sulfate, iron, nickel and zinc had the greatest effects, although the authors also suggest there may be additional toxic components of $PM_{2.5}$ whose concentrations and effects also vary between locations. More recently, Levy *et al.* [89] conducted a meta-analysis which focused on the main components of ambient $PM_{2.5}$; namely elemental carbon, organic carbon matter, sulfates and nitrates. This meta-analysis suggested organic carbon matter, sulfates and nitrates may be more toxic than $PM_{2.5}$ per unit concentration.

The effects of carbon black, ultrafine carbon black and transistion metal salts were also investigated in-vivo and in-vitro by Wilson *et al.* [108]. In this study, the effects of cupric sulfate ($CuSO_4$), ferrous sulfate ($FeSO_4$), and ferric chloride ($FeCl_3$) both alone and in combination with ultrafine carbon black, were investigated in-vitro, first using a cell free environment to measure the increase of reactive oxygen species and later by exposing macrophage cell cultures, and in-vivo via installation into rats'

lungs. In the cell-free environment, the combined effect of ultrafine carbon black and metal salts enhanced the reactivity in a potentiative manner, with the greatest effect observed for $FeCl_3$. In the macrophage cell cultures, the additional effect of metal salts diminished, thought to be due to them being effectively removed by the cells. A potentiative effect was also observed in rats for ultrafine carbon black with $FeCl_3$, thought to be caused by the potentiative effect of metals on epithelial cells whilst carbon black particles affected both epithelial cells and macrophages. The authors also observed that ultrafine carbon black had a significantly greater effect than carbon black across all tests, which suggests particle size impacts toxicity.

Other studies have proposed size is important. Smaller particles are more likely to deposit in smaller airways deeper in the lungs, which also affects particle clearance [4], and there are some observed effects of UFPs in asthmatic adults when there was no consistent effect for $PM_{2.5}$. In an in-vivo study conducted on rats, Oberdörster *et al.* [109] found a greater inflammatory response to ultrafine particles than larger particles of the same low solubility particle titanium oxide (TiO_2). Similarily, two studies investigated the effect of other low toxicity low solubility particles in-vivo [99, 100] and in-vitro using epithelial vells [99]. Here, surface area concentration was found to be a better indication of inflammatory response than mass concentration. Therefore the effect of smaller particles, especially UFPs, may be linked to their larger surface area per unit mass [99, 100, 109], and because their smaller size also allows them to enter the interstitium, or the space around the alveoli, more easily [109]

There is also some suggestion that chemical species might adsorb onto the surface of particles, effecting the particle toxicity. For example, polycyclic aromatic hydrocarbons (PAHs) adsorbed onto particles may disrupt the structure of DNA and stimulate cell death by activating tumour suppressor protein p53 [103]. Peixoto *et al.* [103] also identified PAHs and transition metals as being of particular interest, and that particle composition and resulting toxicity relate to particle source.

Dejmek [110] also considered the effect of PAHs, specifically the carcinogenic fraction (c-PAHs). They observed some effect of particulate exposure in early pregnancy on fetal growth, and hypothesised that the impact of airborne particles may depend on the proportion of c-PAHs adsorbed on the particle surfaces. PAHs are themselves a pollutant of interest, as some are potent carcinogens [104]. The health effects caused by PAHs, whilst not necessarily attributable to $PM_{2.5}$, are related to particle exposure, as PAHs in the air frequently attach to particles [104] and the WHO state that primary exposure to PAHs occurs via inhalation of particles [104]. This also highlights that, due to the complex nature of $PM_{2.5}$, it is important to consider double counting when measured alongside other airborne pollutants.

Finally, Baxter *et al.* [111] proposed an alternative explanation for the observed heterogeneity in epidemiological studies of $PM_{2.5}$, by exploring the factors relating to the process of pollutant transport from outside. Some predictors of variability of health effect estimates were median number of rooms in owner occupied residences, percentage of duplex homes, percentage of multi-family homes, percentage of homes using oil heating, percentage of homes using gas heating, and heating degree days. These housing characteristics may be surrogates for building permeability and particle infiltration, or may be associated with socio-economic status, itself a confounding factor. Additionally, the univariate models were not statistically significant, although the multivariate model explained 11-13% of the observed variability [111]. Building characteristics may explain some but not all of the heterogeneity observed in epidemiological literature, and so particle size and chemical composition may still be significant.

2.1.3.2 Indoor vs Outdoor $PM_{2.5}$

Some research has investigated indoor $PM_{2.5}$ composition, however there are fewer studies that specifically investigate the health effects of indoor $PM_{2.5}$, especially

within the residential environment. Schneider *et al.* [47] investigated the health effects of indoor $PM_{2.5}$ using a literature review. Of the 70 studies found, only 8 were included in the final literature review as the authors judged the others were 'non-informative' or 'inconclusive'. The included studies employed three different approaches: controlled human exposure studies ($n = 5$), cross-sectional office studies ($n = 2$) and a longitudinal study in the elderly population ($n = 1$). It is unlikely that these considered cooking source $PM_{2.5}$, as all but the longitudinal study focused on $PM_{2.5}$ in the office environment. The longitudinal study involved monitoring $PM_{2.5}$ concentrations in an elderly retirement centre and measuring heart rate variability. Whilst some heart rate variability was observed, the authors also stated that this may be explained by confounding factors. It should also be noted that this review dates from 2003, and does not discuss cooking as an indoor $PM_{2.5}$ source.

Elsewhere, some evidence suggests indoor particulates are chemically and physically different from outdoors [15]. Processes that can influence the indoor/outdoor particle ratio include: (i) building air exchange rates; (ii) particle penetration through infiltration pathways; (iii) indoor particle generation including combustion, cooking and household activities; (iv) particle generation or alteration due to secondary impacts; and (v) particle deposition or re-suspension [15]. Additionally, examples of specific components of $PM_{2.5}$ with indoor sources include small quantities of metals in aerosols from laser printers, organic and elemental carbon (OC and EC) from combustion processes, and PAH, aluminium, silicon, phosphor, sulfate and chlorine from burning beech wood in a household oven [15]. Incoming organics from outdoors can shift from gas to particulate phase through sorption to indoor $PM_{2.5}$, and for semi-volatile organic compounds (SVOCs) this is dependent on indoor temperature and availability of $PM_{2.5}$ [112]. These processes can make it difficult to separate indoor and outdoor contributions to the indoor $PM_{2.5}$ concentration [112].

Polidori *et al.* [113] measured higher concentrations of particulate organic matter in the $PM_{2.5}$ size fraction indoors compared to outdoors, $9.8\,\mu g/m^3$ compared to $4.9\,\mu g/m^3$. In measurments in 173 homes from Houston, TX, Los Angeles County, CA and Elizabeth, NJ, $71-78\%$ of particulate organic matter found indoors was emitted or formed indoors. Polidori *et al.* [113] concluded that this is substantial enough to alter the concentration, composition and behaviour of indoor $PM_{2.5}$. They also identified a shift in PAHs from gas-phase to particle-phase with outdoor-to-indoor transport [113]. Similarly, research from the Research Triangle Park panel study identified 4 key indoor and outdoor sources of PM by considering 19 species [114]. Species considered included EC, OC and the first component of OC, OC-1, along with various elements and transition metals. The identified indoor sources included, environmental tobacco smoke (ETS), personal care, a copper-factor mixed with indoor soil, and cooking, which contributed a high abundance of OC and OC-1 and some calcium.

In indoor and outdoor measurements in houses in Antwerp, the most abundant elements in both indoor and outdoor air were found to be sulfur

Investigating the health effects of $PM_{2.5}$ was not a primary aim of this work, as it falls outside the scope of built environment research. The existing literature shows epidemiological evidence of health effects of ambient $PM_{2.5}$, but further medical research is needed to consider the toxicology of $PM_{2.5}$ from indoor sources including emissions from cooking.(S), silicon (Si), and calcium (Ca), followed by iron (Fe), potassium (K), aluminium (Al) and chlorine (Cl) [115]. Unlike the previous study, here the authors attributed most indoor occurring elements to infiltration from outdoors, with the exception of K which was associated with smoking indoors. The location of the sampling device in the living room may have had an effect on these measurements.

2.1.4 Composition of Cooking Source $PM_{2.5}$

The first consideration is particle size. Multiple studies mentioned previously identified that smaller particles may be more toxic than larger particles [4, 108, 109] with the additional thought that this may be attributable to their increased surface area per unit mass. In a review of pollutant emissions from cooking, Abdullahi *et al.* [34] identified that particle emissions are dominated by the ultrafine size fraction. The same conclusion was drawn by [116] whose review also identified cooking particle emissions were mainly UFPs. Here the authors also identified that food type might influence the particle emissions, observing higher concentrations and larger size particles emitted from cooking fatty foods [116].

Other experimental studies have observed predominantly UFP size fraction particle emissions from cooking. Examples include particle emissions from gas cooking in a typical Chinese food stall, where up to 80% of the total particle count was in the ultra-fine range [117], measurements of particle emissions from Indian, Chinese and Italian cooking styles where UFP concentrations in the kitchen increased by a factor of up to 550 [118], and the initial findings from the HOMEChem test house where particles emitted during cooking with the gas stove, oven or electric hot plate were dominated by those with diameter $< 20\,\text{nm}$ [106]. Wang *et al.* [119] found that accumulation mode particles, that is particles with diameter $0.1 - 2.5\,\mu\text{m}$, emitted from cooking contributed more to $PM_{2.5}$ mass than UFPs, though particle counts were higher for UFPs. This may highlight a potential problem with current measurement standards where mass concentration is the standard metric to record $PM_{2.5}$ concentrations.

A second consideration is the chemical composition of particles, with particular emphasis on metal content. In their measurements in the Chinese food stall, See & Balasubramanian [117] measured metal concentrations of $1.5\,\mu\text{g/m}^3$ outside of cooking compared with $15.6\,\mu\text{g/m}^3$ during cooking, a factor 11 increase. The most abundant metals were copper (Cu), Fe and Zinc (Zn) [117], of which Fe and Zn

were previously identified as potentially toxic components of $PM_{2.5}$ [50]. Torkmahalleh *et al.* [120] also identified Fe as one of the most abundant heavy metals emitted when grilling ground beef, along with titanium, strontium, barium and lithium (Ti, Sr, Ba and Li). However, of the 31 elements considered, the highest concentrations were observed for sodium (Na), and the type of pan appeared to influence the elemental composition [120]. In a controlled study of emissions from heating corn oil, the five most abundant trace elements were Fe, Ti, Sr, Li and Ba, and Fe, Ti, Zn, Ba and Cr (chromium) when salt was added [121]. Another study found increased emissions of lead, molybdenum (Mo), chromium and zinc from heating corn oil mixed with salt compared to pure corn oil, and higher emissions of Cu, Rb (rubidium) and Ce (cerium) from heating the pure corn oil [122]. The most abundant elements observed were Ba during tests grilling meat, and Ti during heating oil with salt [122]. Within the HOMEChem project, concentrations of inorganic compounds nitrate (NO_3), chloride and ammonium (NH_4) increased during cooking, but sulfate (SO_4) concentrations, another inorganic component of $PM_{2.5}$, followed the ambient trend [106]. These inorganic components still constituted $< 3\%$ of the total particulate mass [106]. Nevertheless, NO_3 and SO_4 are both potentially toxic components of ambient $PM_{2.5}$ so it is interesting that cooking may be an indoor source of the former, but not the latter. The existing evidence indicates that the chemical composition of $PM_{2.5}$ from cooking sources is variable, which relates to the complexity of cooking, however there is some evidence of Fe, Zn and NO_3 emissions from some cooking which have previously been identified as potentially harmful.

Levy *et al.* [89] previously found that organic carbon matter may be more toxic than $PM_{2.5}$ per unit concentration. Multiple studies have measured emissions of particulate organic matter and OC from cooking sources. In their review, Abdullahi *et al.* [34] reported that cooking fumes contain OC and EC in the $PM_{2.5}$ size fraction, and that cooking techniques using oil release higher concentrations of OC and EC than those using water. OC was observed to constitute 50.0% of the PM emitted

during grilling ground beef (beef mince) [120]. A similar contribution was observed for heating corn oil, 60.8% OC, and corn oil mixed with salt, 56.3% OC [121]. There is some suggestion in the literature that particulate organic matter emitted during cooking is predominantly composed of fatty acids linked to cooking oil [34, 106, 119]. In their review, Wang *et al.* [119] cited one study in which $> 50\%$ of the $PM_{2.5}$ mass was classified as organic compounds. Finally, Farmer *et al.* [106] found that size distribution, concentration and bulk mass spectral organic aerosol all varied with meal type and preparation method, but was predominantly organic aerosol. The dominant ions were consistent with fatty acids found in cooking oils [106], once more indicating the importance of cooking method and food types.

Finally, there is some previous evidence that PAHs might sorb onto indoor particles, which could then form an exposure pathway for PAHs, themselves a potentially harmful pollutant class [103, 104, 110]. Abdullahi *et al.* [34] report PAHs contribute to cooking source $PM_{2.5}$, especially for Chinese cooking style. Buonanno *et al.* [123] measured a larger fraction of volatile compounds when cooking vegetable foods than fat rich foods. In addition they proposed an approach to investigate the *chemical* and *physical* health effects of particles separately, where the physical effects are related to surface area of non-volatile matter, and the chemical effects from volatile mass size distribution [123].

2.1.4.1 Health Effects of Cooking

Elsewhere the literature includes investigations into possible associations between cooking behaviour and observed health effects, although they do not explicitly consider $PM_{2.5}$.

Cooking fumes are an important risk factor for lung cancer [119], with suggestions that inhaling carcinogens whilst cooking meat may increase lung cancer risk among smokers [119]. Exposure to cooking fumes is associated with increased risk of lung cancer in both smokers and non smokers [28]. Much of this research has been

conducted in Chinese and Taiwanese women, where increased risks of developing lung cancer were observed in non-smoking populations [28, 29, 124]. Chiu *et al.* [124] observed disproportionately high incidence of lung cancer in women in Hong Kong, and suggested domestic air pollution might be a risk factor. They also listed influencing changes as (i) decreased use of firewood and kerosene as cooking fuel; (ii) increased use of exhaust fans and cooker hoods in kitchens; and (iii) wider use of foreign domestic helpers in recent decades [124]. Ko *et al.* [29] observed increased risk of developing lung cancer associated with stir-frying amongst never-smoking Taiwanese women, the same association was found for frying but not for deep fat frying. And Wang *et al.* [125] found that both cooking and smoking were seemingly independent risk factors for lung cancer in Hong Kong females. Similar findings have been observed in a meta-analysis of lung cancer risk among cooking adults [28]. Exposure to cooking fumes was associated with increased risks for smokers and non-smokers, with more effect in women than men, although the authors observe that more studies focus on women [28].

Hecht *et al.* [126] measured significantly higher levels of various biomarkers in women who regularly engaged in home cooking. However, this study also identified that the health risks associated with cooking may not only be attributed to $PM_{2.5}$, as they measured recorded levels of benzene that where three times higher in participants who cooked [126]. Benzene is itself a known carcinogen [104, 126].

Brain activity was monitored along with UFP, PM and CO_2 concentrations whilst participants fried ground beef in sunflower oil using an electric stove in an unventilated space [127]. No statistically significant changes in brain activity were observed before and immediately after exposure, however some differences were observed 30 minutes after exposure. The authors hypothesised that these changes relate to UFPs translocating to the frontal and temporal lobes of the brain [127], however further research is needed to ratify these findings and investigate the

TABLE 2.5: Existing $PM_{2.5}$ guidelines and standards

Body	Year	Guideline ($\mu g/m^3$)	Statistic	Location	Details	Reference
WHO	2005	10	Annual mean	Ambient		[128]
WHO	2005	25	24-hour mean	Ambient		[128]
US EPA	2015	12	Annual mean[a]	Ambient	Primary	[129]
US EPA	2015	15	Annual mean[a]	Ambient	Secondary	[129]
US EPA	2015	35	24-hour mean[b]	Ambient	Primary & Secondary	[129]
EU & UK	2008	25	Annual mean[c]	Ambient		[6, 130]
EU & UK	2008	20	Annual mean[d]	Ambient		[6, 130]
WHO	2010	10	Annual mean	Indoor		[104]
WHO	2010	25	24-hour mean	Indoor		[104]
IWBI	2016	15	Unclear	Indoor		[131]
IWBI	2016	35	Unclear	Indoor	Commercial Kitchens	[131]

[a] averaged over 3 years; [b] 98th percentile, averaged over 3 years; [c] Limit value, by 2015 ; [d] Limit value, by 2020

potential consequences for health.

In summary, multiple components of cooking source $PM_{2.5}$ that may be harmful to health and health risks associated with exposure to cooking fumes have been identified in the literature. In particular, UFPs, OC, particulate organic matter, Fe, Zn and NO_3 may be important. The risks associated with exposure to cooking fumes may also be attributed to pollutants other than $PM_{2.5}$, however, mitigation methods to decrease exposure to cooking source $PM_{2.5}$ may also reduce exposure to these pollutants. It is unclear from current research whether cooking source $PM_{2.5}$ in the indoor environment has equal toxicity to ambient $PM_{2.5}$, however the findings present here indicate that it is probably harmful. Further research is needed to establish the magnitude of health risks associated with exposure to $PM_{2.5}$ in cooking fumes.

2.1.5 Guidelines and Standards

Multiple organisations issue guidelines or standards for $PM_{2.5}$ concentrations, see Table 2.5. These are typically targets for improving outdoor air quality, however, some also consider the indoor environment. Most of these guidelines and standards are based on average mass concentration over a set time period.

In 2005 the WHO issued air quality guidelines for ambient particulate matter, ozone, NO_2 and SO_2 [128]. These guidelines consider the global situation, and so

individual countries are recommended to develop their own standards based on local conditions [128]. The WHO recommend 24-hour mean ambient $PM_{2.5}$ concentrations should remain below $25\,\mu g/m^3$ and annual mean concentrations below $10\,\mu g/m^3$, see Table 2.5. The WHO recognise the absence of a clear effect threshold below which no adverse health effects would be anticipated, and that it is unlikely any $PM_{2.5}$ guideline or threshold will offer complete protection [128]. The annual guideline was set based on two major sets of studies, the American Cancer Society study [55] and the Harvard Six-Cities data [49, 55], where robust associations were found between long term $PM_{2.5}$ concentrations and mortality [128]. Neither study is recent (pre-2002) and therefore there may be reason to consider revising these guidelines. To help countries track progress, the WHO also set three interim target values for annual mean $PM_{2.5}$. These are $35\,\mu g/m^3$, $25\,\mu g/m^3$ and $15\,\mu g/m^3$, and were estimated to be associated with increased risk of long term mortality of 15%, 9% and 3% compared to the guideline $10\,\mu g/m^3$ [128]. The rationale for the 24-hour mean guideline is unclear. It appears to be based on the PM_{10} guideline of $50\,\mu g/m^3$ and an assumed $PM_{2.5}/PM_{10}$ ratio of 0.5 [128]. Again, interim targets were set at $75\,\mu g/m^3$, $50\,\mu g/m^3$ and $37.5\,\mu g/m^3$, with associated increased risk of short term mortality of about 5%, 2.5% and 1.2% respectively [128]. Both the short term and long term WHO guidelines are based on mortality rates and do not also consider associated morbidities.

Different regional and national organisations set their own standards. There are key differences between the WHO guidelines and local standards. The WHO set guidelines based purely on the health based evidence, whereas local standards take into account local constraints, capabilities and public health priorities to set the lowest achievable target concentrations [128].

One such example standard is the National Ambient Air Quality Standards (NAAQS) set by the United States Environmental Protection Agency (US EPA). These are not directly applicable to the UK, the primary focus of this work, but are

considered here as the US EPA periodically review and revise these standards based on the latest evidence [129]. Of the standards set out in Table 2.5, only the values indicated as primary are directed at limiting effect on human health, whilst the secondary standard is to provide public welfare protection for example protection against decreased visibility [129].The current NAAQS 24-hour and annual mean concentrations are higher than the WHO guidelines, see Table 2.5.

In 2002, Green *et al.* [132] challenged the NAAQS over three key assumptions that the authors deemed to be problematic. These were that: (i) all forms of ambient $PM_{2.5}$ have identical toxic potencies; (ii) average mass based concentrations over 24-hours or a year are the best measures for public health; and (iii) decreasing these concentrations will decrease mortality rates at a reliably constant rate. Section 2.1.3 explored some evidence to suggest that all ambient $PM_{2.5}$ is not equally toxic, in agreement with the first point from Green *et al.*, who also considered toxicological evidence of the effect of $PM_{2.5}$ composition. With regards the second question about the most appropriate measures, the evidence discussed in Section 2.1.3 suggests that surface area may be a better predictor than mass, or particle count as a better indicator of surface area. However, the reference method for measuring $PM_{2.5}$ concentrations was and remains mass based [133], and the majority of epidemiological evidence continues to report mass concentrations. Measurement methods are discussed in more detail in Section 2.2. In reference to the final point made by Green *et al.* [132], the NAAQS as they existed in 2002 set a limit of $15\,\mu g/m^3$ annual mean and $65\,\mu g/m^3$ daily mean. These have since been lowered to $12\,\mu g/m^3$ and $35\,\mu g/m^3$. However, in two 2017 papers, health effects were observed at concentrations below the current daily [53] and annual [88] targets, and there is no evidence of an effect threshold [66]. Green *et al.* [132] did raise interesting questions, questioning the possibility of socioeconomic status as a confounding factor in epidemiological evidence, and suggesting that reduction attempts should aim to target harmful components of $PM_{2.5}$.

In the UK, ambient $PM_{2.5}$ targets are set as part of the Air Quality Standards Regulations 2010 (AQSR) [6], themselves set inline with European Directive 2008/50/EC [130]. Both sets of standards set absolute limit values for 2015 and 2020, see Table 2.5. However, these values are only for annual mean concentrations and at $25\,\mu g/m^3$ and $20\,\mu g/m^3$, which are higher than the values given by the NAAQS and the WHO. However, the AQSR and EU limit values are also the absolute upper limits, with lower targets set based on the 2010 Average Exposure Indicator (AEI) values with a lowest target of $8.5\,\mu g/m^3$ [6, 130]. These targets are more complex, partially because they are politically driven, considering the health risks in combination with the feasibility of achieving the target concentrations.

With regards the indoor environment, a 2010 WHO report published guidelines for indoor concentrations of select pollutants [104]. The panel concluded there was no "convincing evidence" that indoor $PM_{2.5}$ and outdoor $PM_{2.5}$ differed in toxicity, and in additional indoor $PM_{2.5}$ concentrations are often higher than outdoor in the presence of indoor source [104]. Therefore they recommended the 2005 guidelines as applicable to indoor.

Finally, the International Well Building Institute (IWBI) set air quality criteria as part of the Well Building Standard [131]. This is not a compulsory standard, instead it is commercially driven and aims to encourage holistic approaches which put occupant well-being at the centre of building design. The $PM_{2.5}$ concentration targets are set out to be $15\,\mu g/m^3$, or $35\,\mu g/m^3$ in commercial kitchens. However, the averaging time and measurement methods are unclear, with suggestion that compliance is tested by an assessor conducted spot test. The IWBI cites the NAAQS in reference to their $PM_{2.5}$ criteria, where $15\,\mu g/m^3$ matches with the NAAQS secondary annual mean $PM_{2.5}$ target, and $35\,\mu g/m^3$ corresponds to the 24-hour mean target. The IWBI separately rewards air quality monitoring, including $PM_{2.5}$ at resolution $10\,\mu g/m^3$ or finer, measured by least one monitor per floor in a regularly occupied or communal space, with at least one measurement per hour

[131].

In summary, existing guidelines and standards set different annual and 24-hour mean $PM_{2.5}$ concentration values. Of those considered here, the values set out by the WHO are considered the most suitable to use throughout the body of this work for 3 reasons. Firstly, these are the only truly health based guidelines, the other standards are also commercially and politically driven. Secondly, the WHO deem these guidelines suitable for ambient and indoor air. And finally, with no evidence of a lower effect threshold and growing evidence of increased effect per $\mu g/m^3$ at reduced concentrations, it could be prudent to select the lowest guideline values, which are those set out by the WHO.

2.1.6 Summary

In summary, there is substantial and growing epidemiological evidence of associations between multiple adverse health effects and ambient $PM_{2.5}$ concentrations. Both acute and chronic effects have been observed in the literature primarily for physical health endpoints such as mortality, pulmonary and respiratory, and cardiovascular effects. Toxicological research has identified possible mechanisms for some observed health effects, although more research is needed in this field. There is also limited evidence of possible associations between $PM_{2.5}$ and mental health effects including schizophrenia.

The importance of $PM_{2.5}$ composition has also been raised, as there is evidence of heterogeneity of effects within the epidemiological research. Organic carbon, zinc, iron and nitrates are components of cooking source $PM_{2.5}$ which may be particularly toxic. Additionally, particle size appears to be important. Smaller particles within the ultrafine range may have greater toxicity than larger particles due to their increased surface area.

Concentration-response curves indicate no lower effect threshold concentration for $PM_{2.5}$ mass exposure, and some studies suggest a non-linear relationship, with increased effect at lower concentrations. These findings are important when selecting appropriate guideline or target values. Current guidelines and standards all set mass concentration targets, despite the possible importance of particle count and surface area. Of the existing guidelines and standards available, the WHO annual and daily mean guidelines of $10\,\mu g/m^3$ and $25\,\mu g/m^3$ respectively are selected to use within this thesis.

Investigating the health effects of $PM_{2.5}$ was not a primary aim of this work, as it falls outside the scope of built environment research. The existing literature shows epidemiological evidence of health effects of ambient $PM_{2.5}$, but further medical research is needed to consider the toxicology of $PM_{2.5}$ from indoor sources including emissions from cooking.

2.2 Measuring Domestic $PM_{2.5}$ Concentrations

The most direct method to investigate indoor $PM_{2.5}$ concentrations and resulting exposures is via measurement. This process can also help to identify potential indoor sources. However, the processes and methods of $PM_{2.5}$ measurement are diverse. This section will first examine the existing measurement methods, and then explore published indoor $PM_{2.5}$ concentrations to work towards investigating concentrations within UK housing.

2.2.1 $PM_{2.5}$ Measurement Methods

A wide variety of devices are used to measure $PM_{2.5}$ concentrations. These devices use different methods to either directly measure or indirectly infer particle mass concentrations or particle counts. Gravimetric sampling and light-scattering techniques are amongst the most frequently used measurement methods.

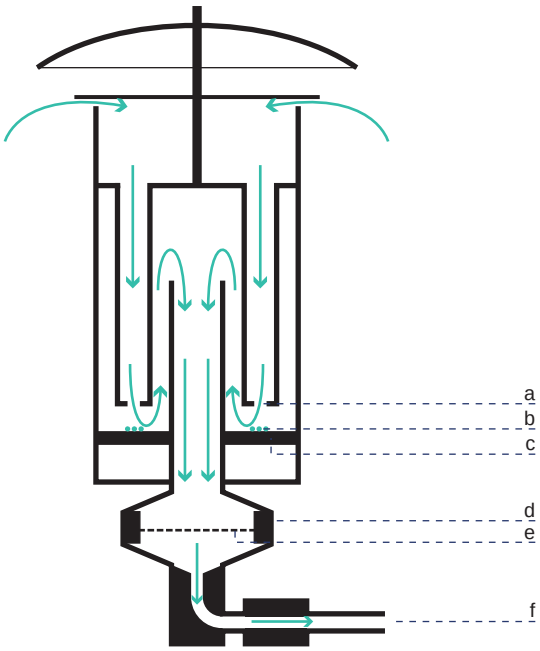


FIGURE 2.1: Diagram of an impactor
(a) sharp angle of streamline (b) larger impacted particles (c) greased impactor plate (d) filter housing (e) filter to collect size fraction of interest (f) connection to pump

2.2.1.1 Gravimetric Sampling

Gravimetric sampling forms the basis of the reference method for measuring ambient $PM_{2.5}$ concentrations [133], and may be the simplest and most established measurement method. Figure 2.1 shows a diagram of an example gravimetric sampling set up, based on a TECORA fine air inlet. The key components are a size selective inlet, filter and a pump to draw the air through the system.

The first stage separates out the required size fraction using a size selective inlet, typically a cyclone or impactor [134]. Both cyclones and impactors separate out the size fraction of interest based on the particle behaviour in the air stream, and therefore depend on the *aerodynamic* diameter [134]. The aerodynamic diameter is defined as the diameter of a spherical particle of unit density with the same aerodynamic properties as the real particle [134]. Particles are separated at the *cut*

point, the diameter at which 50% of the particles are collected [44, 134]. In a cyclone, particles larger than the cut point collect on the walls of the monitor as they fail to follow the helical streamlines [44]. By contrast, in an impactor, the air follows sharply curved streamlines and larger particles unable to follow the streamline are intercepted and collect on an impactor plate [44, 134]. In both situations, the smaller particles of interest continue along the air stream. Figure 2.1 shows a diagrammatic example of an impactor, indicating the streamlines and impactor plate.

The air stream is then drawn through a pre-weighed filter, which collects the remaining particles [44, 134–136]. The filter is removed and post-weighed after a fixed time period of continuous operation [44, 133–136]. The average concentration, $\overline{C}$ ($\mu g/m^3$), is then defined using the change in weight, or the total mass collected, Δm (μg), as per Equation 2.1, where $\bar{q}$ is the average pump flow rate (m^3/s), and T is the total operational time (s). The filter is pre- and post-weighed under specific temperature and relative humidity (RH) conditions [133, 134] to minimise the effect of room conditions on the measured mass.

$$\overline{C} = \frac{\Delta m}{\bar{q} T} \tag{2.1}$$

Key advantages include that this method provides a direct mass measurement and the particles collected on the filter can undergo subsequent chemical analysis [134]. However, this method cannot be used to measure temporal fluctuations in $PM_{2.5}$ concentration as it provides time-averaged data, and it is labour intensive [134]. Additionally, the process of conditioning the filters before post-weighing may cause unintended changes in mass due to adsorption or desorption of volatile or semi-volatile compounds [134, 136], some of which may also be harmful to health [136].

A variation of the gravimetric sampling method employs cascade impactors to measure particle size distribution. Here, rather than a single impactor plate, the air

stream flows through a series of stages with progressively smaller cut points [135]. Particles are retained on collection plates at each stage, and the smallest particles collect on an after filter [135].

2.2.1.2 Optical Measurement Devices

Optical measurement devices infer particle mass or count using light scattering principles [135, 136]. The main advantage of these devices is they provide real time concentration measurements, which can be a useful tool for source identification [137]. Two main categories of optical device are photometers and optical particle counters (OPCs).

Photometers measure the total light scattering effect of all particles and convert this to a mass concentration based on calibration with a known aerosol [137]. Scattered light reflected off the particles in the air stream is detected by a photometer, as they pass through light from a laser [135]. One example of a photometer is the TSI SidePak™ AM510 (henceforth SidePak) which has been previously used to measure $PM_{2.5}$ concentrations from a variety of indoor sources [40, 138]. The SidePak can be fitted with an impactor with cut size 1 µm, 2.5 µm or 10 µm to measure specific size fractions [139].

Alternatively, OPCs measure the light scattering effect of individual particles in the air stream [135–137] to sort the particles into defined size bins [136, 137]. OPCs can directly report size aggregated particle number counts, which can also be converted to estimated mass concentrations based on knowledge or assumptions of particle shape and density [137]. Examples OPCs using these principles previously used in literature include the Grimm 1.108, 1.109 and Mini Laser Aerosol Spectrometer 11-R [137, 140–143]. In OPCs the particles are sorted according to their optical diameter, the diameter of a polystyrene latex (PSL) sphere which would create the same response as the measured particle, as PSL is the standard dust used to calibrate

OPCs [142]. Optical particle diameter is not necessarily equal to the aerodynamic diameter, and it is the latter which is important to health [142].

A limitation of light scattering devices is that this process is only sensitive to particles larger than about 300 nm in diameter [134]. Condensation particle counters (CPCs) are designed to measure particle counts for smaller particles [134, 135]. The particles are drawn in through a saturated chamber where they grow to detectable sizes as water condenses onto them [134, 135]. They are then counted as in an OPC. Before condensing, particles can be sorted by size by electrically charging the particles and separating them by mobility in electric field [134].

In recent years, many consumer grade indoor air quality monitors have been released into the market [137]. These are frequently photometers which report mass concentrations of $PM_{2.5}$ measured using light scattering principles [137]. Singer & Delp [137] tested the response of 7 such consumer grade monitors to particle generating residential activities conducted under laboratory conditions. The response of the consumer monitors varied between devices and sources, however, all seven were found to consistently underestimate mass for events where the majority of emitted mass was from particles with diameter $< 0.3\,\mu m$ [137]. This is consistent with the previous statement that optical light scattering is effective for particles with diameter $> 300\,nm$ [134].

2.2.1.3 Calibration for Optical Devices

A significant limitation to all optical devices is that they do not directly measure mass concentrations. The calibration methods used will affect the estimated mass concentrations, and therefore are potentially a significant source of error.

Optical devices are factory calibrated with a specific test dust, frequently A1 or Arizona test dust [40] or PSL [142]. However, the light scattering effect, and therefore the estimated mass concentrations, varies with particle optical

characteristics including refractive index [40, 138, 141–143], reflectivity [136], density [40, 136, 138, 142, 144], shape [136, 142–144], size distribution [40, 136–138], and composition [136, 137, 143]. In the absence of additional data, optical devices estimate mass concentrations assuming the measured particles have equal properties to the device calibration dust [138].

Previous studies have found OPCs and photometers to both under- and overestimate particle size and mass concentrations. The Grimm OPC 1.108 and 1.109 underestimated mass concentrations of ambient urban aerosol in one study [143], whereas in another study the 1.108 underestimated mass on low concentration days and overestimated on high concentration days [140]. In this latter study, high humidity was thought to be a cause [140]. The effect of relative humidity is considered later. Dacunto *et al.* [40] and Jiang *et al.* [138] compared mass measurements from the SidePak to a gravimetric reference for a variety of indoor and outdoor sources. Both studies found that the SidePak consistently overestimated mass concentrations [40, 138] and Jiang *et al.* [138] also observed some variability in the response to different factory calibrated SidePaks to the same aerosol. Wang *et al.* [136] suggested photometers calibrated with Arizona test dust, such as the SidePak, can be expected to overestimate mass for environmental tobacco smoke (ETS) which is less dense than Arizona test dust. Another $PM_{2.5}$ monitoring device, the personal DataRAM was observed to consistently overestimate mass concentrations, and the degree of overestimation also varied by season and sample type [145].

In OPCs the mass estimate is based on the particle count in each size fraction so it is also important that particles are correctly sized. The process of mass estimation using OPCs requires 4 steps, as outlined by Binnig *et al.* [144], namely: (i) the measured optical diameter, d_{opt}, is converted into an equivalent aerodynamic size, d_{AE}; (ii) for each class size, the particle number is converted into a particle mass by converting d_{opt} into a volume equivalent size d_{VE}; (iii) for each class size, the mass

concentration is weighted using the nominal $PM_{2.5}$ transmission curve, a function of the d_{AE}; and (iv) the total $PM_{2.5}$ mass is calculated as the sum of the masses from all size fractions. As only d_{opt} is directly obtained by the OPC, conversion curves should be obtained for each material [144], however this is complicated, especially for unknown aerosols. Wang *et al.* [136] suggested that OPCs calibrated with PSL are likely to underestimate geometric size as PSL has a higher refractive index than most ambient aerosol; this may also cause mass to be underestimated. The properties of the measured aerosol will affect the accuracy of the reported size distributions. The OPC reported optical diameter for liquid and spherical particles will likely be smaller that the volume equivalent diameter, due to the refractive index, and larger for solid and irregular shaped particles possibly due to enhanced light scattering [142]. In a comparison with an aerodynamic particle sizer, the Grimm OPC 1.108 and 1.109 both recorded similar number mode diameters for mono-disperse PSL [141]. As PSL is the calibration dust, the OPCs would be expected to perform well. All three devices were also tested using polydisperse Arizona test dust, where the OPCs reported lower particle counts for the $0.7 - 2\,\mu$msize fractions [141].

Regards relative humidity (RH), monodisperse ambient aerosols often split into nonhygroscopic and hygroscopic portions at high RH, which can affect particle size [146]. Larger particles ($0.4 - 0.5\,\mu$m) grew more than smaller particles ($0.05 - 0.02\,\mu$m) at high RH [146]. Water can make up about half the atmospheric aerosol mass at 70% RH [146]. Sioutas *et al.* [147] found good agreement in the mass concentrations from a DataRAM photometer and gravimetric sampling at RH below 50%. However, at higher RH the DataRAM overestimated mass at an exponential rate. The low cost Alphasense OPC-N2 (henceforth OPC-N2) was compared to two reference OPCs and a TEOM device (see Section 2.2.1.4). At high RH ($>$ 90%) the OPC-N2 reported higher mass concentrations that the reference OPCs. The authors suggested that the design of the reference OPCs might cause intake air to be warmed, unintentionally drying out the aerosol, which does not

occur in the OPC-N2 [147]. They recommended low cost sensors should use calibration factors to adjust to the dry particle weight if they do not actively dry the inlet stream.

The simplest calibration method makes use of a calibration factor (CF) which is recommended for photometers [40, 138], although there is precedent of use with OPCs [143]. A gravimetric sampling device is collocated with the optical device for the duration of the monitoring period [40, 138, 139]. The average mass concentration from both the optical device, $\overline{C_{opt}}$, and the reference device $\overline{C_{ref}}$ are used to estimate a custom CF either by finding the ratio as in Equation 2.2 [40, 139] or via linear regression [138]. CFs vary with the physical and optical properties of the measured aerosol, and as $PM_{2.5}$ composition varies with source (see Section 2.1.3), no single CF will provide accurate mass concentrations for particles from all sources [137].

$$(CF) = \frac{\overline{C_{ref}}}{\overline{C_{opt}}} \tag{2.2}$$

Two previous studies determined SidePak CFs for $PM_{2.5}$ for a variety of sources [40, 138]. A summary of the results is included in Table 2.6. The CFs vary both between and within sources, although they all measure < 1.0. Notably, the urban outdoor aerosol CFs, based on a series of measurements over a 2 month period, ranged between 0.66 and 0.93, indicating there is no single value suitable for ambient air. Initial documentation for the newer SidePak AM520 also suggested CF $= 0.38$ is appropriate for ambient air [148], which is considerably lower than those measured by Jiang *et al.* [138]. CFs were generally higher for cooking sources ($0.41 - 0.79$) than combustion sources ($0.29 - 0.77$). For 6 of the 8 tests with two sources, the CFs were close to the weighted mean of the individual source CFs [40]. Additionally, Dacunto *et al.* [40] attributed the difference between their CF for burned toast and the one from Jiang *et al.* [138] to a difference in the degree of charring on the bread.

TABLE 2.6: SidePak $PM_{2.5}$ CFs reported in literature

Aerosol Source	n^a	Mean	SD	Uncertainty	Reference
Urban outdoor aerosol	2	0.83			[138]
Urban outdoor aerosol	2	0.66			[138]
Urban outdoor aerosol	2	0.93			[138]
Urban outdoor aerosol	2	0.85			[138]
Urban outdoor aerosol	2	0.66			[138]
Urban outdoor aerosol	2	0.82			[138]
SHS	14	0.29	0.02		[138]
SHS	16	0.28	0.02		[138]
Cigarette smoke	5	0.29	0.03		[138]
Cigarette smoke	11	0.32		0.01	[40]
Double strength incense	5	0.36	0.04		[138]
Double strength incense	5	0.37	0.04		[138]
Double strength incense	5	0.35	0.01		[138]
Incense, stick, 3 types	9	0.35		0.01	[40]
Incense, cone, 3 types	3	0.59		0.02	[40]
Wood chips	5	0.77	0.07		[138]
Fireplace, wood	5	0.44		0.01	[40]
Fireplace, synthetic log	4	0.47		0.003	[40]
Candles	1	0.37		0.02	[40]
Candles	1	0.47		0.04	[40]
Toasting bread	5	0.79	0.07		[138]
Burned toast	4	0.47		0.02	[40]
Fried bacon	7	0.41		0.02	[40]
Fried chicken	11	0.52		0.02	[40]
Fried pork chops	5	0.65		0.02	[40]
Fried salmon	4	0.66		0.01	[40]
Fried hamburger/ground beef	11	0.70		0.02	[40]
Fried bacon & cigarette	4	0.37		0.01	[40]
Fried bacon & cigarette	4	0.35		0.01	[40]
Fried chicken & cigarette	4	0.38		0.01	[40]
Burned toast & cigarette	3	0.38		0.01	[40]
Stick incense T1 & cigarette	4	0.32		0.004	[40]
Cone incense & cigarette	1	0.32		0.01	[40]
Stick incense T3 & cigarette	1	0.29		0.01	[40]
Stick incense T3 & cigarette	1	0.31		0.01	[40]

[a] for [138] n is the number of SidePaks and for [40] n is the number of gravimetric samples

This further indicates the variability in $PM_{2.5}$ characteristics between sources, and highlight a need for custom calibration factors when using optical devices.

As an alternative approach, Dacunto *et al.* [149] also estimated custom calibration curves for a low cost Dylos air quality monitor by comparing measured $PM_{2.5}$ concentrations from a range of sources to those from a collocated gravimetrically calibrated SidePak. They compared the time-resolved concentrations from the well-mixed decay period after emissions had ceased using power law curves. They found that a linear CF, as above, was suitable for fresh emissions, but the CF changed as the aerosol aged.

2.2.1.4 Other Measurement Devices

Many $PM_{2.5}$ measurement methods exist other than gravimetric sampling and optical devices, a summary of which is given by Amaral *et al.* [135]. A select few are of those which are used more frequently are included here.

The first is the tapered element oscillating microbalance (TEOM). This measurement method has the advantage that it can measure continuous mass concentrations [135], and was used by Singer & Delp [137]. Particles in the air stream are drawn in through a size selective inlet and collect on a small filter [134, 135]. This filter is positioned on the end of a tapered quartz or glass wand, which is free to oscillate. The TEOM measures the change in the oscillating frequency of the wand and converts this to a mass concentration [134]. This measurement method is less frequently used, possibly due to the higher cost, but is established in urban monitoring [135].

A second approach is time of flight instruments, such as an aerodynamic particle sizer (APS) [141]. This provides equivalent measurements to OPCs, measuring size resolved particle number concentrations [141]. The aerosol is accelerated through a nozzle, causing the particles to lag behind the air stream due to their intertia. The APS measures the time taken for particles to traverse two laser beams, which it uses to size the particles ad larger particles have a longer time-of-flight [141]. A significant advantage of this method is it measures aerodynamic particle diameters, however liquid aerosols are problematic as droplets deform with acceleration, impact on the acceleration nozzle and their size can be underestimated [141].

2.2.2 Measured $PM_{2.5}$ Concentrations

The most direct method to investigate $PM_{2.5}$ exposures is through measurement. The two standard methods are personal monitoring, where participants wear a portable device for the study duration, and fixed residential monitoring, where

monitors are placed in key micro-environments. With the advent of consumer grade IAQ monitors, a third approach is now also available, using big data from such monitors to evaluate IAQ [150].

2.2.2.1 Personal $PM_{2.5}$ Exposures

Personal monitoring directly measures personal exposures used to estimate dose. Table 2.7 summarises $PM_{2.5}$ concentrations from personal monitoring studies, although it may not be an exhaustive list. Papers report different summary statistics, and so the results are not always directly comparable. Included in Table 2.7 are n, the total number of measurements where indicated in the literature, otherwise the total number of participants; μ the arithmetic mean; SD, the arithmetic standard deviation; GM, the geometric mean; GSD, the geometric standard deviation; the minimum concentration; LQ, the lower quartile or 25th percentile; the median or 50th percentile; UQ, the upper quartile or 75th percentile; and the maximum reported concentration.

Overall the concentrations in Table 2.7 range between $0.1\,\mu g/m^3$, the minimum average concentration reported by Brunekreef *et al.* [151], and $796\,\mu g/m^3$, the maximum concentration reported by Buonanno *et al.* [152]. No single statistic is reported by all studies, making direct comparison between studies difficult. Mean concentrations range between $14.9\,\mu g/m^3$ [153] and $30.5\,\mu g/m^3$ [154], and frequently exceed the WHO 24 hour mean guideline. The corresponding standard deviations are often larger than the mean. $PM_{2.5}$ concentration is always positive, therefore this indicates non-normal distribution. Lai *et al.* [155] reported that from visual inspection, the concentration data appeared log-normally distributed, and therefore reported the geometric mean and standard deviation. The lognormality or near lognormality of pollutant concentrations is established in literature [156], so the geometric mean provides a better estimate of the central tendency of the data than the arithmetic mean. However, existing guidelines are based on arithmetic

means, and so the statistical methods used affect how results can be compared to guidelines. If the data represents a series of tests, particularly gravimetric, where the concentrations are first averaged over either 24 hours or 1 year, to match guidelines, the median and other percentiles can give an indication of overall compliance. In the case of time resolved data, results reported only as a GM and GSD are not directly comparable to existing guidelines.

TABLE 2.7: Measured personal $PM_{2.5}$ exposures ($\mu g/m^3$)

Geographic Location	Measurement Device	Averaging Time	n	μ	SD	GM	GSD	Min	LQ	Median	UQ	Max	Details	Reference & Publ. Year
London, UK	PEM Model 200[a]	12 hour	10	22									children, winter	[157] 2000
London, UK	PEM Model 200[a]	12 hour	10	17									children, spring	[157] 2000
London, UK	PEM Model 200[a]	12 hour	10	15									children, summer	[157] 2000
London, UK	PEM Model 200[a]	12 hour	10	18									children, seasonal average	[157] 2000
West Yorkshire, UK	PEM Model 200[a]	$<$ 24 hour	78			35.5	1.7	10.3				108.3	no smoking, daytime (work)	[158] 2005
West Yorkshire, UK	PEM Model 200[a]	$<$ 24 hour	80			23.6	1.9	6.3				94.0	no smoking, non-work	[158] 2005
Fort Williams, CO, US	pDR-1200[b]	$<$ 24 hour	287			8.0	2.3	1.0	4.6	7.3	13.1	78.3	time reported at home	[159] 2019
Fort Williams, CO, US	pDR-1200[b]	$<$ 24 hour	275			2.4	2.5	0.7	1.1	2.0	4.4	68.6	time reported at work	[159] 2019
Fort Williams, CO, US	pDR-1200[b]	$<$ 24 hour	287			6.8	2.2	0.7	4.3	6.9	10.8	95.0	time reported in transit	[159] 2019
Fort Williams, CO, US	pDR-1200[b]	24 hour	287			7.4	1.9	1.7	4.9	6.6	10.2	51.3	full 24 hours	[159] 2019
Amsterdam, NL	GK 2.05 cyclone[a]	24 hour	337					0.1	8.9	13.6	23.1	261.9	all	[151] 2005
Amsterdam, NL	GK 2.05 cyclone[a]	24 hour	260					0.1	8.2	11.7	16.9	190.4	no ETS	[151] 2005
Helsinki, FI	GK 2.05 cyclone[a]	24 hour	336					2.1	6.9	9.2	13.0	51.3	all	[151] 2005
Helsinki, FI	GK 2.05 cyclone[a]	24 hour	325					2.1	6.9	9.1	12.8	51.3	no ETS	[151] 2005
Battle Creek, MN, US	PEM Model 200[a]	24 hour	118	22.3	25.7	16.2	2.2	3.8				207	all seasons	[154] 2002
East St Paul, MN, US	PEM Model 200[a]	24 hour	107	30.5	38.7	20.6	2.3	2.5				298	all seasons	[154] 2002
Phillips, MN, US	PEM Model 200[a]	24 hour	107	26.5	24.3	20.9	2.0	2.2				211	all seasons	[154] 2002
Raleigh, NC, US	PEM[a]	24 hour	712	23.0		19.2		3.4				142.3		[160] 2003
Ceccano, IT	DustTrak 8534[b]	24 hour	15					13.9	34.6	40.8	54.0	796	15 women	[152] 2015
Oxford, UK	GK 2.05 cyclone[a]	48 hour	41	17.4		13.2	2.0						includes ETS and smoking	[155] 2004
West Yorkshire, UK	PEM Model 200[a]	48 hour	40			30.3	1.6	13.3				66.8	no smoking, time weighted average	[158] 2005
Prague, CZ	DustTrak 8520[b]	236 days	60473	14.9	52.5	8.1	2.4			8.0			1 participant	[153] 2010

[a] indicates gravimetric sampling methods; [b] indicates optical devices with gravimetric calibration

Amongst those studies that report GMs, these ranged between 2.4 $\mu g/m^3$ [159] for time reported at work, and 35.5 $\mu g/m^3$ [158], with corresponding GSDs of around 2.0. Similar range is also found in the median concentrations, from 2.0 $\mu g/m^3$ [159] to 40.8 $\mu g/m^3$ [152]. Between-study variation may indicate the difference between exposures in different environments, and international variation in time-activity patterns.

Most studies included in Table 2.7 follow a similar procedure. Participants typically wear a portable monitoring device throughout the study period, which can be gravitmetric [151, 154, 155, 157, 158, 160] or optical [153, 159]. One study employed a slightly modified method, where participants carried the optical DustTrak monitor to each micro-environment and then placed it within 3 m [152]. The measured concentrations may be less accurate representations of true exposure. Participants also frequently complete time-location-activity-diaries at regular intervals throughout the measurement period [151, 152, 155, 157–160], which researchers use to investigate the relative importance of different micro-environments on overall exposure. In the case of participating children, these diaries may be completed by the children [157] or by their parents [152].

Authors frequently report participants spent most time indoors, especially indoors at home. Buonanno *et al.* [152] reported on average spent $92 \pm 4\%$ time indoors, and $74 \pm 18\%$ time at home. Similar finding were reported by Lai *et al.* [155]: 89.5% time indoors, 69% in homes, 18% in workplace, and by Williams *et al.* [160]: $72 - 75\%$ inside at home, $3 - 5\%$ travelling by car or by bus. Williams *et al.* [160] also found daily mean personal $PM_{2.5}$ exposures were typically higher than corresponding residential or ambient measurements, with moderate correlations between personal and ambient concentrations. They suggested some contribution of a *personal cloud* effect [160]. Elsewhere, high personal $PM_{2.5}$ concentrations were recorded at home and eateries, with average concentrations 70% lower at work, 14% lower in transit, and 39% lower in other locations [159]. In contrast Wheeler *et al.* [157] who

measured personal daytime concentrations for children aged $9-11$ years in London, UK. $PM_{2.5}$ concentrations were highest in the classroom, then home, then personal, then outdoor.

An advantage of optical measurements is that the relative contribution of exposures in each environment can be estimated relatively easily. Buonanno *et al.* [152] estimated that indoor $PM_{2.5}$ contributed $88 \pm 14\%$ to overall exposure, emphasising the potential role of indoor exposure on health. Unique to this study, the concept of *exposure intensity*, see Equation 2.3, was proposed to compare the contribution of exposures in different locations. The highest exposure intensity, 6.9, was recorded for driving or travelling in a car with open windows whilst smoking, and the second highest, 1.9, was for cooking activity [152].

$$\text{exposure intensity} = \frac{\text{daily exposure contribution (\%)}}{\text{daily time contribution (\%)}} \tag{2.3}$$

Authors also investigate potential correlations. Mohammadyan & Ashmore [158] found stronger correlations between non-work personal exposures and residential indoor $PM_{2.5}$ than daytime personal exposures and indoor office $PM_{2.5}$ amongst 40 workers from the University of Bradford. Higher ambient temperatures were also associated with lower concentrations, thought to be linked to window opening behaviour [158]. In a study involving 32 non-smoking adults, significant associations were found between personal and indoor concentrations but not between personal and outdoor concentrations [154]. Additionally, personal and indoor concentrations were more variable than outdoor concentrations [154]. Similarly, Lai *et al.* [155] found good correlation between residential $PM_{2.5}$ concentrations and personal exposures, indicating the importance of exposure at home. Brunekreef *et al.* [151] also found more variability in personal exposures for participants of their study located in Amsterdam than those in Helsinki.

Environmental tobacco smoke (ETS) and cigarette smoking are known sources of

$PM_{2.5}$, which is further corroborated by multiple personal exposure measurements. Brunekreef *et al.* [151] reported lower personal concentrations for participants who did not report exposure to ETS, with personal concentrations similar to background. Lai *et al.* [155] calculated indoor/outdoor ratios which were higher among the smoking group than the non-smoking group. A second source considered is cooking. Buonanno *et al.* [152] identified cooking as having the second highest exposure intensity, and Brunekreef *et al.* [151] observed moderate associations between time spent cooking and personal $PM_{2.5}$ exposure in Amsterdam. Lai *et al.* [155] also reported that 87.2% participants spent some time cooking within the 48 hour measurement period, and those that did spent 1.20 ± 0.79 h cooking. There was a positive correlation between gas cooking and personal $PM_{2.5}$ [155].

The studies in Table 2.7 mostly measure short term personal exposures, for example in one study volunteers were monitored for seven consecutive days in each of four seasons [160], as personal monitoring is invasive and resource intensive. One exception is the study by Braniš & Kolomazníková [153] who conducted a continuous personal monitoring study with a single participant for 10 months. The student participant carried a TSI DustTrak photometer set to log at 5 minute intervals continuously from February to December of the same year, excepting April, resulting in 60473 data points over 236 days. The measured $PM_{2.5}$ mass concentrations were adjusted using custom gravimetric calibration curves developed from collocating the DustTrak with a nearby ambient monitor for 35 days [153], however the exact magnitude of peak concentrations should still be considered estimations. In addition, the participant completed a continuous time-location-activity diary to record her location in different micro-environments every 5 minutes throughout the investigation [153]. The overall mean concentration ($14.9\,\mu g/m^3$) was above the WHO annual mean guideline value, and the large standard deviation ($52.5\,\mu g/m^3$) also indicates very large spread and non-normal distribution. The highest average concentrations were recorded in restaurants and

bars ($\sim$300 µg/m^3) and rooms with wood- or coal-burning stoves ($>$ 100 µg/m^3). The lowest average concentrations were recorded in the natural environment, home B (in a smaller town outside Prague) and home A (in Prague). Despite low concentrations of 9.7 ± 10.0 µg/m^3, exposures in home A accounted for the largest contribution to total exposure at 29.8% [153]. This emphasises the influence home exposure to $PM_{2.5}$ could have on health. In contrast, the second highest contributor to overall exposure was exposure in restaurants and bars. Here, the opposite was true, as she occupied such spaces just 1.2% of the time, but the high average concentrations of 294.4 ± 278.8 µg/m^{3}resulted in these micro-environments contributing 24.3% to total exposure [153]. At the time of the study, smoking was permitted in bars and restaurants in Prague, and so ETS likely caused these high $PM_{2.5}$ concentrations.

2.2.2.2 Residential $PM_{2.5}$ Concentrations

Fixed monitoring is somewhat easier than personal monitoring, and therefore there are more studies in the literature. Personal monitoring studies have frequently identified that indoor $PM_{2.5}$ concentrations at home contribute significantly to personal exposure, where people spend most time [152, 155, 160], and so measuring such concentrations might offer a better estimation of exposure than centralised ambient monitoring.

Table 2.8 summarises $PM_{2.5}$ concentrations measured in housing as reported in literature. Again, this is not an exhaustive list, studies from the UK, Europe and other western countries with similar lifestyles were prioritised.

TABLE 2.8: Measured residential $PM_{2.5}$ concentrations ($\mu g/m^3$)

Location		Measurement	Averaging	n	μ	SD	GM	GSD	Min	LQ	Median	UQ	Max	Details	Reference
Geographic	Residential	Device	Time												& Publ. Year
Detroit, MI, US	in home	Cyclone inlet[a]	24 hour	36	17.9	9.1								winter, non-smoking	[161] 2002
Detroit, MI, US	in home	Cyclone inlet[a]	24 hour	31	32.0	23.2								spring, non-smoking	[161] 2002
Detroit, MI, US	in home	Cyclone inlet[a]	24 hour	24	21.7	14.6								summer, non-smoking	[161] 2002
Detroit, MI, US	in home	Cyclone inlet[a]	24 hour	39	24.1	19.1								autumn, non-smoking	[161] 2002
Oxford, UK	in home	GK2.05 cyclone[a]	48 hour	42	17.3		11.4	2.4						inc ETS & smoking	[155] 2004
Raleigh, NC, US	in home	Harvard Impactor[a]	24 hour	761	19.1		15.3		2.3				119.4		[145] 2006
Beijing, CN	in home	Laser Egg	1 year	4403	38.6	18.4					34.6			all year	[150] 2018
Beijing, CN	in home	Laser Egg	8 months	4403	34.9	15.8					32.7			non-heating season	[150] 2018
Beijing, CN	in home	Laser Egg	4 months	4403	46.1	21.2					40.0			heating season	[150] 2018
Birmingham, UK	living room	TEOM	2.5 day	1	7.9	5.2									[162] 2000
London, UK	living room	Harvard Impactor[a]	5 day	10	29									winter	[157] 2000
London, UK	living room	Harvard Impactor[a]	5 day	10	24									spring	[157] 2000
London, UK	living room	Harvard Impactor[a]	5 day	10	19									summer	[157] 2000
London, UK	living room	Harvard Impactor[a]	5 day	10	23									seasonal average	[157] 2000
Oxford, UK	living room	GK2.05 cyclone[a]	<48 hour	10	17		12	2.2	6				71	ETS, occupied hours	[163] 2000
Oxford, UK	living room	GK2.05 cyclone[a]	<48 hour	9	10		9.0	1.6	6.0				19	no smoking, occupied hours	[163] 2000
Battle Creek, MN, US	living room	PEM Model 200[a]	24 hour	108	10.6	6.6	9.0	1.8	2.3				36.9	all seasons	[154] 2002
East St Paul, MN, US	living room	PEM Model 200[a]	24 hour	97	17.4	20.3	12.2	2.2	1.3				130	all seasons	[154] 2002
Phillips, MN, US	living room	PEM Model 200[a]	24 hour	89	14.2	13.0	11.3	1.9	3.1				90.9	all seasons	[154] 2002
Amsterdam, NL	living room	Harvard Impactor[a]	24 hour	409					2.5	9.6	13.6	24.2	442.1	all	[151] 2005
Amsterdam, NL	living room	Harvard Impactor[a]	24 hour	353					2.5	8.9	12.3	18.5	225.7	no ETS	[151] 2005
Helsinki, FI	living room	Harvard Impactor[a]	24 hour	503					1.8	7.1	9.2	13.0	132.2	all	[151] 2005
Helsinki, FI	living room	Harvard Impactor[a]	24 hour	495					1.8	7.1	9.1	12.8	132.2	no ETS	[151] 2005
West Yorkshire, UK	living room	PEM Model 200[a]	<24 hour	80			19	2.1	3.2					no smoking, occupied hours	[158] 2005
NE Scotland, UK	living room	DustTrak[c]	8-14 hour	148						6	18	77		49% smoking households	[164] 2007
Colchester, UK	living room	Grimm 1.108 or 1.109[b]	24 hour	30	13	6			6				22	1 bed flats, separate kitchens	[165] 2013
Colchester, UK	living room	Grimm 1.108 or 1.109[b]	24 hour	20	18	7			12				26	1 bed flats, open plan kitchens	[165] 2013
Colchester, UK	living room	Grimm 1.108 or 1.109[b]	24 hour	30	11	2			8				14	2-3 bed house, central heating	[165] 2013
Colchester, UK	living room	Grimm 1.108 or 1.109[b]	24 hour	10	191	193			53				619	2-3 bed house, wood burner	[165] 2013
Colchester, UK	living room	Grimm 1.108 or 1.109[b]	24 hour	20	6	2			3				7	student halls, winter	[165] 2013
Colchester, UK	living room	Grimm 1.108 or 1.109[b]	24 hour	30	9	4			4				15	student halls, Fsummer	[165] 2013
Prague, CZ	living room	Harvard Impactor[a]	24 hour	13	49.6				12.0		30.6		203.2		[166] 2013
nr Dublin, IE	living/bed room	SidePak AM510	24 hour	7	10.01	2.69								pre-retrofit	[9] 2017
nr Dublin, IE	living/bed room	SidePak AM510	24 hour	7	17.55	2.60								post-retrofit	[9] 2017
nr Dublin, IE	living/bed room	SidePak AM510	24 hour	8	9.33	2.60								pre-retrofit	[9] 2017
nr Dublin, IE	living/bed room	SidePak AM510	24 hour	8	15.34	3.89								post-retrofit	[9] 2017
nr Denver, CO, US	living area	Dylos DC1700	3-5 days	9	11.2	10.4			2.3		7.3		32.6	tightly constructed homes	[167] 2018
nr Denver, CO, US	living area	Dylos DC1700	3-5 days	1	20.1									conventional home	[167] 2018
Antwerp, BE	living room	Dekati 3-stage Impactor[a]	24 hour	15	36.0				10.9		27.2		105	15 houses	[115] 2009
Antwerp, BE	living room	Harvard Impactor[a]	24 hour	4	41.1				12.1		41.0		69.7	4 houses	[115] 2009
Boston, MA, US	kitchen	Harvard Impactor[a]	12 hour	63	13.9	15.2			3		11.6		128.3	4 non-smoking houses	[168] 2000
Oxford, UK	kitchen	GK2.05 cyclone[a]	48 hour	10	18		13	2.1	5				77	inc ETS & smoking	[163] 2000
Oxford, UK	kitchen	GK2.05 cyclone[a]	48 hour	9	12		11	1.5	5				20	no smoking	[163] 2000
Colchester, UK	kitchen	Grimm 1.108 or 1.109[b]	24 hour	50	56	40			20				130	student halls, electric	[165] 2013
Colchester, UK	kitchen	Grimm 1.108 or 1.109[b]	24 hour	14	46	33			13				130	single bed flats, electric	[165] 2013
Colchester, UK	kitchen	Grimm 1.108 or 1.109[b]	24 hour	16	10	7			5				23	2-3 bed house, gas	[165] 2013

[a] indicates gravimetric sampling methods; [b] indicates optical devices with custom gravimetric calibration; [c] indicates optical devices with general gravimetric calibration

Again no single statistic is reported by all studies, see Table 2.8, however, the arithmetic mean is given by all but Brunekreef *et al.* [151], Mohammadyan & Ashmore [158], and Osman *et al.* [164]. There is also more variation in the averaging period, with more studies reporting concentrations averaged over a few days or more. Reported mean concentrations range between $6\,\mu g/m^3$ measured in student living rooms and $191\,\mu g/m^3$ for living rooms in 2-3 bed houses with a wood burner both in Colchester, UK [165]. Of the 19 results reporting arithmetic means with a 24 hour averaging period, the mean concentration was greater than the WHO guideline $25\,\mu g/m^3$ in 7 cases. A further four results for a 24 hour averaging period instead reported median average concentrations, all of which were below the WHO guideline. Overall, these studies suggest that domestic air quality may be acceptable in many cases, however the studies presented in Table 2.8, with mostly small sample sizes $n < 100$, are unlikely to be representative of the UK housing stock.

None of the seven UK based studies in Table 2.8 intentionally used representative sampling methods, so the concentrations measured are not representative of those found in all UK dwellings. Mean concentrations ranged between $7.9 \pm 5.2\,\mu g/m^3$ and $191 \pm 193\,\mu g/m^3$ ($1 \leq n \leq 48$), with 3 reported mean concentrations exceeding the WHO daily mean threshold. Therefore there is some evidence of high indoor $PM_{2.5}$ concentrations in UK dwellings, but it is inconclusive and warrants further investigation.

Only one study reported long term indoor $PM_{2.5}$ concentrations. Zuo *et al.* [150] used big data from concentrations measured by the consumer grade optical *Laser Egg* in Beijing. They report an overall annual mean concentration of $38.6 \pm 18.4\,\mu g/m^3$, with corresponding values of $46.1 \pm 21.2\,\mu g/m^3$during the 4 month heating season, and $34.9 \pm 15.8\,\mu g/m^3$otherwise. These concentrations are $3 - 4$ times the WHO annual mean guideline. Ambient $PM_{2.5}$ contributed about 42% to indoor $PM_{2.5}$ in the heating season and 52% otherwise [150]. The main

advantage of this method is the large quantity of $PM_{2.5}$ data available, however, the location of the Laser Egg in the dwelling and occupant behaviour are both unknown. Additionally, the authors suggest there may be some sampling bias, as people who choose to buy IAQ monitors might pay more attention to IAQ, and may be more likely to use air purifiers [150].

From Table 2.8, the most common in home monitoring location is the living room or main living area. There is evidence of high correlations between particle concentrations in different rooms throughout the house, with the living room thought to be representative of whole house concentrations [157]. This location is also likely to cause minimal intrusion to occupants [162]. Wigzell *et al.* [163] and Hill *et al.* [169] measured simultaneous time-resolved particle number concentrations (PNCs) in the kitchen, living room and bedroom of UK houses. Both identified concentration peaks in one room were reflected in another room, indicating pollutant transport within the dwelling. Wigzell *et al.* [163] also identified strong correlation between mass concentration in the kitchen and the living room using gravimetric sampling. However, pollutant transport between rooms was not always high, for example in one instance a cigarette smoked in the bedroom did not lead to large increases in PNC in the kitchen and living room [169]. Finally, Wigzell *et al.* [163] observed that the presence of an awake person in the house was associated with higher PNCs in the fine and coarse size fractions.

The relationship between indoor and outdoor $PM_{2.5}$ is variable, and is dependent on the presence of indoor sources. Outdoor particles made up about 50% of indoor concentrations in one study [145] and explained $20 - 30\%$ of the variance in indoor PM_{10} concentrations in another [170]. In residential indoor $PM_{2.5}$ concentrations in Detroit, Michigan, variance was attributed to the proximity to major roads and an industrial plant [161], both ambient $PM_{2.5}$ sources. Morawska *et al.* [15] reported indoor/outdoor (I/O) ratios for naturally ventilated buildings in the range $0.5 - 0.98$ with no indoor sources, and $1.14 - 3.91$ when there were indoor sources

present. Similarly, Lai *et al.* [155] reported a geometric mean 20% higher indoors than outdoors of $PM_{2.5}$ and its constituents. In tightly constructed Colorado houses, I/O ratios were $0.11 - 2.26$, and compared to 3.23 in a single conventional house [167]. Filtered supply air reduced the infiltration of outdoor $PM_{2.5}$ and homes with high I/O ratios were thought to have increased cooking events [167].

Two studies also considered the effect of airtight construction on indoor $PM_{2.5}$ concentrations. The first monitored $PM_{2.5}$ concentrations in 9 tightly constructed homes and a single conventional home near Denver, Colorado [167]. The $3 - 5$ day average concentration was higher in the conventional home than the tightly constructed homes and was less influenced by outdoor concentrations. Before and after the $3 - 5$ day monitoring period, investigators also conducted a controlled cooking test, during which $PM_{2.5}$ concentrations were higher in the tightly constructed homes, and took longer to decay back down to background levels than in the conventional home [167]. A second study in Ireland measured $PM_{2.5}$ concentrations in 15 dwellings before and after energy retrofits [9]. Post-retrofit concentrations were significantly higher in both groups of houses, though the mean concentrations remained below the WHO daily guideline [9]. Both of these studies include small sample sizes ($n = 10$ and $n = 15$) and relatively short monitoring periods ($3 - 5$ days and 24 hours), however, they suggest that energy improvements that increase building airtightness increases the importance of indoor $PM_{2.5}$ sources on IAQ.

Urso *et al.* [171] investigated the impact of key outdoor factors, house characteristics and indoor activities on indoor size fraction $PM_{2.5}$ concentrations using residential gravimetric sampling and time-activity diaries. Ambient $PM_{2.5}$ was a predictor for all size fractions of indoor $PM_{2.5}$, and ETS a predictor for for particles $< 0.5\,\mu m$. Other factors linked to increase in indoor $PM_{2.5}$ concentrations included RH, burning wood in open fireplaces, number of occupants and time spent cleaning. Time spent cooking was not significantly associated with increased

living room $PM_{2.5}$ concentrations, however, using kitchen exhaust ventilation during cooking did reduce concentrations of $PM_{0.5-1}$ [171], which is suggestive of some cooking influence.

Multiple sources identified smoking and ETS as significant contributors to indoor $PM_{2.5}$ concentrations [114, 115, 162, 164–166, 170, 172, 173]. Whilst this clearly identifies a risk of smoking indoors, many risks associated with smoking and passive smoking are culturally established. Reducing indoor ETS exposures does not require a built solution as people can be discouraged from smoke indoors. For these reasons, smoking is not the focus of this thesis.

Other indoor $PM_{2.5}$ sources identified in the literature include cooking [114, 145, 162, 165–167, 170, 172], cleaning [145, 165, 172, 173], personal care [114, 145, 166], indoor combustion including candles, incense and using a fireplace [145, 166, 172], and general indoor activity [114, 165, 166]. Within a two week monitoring study in a 2 bed apartment, 15 distinct $PM_{2.5}$ events were identified [166]. Ranked from most to least significant these were: smoking, smoking, hairspray, incense burning, cooking + incense, cooking, people present, cooking, cooking + oil lamp, frying + candle, cooking + frying, oil lamp, oil lamp, people present, and cooking + baking. Seven of these emission events involve cooking activity of some form. Jones *et al.* [162] identified peak concentrations of around $50 - 90\,\mu g/m^3$associated with cooking activity, and Ozkaynak *et al.* [170] estimated cooking activity contributed $12 - 15\,\mu g/m^3$to daily indoor $PM_{2.5}$ concentrations. Most of these studies employ real-time measurement methods to identify peak concentrations which are then attributed to different activities using time-activity diaries. An alternative approach taken by Zhao *et al.* [114] attributed emissions to different sources using the elemental composition on filter samples. A source with high abundance of OC and OC1, and some Ca, was attributed to cooking, and contributed 53.6% to residential indoor samples.

Despite cooking frequently being identified as a key indoor $PM_{2.5}$ source, only 3

studies included in Table 2.8 measured mass concentrations in residential kitchens. Abt *et al.* [168] intentionally located the measurement device between the kitchen and the living room to target the areas where most activity occurs. $PM_{0.02-0.1}$ concentrations were highest in the evening, when the majority of cooking occurred in homes [168]. Nasir & Colbeck [165] reported higher $PM_{2.5}$ concentrations in kitchens than living rooms in student accommodation and one bedroom flats. In 2-3 bedroom houses in this study, average concentrations were lower in the kitchen than living room, which the authors attributed to a combination of the use of wood burning to heat the living room and large kitchen volumes [165]. Wigzell *et al.* [163] also measured in multiple locations within each house. They observed higher peak PNCs associated with cooking in the kitchen than the living room. These observations of higher kitchen concentrations may partially explain the personal cloud effect observed by Ozkaynak *et al.* [170] and Williams *et al.* [160] who measured higher personal exposures than indoor concentrations. They located residential monitors in the living room, therefore if participants were in close proximity to $PM_{2.5}$ sources, they would be exposed to higher concentrations.

2.2.3 Summary

To conclude, a variety of existing methods exist to measure $PM_{2.5}$ concentrations, of which gravimetric sampling and optical measurement devices are the most frequently used. The nature of the study cost are factors when choosing a method. Gravimetric sampling provides exact mass concentrations and allows for subsequent chemical analyses, however concentrations are time averaged. Optical measurement devices measure real-time mass particle number concentrations, and sometimes particle size distributions. However, any mass concentrations are estimates dependent on custom calibration. Calibration factors vary within and between source types, therefore custom calibration should be conducted using concurrent gravimetric sampling to minimise error.

Existing research has measured personal exposures and indoor residential $PM_{2.5}$ concentrations. There is evidence to suggest individuals may be exposed to concentrations exceeding current guidelines, and in-home exposures contribute significantly to total exposure. Increasing building airtightness to reduce energy demand may increase the effect of indoor pollutant sources on IAQ. Key indoor $PM_{2.5}$ sources include smoking, cooking, cleaning, combustion, personal care and general activity. Cooking has frequently been found to contribute significantly to indoor concentrations and is an activity undertaken by many people, therefore is a source of interest. Proximity to sources may also be important so people located in the kitchen during cooking may be subject to higher risks. However, there are relatively few studies in literature that report $PM_{2.5}$ concentrations measured in kitchens in UK houses, hence the first objective to measure $PM_{2.5}$ in domestic kitchens, see Section 1.3.

2.3 Mitigation Strategies

There are three basic strategies to improve IAQ: (i) indoor and outdoor source control; (ii) ventilation; and (iii) air cleaning technologies [174]. Source control is preferred [23], as prevention is better than a cure. IAQ can be significantly improved by removing pollutant sources from the building. When the sources cannot be eliminated, for example $PM_{2.5}$ emitted from cooking, *source control* mechanisms can be employed to directly remove pollutants at source. Ventilation supplies clean air and extracts contaminated air to dilute the concentrations within a space [23], and air cleaning directly removes pollutants from the air, sometimes without need for ventilation [174].

Again, mitigation strategies can be directly investigated through measurement. Researchers frequently conduct intervention tests, where they measure indoor $PM_{2.5}$ concentrations under normal conditions for a given time period, perform an

intervention, for example providing additional equipment, and repeat the measurements to look for improvements.

Batterman *et al.* [173] provided free High Efficiency Particulate Air (HEPA) filters and window mounted air conditioning (AC) units with filtration low-income households with asthmatic children. A standard intervention group received just the freestanding HEPA filter, an enhanced group received the filter and an AC unit, and a control group received no intervention. The filter and AC were provided to the child's bedroom, where $PM_{2.5}$ was monitored. Overall, pre-intervention concentrations averaged $29 \pm 23\,\mu g/m^3$. After the intervention, $PM_{2.5}$ concentrations were lower in both intervention groups than in the control group, and filters reduced $PM_{2.5}$ levels by $15 \pm 13\,\mu g/m^3$ or 50%. Filter usage was initially high, but dropped in subsequent seasonal visits. Karottki *et al.* [175] conducted a similar study in 27 homes with non-smoking elderly occupants. Recirculating HEPA units provided to living and bedrooms reduced median $PM_{2.5}$ concentrations from median $8\,\mu g/m^3$ to median $4\,\mu g/m^3$ in consecutive two-week monitoring periods. Both studies show filtration can effectively reduce indoor $PM_{2.5}$ concentrations. However filters become dirty over time, and can themselves become a $PM_{2.5}$ source [23].

As part of their investigation in tightly constructed houses, Militello-Hourigan & Miller [167] measured PNCs during a controlled cooking test. The test was conducted first under normal ventilation conditions, and later with temporary boost ventilation activated. In the tightly constructed homes, results suggested the temporary boost ventilation was ineffective at reducing particle counts, however in the conventional home, they observed an 85% decrease from using the cooker hood. Dobbin *et al.* [176] found leaving the cooker hood running for an additional 15 minutes after cooking reduced integrated concentrations by a similar amount as increasing the flow rate by about 45 L/s.

Militello-Hourigan & Miller [167] and Dobbin *et al.* [176] both also modelled further

scenarios as part of their investigations. Dobbin *et al.* [176] predicted a 1 hour integrated exposure, which was lower when the fan was run for the first 15 minutes after cooking. Militello-Hourigan & Miller [167] predicted that for concentrations to return to below $35\,\mu g/m^3$ in 1 hour, the boost ventilation would have to run at $632\,L/s$ for 20 minutes, or $203\,L/s$ for the entire hour. In contrast the existing ventilation system would take > 2 hours. Running the cooker hood on high was predicted to reduce concentrations to $35\,\mu g/m^3$ in 20 minutes [167]. This highlights the benefits of extracting polluted air at source, however none of the tightly constructed homes were fitted with directly exhausting cooker hoods.

In 352 California homes, measured indoor NO_x, NO_2 and peak 1-hour CO concentrations were higher in homes with gas cooking, and increased amounts of gas cooking [177], and so were associated with cooking. The same study found that in homes that cooked 4 hours or more, self-reported use of kitchen extract ventilation was associated with lower concentrations of these pollutants. Similar results were also predicted using modelled CO, formaldehyde and NO_2 concentrations [38]. Although $PM_{2.5}$ was not modelled or measured in these studies, the predicted lower pollutant concentrations in households using cooker hoods or kitchen ventilation is likely to also apply to $PM_{2.5}$.

2.3.1 Kitchen Ventilation

Targeted kitchen exhaust ventilation is commonly accepted to be the most direct means of reducing cooking source pollutants, as demonstrated previously. Some ventilation systems, such as cooker hoods, provide *source control* as they extract pollutants at source before they mix with the environment. Pollutants can be diluted by the infiltration of ambient air through adventitious openings and by PPV. Infiltration is a poor dilution mechanism because it gives low, uncontrolled airflow rates, and because it is generally desirable to eliminate it to save energy. Accordingly, PPV is the most appropriate mechanism.

In England, statutory Approved Document F (ADF) [16] prescribes intermittent kitchen ventilation rates of 30 L/s adjacent to a hob (assumed to be through a cooker hood) or 60 L/s elsewhere, or a continuous ventilation rate of 13 L/s. In new dwellings these are obligatory, whereas it is only necessary to maintain existing ventilation systems when refurbishing other dwellings. These ventilation rates were chosen to remove moisture, with the further expectation that they will dilute NO_2 and CO emitted by gas cooking. $PM_{2.5}$ and other pollutants generated during cooking were not considered.

Other existing ventilation requirements are included in Table 2.9 for comparison. The European standard EN 16798-1:2019 provides recommended ventilation rates for use in the absence of national standards [178]. Its kitchen extract airflow rate is dependent on the number of main rooms in the dwelling and occupant expectations. For example, for a normal dwelling with 4 main rooms, the specified kitchen extract is 35 L/s. However, the standard is unclear whether this is an intermittent or a constant airflow rate, but the ventilation rate can be halved if there is also a cooker hood present. The prescribed ventilation rate varies with dwelling size and so is not directly comparable to those in ADF.

In the US, the American Society of Heating, Refrigerating and Air-Conditioning Engineers (ASHRAE) recommends higher kitchen exhaust ventilation rates in their Standard 62.2 [179], and gives different requirements for enclosed and nonenclosed kitchens. The recommended intermittent general extract rate of 150 L/s is more than twice the 60 L/s prescribed by ADF. An additional continuous extract is prescribed for enclosed kitchens, at $5\,h^{-1}$.

Within the field of IAQ, the principle of *ventilation equivalence* can be defined as the provision of the equivalent volume of air, and IAQ equivalence the provision of equivalent IAQ [180]. To simplify the concept outlined by Sherman *et al.* [180], an alternative ventilation pattern can be used if it provides the same average concentration of a generic contaminant. For pollutants with constant indoor

TABLE 2.9: Existing kitchen ventilation standards

Standard	Location	Kitchen Geometry	Extract Rate	Intermittent?	Other details	Reference
ADF	England and Wales	Any kitchen	30 L/s	Intermittent	Adjacent to hob	[16]
ADF	England and Wales	Any kitchen	60 L/s	Intermittent	Elsewhere in kitchen	[16]
ADF	England and Wales	Any kitchen	13 L/s	Continuous	Minimum high rate	[16]
EN 16798-1:2019	Europe	Any kitchen	20 L/s	?	for 1 main room*	[178]
EN 16798-1:2019	Europe	Any kitchen	25 L/s	?	for 2 main rooms*	[178]
EN 16798-1:2019	Europe	Any kitchen	30 L/s	?	for 3 main rooms*	[178]
EN 16798-1:2019	Europe	Any kitchen	35 L/s	?	for 4 main rooms*	[178]
EN 16798-1:2019	Europe	Any kitchen	40 L/s	?	for 5+ main rooms*	[178]
ASHRAE 62.2	US	Enclosed Kitchen	50 L/s	Intermittent	Vented range hood	[179]
ASHRAE 62.2	US	Enclosed Kitchen	150 L/s	Intermittent	Other exhaust inc downdraft	[179]
ASHRAE 62.2	US	Enclosed Kitchen	5 h^{-1}	Intermittent	Other exhaust inc downdraft	[179]
ASHRAE 62.2	US	Non-enclosed	50 L/s	Intermittent	Vented range hood	[179]
ASHRAE 62.2	US	Non-enclosed	150 L/s	Intermittent	Other exhaust inc downdraft	[179]
ASHRAE 62.2	US	Enclosed Kitchen	5 h^{-1}	Continuous	General exhaust, occupied hours	[179]

*also gives multiplying factors for different building categories:
(I) 1.4 sensitive or vulnerable populations (II) 1 normal (III) 0.7 allowed in renovation (IV) 0.5 only if additional cooker hood

emissions ventilation equivalence can be defined mathematically. However this is not applicable to episodic activity, such as cooking, which is better addressed with targeted task ventilation [180].

2.3.1.1 Cooker Hood Performance

Both the ventilation requirements set out in ADF and ASHRAE Standard 62.2 require lower ventilation rates through a cooker hood, also known as a *range*, *exhaust* or *extractor* hood, based on the assumption that a significant proportion of cooking fumes are removed before mixing with the kitchen air. ADF does not currently require or use a performance metric for cooker hoods, but seemingly assumes the close proximity to the source means it is twice as effective. A cooker hood *capture efficiency* (CE) has been defined as the percentage of an emitted pollutant extracted, either directly or during operation [181].

Cooker hood CEs have not been measured in UK dwellings, but elsewhere they ranged 12% [181] and 98% [182]. The CE of a hood has been found to vary with the airflow rate, installation height, hood capture volume, and the fraction of the stovetop covered by the hood [181, 183].

Multiple methods have been used to measure CEs. Singer *et al.* [183] measured the performance of 15 hoods as installed in California residences. Some hoods had operational airflow rates significantly below the manufacturer specified rates, and

the ratio of measured to nominal airflow at the highest fan setting was $28 - 100\%$. CEs were assessed using CO_2 as a marker for burner pollutants, which was measured in the exhaust stream. 4 burner arrangements were tested, namely: oven only, single front burner, single rear burner, and 1 front and 1 back burner. Cooktop coverage, defined as the fraction of the burner area covered by the exhaust device, was between $< 50\%$ and $> 100\%$. CEs were higher in the back burner tests for many devices. Generally, higher flow rates were associated with higher CEs, although there was wide variation in CE between different hoods operating at the same flow rate.

Lunden *et al.* [181] performed scripted cooking procedures and determined direct CEs by comparing CO_2 mass flow through the hood to CO_2 produced at the burner, and indirect particle CEs by comparing particle concentrations in the room for repeat cooking events with or without a cooker hood. The direct approach was deemed unsuitable for particles, as additional particle losses in the hood and duct-work would cause bias. Overall, the findings were similar to those reported by Singer *et al.* [183], with lower CEs at low flow rates ($34 - 38\%$) than high ($54 - 72\%$). Particle and CO_2 CEs were similar when over 80%, but deviated at lower capture efficiencies and for different cooking procedures. Therefore, the authors concluded that CEs determined using burner CO_2 would only be applicable to particles under certain circumstances [181].

Three further studies found similar results. Rim *et al.* [182] measured whole house PNC reductions of $31 - 94\%$ when using the front burner, and $54 - 98\%$ on the back burner. Particle reduction efficiency was larger for higher hood flow rates and larger size fractions. Sjaastad & Svendsen [184] also found positioning the hood and stove in the corner of the room was beneficial in comparison to the centre (island hood) or at the wall. Finally, physical features that entrain pollutants towards the device, and increase the capture volume, may increase the reduction potential of a hood. Two such features found to reduce room particle concentrations were a *damp buffer*

(where the sides that frame the hood protrude below its central horizontal plate) and side barriers [185].

A new ASTM standard prescribes a steady state capture efficiency test method that only considers pollutants removed before they mix with room air [186, 187]. A tracer gas is emitted at a defined rate into a plume from a heated surface. The test is conducted in a chamber with defined kitchen cabinetry geometry, kept under constant conditions until steady state conditions are achieved. A minimum of 10 tracer gas measurements then are taken over $\geq$ 10 minutes in each of three locations, the exhaust stream, C_e, the mixed chamber air , C_c, and the chamber inlet C_i, to calculate CE using Equation 2.4.

$$\mathrm{CE} = \frac{C_e - C_c}{C_e - C_i} \tag{2.4}$$

This method aims to reduce error, including avoiding measurement biases when the same instrument is used for all three measures [186], and because all measurements are taken simultaneously between test variability is avoided. The current procedure is only suitable for wall mounted cooker hoods, although an equivalent procedure is being developed for island hoods and downdraft units [186]. This method uses on tracer gas concentrations rather than particulates, an unimportant distinction when comparing and rating the performance of different hoods. However, this may be a source of error if the measured CEs are used to predict particle concentrations in dwellings.

A series of trials tested the ASTM procedure with 8 cooker hoods [186, 188]. CE increased with higher flow rate and lower mounting height, and CEs varied between models and configurations at the same flow rate [186]. At high flow rates ($>$ 140 L/s) the measured CEs were over 80% for all tested hoods [188]. The authors stated it is impractical to require all hoods to have flow rates of this magnitude as there would be associated energy penalties and increased noise, making it less likely that the hood would be used [188]. Kim *et al.* [186] suggested

CE might be a better performance metric than flow rate. However, cooker hoods also reduce internal concentrations by increasing the building air exchange rate [176], therefore, both CE and flow rate are important to cooker hood performance.

2.3.2 Summary

In summary, the key mitigation strategies appropriate to indoor air are source control, ventilation and air cleaning. With regards to cooking source emissions, complete source removal is not possible, however source control can be achieved using targeted task ventilation such as a cooker hood.

Current UK ventilation requirements in ADF were set to control humidity, and are lower than those set elsewhere. ADF also allows a lower extract rate for ventilation adjacent to the stove, here presumed to be through a cooker hood. However, this and other standards do not prescribe additional performance metrics for cooker hoods. Previous work does not investigate the performance of existing ventilation in UK kitchens with regards $PM_{2.5}$ concentrations, hence the objective of this work to investigate the potential to reduce kitchen concentrations using existing ventilation through a simple intervention.

One such performance metric is the capture efficiency (CE), here defined as the proportion of an emitted pollutant directly extracted by the hood before mixing with room air. Multiple CE test methods exist in literature, and a direct steady state approach using tracer gas is considered preferable, although it may overestimate CEs for particles at lower exhaust flow rates. Factors that impact hood CEs include flow rate, burner coverage, position in room, mounting height and hood geometry. The benefits of using a cooker hood are established but existing empirical performance data is mostly from the USA, hence the objective to investigate the potential to reduce emissions using a cooker hood.

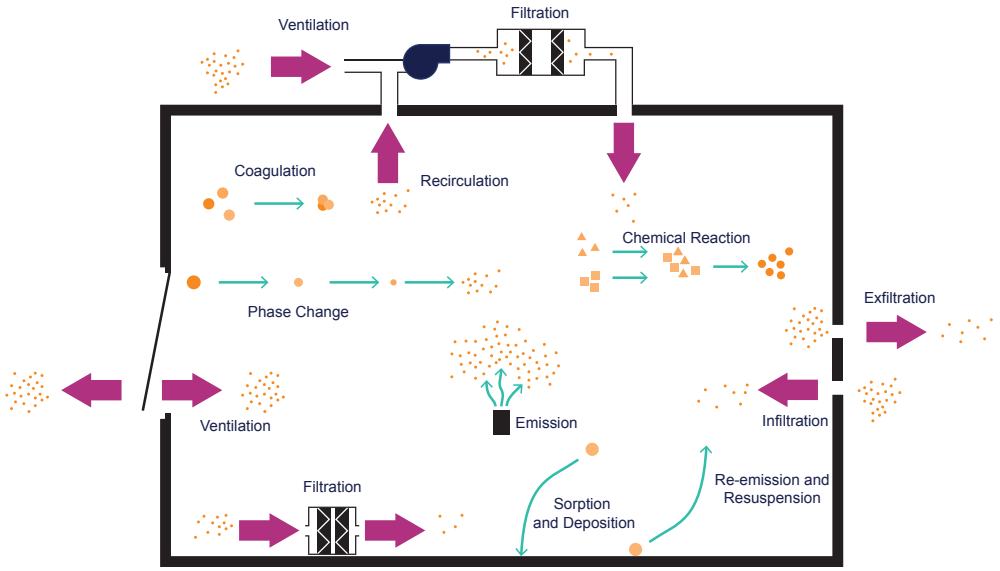


FIGURE 2.2: Processes that affect particle concentrations in the indoor environment

2.4 Simulation and Modelling

Large scale in situ monitoring is often invasive, and cost and time prohibitive. In addition, this method cannot be used to test the effect of changes to the built environment without possible ethical implications. An alternative is to model stocks of dwellings to estimate concentrations or exposures at the population level and predict the impact of interventions.

2.4.1 Factors Affecting Indoor $PM_{2.5}$ Concentrations

The processes that influence indoor $PM_{2.5}$ concentrations are outlined in Figure 2.2, and some have previously been mentioned.

First, ventilation. Particles can enter and leave the building via ventilation, including intentional, purpose-provided ventilation (PPV) and unintended

infiltration and exfiltration through gaps and cracks in the building fabric. Some ventilation methods can have a filtering effect so the concentration is lower indoors than outdoors; for example, in a California house with no indoor sources and mechanical extract-only purpose provided ventilation, indoor $PM_{2.5}$ concentrations were 70% lower than outdoor concentrations [189]. The degree of filtering, represented by non-dimensional penetration coefficient P, relates to the ventilation mechanism. Open windows provide no filtration ($P = 1.0$) [44], mechanical ventilation supply can provide filtration ($P < 1.0$) [189], and infiltration typically filters out some particles ($P < 1.0$) [44]. $PM_{2.5}$ can also be filtered indoors without ventilation using free-standing units, as explored in Section 2.3.

Indoors, $PM_{2.5}$ forms through primary processes, where particles are emitted from sources such as cooking or smoking, or secondary processes, where particles form as a result of chemical reactions in the air [106]. Once formed, $PM_{2.5}$ mixes with room air.

Pollutants such as semivolatile organic compounds (SVOCs) can change between particle and gas phases, and such phase change can cause indoor $PM_{2.5}$ concentrations to increase or decrease [112]. Smaller particles colliding in the air can coagulate to form larger particles [190], this can lead them to grow to sizes larger than 2.5 µm. Particles deposit onto indoor surfaces [106], and deposition rate varies with particle size and surface characteristics [190]. He *et al.* [190] found that ventilation and deposition were the most significant particle decay mechanisms. Deposited particles can also be re-suspended [44], and activities sometimes considered indoor $PM_{2.5}$ sources may be re-suspending rather than emitting $PM_{2.5}$, for example indoor activity [114, 165, 166] and some forms of cleaning [191].

2.4.2 Simulation Approaches

A frequent modelling approach uses building simulation tools such as CONTAM or the Generic Contaminant model within EnergyPlus™, to model multi-zone

representations of buildings with a large number of inputs. Previous studies have used similar approaches to estimate indoor concentrations [37], or exposures [39, 191–194], of $PM_{2.5}$ [37, 39, 191, 193] and other pollutants [169, 192–196], for all [39, 192, 193] or part [37, 191, 194] of the English housing stock. They often create archetypal rooms or houses that are simulated either deterministically (with a single set of input parameters), or probabilistically (varying the input parameters within a range of known values). The predictions are then extrapolated to the housing stock scale. Additionally, these models have been used to predict concentrations or exposures for future scenarios [39, 191–193], which is useful for testing the potential impacts of retrofit policies. A few such models and their potential limitations are considered in greater detail.

Shrubsole *et al.* [191] used deterministic modelling to predict indoor $PM_{2.5}$ concentrations in the London housing stock for present day (2010) and future (2050) scenarios. The 2050 scenario modelled the 2010 stock with energy efficiency refurbishments including reduced fabric permeability and installation of mechanical ventilation with heat recovery (MVHR) systems. Representative multi-zone archetype dwelling forms were modelled in CONTAM [197], a validated indoor air quality model [191]. $PM_{2.5}$ sources included outdoors, cooking, smoking, sweeping, vacuuming, dusting and washing or showering, with fixed emission or resuspension rates and schedules for each indoor source. For example, cooking emitted $PM_{2.5}$ at 1.6 mg/min for 15 minutes each morning, 30 minutes each evening and 30 minutes at weekend lunchtimes. The emission schedules were assumed due to a lack of available data. Occupants exposures were estimated using time weightings for the living room, kitchen and bedroom, which differed for occupants who spend time in the kitchen, labelled *cooks*, and *non-cooks* who do not. The predicted 2010 annual average exposures were higher in households with smoking (overall average exposure 54.1 $\mu g/m^3$) than those without (22.0 $\mu g/m^3$), and were higher for cooks (non-smoking scenario 54.2 $\mu g/m^3$) than non-cooks (15.5 $\mu g/m^3$). Concentrations were lower in the future scenarios due to reduced

ingress of external $PM_{2.5}$ and improved ventilation using the MVHR system. The authors also conducted a sensitivity analysis by re-running the model with minimum and maximum values for each input in turn. The model was most sensitive to $PM_{2.5}$ deposition rate, $PM_{2.5}$ emission rate and window opening.

Two further studies used a similar deterministic modelling approach and assumptions to estimate I/O ratios for outdoor source $PM_{2.5}$ in London [198], and the impacts of energy efficiency retrofitting measured on indoor $PM_{2.5}$ [39]. Both use EnergyPlus™ rather than CONTAM to create a coupled dynamic thermal and contaminant model [198]. One advantage of this method is that window opening behaviour can be coupled to internal temperatures. I/O ratios modelled without internal $PM_{2.5}$ sources were higher in the summer months when windows were openable, higher in detached and semi-detached properties with higher permeability, and lower in flats [198]. A differential sensitivity analysis, similar to Shrubsole *et al.* [191], was selected over a Monte-Carlo analysis as little information was known about the distribution of input parameters [198]. The second study built on this approach to test the effect of wall insulation, loft insulation, floor sealing and double glazing on indoor $PM_{2.5}$ concentrations in the English housing stock [39].

Hamilton *et al.* [193] combined a similar multi-pollutant deterministic model using CONTAM, and subsequent Monte-Carlo analysis to estimate the health effects of energy and ventilation retrofits. Three different future ventilation scenarios were tested for the same energy retrofits, namely ADF compliant PPV through extract fans and trickle vents in all houses, the same PPV provided in dwellings recorded as having signs of inadequate ventilation in the English Housing Survey, and no additional PPV except to repair existing extract fans or trickle vents. The first scenario reduced indoor concentrations of indoor source pollutants, and resulted in 2241 QALYs gained per 10000 persons due to the increased ventilation rates. Scenario 2 and 3 both increased indoor exposure to indoor source $PM_{2.5}$, and

predicted 539 and 728 QALYs lost per 10000 persons over 50 years. Assuming indoor source $PM_{2.5}$ had no adverse health effect, to test the effect of toxicity, reduced the overall net health effect by 78%, and therefore the authors highlighted the need for more empirical studies to establish the dose-response relationship for such $PM_{2.5}$ [193].

Deterministic models use a limited number of simulations which reduces total computational time. However, they do not incorporate uncertainty in the inputs or outputs, and so the results are typically presented as an overall mean concentration or exposure for the stock, rather than producing distributions. The differential sensitivity analysis approach highlights the importance of different inputs, but is limited as it gives equal weights to the upper and lower bounds of the distribution, where values are more likely to be closer to the mean [198].

Das *et al.* [37] proposed an enhanced approach using a base model similar to Shrubsole *et al.* [191], but with inputs sampled from appropriate probability distributions. They used a Latin Hypercube sampling technique, using a series of mini Latin Hypercubes to generate inputs to the CONTAM model. Sampling continued until the change in mean and standard deviation was less than 0.2%, thereby producing multiple sets of model inputs with corresponding outputs. This allows a sensitivity analysis based on correlation and regression analysis. For single-storey English flats, the three most important variables for predicting indoor exposure to indoor source $PM_{2.5}$ were the opening area of the kitchen window, indoor $PM_{2.5}$ emission rate and indoor temperature [37].

Das *et al.* [37] still used a fixed emission schedule for cooking and smoking similar to those used previously, and so duration of emissions was not included in the sensitivity analysis. Additionally, in all the above studies, exposure was estimated based on a ratio of time spent in the kitchen, living room and bedroom. This offers a simple solution, however it may underestimate exposures by not accounting for the fact that cooks will mainly occupy the kitchen during cooking, when

concentrations are likely to be highest.

The EXPAIR [194] and INDAIR-2 [194, 196] models are probabilistic models which mathematically compute pollutant concentrations in each of 3 compartments to model residential microenvironments. The INDAIR-2 model includes variability in cooking and smoking behaviour, and EXPAIR was designed to estimate population exposures using probabilistic time-activity-location profiles [194]. Probabilistic time-activity-location profiles were developed for 3 population groups: children aged $4 - 15$, office workers and adults aged 65+. The model was applied to Leicester to predict NO_2 exposures, but not for $PM_{2.5}$ [194]. A limitation of this model is the availability time-activity data. Here the authors used the BBC Survey for Daily Life data from 1998, however it is reasonable to assume population behaviour may have changed in the last two decades.

Finally, the complex models described here can predict indoor pollutant concentrations or exposures, but are computationally expensive, which limits their ability to test multiple scenarios. Therefore, they are useful for testing the efficacy of existing or proposed regulations, but not for searching for optimal solutions to a problem, such as identifying appropriate ventilation rates for domestic kitchens.

2.4.3 Simplified Mathematical Modelling

Alternatively, Salthammer [10] proposed a statistical approach to generate a probability density function (PDF) of pollutant concentrations using a simple, mathematical, mass balance model and Monte-Carlo sampling. The basis of mass balance principle is that the total emitted pollutant mass must be accounted for, as mass cannot be created or destroyed [44]. Ott *et al.* [44] derived different forms of the mass balance equation starting from Equation 2.5.

$$\begin{pmatrix} \text{Mass generated} \\ \text{between Time} \\ t = 0 \text{ and } t = T \end{pmatrix} - \begin{pmatrix} \text{Mass lost} \\ \text{between Time} \\ t = 0 \text{ and } t = T \end{pmatrix} = \begin{pmatrix} \text{Mass inside} \\ \text{chamber at} \\ \text{Time } t = T \end{pmatrix} \tag{2.5}$$

These principles can then be used to derive Equation 2.6, a form of the mass balance equation frequently found in literature (see He *et al.* [35], Dacunto *et al.* [40], and Olson & Burke [199]), which gives the concentration, C, as a function of time, t. A significant limitation of this equation is that all variables must be constant over time. If any of these change during the period of interest, the equation is reset with a new initial concentration. For a full derivation of Equation 2.6 see Appendix B.

$$C(t) = \frac{\lambda_T P C_a}{\phi} + \frac{(1-\eta)g}{\phi V} + \left(C(0) - \frac{\lambda_T P C_a}{\phi} - \frac{(1-\eta)g}{\phi V} \right) e^{-\phi t} \tag{2.6}$$

$C(t)$	Pollutant concentration at time t	[M/L^3]
$C(0)$	Initial concentration	[M/L^3]
C_a	Ambient concentration	[M/L^3]
P	Penetration coefficient	[-]
V	Volume	[L^3]
g	Emission rate, considered constant	[M/T]
η	Cooker hood capture efficiency	[-]
t	time	[T]
ϕ	Total decay rate, often split into:	[T^{-1}]
λ	Decay due to ventilation and	[T^{-1}]
k	Decay due to all other mechanisms	[T^{-1}]
	(L length, M mass, T time)	

Under constant conditions, as t becomes large, $e^{-\phi t}$ tends to 0. This scenario is the *steady state* condition, under which Equation 2.6 simplifies to Equation 2.7, which defines the steady state concentration C_{ss}.

$$C_{ss} = \frac{\lambda_T P C_a}{(\lambda_T + k)} + \frac{(1-\eta)G}{(\lambda_T + k)V} \tag{2.7}$$

Salthammer [10] used a version of Equation 2.7 to predict indoor formaldehyde levels emitted from building materials. The model assumes that the pollutant is well mixed throughout the room once released, and that the environmental conditions remain constant. If each input is considered to be a random variable with a known distribution, sampling from these distributions and performing multiple calculations gives a PDF of steady state concentrations. The resulting distribution can be used to predict what percentage of the stock experience concentrations exceeding relevant guidelines, and could also be iteratively used to set ventilation requirements.

This method is useful for constantly emitting pollutants. For intermittent sources, such as cooking, this method can also be used to predict steady state concentrations. However, the predicted concentrations are not useful if it is unlikely the steady state concentration will be reached. Additionally, current guidelines set mean concentrations, so a model should predict mean concentrations over the same time period to allow direct comparison.

2.4.4 Summary

Models can be a useful tool for predicting pollutant concentrations and exposures, and the effect of changes to the building stock. Probabilistic modelling is favourable as it accounts for uncertainty in the model inputs and can produce distributions of outputs, useful for assessing the percentage of the population or stock predicted to be at risk. Complex modelling are computationally expensive and are not designed to predict optimal building conditions. An alternative simplified mathematical model predicted steady state concentrations from a constantly emitting source in a single zone. This approach could be modified for

use with intermittent cooking $PM_{2.5}$ emissions. Previous research considered here has used models to predict $PM_{2.5}$ concentrations in dwellings, but not to test the merits of ventilation rates currently prescribed in ADF, hence the objective to do so using statistical modelling.

Sensitivity analyses for existing models revealed that models are highly sensitive to the emission rates used for indoor $PM_{2.5}$, however, all of the models considered in detail used the same emission rate of 1.6 ± 0.6 mg/min. This emission rate was derived from data published in 1996 [170], and therefore the measured variability of cooking emissions warrants further consideration, an objective of this work, see Section 1.3.

2.5 $PM_{2.5}$ Emission Rates

$PM_{2.5}$ concentrations in stocks of houses can be predicted using modelling approaches, where nationwide monitoring campaigns might be invasive, expensive and time restricted. However, these models are sensitive to the emission rates used. Therefore, this section considers existing knowledge regarding $PM_{2.5}$ cooking emission rates and the techniques used to measure them.

2.5.1 Cooking Source Emission Rates

Numerous studies investigated cooking emissions from cooking single ingredients, or general emissions from uncontrolled cooking events. The measurement techniques are explored in Section 2.5.3. Table 2.10 gives a summary of cooking emission rates reported in literature. Herein, grilling refers to food being cooked using only radiant heat from above, which is the UK definition, and is equivalent to broiling in the US.

TABLE 2.10: Cooking emission rates (g) and source strengths (G)

Cooking Process	Event	Fuel	n	g (mg/min)		G (mg)		Reference
				μ*	SD*	μ*	SD*	
ALL	All activity		836	1.72		30		Chan *et al.* [36]
	All activity		836	0.62^{M}		12^{M}		Chan *et al.* [36]
	All activity		836	0.67^{G}	0.065^{G}	13^{G}	3.7^{G}	Chan *et al.* [36]
	Multiple events		43	29	63			Olson & Burke [199]
	Unspecified		149	14	17			Olson & Burke [199]
	Cooking		37	0.11^{M}	0.99			He *et al.* [35]
	All cooking		411	36	98			Olson & Burke [199]
	Full Meal	Gas	1	2.45				Fortmann *et al.* [32]
	Burned food		6	470	530			Olson & Burke [199]
STOVETOP	Stove		4	0.24^{M}	1.29			He *et al.* [35]
	Stovetop		40	17	35			Olson & Burke [199]
	Boiling	Elec	15	1	0.21			Nasir & Colbeck [165]
Frying	Frying		4	2.68^{M}	2.18			He *et al.* [35]
	Frying		105	60	92			Olson & Burke [199]
	Frying	Elec	10	1.47	0.75			Nasir & Colbeck [165]
	Salmon		1	1.6		18.3		Dacunto *et al.* [40]
	Chicken breast		1	15.2		289		Dacunto *et al.* [40]
	Chicken		6	2.5	0.9	62.2		Dacunto *et al.* [40]
	Bacon	Gas	1	3.88				Fortmann *et al.* [32]
	Bacon	Elec	1	1.45				Fortmann *et al.* [32]
	Bacon	Gas	1	3.07				Fortmann *et al.* [32]
	Bacon		6	3.5	2.8	36.6	27.7	Dacunto *et al.* [40]
	Pork chops		2	2.1	0.3	24.3	7.1	Dacunto *et al.* [40]
	Sausage links		1	5.4		54		Dacunto *et al.* [40]
	Fry Beef	Gas	1	0.72				Fortmann *et al.* [32]
	Fry Beef	Gas	1	1.25				Fortmann *et al.* [32]
	Fry Beef	Gas	1	4.28				Fortmann *et al.* [32]
	Fry Beef	Gas	1	0.45				Fortmann *et al.* [32]
	Fry Beef	Gas	1	2.99				Fortmann *et al.* [32]
	Ground beef		2	0.17	0.05	1.9	0.8	Dacunto *et al.* [40]
	Hamburger patty		4	4.3	3	40.6	27.8	Dacunto *et al.* [40]
	Stir Fry	Gas	1	3.36				Fortmann *et al.* [32]
	Stir Fry	Gas	1	1.54				Fortmann *et al.* [32]
	Stir Fry	Elec	1	2.92				Fortmann *et al.* [32]
	Stir Fry	Gas	1	3.70				Fortmann *et al.* [32]
	Stir Fry	Gas	1	9.10				Fortmann *et al.* [32]
	Chicken Stir Fry		2	0.4		5.7		Dacunto *et al.* [40]
	Tortillas	Gas	1	4.22				Fortmann *et al.* [32]
	Tortillas	Elec	1	10.3				Fortmann *et al.* [32]
	French fries (deep fried)	Gas	1	1.10				Fortmann *et al.* [32]
	French fries (deep fried)	Gas	1	0.77				Fortmann *et al.* [32]
	French fries (deep fried)	Elec	1	4.37				Fortmann *et al.* [32]
	French fries (deep fried)		2	0.34	0.03	8.4	2.4	Dacunto *et al.* [40]
Oven	Oven		38	10	12			Olson & Burke [199]
	Oven		6	0.03^{M}	0.03			He *et al.* [35]
	Cooking pizza		1	1.59^{M}				He *et al.* [35]
	Frozen pizza (oven)		1	0.1		1.4		Dacunto *et al.* [40]
	Bake Lasagna	Gas	1	4.19				Fortmann *et al.* [32]
	Bake Lasagna	Gas	1	1.48				Fortmann *et al.* [32]
	Bake Lasagna	Elec	1	0.43				Fortmann *et al.* [32]
	Pork Roast	Gas	1	0.15				Fortmann *et al.* [32]
	Pork Roast	Gas	1	0.025				Fortmann *et al.* [32]
	Pork Roast	Gas	1	1.11				Fortmann *et al.* [32]
Grill	Grilling		6	2.78^{M}	17.8			He *et al.* [35]
	Grilling		3	173	92			Olson & Burke [199]
	Grilling	Elec	10	1.7	0.27			Nasir & Colbeck [165]
	Grill Fish	Gas	1	5.30				Fortmann *et al.* [32]
	Grill Fish	Elec	1	3.81				Fortmann *et al.* [32]
	Grill Fish	Gas	1	2.28				Fortmann *et al.* [32]
Toasting	Toasting		18	0.11^{M}	0.37			He *et al.* [35]
	Toaster oven		7	51	27			Olson & Burke [199]
	Toast		3	9.5	10.8	72.5	71.2	Dacunto *et al.* [40]
Other	Microwave		18	0.03^{M}	0.11			He *et al.* [35]
	Microwave		20	11	11			Olson & Burke [199]
	Popcorn (microwave)		1			2.5		Dacunto *et al.* [40]
	Kettle		25	0.03^{M}	0.31			He *et al.* [35]
	Oven Cleaning	Elec	1	2.94				Fortmann *et al.* [32]

*arithmetic mean and standard deviation unless indicated otherwise; ^{M}indicates median value; ^{G}indicates geometric mean or standard deviation

The emission rates in Table 2.10 are highly variable, ranging from 0.03 mg/min from roast pork with extract ventilation [32] to 470 mg/min for burned food [199]. For comparison with the 1.6 ± 0.6 mg/min used in previous models, only 12 of the 66 emission rates in Table 2.10 fall within 2 standard deviations of this mean. Where the SD is reported is is often a similar magnitude or larger than the mean. This is especially true for the less specific events [35, 199]. Emission rates are always positive, so these SDs indicate non-normal distributions.

The variation is observed both within and between cooking types. For example, Fortmann *et al.* [32] reported emission rates of $0.45 - 4.28$ mg/min for frying beef mince (or ground beef) under differing conditions, by comparison, Dacunto *et al.* [40] found a lower $g = 0.17 \pm 0.05$ mg/min for the same source. Reasons for this variability may be of interest when building a probabilistic model. The reported emission rates are frequently based on small sample sizes and presented as a mean and standard deviation, but the shape of the distribution is not explored. Therefore, there is need for a method to measure $PM_{2.5}$ cooking emission rates and develop probability distribution functions suitable to input into a probabilistic model.

2.5.2 Factors that Influence Emission Rates

The variability in cooking emissions relates to the complex nature of cooking. Five main factors can affect cooking emissions.

Firstly, the cooking method has a clear effect on the emission rate. Dry, water based, and oil based cooking process all have different emission characteristics, and oil based methods, such as frying, have the highest emission rates [200]. Similarly, burned food, grilling/broiling, and frying have been found to have the highest emission rates [32, 35, 199]. Higher emissions have been found from stir-frying than pan frying, which wa attributed to higher temperatures [181]. Higher particle numbers and mass concentrations at higher concentrations have been found some

[201], but not by all; [46] possibly because the oil *smoke point*, the temperature at which the oil visibly smokes, was not reached.

Secondly, there is evidence that ingredients influence $PM_{2.5}$ emissions, and oil type is perhaps the most significant [123, 201]. The oil smoke temperature is important, but so are the composition and water content [200, 202, 203]. Emission rates from the heating of different cooking oils have been found to vary [202, 204]. Corn, coconut, and olive oils were found to have higher emission rates that heated soybean, safflower, canola, and peanut oils [202]. This difference was mostly related to the smoke point of the oils, except for olive oil, which generated $PM_{2.5}$ at the same temperature as corn and coconut oils, despite having a higher smoke point. Elsewhere, corn and soybean oils were found to have a lower emission rate than rapeseed and sunflower oils [204].

Thirdly, the effects of non-essential additives, such as seasonings, on emissions from heating oil were investigated by Torkmahalleh *et al.* [205]. In controlled laboratory tests, the addition of sea and table salt to canola oil reduced the $PM_{2.5}$ emission rate. A similar test reduced emission rates by 56% when salt was added to corn oil [121]. In contrast, higher particle emissions were found for grilling salted meat than unsalted meat [206].

Fourthly, the food type is important. A positive correlation was found between the fat content of foods and their emission rate [116, 123, 201]. Additionally, the water content of foods could impact particle size distribution, as observed when grilling ground beef [120].

Finally, there is some evidence that the cooking equipment used is influential. For example, adsorbed organic matter on the surface of pans may generate particulate matter when heated [200]. Heating an empty pan has been found to emit ultra-fine particles (UFP) but not $PM_{2.5}$ [206]. Additionally, higher emission rates have been reported when cooking using gas burners rather than electric hobs (also known

as a cooktop) [31, 200]. However, this finding is not universal, possibly due the confounding effect of hob temperature [201].

2.5.3 Measurement Methods

Existing measurement methodologies of emission rates from domestic cooking can be divided into five categories, each of which has its own benefits and frequency in the literature.

The first method is personal monitoring, which is generally rare. Olson & Burke [199] used this method to determine so called *personal* emission rates using measurements of $PM_{2.5}$ concentration made during four, seven-day monitoring periods for 37 participants. Concentration peaks from cooking were identified and cross-referenced with participant diaries, and the emission rate calculated using a mass balance approach that considered a house to be a single well-mixed space. A total of 411 cooking events found that emission rates ranged between 0.6 mg/min and 1496 mg/min ($\mu = 36$ mg/min). The authors made it clear, however, that these *personal* emission rates are high, due to the proximity of the sensor to the source. Additionally, the true emission rates are likely further overestimated by the assumption of the whole house being well-mixed. The cooking methods were recorded, but details of the food were not, reducing the utility of the data for use in modelling.

The second approach uses monitors located in places of interest, frequently in dwellings. He *et al.* [35] measured $PM_{2.5}$ mass concentrations for 48 hours in 15 houses in Brisbane, Australia. Elevated concentrations were linked to events using occupant diary entries, and catalogued into 21 activity types. An air exchange rate (AER) test and controlled cooking test were conducted in each house. The latter consisted of frying half an onion in vegetable oil on *high* heat, both under minimum and normal ventilation conditions. This provided an opportunity for direct comparison between houses. Emission rates were determined using a mass balance

relationship for a total of 106 cooking events (153 total), where emission rates ranged from 0.03 mg/min to 2.78 mg/min ($\mu = 0.11$ mg/min). The lack of control over ventilation rates and emission periods led to large errors. Nasir & Colbeck [165] followed the same method in English dwellings and estimated emission rates for oven grilling, $g = 1.70 \pm 0.27$ mg/min ($n = 10$), boiling, $g = 1.00 \pm 0.21$ mg/min ($n = 15$), and frying, $g = 1.47 \pm 0.75$ mg/min ($n = 10$). Chan *et al.* [36] developed an algorithm to identify emission periods and estimate emission rates from 224 days This method did not require occupant activity diaries, however this means the source types are unknown. Overall 836 distinct events were identified, with $\mu = 1.72$ mg/min. In a non-domestic study of this kind, See & Balasubramanian [117] monitored fine and ultrafine particles in a typical Chinese food stall in Singapore. Studies of commercial cooking are useful for understanding the nature of particle emissions from cooking, conversely, concentrations and emission rates reported may be less applicable due to differences in cooking behaviour and duration, and quantities of food prepared.

The third method uses managed tests conducted in semi-controlled conditions in the field, typically in a residential setting; see Dacunto *et al.* [40] and Fortmann *et al.* [32]. Both studies investigated emission rates from a variety of sources, and conducted at least some tests in a residential setting, using gas and electric stoves. They used different mass balance relationships to estimate emission rates. The AERs were measured using tracer gas decay, and the test environments flushed with outside air between tests. Dacunto *et al.* [40] conducted 66 measurements in 3 locations, including both cooking and non-cooking sources, with duplicates of most, but not all tests. The lowest emission rate of 0.1 mg/min was from oven cooking frozen pizza, whilst the highest cooking emission rate of 15.2 mg/min was from fried chicken breast. Fortmann *et al.* [32] conducted 32 cooking tests using a variety of foods and cooking methods based on the US diet, under multiple ventilation conditions, in a single location. 7 tests were repeated, but comparing changes using gas and electric heat sources. Just 4 tests were duplicated exactly to

investigate variability, the results of which suggested high variability that exceeded 100% and a need for more repetitions. Estimated emission rates ranged from 0.025 mg/min, for pork roast with exhaust ventilation, to 10.3 mg/min, for frying tortillas on an electric range. The range of emission rates from both of these studies highlights the need to understand the population diet when choosing food sources to investigate, whilst the low repeatability - indicated in the repeatability tests [32] and large standard deviations [40] - calls for more repetitions.

The fourth measurement method employs large scale chambers to obtain a further level of control over ventilation rates and mixing conditions, pollutant concentrations in the supply air, and internal materials to minimise sink effects. By creating a test chamber within a building, the effect of outside air and natural ventilation processes is reduced, and over-pressuring the test chamber controls the direction of background ventilation [14]. Afshari *et al.* [14] investigated concentrations of fine and ultrafine particles from 10 cooking and non-cooking sources, under identical conditions, in a full-scale test chamber. Lee & Wang [207] found $PM_{2.5}$ emission rates of $0.72 - 1.83 mg/min$, for 5 mosquito coils tested in a large environmental chamber. Temperature, relative humidity, AER, mixing conditions and leakage were all controlled during these tests, which reduces error in the estimated emission rates. Finally, Pagels *et al.* [208] investigated chemical composition and mass emission of candle smoke particles. These are not cooking sources, so only the method is of interest, which used a positively pressurised stainless steel chamber, with controlled relative humidity, temperature, ventilation rates and mixing conditions, and filtered supply air [208].

The fifth and final method uses small scale chambers. Géhin *et al.* [209] utilised a hexagonal test chamber, volume $2.36 \pm 0.05 m^3$, first designed for vacuum cleaner efficiency characterisation, to investigate fine and ultrafine particle emission rates for cooking and non-cooking sources. The control measures used in large chamber tests were used plus an antistatic coating on internal surfaces and an upward current

of filtered air to reduce sink effects. Torkmahalleh *et al.* [202] measured emissions from heating cooking oils in a laboratory fume hood operating at 65 m^3/h (80 h^{-1}), with 5 repetitions of each test. Mixing conditions were tested using SF_6 and a grid with points located 10 cm apart. The small volume meant the air was very well mixed, but the method of heating the cooking oils in a beaker on a hot plate was far abstracted from real cooking methods, and so the resulting emissions may not reflect those found in a domestic kitchen. This may also be true for the emissions from Géhin *et al.* [209] where the forced airflow conditions may have impacted the emissions from the combustion source.

2.5.4 Calculation Methods

Measurements of $PM_{2.5}$ concentrations, or indeed any pollutant concentration, can be used to compute pollutant source strength and emission rates if other conditions are known. Multiple calculation methods are used in literature, but most are based on the mass balance principle introduced in Section 2.4.3. Existing emission rate estimation models each rely on different assumptions about the source emission characteristics and chamber conditions, and frequently use an equation similar to Equation 2.6, repeated here (Equation 2.8) for ease.

$$C(t) = \frac{\lambda P C_a}{\phi} + \frac{(1-\eta)g}{\phi V} + \left(C(0) - \frac{\lambda P C_a}{\phi} - \frac{(1-\eta)g}{\phi V} \right) e^{-\phi t} \tag{2.8}$$

$C(t)$	Pollutant concentration at time t	[M/L^3]
$C(0)$	Initial concentration	[M/L^3]
C_a	Ambient concentration	[M/L^3]
P	Penetration coefficient	[-]
V	Volume	[L^3]
g	Emission rate, considered constant	[M/T]
η	Cooker hood capture efficiency	[-]
t	time	[T]
ϕ	Total decay rate, often split into:	[T^{-1}]
λ	Decay due to ventilation and	[T^{-1}]
k	Decay due to all other mechanisms	[T^{-1}]
	(L length, M mass, T time)	

This equation is simplified to suit the measurement conditions. Firstly, the penetration coefficient, P, is often unknown and so the background concentration C_b is used where $C_b = \frac{\lambda_T P C_a}{\phi}$ and can be obtained by measuring concentrations in the space in the absence of internal sources [35, 40]. Secondly, during emission rate measurements, a cooker hood is not normally used and so the capture efficiency, η is presumed 0. Therefore, Equation 2.6 simplifies to Equation 2.9.

$$C(t) = C_b + \frac{g}{\phi V} + \left(C(0) - C_b - \frac{g}{\phi V} \right) e^{-\phi t} \tag{2.9}$$

2.5.4.1 Steady State

The simplest method, as used by Torkmahalleh *et al.* [202, 205], is the steady state approach. Under constant conditions, as t becomes very large relative to $1/\phi$, the pollutant concentration tends towards an asymptote with the value of the steady state concentration, C_{ss}, see Figure 2.3. Under these conditions, the time dependent component of Equation 2.9 tends to zero, and so the steady state concentration is given by Equation 2.10.

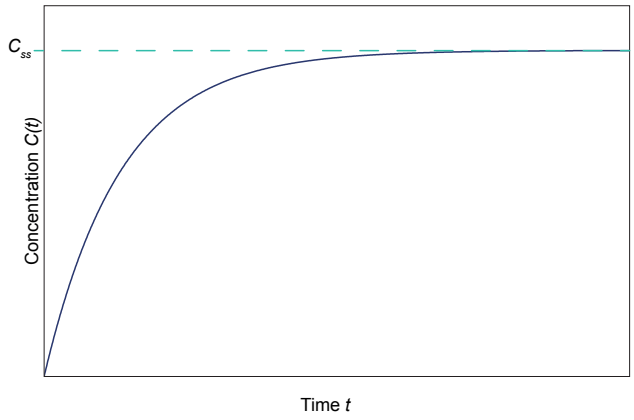


FIGURE 2.3: Steady state concentration plot

$$C_{ss} = C_b + \frac{g}{\phi V} \tag{2.10}$$

Torkmahalleh *et al.* [202, 205] used a small chamber volume, to reduce the time taken for the chamber concentration, C_{in}, to reach steady state, and assumed $C_b = 0$ so that g can be found using Equation 2.11.

$$g = \phi V C_{in} \tag{2.11}$$

This calculation method is simple, however it only works for constant sources and where ϕV is sufficiently small that near steady state conditions will be met. The required measurement conditions may affect the observed emissions, and so this is not the optimal method for $PM_{2.5}$ emissions.

2.5.4.2 Peak Estimation

For a source which emits at a constant rate for a limited period and then ceases to emit, known as a rectangular source function, concentrations are unlikely to reach steady state, instead increasing during the emission period, duration T, at the end of which the concentration reaches a maximum value, $C(T)$ or C_p. After this point,

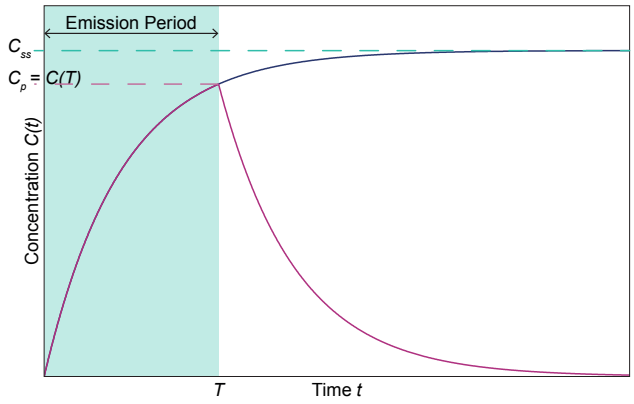


FIGURE 2.4: Rectangular source function plot

when the source stops emitting, concentrations begin to decay, see Figure 2.4. Therefore, from Equation 2.9, two equations can be determined, for the emission period and the decay period. This method is similar to those used by Dacunto *et al.* [40], Olson & Burke [199] and He *et al.* [35] amongst others.

First, during the decay period, $g = 0$, and $t \geq T$, so:

$$C(t) = C_b + (C_p - C_b)e^{-\phi(t-T)} \tag{2.12}$$

Ventilation rates can be measured however other factors such as deposition are more difficult to quantify. Therefore, when measuring emission rates, it is useful to first determine the constant decay rate using Equation 2.12 as follows:

$$\frac{C(t) - C_b}{C_p - C_b} = e^{-\phi(t-T)} \tag{2.13}$$

$$ln(C(t) - C_b) - ln(C_p - C_b) = -\phi(t - T) \tag{2.14}$$

$$ln(C(t) - C_b) = ln(C_p - C_b) - \phi(t - T) \tag{2.15}$$

Equation 2.15 is now a linear equation of form $y = mx + c$, where the gradient is the decay rate, ϕ and the y-intercept, the difference between the peak concentration and

the background concentration, $C_p - C_b$. With this method, a log-linear regression can determine the decay rate from monitored concentrations if: (i) the decay is constant or can be assumed constant; (ii) there are no emissions; and (iii) the background concentration is, or can be assumed, constant.

Once the decay rate has been determined, and with the known peak concentration, C_p, occurring at time, T, the duration of the emission period, from Equation 2.9 g can be found:

$$C_p = C_b + \frac{g}{\phi V} + \left(C(0) - C_b - \frac{g}{\phi V} \right) e^{-\phi T} \tag{2.16}$$

$$g = \phi V \left[\frac{C_p - C_b - (C(0) - C_b) e^{-\phi T}}{1 - e^{-\phi T}} \right] \tag{2.17}$$

This approach assumes full-mixing occurs instantaneously within the space, which is rarely observed. Instead, for cases of non-instantaneous mixing, Ott *et al.* [44] define three phases, the α-, β- and γ-period, to account for this delay in mixing. The α-period is the source emission period, during which the air is poorly mixed; the β-period, the time immediately following the end of emissions, while the air is still mixing; and the γ-period, once the air is fully mixed within the volume and concentrations begin to decay back down towards background levels [44]. Ott *et al.* [44] suggest that when the observed peak concentration occurs after the end of the α-period, it indicates the end of the β-period. From this time, the air should be well mixed and the decay smooth, as expected during the γ-period. The decay rate can be determined as before, using the γ-period, and is then projected back through the β-period to time $t = T$ to find the *theoretical peak* concentration. This method, or one like it, is used by Dacunto *et al.* [40] and Olson & Burke [199], and has many advantages, including: (i) the *theoretical peak* approach accommodates for instances where mixing is not instantaneous, an assumption of the mass balance model; (ii) it is not necessary to know the ventilation or decay rates, as the total decay rate can

be determined by monitoring the pollutant concentration decay; and (iii) it is established in the literature.

Of course, this method also has limitations, namely: (i) there may not be a constant emission rate and rectangular source function, as assumed by the model; (ii) the *theoretical peak* approach assumes further increases in concentrations after the end of the emission period are only due to the time taken for good mixing to occur, it does not consider that other factors, such as continued emissions, coagulation or chemical reactions between airborne pollutants, may cause this further increase; (iii) the decay must be first order exponential, that is ϕ is not a function of t; and (iv) the decay rate is assumed constant throughout the α-, β- and γ-periods.

2.5.4.3 Area-under-the-curve

This second method is an alternative, exact, method proposed by Ott *et al.* [44]. The decay rate can be determined using the monitored decay as for the *peak estimation* method. Ott *et al.* [44] start with the principles of the mass balance outlined in Section 2.4.3 Equation 2.5, which can be expressed as:

$$\int_0^T g(t)\mathrm{d}t - \int_0^T \phi V C(t)\mathrm{d}t = VC(T) \tag{2.18}$$

$g(t)$	emission rate as function of time	[M/T]
V	volume of interest	[L^3]
$C(t)$	concentration as function of time	[M/L^3]
t	time	[T]
T	duration of emissions	[T]
ϕ	total decay rate	[T^{-1}]
$C(T)$	concentration at end of emissions	[M/L^3]

Differentiating Equation 2.18 gives a first order differential equation to relate the change in concentration over time to the emission rate and the concentration at time t:

$$\frac{1}{\phi}\frac{dC(t)}{\mathrm{d}t} + C(t) = \frac{g(t)}{\phi V} \tag{2.19}$$

This method does not make assumptions about the function $g(t)$, however it does not determine this function [44]. Instead an average emission rate, $\overline{g(t)}$, is calculated. Averages are taken of all components of Equation 2.19, by dividing by averaging time T, and integrating between $t = 0$ and $t = T$ to give:

$$\frac{1}{\phi T}\int_0^T \frac{dC(t)}{\mathrm{d}t} + \frac{1}{T}\int_0^T C(t) = \frac{1}{\phi V T}\int_0^T g(t) \tag{2.20}$$

Where the average concentration, $\overline{C(T)}$, and the average emission rate $\overline{g(t)}$ are:

$$\overline{C(T)} = \frac{1}{T}\int_0^T C(t)\mathrm{d}t \tag{2.21}$$

$$\overline{g(T)} = \frac{1}{T}\int_0^T g(t)\mathrm{d}t \tag{2.22}$$

Assuming no background $PM_{2.5}$ such that the initial concentration, $C(0) = 0$:

$$T\overline{g(T)} - \phi V T\overline{C(T)} = VC(T) \tag{2.23}$$

Equation 2.23 has 3 components which directly correspond to the three components of the original mass balance Equation 2.5:

$$T\overline{g(T)} \leftrightarrow \text{(Mass generated between Time } t = 0 \textit{ and } t = T\text{)}$$

$$\phi VT\overline{C(T)} \leftrightarrow \text{(Mass lost between Time } t = 0 \textit{ and } t = T\text{)}$$

$$VC(T) \leftrightarrow \text{(Mass inside chamber at Time } t = T\text{)}$$

Finally, Equation 2.23 can be rearranged to find the average emission rate, $\overline{g(T)}$, using only: the decay rate, ϕ, chamber volume, V, average concentration, $\overline{C(T)}$ and final concentration, $C(T)$, at the end of the emission period, duration T using Equation 2.24.

$$\overline{g(T)} = \phi V\overline{C(T)} + \frac{VC(T)}{T} \tag{2.24}$$

This second method has advantages: (i) it is an exact approach; (ii) it does not assume a constant emission rate, instead determines the average emission rate over the α-period; (iii) by taking the average concentration over the α-period, the concentrations throughout this period are considered, not just the final (assumed peak) concentration; and (iv) the resulting Equation 2.24 is simpler than Equation 2.17, and all the terms can be obtained from experimental data.

Given these advantages, it is perhaps surprising that this method is not more widely used. For many $PM_{2.5}$ sources it is reasonable to assume a constant emission rate. Additionally this method may be less popular due to limitations including as it assumes instantaneous full-mixing, and, unlike the *theoretical peak estimation* approach, there is no means to accommodate for when this does not occur. Additionally, Equation 2.24 assumes C_b and $C(0) = 0$, although it could be adapted if this is not the case.

2.5.4.4 Source Strength

Finally, in modelling, emissions are most frequently characterised as the rate of mass emitted over time, to use in conjunction with activity schedules. Sometimes it is useful to consider the total mass emitted, otherwise known as the *source strength*, G, when comparing emissions from sources of different duration. If the $C(t)$, is a function of the emission rate, $g(t)$ and time, t, integrating this function over the emission period from $t = 0$ to $t = T$ gives the *area-under-the-curve*. This area is equivalent to the total mass emitted over this time. The *area-under-the-curve* method derived in the previous section is based on this principle, therefore the source strength is simply the product of the average emission rate, $\overline{g(T)}$ and the duration of the α-period:

$$G = \overline{g(T)}T \tag{2.25}$$

Or for constant emission rate, g:

$$G = gT \tag{2.26}$$

2.5.4.5 Comparison of Methods

Figure 2.5 shows a comparison of the three calculation methods. The orange line in each plot is the measured concentration profile, whereas the blue line represents a predicted emission and decay curve for the constant emission rate g, or average emission rate $\overline{g(t)}$, predicted in each case. This demonstrates how, for this source, both the *peak* and *theoretical peak* estimation methods could overestimate the source strength, as the emission rate appears non constant. As the emissions continue to increase after the end of the α-period, full-mixing is not instant, which would increase the error associated with the *area-under-the-curve* method.

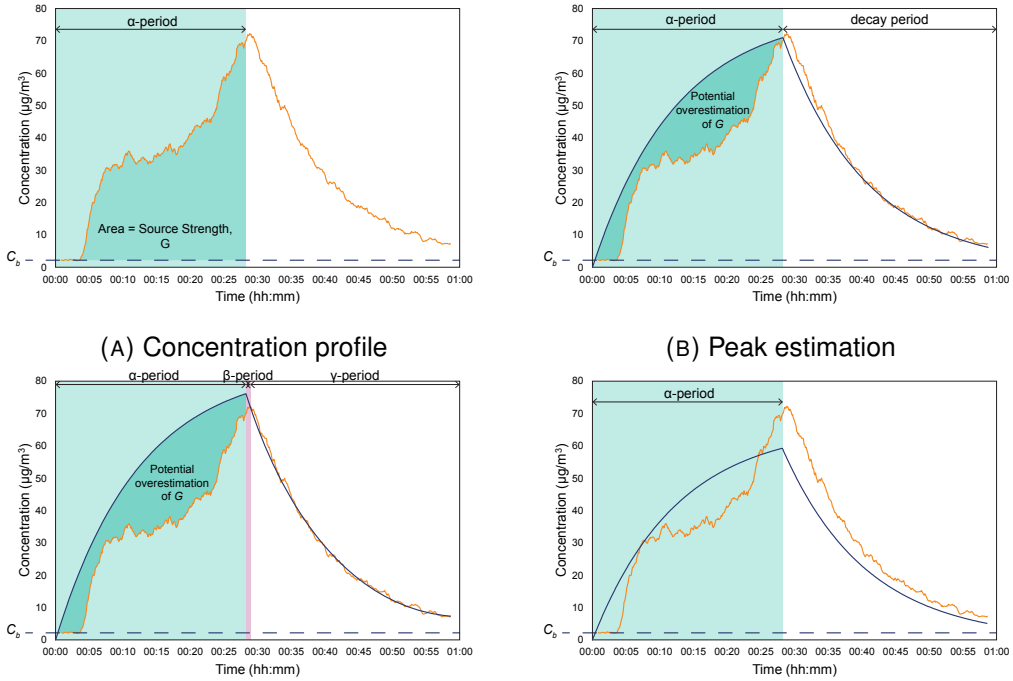


FIGURE 2.5: Comparison of emission rate and source strength calculation methods

2.5.5 Summary

In summary, cooking $PM_{2.5}$ emission rates in the literature vary widely, with some evidence that factors including cooking style, oil type, food type, and temperature might influence these cooking emissions. The objectives to measure emission rates suitable for use in probabilistic modelling and to measure the effect of equipment and ingredients on $PM_{2.5}$ emissions were identified due to the variation observed in literature and as models have previously been found to be sensitive to emission rates.

Multiple measurement methods have been used to estimate $PM_{2.5}$ emission rates, from personal monitoring to controlled tests in a small test chamber, hence the objective to develop an appropriate test method. Of these, controlled tests either conducted in situ or in a full scale chamber allow the most repeatable test methods. However, given the variability of cooking behaviour, a very large number of tests

may be needed to produce representative cooking emission rates.

Additionally, multiple calculation methods have been established based on the mass balance model. The peak estimation approach and area-under-the-curve method both have advantages. The choice of which method is most appropriate in any scenario may depend on the emission characteristics of the cooking source.

2.6 Summary

The aims and objectives listed in Section 1.3 were identified through the consideration of the existing literature. In addition, the current knowledge of health effects associated with $PM_{2.5}$ exposure were considered to create the basis for investigating $PM_{2.5}$ behaviour in buildings.

Firstly, the epidemiological and medical literature explored in Section 2.1 shows there is established evidence of relationships between short and long term exposure to $PM_{2.5}$ and multiple morbidities and mortality. This is the basis for considering exposure to $PM_{2.5}$ in dwellings where people spend most time. However, there is some evidence of heterogeneity in the observed health effects, which may related to differing $PM_{2.5}$ composition. This area of research is less well established and warrants further research, especially with regards the toxicology of $PM_{2.5}$ from indoor sources, however it falls outside the cope of this investigation.

The WHO set guideline daily and annual mean $PM_{2.5}$ concentrations, which they set for both indoor and outdoor $PM_{2.5}$, despite no evidence of a lower effect threshold in the literature. As a result, the WHO guidelines were identified as an appropriate metric within the context of this work, as they are the only existing guidelines based purely on minimising risk to health.

Existing literature on indoor domestic $PM_{2.5}$ concentrations, primarily in locations similar to the UK, examined in Section 2.2, showed evidence that indoor $PM_{2.5}$ concentrations might be elevated compared to outdoors. This is especially true for

environments with significant indoor sources such as cooking. However, there is limited concentration data measured in the English housing stock, with the most recent data published in 2013 [165]. Therefore, a primary objective of this thesis was identified to measure $PM_{2.5}$ concentrations in domestic kitchens, to add to existing data and to investigate the effect of cooking on temporal $PM_{2.5}$ concentrations.

Section 2.2 also identified challenges faced when measuring $PM_{2.5}$, which should be considered in experimental design. Gravimetric sampling techniques offer the most reliable test method, as mass collected on the filter is used to directly obtain mass concentrations. However, this method does not provide time resolved concentration data. Alternatively, multiple optical monitors established in literature can be used to infer temporal mass concentrations, but they are limited as they require custom calibration from concurrent gravimetric sampling.

Of the multiple mitigation methods identified in Section 2.3, source control was identified as preferable, and ventilation is frequently used to dilute concentrations of emitted pollutants. For cooking source pollutants, complete source removal is not feasible, instead targeted kitchen ventilation can be used to remove emitted pollutants at source, before they mix with room air. One such targeted ventilation device is a cooker hood. Current English building regulations specify ventilation rates for kitchens set to control relative humidity. ADF also specifies reduced ventilation rates when extracting air close to the source, such as through a cooker hood, although they do not specify a capture efficiency. Therefore, the further project objectives were identified (i) to test the ability to reduce $PM_{2.5}$ concentrations in domestic kitchens using existing ventilation mechanisms; (ii) to measure the reduction potential of a cooker hood; and (iii) to investigate the effect of current ADF requirements by predicting $PM_{2.5}$ concentrations in the English housing stock.

Stock scale indoor air quality models were introduced in Section 2.4 as an alternative to time consuming and costly measurement campaigns. Previous

models have used probabilistic or deterministic techniques coupled with mathematical modelling or simulation software to predict indoor pollutant concentrations for entire stocks of dwellings. A simplified mathematical steady state model was identified to adapt to approach the objective to synthesise probability density functions of $PM_{2.5}$ concentrations in English domestic kitchens and investigate the current ADF requirements.

Existing models have been found to be sensitive to emission rates. Accordingly, literature on cooking source $PM_{2.5}$ emission rates was examined in Section 2.5. Measured emission rates from cooking were found to vary and factors that might affect emission rates were identified to include food type and ingredients, fuel type, and cooking method and temperature. However, multiple measurement methods have been used, frequently with small sample sizes from which it is difficult to establish the distribution of emission rates suitable for probabilistic modelling. For this reason, the objective was identified to develop a method to measure emission rates from cooking sources suitable for use in statistical modelling. Furthermore, few studies included cooking full meals or the effect of equipment and ingredients on their emissions, hence the final objectives to measure $PM_{2.5}$ emission rates for cooking full meals and the effect of equipment and ingredients.

Chapter 3

Kitchen Intervention Test: $PM_{2.5}$ Concentrations and Reductions Measured in Domestic Kitchens

Work from this chapter was previously presented as a long presentation and paper at the AIVC conference 2018 in:

O'Leary, C. *et al. An intervention study of $PM_{2.5}$ concentrations measured in domestic kitchens* in *39th Air Infiltration and Ventilation Centre Conference* (Antibes Juan-Les-Pins, France, 2018), 1063–1072

3.1 Introduction

Cooking has been previously identified as a key indoor source of $PM_{2.5}$, see Section 2.2.2 for more details, therefore this study focuses on domestic kitchens. This study aimed to investigate $PM_{2.5}$ concentrations in domestic kitchens, to investigate whether there might be potential for high $PM_{2.5}$ concentrations in domestic kitchens, and to assess the degree to which existing ventilation strategies can be used to mitigate these concentrations. The aims were achieved using a two

TABLE 3.1: Dwelling and kitchen details

House	A	B	C	D	E
Dwelling Type	Semi-D	Semi-D	Semi-D	Mid Terrace	Semi-D
Total Occupants	6	4	3	3	6
Kitchen	Open	Enclosed*	Open	Open	Enclosed*
Kitchen Volume	77 m^3	24 m^3	21 m^3	41 m^3	27 m^3
Stove type	Gas	Gas	Gas	Electric	Gas
Oven Type	Electric	Gas	Electric	Electric	Electric
Week 1 Start	20 Oct 2016	12 Feb 2018	15 Oct 2018	03 Oct 2018	22 Oct 2018
Week 2 Start	01 Dec 2016	14 Mar 2018	24 Oct 2018	11 Oct 2018	11 Nov 2018

Semi-D = Semi-detached; *Internal doors normally left open

week intervention test to investigate: (i) what $PM_{2.5}$ concentrations might be present in residential kitchens and (ii) the extent to which these concentrations can be reduced by encouraging occupants to use existing ventilation mechanisms.

3.2 Method

Five multiple occupancy, student dwellings were selected to participate in this study. All households were non-smoking, both conventional and electronic-cigarettes (e-cigarettes), to eliminate tobacco smoke or e-cigarettes as a confounding $PM_{2.5}$ source and to focus on cooking source $PM_{2.5}$. Student houses were used due to their availability and because there are often of poor quality so might represent a worst case scenario. One student from each house aided the data collection process and also used the data within their own undergraduate dissertation. This student was identified as *lead occupant*, who took responsibility for communicating information to the remaining occupants. Additional houses were included in the initial data set, but later rejected for reasons including cigarette smoking indoors, e-cigarette use, and the cooking log not being returned. All data was collected in the heating seasons between October 2016 and November 2018, see Table 3.1 for details of the measurement periods.

Full details of all five houses are given in Table 3.1. The occupants of student houses can be in fuel poverty [211], and so occupants may be less inclined to use ventilation methods in an attempt to conserve heat. This represents a worst case behaviour

scenario. Furthermore, at least Houses A, B and E were expected to be leaky as they were built before 1930 [212], which represents a best case infiltration scenario.

The cooking facilities varied between houses, as seen in Table 3.1. In addition to an electric or gas stove-top and oven, student kitchens would be expected to have a toaster, kettle and microwave. Example photos of all appliances in House E can be found in Appendix C.1. UK ovens typically have a grill option, where food is cooked using only radiant heat from the upper heating element at a high temperature, therefore *grill* or *grilling* refers to this cooking process.

3.2.1 Equipment

A SidePak™ AM510 Personal Aerosol Monitor (SidePak) (TSI Inc., Shoreview, MN, USA), fitted with a 2.5 µm impactor, was used to monitor $PM_{2.5}$ concentrations. The SidePak is a photometer that infers PM mass concentration through light scattering and requires custom calibration [139]. More details were given in Section 2.2.1. Concentrations were time averaged and logged at 1 minute intervals because the SidePak has a limited storage capacity. The calibration factor (CF) was set to the default 1.0, as no concurrent gravimetric sampling was available, therefore all reported mass concentrations are approximations. In addition, an IAQ-Calc Indoor Air Quality Meter (IAQ-Calc) (Model 7545, TSI Inc., Shoreview, MN, USA) monitored indoor temperature, CO_2 and CO concentrations, and relative humidity (RH). RH was the parameter of most interest from the IAQ-Calc. As discussed in Section 2.2.1, high RH may affect the $PM_{2.5}$ mass measurements recorded by the SidePak [40, 146]. RH was monitored to identify periods of high humidity. In addition, an HOBO data logger (HOBO) (U10-001, ONSE, Cape Cod, MA, USA) was used to monitor external temperature at the location. It was set to log temperatures at 1 minute intervals, and can run for up to 28 days, so in most cases was left in location between the two weeks of measurements.

The SidePak and IAQ-Calc were issued to the lead occupant for each dwelling, to be positioned together, in the kitchen, out of direct sunlight and away from heat sources. Lead occupants were offered specific advice on where to locate the equipment on request. The aim was to measure $PM_{2.5}$ concentrations in mixed air, and therefore the equipment was positioned away from sources of $PM_{2.5}$ and water vapour, such as the stove and the kettle. The exact location varied between dwellings, and was restricted by the need for both devices to be connected to a power supply. In houses with an open plan kitchen (House A, C and D) the measurement location was within the kitchen area of the room. The HOBO was located in a secure location outside the property, out of direct sunlight, preferably on the north façade of the building, to measure the dry-bulb temperature without any radiant effect from the sun. Lead occupants were advised to place the HOBO in a secure container to protect it from rain.

3.2.2 Monitoring

The lead occupant for each household was briefed on the aims of the study and instructed how to use the equipment and how to brief the other occupants. During Week 1, only this occupant was aware that $PM_{2.5}$ concentrations related to cooking were being investigated. All occupants were aware that measurements were being taken in the kitchen, and were asked to record cooking behaviour in a household cooking log. This included a record of cooking start and end times, and notes describing the cooking activity taking place. A blank example cooking log is included in Appendix C.2. The lead occupant was asked not to mention ventilation use during Week 1, in order to measure *typical* conditions.

At the end of Week 1, the lead occupant returned the SidePak and IAQ-Calc for the data to be downloaded. Plots of $PM_{2.5}$ concentration over the week were shared with the lead occupant, and the initial findings discussed. They were given an overview of what the measured concentrations might mean and verbal instructions

on how to direct the other occupants before commencing Week 2. A follow up email was shared with the lead occupant upon request, which contained a summary of these instructions. A sample email and plot are included in Appendix C.3. Occupants were informed of the health risked associated with exposure to elevated $PM_{2.5}$ concentrations and shown the concentrations from Week 1 with reference to the WHO daily and annual mean guidelines [128]. They were then informed that cooking is a known indoor source of $PM_{2.5}$, with reference to peak concentrations within the Week 1 data which corresponded to cooking events, and given suggestions of how they might use ventilation to improve the indoor air quality. Ventilation methods were suggested, including opening windows or external doors, or using an extract fan, both during or after cooking, but no specific direction or indication of preference was given. The lead occupant was provided with a new cooking log for Week 2, with an additional column to tick when different ventilation strategies were used during a cooking event. An example blank log is included in Appendix C.2.

After the end of Week 2, the lead occupant was asked to investigate any mechanical ventilation system present in the kitchen, to help identify whether the cooker hood extracted or recirculated air. They were also asked to provide the kitchen information presented in Table 3.1. This investigation was conducted after measurements so the intervention would approximate the effect of a public health message to encourage non-expert members of the public to use more ventilation during cooking.

3.2.3 Statistical Analyses

The monitoring data was collated and analysed using bespoke MATLAB [213] code[1]. Each monitoring period was truncated to include only the first 7 full days, missing $PM_{2.5}$ data points were removed, and data points corresponding to

[1]The code is available under a creative commons license from DOI: 10.13140/RG.2.2.27857.48483

RH $>$ 70% were identified. Further analysis was conducted twice, both including and excluding these high RH data points, to assess whether the confounding effect of RH might impact the results of the investigation. To investigate the effect of cooking on $PM_{2.5}$, concentrations were split into cooking and non-cooking periods using the occupant recorded cooking event times.

A series of statistical tests were conducted on the $PM_{2.5}$ data sets for each house to investigate whether concentrations were significantly different in the Week 2 to Week 1. The five houses were considered separately, as five individual case studies, rather than a single larger study. Initial investigations into the concentration distributions suggested that they were not normally distributed, and therefore non-parametric tests would likely be required. Visual inspection of Quantile-Quantile (Q-Q) plots comparing the log-transformed concentrations to a normal distribution were used to check for log-normality. The full details of these are included in the results, however they indicated that this transformation would not allow for subsequent parametric tests.

The non-parametric Mann-Whitney U or Wilcoxon's Rank-sum test (MW test) was selected to test whether the data in Week 1 and Week 2 was from continuous distributions with equal medians [214]. This test is appropriate as the concentration distribution is unknown. When the MW test indicated a statistically significant difference ($p \leq 0.05$), the *effect size* (r) was calculated using Equation 3.1, where z is the MW z-score and N is the total number of data points [214]. The effect size is a measure of the size of the difference between two samples [215], and is an assessment of how much change may be expected from implementing the intervention elsewhere. An effect size < 0.3 corresponds to a small effect, $0.3 - 0.5$ a medium effect, and > 0.5 a large effect [214].

$$r = \frac{z}{\sqrt{N}} \tag{3.1}$$

The MW test assumes the data sets are independent. In this case a change has been applied, therefore a dependent test might appear more appropriate. However, there are a few reasons why this is not true. Firstly, for a true paired test, the changes would have to be applied directly to Week 1, instead they are applied in a subsequent week. Additionally, because the houses are considered as a series of case studies rather than a single test, the aim was to compare all the time-resolved concentrations measured during Week 1 to those from Week 2, for each house in turn, and draw a separate conclusion for each house. A dependent test would consider a single test statistic for each house in Week 1 and 2, and compare the two resulting data sets. Finally, statistical tests for dependent data sets require paired values, but given the variability of cooking behaviour and additional factors such as weather conditions, there is no logical way to pair the temporal concentrations in Week 1 and 2. For these reasons the Week 1 and Week 2 data sets were considered independent.

The MW test was also used to investigate whether $PM_{2.5}$ concentrations were significantly higher during cooking periods over the first week of monitoring. This was achieved by comparing all concentrations measured during recorded cooking periods to those measured at other times, for each house in turn.

3.2.3.1 Comparison to Guidelines

To set the measured concentrations into a wider context, and give some indication of the potential associated risk, it is useful to compare them to existing guidelines. This comparison is not simple. The seven day monitoring period does not correspond to either averaging time for the WHO $PM_{2.5}$ guidelines. Therefore two comparisons were made. Firstly, the mean over 7 days was compared to the WHO annual guideline $10\,\mu g/m^3$. Secondly, the data was parsed into 7 consecutive 24 hour periods, and the mean concentration for each of these is compared to the WHO daily mean guideline $25\,\mu g/m^3$.

However, the raw data was measured with CF $= 1.0$. As discussed in Section 2.2.1 a custom CF from concurrent gravimetric sampling is required for the SidePak to provide accurate mass concentrations [139]. Based on published SidePak CFs for $PM_{2.5}$ from a variety of cooking sources listed in Table 2.6, it is likely that the calibration factor is in the range $0.41 - 0.70$ [40, 138]. The raw SidePak concentrations likely overestimate the true mass values. Therefore, these CFs were applied to the weekly and daily mean concentrations, to estimate a concentration range for each house. The comparison to guidelines was then repeated to investigate the possible effect of the CF.

3.2.3.2 Other Parameters

Other IEQ parameters were also investigated to explore the overall environmental quality within each kitchen, and to assess how this changed over the two weeks. One purpose of this further analysis was to check if there is any indication that any of the potential behavioural changes would be sustained past the end of the study. The distribution of CO_2 concentration, RH and internal temperature (t_i) were plotted, as was the external temperature (t_{ext}) to allow comparison with t_i. Here, comparisons were made by visual inspection only, as these parameters were not the primary focus of the investigation.

A rudimentary investigation of the frequency and duration of cooking events and ventilation use as reported in the cooking logs was also conducted. Cooking duration, frequency and type could all effect the kitchen $PM_{2.5}$ concentrations, and therefore are potential confounding factors. The frequency of ventilation use during cooking might also indicate behavioural change as a result of the intervention.

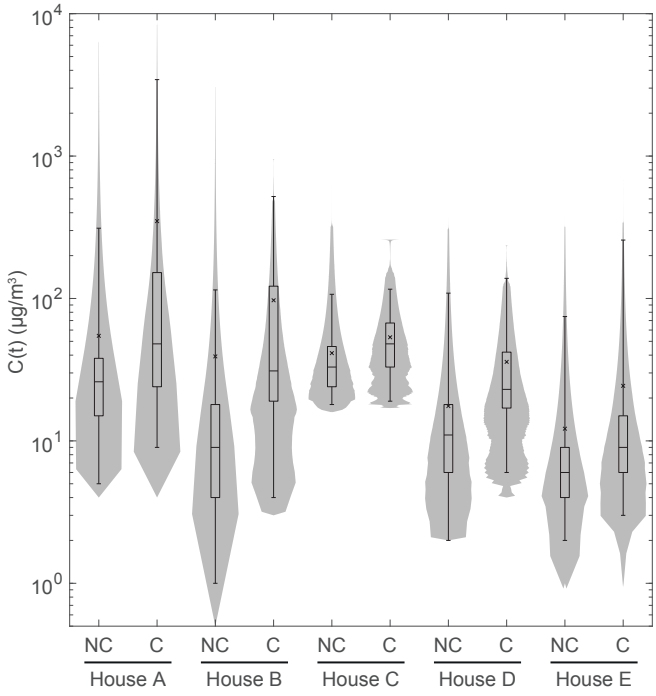


FIGURE 3.1: Week 1 $PM_{2.5}$ concentration violin plot during (C) and outside of (NC) recorded cooking events (CF $= 1.0$)

3.3 Results

3.3.1 Influence of Cooking on Concentrations

Figure 3.1 and Table 3.2 show the Week 1 $PM_{2.5}$ distributions during and outside of recorded cooking periods. The shaded *violins* in Figure 3.1 show the shape of the probability density function. The overlaid boxes show the lower quartile (25th percentile), median and upper quartile (75th percentile) of recorded concentrations, and the whiskers the 2nd and 98th percentiles. In addition, the arithmetic mean concentration for each week is marked with an $\times$. These distributions show that concentrations are higher during cooking periods, and the MW test indicates a statistically significant *small* effect in all houses (Table 3.2). As shown in Table 3.2, the MW p–values were very small in all houses, possibly due to the large number of data points in the test samples. Here n ranged between 165 and 9916.

TABLE 3.2: Week 1 $PM_{2.5}$ concentration summary statistics outside of (NC) and during (C) recorded cooking events (CF $= 1.0$) ($\mu g/m^3$)

Centile/	House A		House B		House C		House D		House E	
Statistic	NC	C	NC	C	NC	C	NC	C	NC	C
2%	5	9	1	4	18	19	2	6	2	3
25%	15	24	4	19	24	33	6	17	4	7
50%	26	48	9	31	33	48	11	23	6	12
75%	38	152	18	122	46	67.25	18	42	9	19
98%	312	3443	115	520	107	116	109	139	70	510
μ	54.7	349	39.2	97.1	41.3	53.4	17.6	35.9	12.0	36.8
σ	293	1169	290	145	39.2	31.9	26.1	33.5	35.3	92.3
GM*	25.8	73.9	9.36	42.3	35.1	47.0	11.2	25.7	6.72	14.0
GSD*	2.46	4.50	3.05	3.60	1.64	1.64	2.38	2.24	2.23	2.98
$n^{\dagger}$	9547	534	9722	352	9916	165	9481	600	8483	1598
MW p	2.5×10^{-62}		2.6×10^{-97}		9.7×10^{-16}		1.8×10^{-109}		2.5×10^{-116}	
MW r	-0.17		-0.21		-0.08		-0.22		-0.23	
MW effect	small		small		small		small		small	

*Calculated with any $0\,\mu g/m^3$ values removed; † total number of data points

Figure 3.2 shows a sample plot of concentrations from House A Week 1. The concentrations corresponding to cooking events recorded in the household cooking log are highlighted in orange. Most concentration peaks are within these cooking periods, indicating cooking does correspond with elevations in $PM_{2.5}$ concentration. A similar pattern occurred in the other houses, see Appendix C.4. Elevated concentrations outside of the recorded cooking events suggests there were additional $PM_{2.5}$ sources within the kitchen, either unrecorded cooking activity or other sources.

3.3.2 Intervention Impact on $PM_{2.5}$ Concentrations

Table 3.3 and Figure 3.3 show the key $PM_{2.5}$ statistics for Week 1 and Week 2 across all houses. These are for both cooking and non-cooking periods with high RH data points included. The shape of the distributions is similar for all houses except House C (see Figure 3.3) which shows a more substantial positive skew and less variation in concentration than the other houses.

Visual inspection of Figure 3.3 suggests $PM_{2.5}$ concentrations differed between the two weeks in all houses. In House A, the median and mean concentrations appear

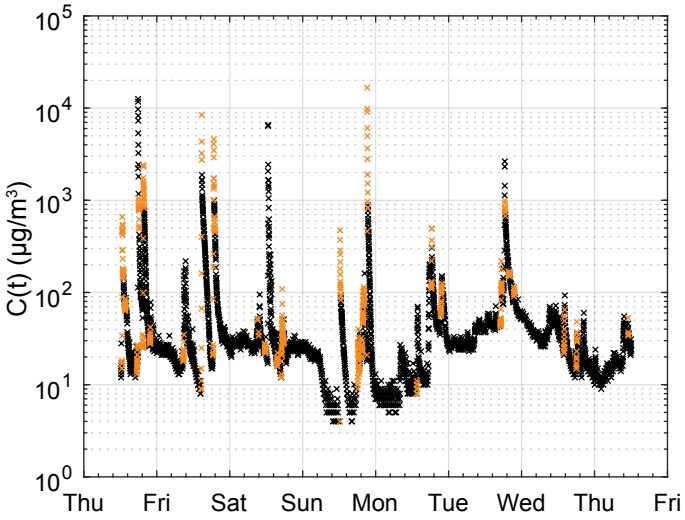


FIGURE 3.2: Temporal concentrations measured in House A **orange** points indicate concentrations during occupant recorded cooking periods

similar across both weeks, although there is more spread in Week 2. In House B, the mean is lower in Week 2, however the median is higher and there is less spread. $PM_{2.5}$ concentrations appeared lower during Week 2 in House C, where the median and the mean decreased. In House D, the median decreased, whereas the mean increased, which appears to be due to greater positive skew in Week 2. Finally, in House E, the overall spread is less in Week 2, but the interquartile range is greater, as are the mean and the median.

TABLE 3.3: Week 1 and Week 2 $PM_{2.5}$ concentrations, summary statistics (CF $= 1.0$) ($\mu g/m^3$)

Centile/	House A		House B		House C		House D		House E	
Statistic	W1	W2	W1	W2	W1	W2	W1	W2	W1	W2
2%	5	3	1	3	18	17	2	4	2	2
25%	16	15	5	7	24	22	6	6	5	5
50%	26	30	10	13	33	27	12	9	7	13
75%	40	54	19	19	47	34	20	15	10	25
98%	485	542	194	55	108	69	115	178	98	74
μ	70.3	73	41.2	28.6	41.5	30.5	18.6	27.8	14.1	19.5
σ	398	249	286	193	39.1	16.4	27.0	125.0	43.8	38.0
GM*	27.3	29.3	9.86	13.0	35.3	28.5	11.8	10.7	7.16	12.0
GSD*	2.64	3.12	3.17	2.33	1.65	1.41	2.42	2.54	2.36	2.60
$n^{\dagger}$	10081	10081	10074	10081	10081	10081	10081	10081	10081	10081
MW p	1.6×10^{-21}		1.2×10^{-108}		1.1×10^{-217}		6.4×10^{-47}		0	
MW r	-0.07		-0.16		0.22		0.10		-0.29	
MW effect	Small		Small		Small		Small		Small-Medium	

*Calculated with any 0 $\mu g/m^3$ values removed; † total number of data points

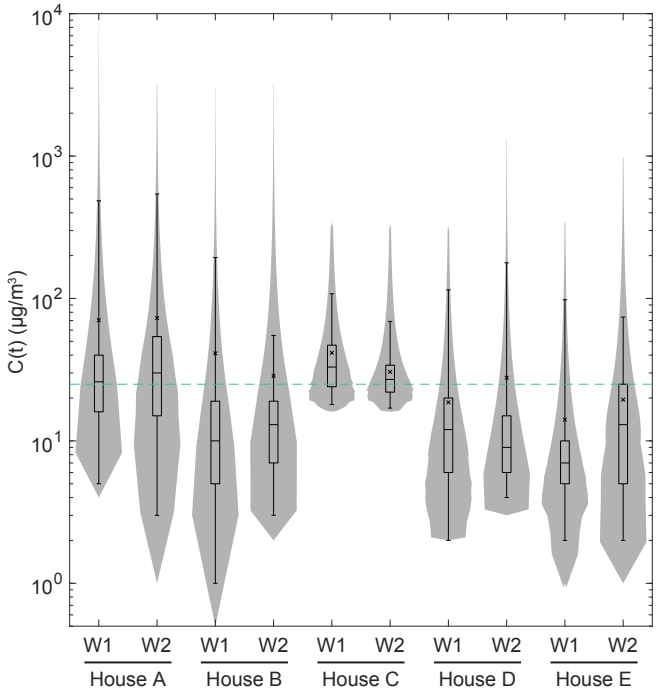
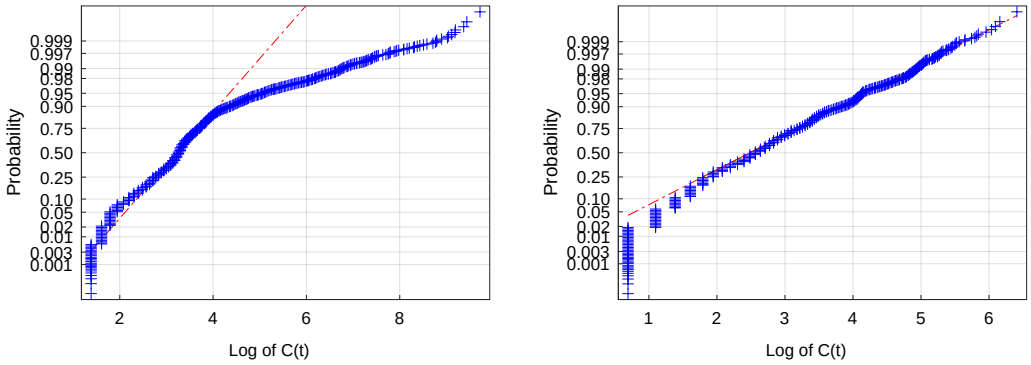


FIGURE 3.3: $PM_{2.5}$ concentrations in Week 1 and Week 2

Figure 3.4 contains two example Q-Q plots comparing the log-transformed concentration distributions to a normal distribution. The Q-Q plots for all other houses are contained in Appendix C.4. As can be seen in Figure 3.4, the distribution appears close to log-normal in some measurement weeks, but this was not universal. Therefore, a log-transformation would not consistently produce distributions suitable for use with parametric tests, and so this process was rejected in favour of non-parametric tests.

The MW test found significant changes in the median between weeks for all five houses ($p \leq 0.05$). The p–values and corresponding effect sizes suggest there was a *small* effect in all houses, see Table 3.3. In Table 3.3, a positive effect size indicates lower concentrations in Week 2. Therefore, whilst there was a statistically significant difference in concentrations between weeks across all houses, concentrations were only found to decrease in House C and House D. Again, p was very small in all five houses, likely due to the large sample sizes. Here n was $10074 - 10081$, see Table 3.3.



(A) House A Week 1 (B) House D Week 1

FIGURE 3.4: Sample Q-Q plots to check for log-normality of distributions

3.3.2.1 Effect of High Relative Humidity

Table 3.4 and Figure 3.5 shows the same statistics as Table 3.3 and Figure 3.3 but for the data sets where $PM_{2.5}$ concentrations corresponding with RH $>$ 70% removed. Here the results are largely the same, where the medians are significantly different Week 1 to Week 2 (MW test) and there was a small but significant reduction in House C and D, whilst the other houses experienced a small but significant increase. These results do not contradict those where high RH data points were included therefore RH does not appear to have significantly influenced the results of this study.

3.3.3 Comparison to Guidelines

Table 3.5 shows the overall mean concentration and total days where the mean concentration was less than the WHO daily mean guideline for each house and week. These results are presented for the original CF $=$ 1.0, and adjusted by CF $=$ 0.70 and CF $=$ 0.41. The overall mean concentration exceeded the WHO annual mean guideline in all houses and weeks when CF $=$ 1.0 or CF $=$ 0.70. When CF $=$ 0.41, the mean concentration was below the guideline in House D Week 1 and House E Weeks 1 and 2. Crucially for all three CFs, the average concentration did

TABLE 3.4: Week 1 and Week 2 $PM_{2.5}$ concentrations, summary statistics with points corresponding to RH $> 70\%$ removed (CF $= 1.0$) ($\mu g/m^3$)

Centile/	House A		House B		House C		House D		House E	
Statistic	W1	W2	W1	W2	W1	W2	W1	W2	W1	W2
2%	5	3	1	3	18	17	2	3	2	2
25%	15	15	5	7	24	22	6	5	5	5
50%	26	30	9	13	33	27	11	8	7	13
75%	40	54	18	19	47	34	19	12	9	24
98%	490	542	106	53	108	69	116	120	81	73
μ	70.6	72.8	26.8	28.4	41.5	30.5	18.1	26.8	12.6	19.2
σ	400	249	147	194	39.1	16.4	26.5	137.8	36.6	31.9
GM*	27.2	29.2	9.30	12.9	35.3	28.5	11.4	9.6	6.95	12.0
GSD*	2.65	3.12	2.93	2.32	1.65	1.41	2.41	2.44	2.27	2.59
%$>70\%$ RH	2.1	0.2	2.6	0.6	0.0	0.0	4.9	31	1.7	0.3
$n^{\ddagger}$	9957	10064	9818	10023	10081	10081	9605	7110	9912	10058
MW p	2.3×10^{-22}		1.9×10^{-132}		1.1×10^{-217}		2.3×10^{-73}		0	
MW r	-0.07		-0.17		0.22		0.14		-0.30	
MW effect	Small		Small		Small		Small		Medium	

*Calculated with any $0\,\mu g/m^3$ values removed; ‡ total number of data points

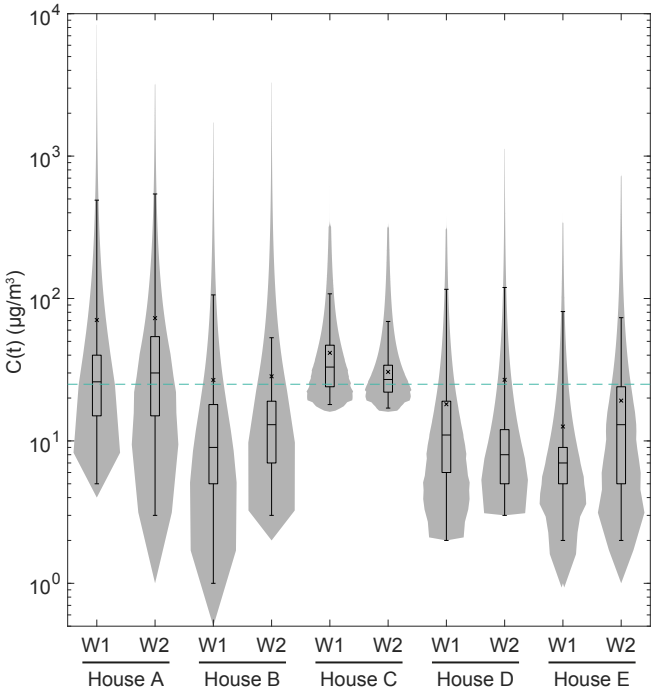


FIGURE 3.5: $PM_{2.5}$ concentrations in Week 1 and Week 2 with points corresponding to RH $> 70\%$ removed

TABLE 3.5: Overall mean concentrations ($\mu g/m^3$) and days below $25\,\mu g/m^3$

		House A		House B		House C		House D		House E	
	CF	W1	W2	W1	W2	W1	W2	W1	W2	W1	W2
Overall Mean	1.0	70	73	41	29	42	31	19	28	14	19
($\mu g/m^3$)	0.70	49	51	29	20	29	21	13	19	10	14
	0.41	29	30	17	12	17	13	8	11	6	8
RSD* (%)	-	566	341	695	674	94	54	145	450	311	195
RSD* in 24-hour mean (%)	-	51	62	166	86	42	5	42	112	55	58
Days below WHO-24	1.0	1	1	6	3	1	0	6	5	6	5
	0.70	1	1	6	6	3	7	7	6	7	6
	0.41	3	3	6	6	6	7	7	6	7	7

*relative standard deviation

not drop from above the guideline in Week 1 to below the guideline in Week 2 in any house.

The overall (7-day) mean concentrations also equal the overall mean of the 24-hour mean concentrations. However, the relative standard deviation (RSD) of 24-hour mean concentrations, included in Table 3.5, was substantially smaller than the overall RSD. This metric gives an indication of the day-to-day variation in $PM_{2.5}$ concentrations, and as it is relative, it is not affected by the CF. Some houses experienced large day-to-day variation, for example in House B Week 1 the RSD $= 166\%$. This may also indicate non-normal distributions but as the sample size was small ($N = 7$ days), it is difficult to draw definitive conclusions. Other houses were much more consistent day-to-day, especially House C Week 2 where RSD $= 5\%$.

At CF $= 1.0$, Houses A and C performed worst compared to the 24-hour guideline, where the concentrations were below the guideline on a maximum of 1 of 7 days, see Table 3.5. Adjusting the concentrations using a CF did improve the performance. For CF $= 0.70$, the improvements were noticeable in most houses, where Week 2 24-hour mean concentrations remained below the WHO guideline on at least 6 days in Houses B - E. However, even with the lower CF $= 0.41$, the 24-hour mean was below the guideline on just 3 days in each Week 1 and 2 in House A.

The CF also had an effect on the success rate of the intervention according to this metric. For example, in House C, when CF $= 1.0$, more days complied in Week 1

TABLE 3.6: Recorded cooking and ventilation behaviour

	Total Cooking Time (minutes)		No. Cooking Events			
			Total		With Ventilation	
House	W1	W2	W1	W2	W1	W2
A	554	525	34	28	7	21
B	401	697	30	47	5	26
C	205	158	27	18	2	13
D	673	457	34	35	2	22
E	1826*	1074	83	72	-	55

*Includes 1 overnight slow cooker event, reduces to 1096 minutes when removed

(1 day) than Week 2 (0 days), however the opposite was true with CF = 0.70 (3 vs 7 days) and CF = 0.41 (6 vs 7 days), see Table 3.5. This is because there were many near misses in Week 2 with the original CF.

3.3.4 Cooking and Ventilation Behaviour

The concentrations increased in Week 2 in 3 of the 5 dwellings. One reason might be differing occupant cooking patterns. Therefore, Table 3.6 outlines the number of cooking events and the total time spent cooking for each house and week.

Table 3.6 also indicates the total number of cooking events during which occupants recorded ventilation use during Week 2. Occupants were not instructed to record ventilation use in Week 1, however some ventilation use was indicated in all but House E (see Table 3.6). In these houses ventilation use increased substantially during Week 2. This indicates the intervention did modify behaviour even if it did not consistently reduce $PM_{2.5}$ concentrations, although it is possible that not all ventilation use was recorded in Week 1.

Figure 3.6 shows the total duration of cooking events in each house divided according to the cooking method used. Across all houses, oven cooking, boiling and multiple method cooking events contributed most to the total cooking time. It is also interesting to note that in House A, the total time spent cooking decreased in Week 2, but the time spent frying increased. This may have contributed to the increased $PM_{2.5}$ concentrations observed in Week 2. As this study involves a

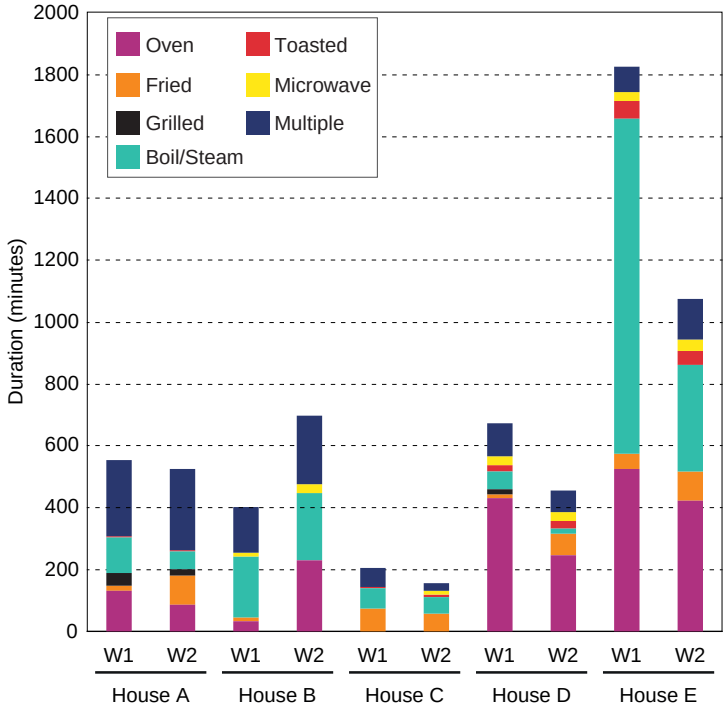
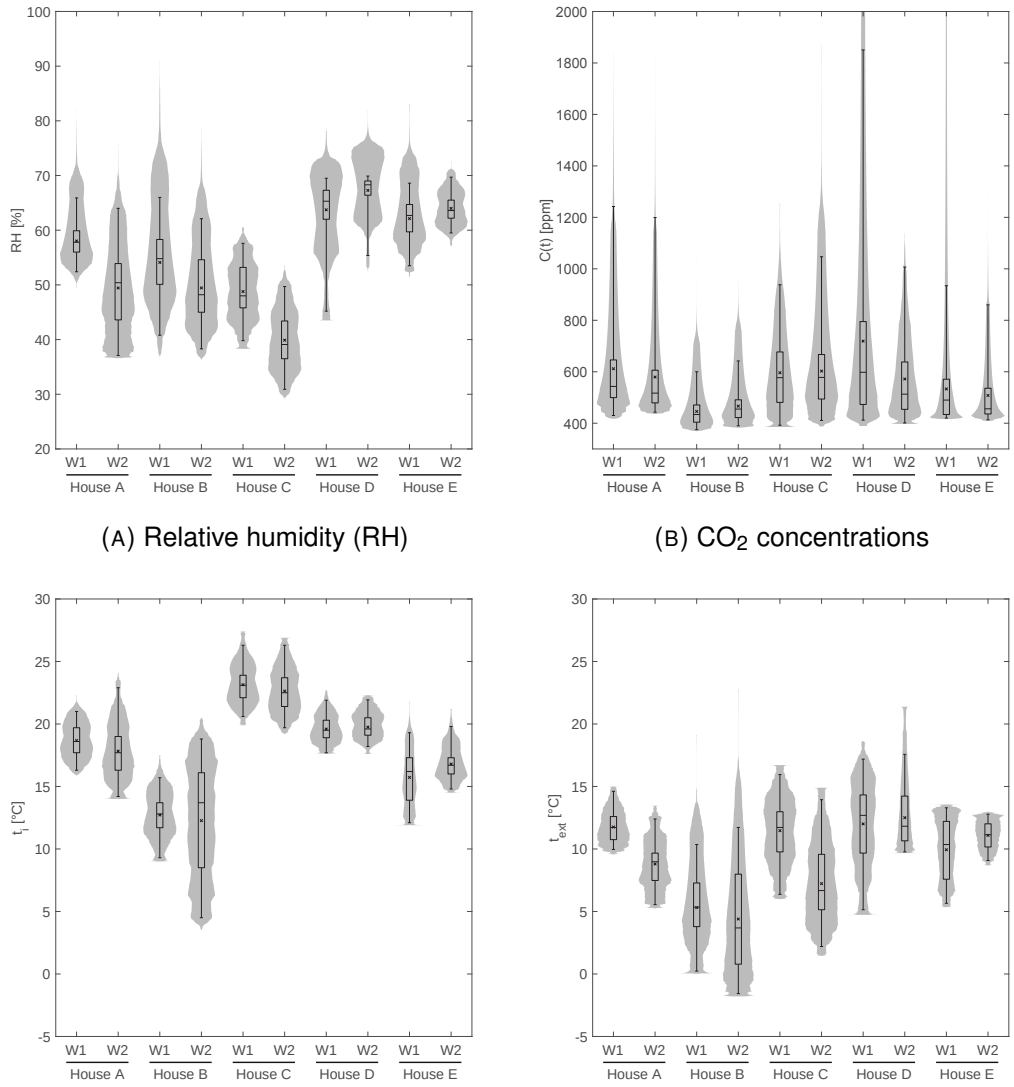


FIGURE 3.6: Cooking method duration

relatively small non-representative sample of five student houses, the relationship between cooking method and $PM_{2.5}$ concentrations was not explored in more detail.

3.3.5 Other Indoor Environmental Parameters

Parameters measured by the IAQ-Calc and HOBO data logger were not the primary target of this investigation, however a brief overview is presented here to give a broader picture of the indoor environment in each kitchen. Figure 3.7 shows the temperature, RH and CO_2 distributions. The external temperature (t_{ext}) is also included, despite not being a measure of the indoor environment, to allow comparison to the internal temperature (t_i). CO is not included as there was very little fluctuation in CO concentration.



(A) Relative humidity (RH)

(B) CO_2 concentrations

(C) Internal temperature

(D) External temperature

FIGURE 3.7: Distributions of other environmental parameters

The measured temperatures give an indication of heating use in each household. Here House C is something of an outlier. The internal temperatures were highest in this kitchen, despite similar t_{ext} values to those measured at other houses. The elevated t_i may also explain the lower RH also measured in this house. There was no clear and consistent effect of the intervention on t_i. In House B, t_i was more varied in Week 2, however the same was true for t_{ext}.

In Houses A, B and C, RHs were lower in Week 2 (see Fig 3.7a). This may indicate an unintended benefit of the intervention in some cases. However, RH increased in Houses D and E, so this effect was not universal. It should also be noted that the lead occupant from House D reported the presence of a bathroom immediately adjacent to the kitchen. They previously found this caused humidity and damp in the kitchen, especially after occupants used the shower. This is observed in the data presented in Figure 3.7a where the RH distribution appears bi-modal with a high frequency of RHs at or above 70%.

CO_2 was the only parameter, other than $PM_{2.5}$, which appeared closer to being log-normally distributed. Overall, CO_2 concentrations appear similar between weeks in each house, except for House D (see Fig 3.7b). Here, the concentrations were more consistent in Week 2, and there was a reduction in the mean concentration.

3.4 Discussion

The concentrations reported here are similar to those measured in previous studies. Stranger *et al.* [115] measured indoor $PM_{2.5}$ concentrations with medians $27.2\,\mu g/m^3$ and $41.1\,\mu g/m^3$ across two sets of dwellings in Antwerp, Belgium. Wallace *et al.* [145] measured concentrations of similar magnitudes in 36 North Carolina residences, with an overall mean of $29.8\,\mu g/m^3$ and a maximum average $56.8\,\mu g/m^3$ in non-smoking households. Similar mean concentrations, ranging from $1.6\,\mu g/m^3$ to $64.1\,\mu g/m^3$ were measured across 18 California apartments by Chan *et al.* [36]. These concentrations are similar in magnitude to the mean concentrations of $14.1\,\mu g/m^3$ to $72.9\,\mu g/m^3$ reported in Table 3.3, although overall the concentrations measured here may be slightly higher. Both Stranger *et al.* [115] and Wallace *et al.* [145] measured $PM_{2.5}$ concentrations using gravimetric sampling techniques, so reported true mass concentrations. Chan *et al.* [36] measured concentrations with a DustTrak photometer, and corrected the concentrations with an estimated CF $= 0.4$, so their concentrations are mass estimates. A common

feature of these studies that differs from this investigation is that the sampling location was not exclusively in the kitchen. When sampling in the kitchen, concentrations may be expected to be higher due to closer proximity to the source.

In a study of 15 domestic kitchens during autumn and winter in Brisbane, the overall mean concentrations were 15.5 $\mu g/m^3$ during activity and 11.1 $\mu g/m^3$ otherwise [216]. These concentrations are lower than those in Table 3.3, but many factors may contribute to this difference, including household and dwelling characteristics, activity types, and the SidePak CF. Additionally, these studies are not of UK houses, and dwelling characteristics and occupant behaviour may vary significantly between countries.

Two studies included in Table 2.8 reported $PM_{2.5}$ concentrations measured in UK domestic kitchens: Wigzell *et al.* [163] in Oxford and Nasir & Colbeck [165] in Colchester. First, Wigzell *et al.* [163] measured $PM_{2.5}$ concentrations in non-smoking kitchens. They reported 48-hour gravimetric concentrations $\mu = 12\,\mu g/m^3$, GM $= 11\,\mu g/m^3$ and GSD $= 1.5$ ($n = 9$). The GM and GSD are similar magnitude to some of those reported in Tables 3.3 and 3.4, though their μ is lower than those measured here. Nasir & Colbeck [165] measured 24-hour kitchen $PM_{2.5}$ concentrations in student dwellings ($N = 4$), one-bedroom flats ($N = 1$) and 2-3 bedroom houses ($N = 2$) in using custom calibrated OPCs. They reported $\mu = 56 \pm 40\,\mu g/m^3$ ($n = 50$), $\mu = 46 \pm 33\,\mu g/m^3$ ($n = 14$) and $\mu = 10 \pm 7\,\mu g/m^3$ ($n = 16$) respectively. Their mean concentrations are of similar magnitude to those for Week 1 in House B (41.2 $\mu g/m^3$), House C (41.5 $\mu g/m^3$), House D (18.6 $\mu g/m^3$) and House E (14.1 $\mu g/m^3$). However, it is not entirely appropriate to make direct comparisons to either study due to the differing averaging times and the absence of a custom CF in the current work.

Concentrations exceeded the WHO 24-hour mean guideline on 0% - 100% of days, assuming CFs measured for similar cooking sources elsewhere are applicable to this study. Concurrent gravimetric sampling would allow better analysis in

comparison to guidelines, as the mean concentration for each filter period would be a true mass concentration. Ideally future work should include a gravimetric sampling procedure with at least 1 filter per 24-hour period to allow the mass values to be directly compared to the WHO daily mean guideline without need for the optical measurements. The limitation of this calibration technique for OPCs is that the CF has been found to vary with particle characteristics and source type, and the range of cooking activities experienced within a single dwelling mean that even with a CF for each day, the full variation of CFs may not be captured. Mass concentrations measured by the SidePak are adjusted by multiplying them by the CF, so the average concentration over the entire period is the same from both measurement methods. The same is true for other OPCs. The peaks and troughs in the temporal concentrations are adjusted by the same scale factor, but current methods do not account for differing particle characteristics across the range of concentrations measured. This is an ongoing challenge for measuring time resolved particle mass concentrations.

The potential for the intervention to reduce concentrations was limited by three key factors: (i) the ventilation systems available; (ii) occupant decisions on how and when to use ventilation; and (iii) uncontrolled variables including variation in cooking behaviour between measurement periods. All of the houses included in this study were fitted with cooker hoods that extract air to outside. Subsequent investigation in House A revealed damaged duct-work, see Appendix C.1, so it is likely some air was recirculated to the kitchen without filtration. The performance of the installed cooker hoods was not measured, so the operational flow rates or capture efficiencies are not known. It would be beneficial to include such investigation in future work to compare installed performance to the requirements set out in ADF.

In the two houses where concentrations were reduced in the second week, House C and D, the median concentration was 18% and 25% lower in Week 2, respectively.

However, whilst in House C the mean concentration also reduced, by 26%, in House D the mean concentration was 50% higher. Current guidelines are set for mean concentrations over set averaging periods (24 hours or 1 year). However the measured $PM_{2.5}$ concentration distributions suggest the arithmetic mean may not be the most representative statistic. In House D, with CF = 1.0, concentrations remained below 10 $\mu g/m^3$ 44% of the time in Week 1, compared to 60% of the time in Week 2. A similar pattern was observed for the daily mean guideline 25 $\mu g/m^3$, which concentrations remained below 83% of the time in Week 1 and 87% of the time in Week 2. Only reporting mean $PM_{2.5}$ concentrations causes some of this resolution to be lost. However, current guidelines are based on existing health research, see Section 2.1.5, which depends on available measurement methods and $PM_{2.5}$ concentration data, see Section 2.1.1.6. The gravimetric reference method [133] is the standard method to measure outdoor $PM_{2.5}$, and only provides time-averaged or mean concentrations. For this reason, it is likely there is currently insufficient epidemiological evidence to change the guideline values.

Across all houses, occupants recorded using ventilation during the majority of cooking events in Week 2. In those houses (A and B) where ventilation was also noted in Week 1, there was a substantial increase in ventilation use during Week 2. However, occupants knew that these measurements would only continue for one week, and therefore may have made compromises during Week 2, which may not be sustained over time. Figure 3.7c does not indicate that the intervention had a detrimental impact on internal temperatures. In some houses (A, B and C) t_i decreased in Week 2, however t_i appeared to increase in the remaining houses. In all cases, the shift in t_i was in the same direction as the shift in external temperatures, and the magnitude of the change was smaller for t_i in all houses other than House E where it was similar to that in t_{ext}. It is not possible to eliminate external temperature changes caused the change in internal temperatures, and therefore it is not possible to definitively conclude whether the intervention had an unintended impact on the indoor thermal environment. However, all

measurements were conducted in the heating season, therefore the heating system may have made up any additional ventilation heat losses. No attempt was made to monitor home energy use during this investigation, but, especially with the growing popularity of smart meters, it would be valuable to include energy use as an additional parameter in future work. If occupants can directly observe that using their cooker hood or opening windows during cooking is increasing their energy bills, they might be less inclined to continue taking these measures despite the health benefits.

Cooking behaviour is semi-random, where the frequency, duration and emission rates all vary. Table 3.6 shows how the total time spent cooking varied between weeks in all houses. In House B the total time spent cooking increased during the Week 2. This may have directly impacted the measured $PM_{2.5}$ concentrations, especially as oven use also increased in Week 2. During Week 1, one of the main peaks in $PM_{2.5}$ concentrations corresponded to use of the oven, see Figure C.10. This was identified and discussed with the lead occupant during the preparation for Week 2. In Week 2, the occupants appeared to respond to this finding by using the cooker hood on 10 of the 11 occasions the oven was in use. Nevertheless, the two highest peak $PM_{2.5}$ concentrations during Week 2 corresponded to events using the oven, reaching $6500\,\mu g/m^3$ and $2656\,\mu g/m^3$ (CF $=$ 1.0). On both occasions the cooker hood was switched on and the external door was opened. It is possible that using multiple ventilation systems in combination, in this way, may not lead to increased benefits. Similar behaviour was demonstrated in other households, where windows or doors were opened as well alongside using the cooker hood. This may also encourage $PM_{2.5}$ transport into adjacent rooms. Ideally, the kitchen should be negatively pressurised, achieved when using an only using an extractor fan, to prevent this from occurring. With regards ventilation use in Week 2, there was no clear correlation between cooking method and whether ventilation was used.

In House C, cooking behaviour was relatively consistent across both weeks. Here the $PM_{2.5}$ distributions were the most consistent between weeks and demonstrated the least spread. The success of the intervention in this case may be partially attributed to the consistency in behaviour. $PM_{2.5}$ emissions may have been more consistent across the two weeks, reducing the effect of this confounding factor. Although the occupants did spend less time cooking during Week 2. However, the mean and median concentrations remained above $25\,\mu g/m^3$ during both weeks (CF = 1.0), so whilst there was a statistically significant reduction in concentrations, the concentrations remained high. The concentration always remained above $10\,\mu g/m^3$, with a minimum $16\,\mu g/m^3$ measured in each week (CF = 1.0), the reason for which is unclear.

A better test of the ventilation systems available might be to design the cooking schedules in advance, so the same cooking behaviour is replicated in both weeks. However, the measured concentrations would be less representative of normal conditions in each test house. Alternatively, it might be possible to isolate concentrations attributed to similar cooking events in each week, and directly compare the results. The current method of parsing the data purely based on recorded cooking event times, as shown in Section 3.3.1, only captures some of the elevated concentrations and therefore a better method might be preferable, such as those used by He *et al.* [35], Nasir & Colbeck [165] or Chan *et al.* [36]. Additionally, this method would still require some consistency in cooking behaviour, and would not assess the overall performance, which was the purpose of this investigation. Future work could also attempt to estimate $PM_{2.5}$ emission rates using a method similar to those used by He *et al.* [35], Nasir & Colbeck [165] or Chan *et al.* [36], but only if concurrent gravimetric sampling were available. Gravimetric sampling was not available in the current study, so there would have been no merit to calculating emission rates from the concentrations measured here. The lack of CF would result in infinite uncertainty in the estimated emission rates.

3.5 Conclusions

The results of this investigation suggest that $PM_{2.5}$ concentrations in the kitchens of multiple occupancy student dwellings may be elevated above ambient, and that they may be reduced by advising occupants use additional ventilation during cooking. However, in 3 of 5 cases, the intervention did not reduce concentrations in Week 2, and in the two houses where concentrations were reduced, the effect was small. This intervention was restricted by the ventilation methods available in each kitchen. Assuming these met requirements, the existing building regulations may to be need reviewed to assess whether they prescribe sufficient ventilation and appropriate strategies.

This investigation also highlights multiple challenges in measuring $PM_{2.5}$ related to cooking, including but not restricted to: (i) the variability of human cooking behaviour; and (ii) difficulties regarding $PM_{2.5}$ measurement techniques.

This study also highlighted flaws in the method which might be corrected in future work. Future intervention studies should include concurrent gravimetric sampling, ideally with a filter change at least once every 24 hours. This would facilitate direct comparison with $PM_{2.5}$ guidelines and the measured concentrations could also be used to estimate emission rates using a method similar to that found in previous studies. It would be beneficial to also measure the installed flow rate of existing mechanical extract ventilation systems, to allow comparison to ADF requirements, and to include household energy use as an additional indicator of the longevity of resulting change. The current study was based on a relatively small sample, therefore it would be beneficial to use a larger representative sample in future. This would be costly and take considerable time, therefore mathematical modelling might offer an alternative method to predict the effect of ventilation systems.

Chapter 4

A Method to Measure $PM_{2.5}$ Emission Rates

Work from this chapter was previously presented as a short presentation, poster and paper at the AIVC conference 2017, in:

O'Leary, C. & Jones, B. *A Method to Measure Emission Rates of $PM_{2.5}$s from Cooking* in *38th Air Infiltration and Ventilation Centre Conference* (Nottingham, UK, 2017), 463–472.

The experimental data included in Experiment Phase 1 was collected prior to commencing this PhD during a placement in the Engineering Research Placement Scheme (ERPS) at the University of Nottingham, in summer 2015.

4.1 Introduction

The previous chapter highlighted some of the difficulties of measuring the effect of ventilation on indoor $PM_{2.5}$ concentrations, and that residential $PM_{2.5}$ concentrations can be highly variable. Probabilistic or deterministic stock models were introduced in Section 2.4 as an alternative method to investigate indoor $PM_{2.5}$ concentrations at stock levels. These models are highly sensitive to emission rate

inputs [37, 191]. Therefore, it is important to establish emission rates from cooking sources in an appropriate form to be used as a model input.

Studies frequently report emission rates based on small sample sizes (Table 2.10), from which it is not possible to produce the probability distribution functions (PDFs) required for probabilistic modelling. Section 2.5.3 introduced a range of methods previously used to measure $PM_{2.5}$ emission rates. These range from small scale controlled test chambers to personal monitoring techniques. Each method has associated benefits and drawbacks, including cost and time considerations. Therefore the aim of this chapter was to develop a method to measure $PM_{2.5}$ cooking emissions using toasting bread as a simple, repeatable cooking source, from which it is possible to derive PDFs of emission rates.

4.2 Experiment Phase 1

4.2.1 Method

The first phase of tests followed a controlled field test method similar to that used by Fortmann *et al.* [32] and Dacunto *et al.* [40]. Tests were conducted in a residential kitchen, volume 35.4 m^3 (see Figure 4.1) in Nottingham, UK, during July 2015. It was not possible to precisely control the ventilation conditions, however, all windows and internal and external doors were closed during testing, and the installed wall mounted kitchen extractor fan was used to stabilise the ventilation rate. The ventilation rate was not measured and mixing conditions were not investigated.

A SidePak™ AM510 Personal Aerosol Monitor (SidePak) was used to monitor $PM_{2.5}$ concentrations, at height 1.1 m, 1 m from the source; see Figure 4.1. The SidePak was used in Chapter 3, and the limitations of this optical device were introduced and discussed in Sections 2.2.1.3 and 3.4. Concentrations were time-averaged and

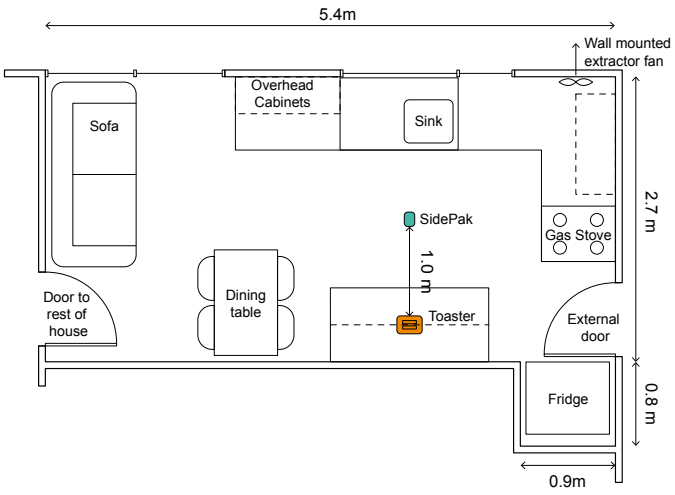


FIGURE 4.1: Kitchen plan

reported at 1 minute intervals following Ott *et al.* [44]. The calibration factor (CF) was set at default 1.0 as no concurrent gravimetric sampling was available.

The test consisted of toasting 2 slices of bread (medium sliced white or wholemeal Hovis 800 g loaf) in a supermarket branded toaster (see FigureD.1) on the highest setting (6). The toaster was new at the start of testing and was not cleaned between tests. The test was chosen to be simple and repeatable; the toaster had a predefined cooking time and temperature, and the bread slices were consistent in weight and geometry. The start and end times of the toasting period were recorded. The room was monitored under constant conditions for at least 25 minutes between tests to allow $PM_{2.5}$ concentrations to return to background levels. The test was repeated a total of 40 times, evenly split between white and wholemeal bread.

An additional six *plume tests* were conducted in the same location in October 2016 to further investigate the concentration profile during the emission period. During these tests, a sample tube attached to the SidePak inlet was positioned in the plume, 20 cm above the toaster, see FigureD.2, and concentrations were logged at 1 second intervals. Here the decay period was not monitored and the room was flushed with outside air between tests.

4.2.1.1 Emission Rate Calculation

The method used to determine emissions rates was based on the *theoretical peak* mass balance model introduced in Section 2.5.4.2 and used previously by Dacunto *et al.* [40], Ott *et al.* [44], and Olson & Burke [199]. The emission rate, g (μg/s) was determined with Equation 4.1 (from Equation 2.16) using the theoretical peak concentration, C_{Tp} (µg/m^3), background concentration, C_b (µg/m^3), initial concentration, $C(0)$ (µg/m^3), duration of emissions, T (s), mixing volume, V (m^3), and the total decay rate, ϕ, due to ventilation and other mechanisms (s^{-1}).

$$g = \phi V \left[\frac{C_{Tp} - C_b - (C(0) - C_b)e^{-\phi T}}{1 - e^{-\phi T}} \right] \tag{4.1}$$

The decay rate and theoretical peak concentration were not directly measured, and so were determined as follows. The recorded concentrations were split into the emission or α–period and the decay period using the recorded cooking times. The decay period was then further split into the β– and γ–periods. During the β–period the emitted $PM_{2.5}$ is not yet fully mixed with room air whereas during the γ–period the air is well mixed and smooth decay occurs [44]. The end of the β–period and start of the γ–period was identified as the time within the decay period when the peak concentration occurred. For an example diagram illustrating the α–, β– and γ–periods see Figure 2.5c. The total decay rate, ϕ, was determined using log-linear regression of the $PM_{2.5}$ concentrations during the γ–period. The estimated value of ϕ was extrapolated through the β–period to estimate C_{Tp}, the predicted concentration at the start of the β–period or end of the α–period. This method is similar to that proposed by Ott *et al.* [44].

A custom MATLAB [213] code[1] was used to process the data following the method above. Tests were eliminated where: (i) $R^2 < 0.7$ for the log-linear regression of the decay concentrations; (ii) the emission rate was negative; or (iii) the emission rate

[1]The code is available under a creative commons license from DOI: 10.13140/RG.2.2.22824.32003

could not be estimated, assumed to indicate a problem with the test data. Outliers were tested using *Chauvenet's Criteria* [217], and removed accordingly. Additionally, the non-parametric Mann-Whitney or Wilcoxon's Rank-sum test (MW test) was used to test for a statistically significant difference in the emission rates from toasting wholemeal or white bread, as visual inspection of the distributions of g appeared non-normal.

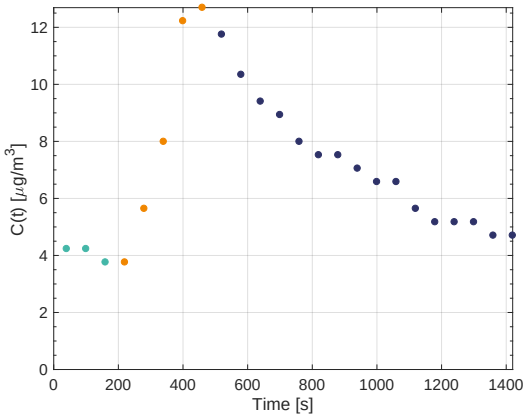
4.2.1.2 Calibration Factors

Concurrent gravimetric sampling to determine custom CFs (see Section 2.2.1.3) was not available. Two CFs have been published for toasting bread; Dacunto *et al.* [40] reported CF $= 0.47$ for burned toast ($n = 3$) and Jiang *et al.* [138] measured CF $= 0.79$. Dacunto *et al.* attributed the difference to levels of charring, referring to a previous *char index*. It was not possible to obtain details of this char index for comparison, so in absence of further information the data set was processed twice, applying each CF in turn.

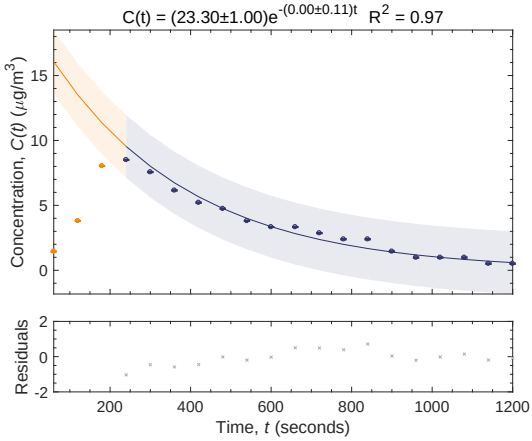
4.2.2 Results

Figure 4.2 shows the concentrations over time for a sample test (Fig 4.2a) and the corresponding decay concentrations with the fitted decay curve (Fig 4.2b). The α–period is highlighted in teal, the β–period in orange and the γ–period in blue. Plots for further tests are included in Appendix D.1.1. 30 of the original 40 tests produced positive emission rates, with $R^2 > 0.7$ for the decay and passed *Chauvenet's Criteria*. The eliminated tests are indicated in Appendix D.1.1.

Figure 4.3 and Figure D.5 show the distribution of emission rates for CF $= 0.47$ and CF $= 0.79$ respectively. The shape of the distribution is identical for both CFs, and shows a significant positive skew, where the median is much smaller than the mean,



(A) Concentrations over time



(B) Decay concentrations and fitted model

FIGURE 4.2: Example test concentrations
(CF $= 0.47$, wholemeal bread)
teal $= \alpha$–period; **orange** $= \beta$–period; **blue** $= \gamma$–period

see Table 4.1. This is attributed to a few tests with a high emission rate. Table 4.1 also shows the substantial impact the CF had on the emission rate.

The distribution of emission rates for *all* tests, *wholemeal* bread tests and *white* bread tests are shown in Figure 4.4 for CF $= 0.47$. The equivalent plot for CF $= 0.79$ is in Figure D.6. Here the ends of the whiskers indicate the minimum and maximum emission rates. The MW test indicated the white bread and wholemeal bread emission rates were not significantly different at the $p < 0.05$ significance level.

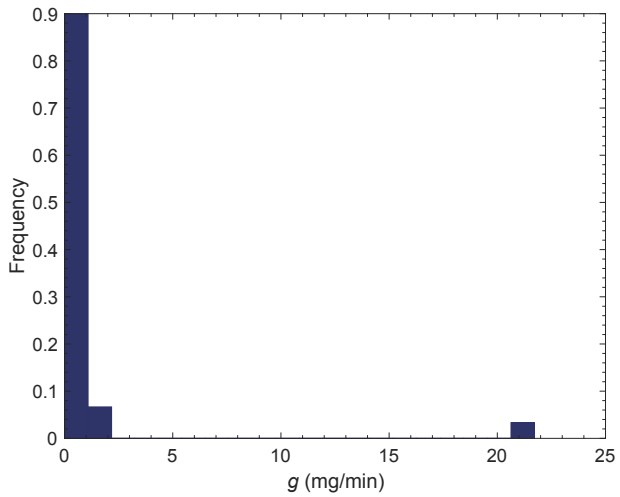


FIGURE 4.3: Phase 1 emission rates histogram CF $= 0.47$

TABLE 4.1: Phase 1 emission rates (mg/min)

	CF $= 0.47$			CF $= 0.79$		
	All	Wholemeal	White	All	Wholemeal	White
Min	0.003	0.003	0.02	0.02	0.02	0.03
LQ	0.04	0.03	0.04	0.06	0.05	0.06
Median	0.06	0.06	0.07	0.10	0.09	0.11
UQ	0.20	0.12	0.20	0.33	0.21	0.33
Max	22	22	0.47	37	37	0.78
Mean	0.9	1.7	0.13	1.6	2.9	0.22
SD	3.9	5.6	0.13	6.6	9.4	0.23
GM	0.09	0.09	0.08	0.16	0.17	0.14
GSD	5.5	9.1	2.8	5.1	7.9	2.8
N	30	15	14	30	15	14

4.2.2.1 Plume Test Results

The peak concentration in the initial tests occurred after the end of the α–period, see Figure 4.2b. The reasons for this were unclear and the 1 minute resolution of the recorded data made it difficult to conduct further investigations, especially given the toasting time of ~3 minutes. The theoretical peak calculation method assumed this lag was due to the time taken for the $PM_{2.5}$ to fully mix within the space. However, emissions may have continued after toasting ended as the toast and toaster were not sealed at the end of the α–period. Alternatively the particle composition and chemistry may have changed over time. Hence the need to conduct follow up plume

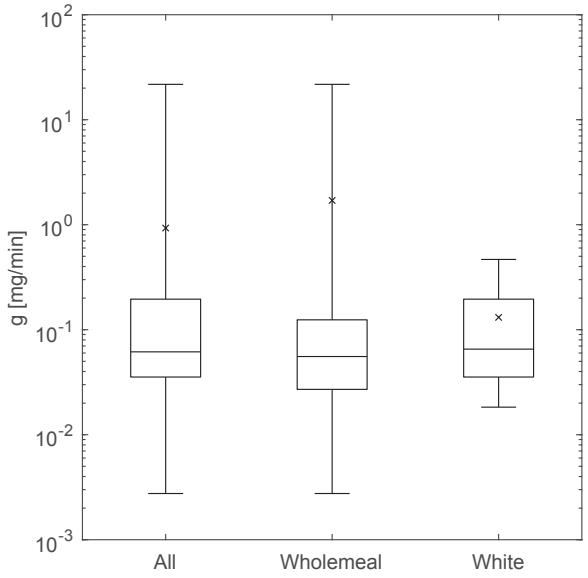


FIGURE 4.4: Phase 1 emission rates distribution comparison
CF $= 0.47$

tests.

The results of the plume tests are shown in Figure 4.5, which depicts the $PM_{2.5}$ during the α–period of all 7 tests. Here $PM_{2.5}$ does appear to have been emitted during toasting. The rate of emissions, indicated by the gradient of concentrations over time, does not appear constant throughout the α–period, suggesting the source function was not rectangular. However, the peak concentration did occur at the end of the α–period, therefore the peak estimation method might still provide a reasonable estimation of g.

4.3 Experiment Phase 2

This second phase of tests was developed in response to the problems encountered in Phase 1. It combined conditions similar to field tests, such as the method used by Dacunto *et al.* [40], and large scale chamber tests, similar to the method used by Afshari *et al.* [14].

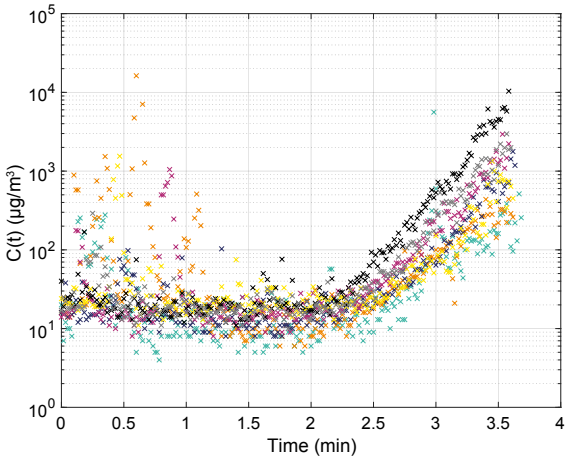


FIGURE 4.5: Plume test concentrations (CF $= 1.0$)

4.3.1 Method

Tests were conducted in a chamber with dimensions $2.62 \times 3.49 \times 2.35$ m in Nottingham, UK, during May 2017; see Figure 4.6. The test chamber was re-purposed for air quality measurements by laying plastic sheeting on the floor and removing unnecessary furniture and fixings. Custom boards were installed into the open window and door to control the ventilation conditions. A low level 0.1×0.1 m opening was located in the doorway, and an extractor fan (XF100S 15W, Manrose, Reading, UK) with a target exhaust flow rate of $85\,m^3/h$ ($23\,L/s$) was fitted into the window as pictured in Figure D.7. A desk fan was used to aid mixing within the chamber as indicated in Figure 4.6.

Two SidePaks measured $PM_{2.5}$ mass concentrations at 1 second intervals, to improve resolution from the 1 minute interval in Phase 1. Both SidePaks were mounted on tripods at height 1.1 m. The first, SidePak_F, was centrally located (Position 0 in Figure 4.6) whereas the second, SidePak_R, was moved between all 9 positions between tests to investigate the chamber mixing conditions. Only the concentrations from SidePak_F were used to calculate emission rates.

In addition, two IAQ-Calc Indoor Air Quality Meters (Model 7545, TSI Inc., Shoreview, MN, USA) were used to monitor indoor and outdoor temperature, CO_2

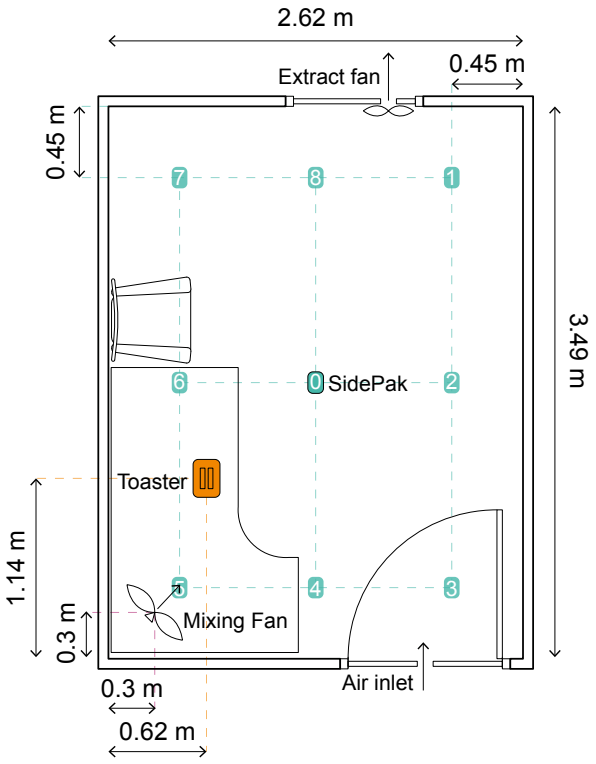


FIGURE 4.6: Pod plan

concentration, and relative humidity. Chamber RH was maintained below 70% throughout testing, to minimise its influence on SidePak performance. An electric oil-filled radiation was used to heat the chamber and maintain the RH when required.

Both SidePaks measured background $PM_{2.5}$ concentrations for a 5 minute period at the start of each test day to establish a value for C_b. The SidePaks were then moved indoors, the boards fitted to the window and doorway, and the extractor fan turned on before commencing testing. As in Phase 1, each test consisted of 2 slices of bread being toasted in the same toaster and setting. The toaster was located on a table, see Figure 4.6. Unlike Phase 1, all tests were conducted using wholemeal bread (Hovis, medium slice 800 g) as there was not a statistically significant difference in emissions from toasting white and wholemeal bread during Phase 1. The start and end of the α–period were recorded when the toaster was switched on and when

toasting ended, i.e. when the toast "popped up". A box was placed over the toast and toaster at the end of the α–period to prevent further $PM_{2.5}$ emissions into the chamber. $PM_{2.5}$ concentrations were then monitored under constant conditions for a 20 minute decay period. The room was then flushed with outside air to return chamber $PM_{2.5}$ concentrations to background levels.

4.3.1.1 Mixing Conditions

As stated previously, SidePak$_F$ remained in a fixed location throughout testing, whereas SidePak$_R$ was moved between tests. This was to check the main sampling location was providing a representative $PM_{2.5}$ concentration for the test chamber. SidePak$_F$ and SidePak$_R$ were collocated during the first test of each test day. For each subsequent test SidePak$_R$ was moved sequentially through Positions $0-8$. Jiang *et al.* [138] reported some variation in CF between SidePaks, therefore, when both SidePaks were in Position 0, a relative CF (rCF) was calculated using Equation 4.2 to adjust the concentrations reported by SidePak$_R$ ($\overline{C(T)_R}$) to match those from SidePak$_F$ ($\overline{C(T)_F}$). The rCF was applied to concentrations reported by SidePak$_R$ in other positions until both SidePaks were again collocated. In total 8 relative CFs were calculated.

$$\text{rCF} = \frac{\overline{C(T)_F}}{\overline{C(T)_R}} \tag{4.2}$$

The concentrations reported by each SidePak were overlaid for each test in turn, to allow visual comparison of the two data sets. In addition, the concentrations reported by both SidePaks were compiled and the overall distributions of concentrations for each SidePak compared for each location in turn. This comparison involved two tests, first, the Q-Q plots were produced to compare SidePak$_F$ to SidePak$_R$ in each position, and second a MW test was used to check for statistical significance.

4.3.1.2 Calibration Tests

Gravimetric sampling was not available during the initial Phase 2 test period in May 2017 and so the CF was initially set at 1.0. Subsequent tests conducted in the same test chamber in March 2018 also included gravimetric sampling. These tests were conducted under minimum ventilation conditions, where the door and window remained closed throughout to ensure a detectable mass deposit on the filters. The same two SidePaks used for the main Phase 2 tests were used to monitor $PM_{2.5}$ at 1 second intervals. Both SidePaks were collocated in the centre of the test chamber (Position 0, Figure 4.6) along with a MiniVol Tactical Air Sampler (MiniVol) (Airmetrics, Eugene, OR, USA) used for gravimetric sampling. The MiniVol pump drew air through the installed $PM_{2.5}$ impactor and a pre-weighed 47 mm Millipore fluoropore™ PTFE membrance filter at 8 L/min. Three calibration tests were conducted; tests 1 and 3 consisted of 5 repetitions of toasting 2 slices of bread (Hovis, wholemeal 800 g medium slice) followed by a short decay period, whereas test 2 consisted of a single repetition of toasting 2 slices of bread followed by a long decay period of ~1.5 hours. Tests 1 and 3 were designed to be high concentration tests, and test 2 a low concentration test. The start and end time were recorded for each filter. The toast and toaster were sealed in a container between α–periods as in the main Phase 2 tests. A single filter was used for each test and the chamber was flushed with outside air between tests.

$$\overline{C(T)} = \frac{m}{qT} \tag{4.3}$$

Filters were pre and post weighed by a researcher from the University of Chester under controlled conditions to determine the total mass (m) collected on each filter in μg. The average mass concentration $\overline{C(T)}$ in $\mu g/m^3$ from each filter was determined using Equation 4.3, where q is the pump flow rate in m^3/s, and T the total time the filter was in use. SidePak concentrations were parsed according to the

filter times and the mean concentration calculated. Calibration factors were then determined separately for each SidePak using regression analysis with the intercept forced through the origin. In addition, a combined calibration factor was determined using linear regression across both SidePaks.

4.3.1.3 Data Analysis

Calibration factors were determined using Microsoft Office Excel 2013. All other analysis was conducted in MATLAB[2].

To calculate emission rates, first the combined custom CF (see Section 4.3.1.2) was applied to the $PM_{2.5}$ concentrations. The MATLAB code used to estimate emission rates in Phase 1 was updated to accommodate the noisy data resulting from the 1 second time step.

In Phase 1 the end of the β–period was identified by the peak concentration. The 1 second logging interval in Phase 2 resulted in noisy data that required an alternative approach. Here the MATLAB code searched for the smooth decay period using the R^2 value as a test for the goodness of fit of the decay. The data was isolated for each test in turn, and the α–period removed based on recorded test times. First the times for this decay data were zeroed using the start of the decay and C_b subtracted from the concentration. The log-linear regression was then conducted for the entire decay period (β and γ) and the R^2 recorded. An iterative process illustrated in Figure 4.7 then began. Starting at the beginning of the decay period a single data point was removed and the regression repeated for the updated data set. The R^2 values were compared and the decay data with the better fit (higher R^2) retained for the next comparison. This process continued until the same data set was retained for a number of iterations equal to 25% of the total number of data points in the entire decay period. The data set that had been

[2]The code is available under a creative commons license from DOI: 10.13140/RG.2.2.14016.28167

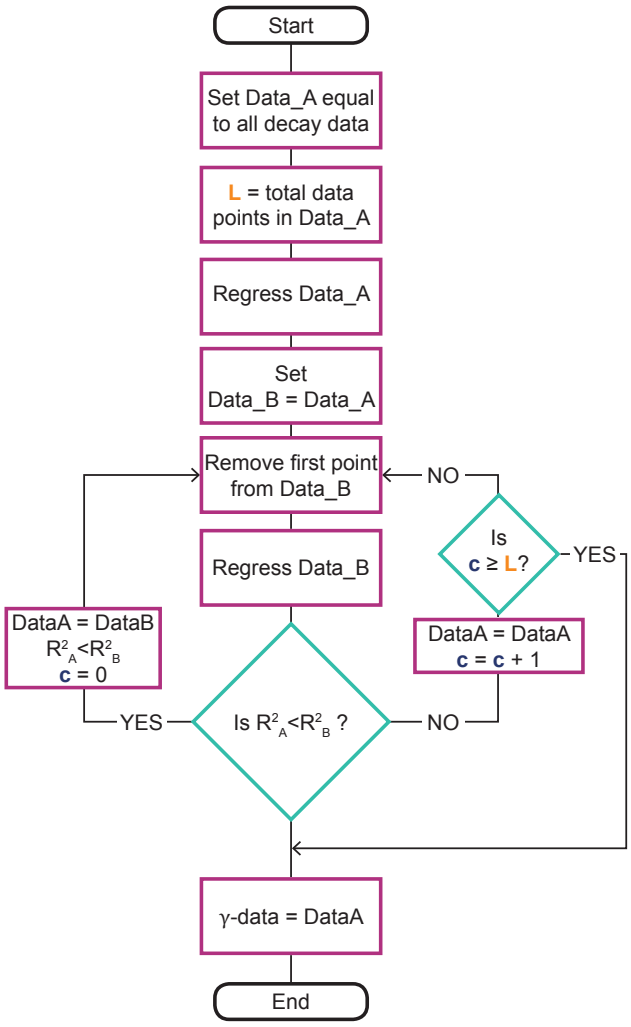


FIGURE 4.7: Flow chart of method to identify γ–period

retained at this point was recorded as the γ–period, the start of which indicated the start of the smooth decay period.

Subsequent stages followed the same method as for Phase 1: the decay rate was extrapolated back through the β–period to predict C_{Tp}, and the emission rate calculated using Equation 4.1.

The decay rates determined by the MATLAB code were summarised for comparison to the expected ventilation rate from the installed fan. Finally, the resulting emission rates were used to produce an empirical cumulative distribution function suitable

for use in probabilistic modelling and the shape of this distribution was investigated.

4.3.2 Results

4.3.2.1 Mixing Conditions

The relative CFs, see Section 4.3.1.1, used to correct the SidePak$_R$ concentrations to match those from SidePak$_F$ ranged between 0.82 and 0.94 with $\mu = 0.89 \pm 0.04$. With the rCFs applied, the MW test found a significant difference between SidePak$_F$ and SidePak$_R$ in Positions $2-8$ but not in Position 1, nor when they were collocated in Position 0. However, the statistical significance tests do not fully describe the mixing conditions in the chamber. Figure 4.8 and Figure 4.9 show Q-Q plots comparing SidePak$_F$ and SidePak$_R$ in each location and the time-resolved $PM_{2.5}$ concentrations during a sample test in each location. The Q-Q plots show good agreement between both SidePaks at lower concentrations. At higher concentrations SidePak$_F$ measured higher concentrations than SidePak$_R$. Some explanation for this difference can be found in Figure 4.9, where SidePak$_F$ appears more sensitive to the concentration increase due to toasting, but both SidePaks showed good agreement during the decay period. There was also a similar, smaller discrepancy when the SidePaks were collocated. The reason for this discrepancy is unclear, but may relate to noise in the data.

Overall, these tests indicate good mixing in the smooth decay period but not before. The time taken to achieve good mixing was accounted for by theoretical peak calculation method by using the γ–period data to estimate the theoretical peak.

4.3.2.2 Calibration Factor

Figure 4.10 shows a plot regressing the average concentrations from the gravimetric sampling against the average concentrations measured by each SidePak both seperately and combined. The resulting CFs, R^2 and standard errors (SE) are

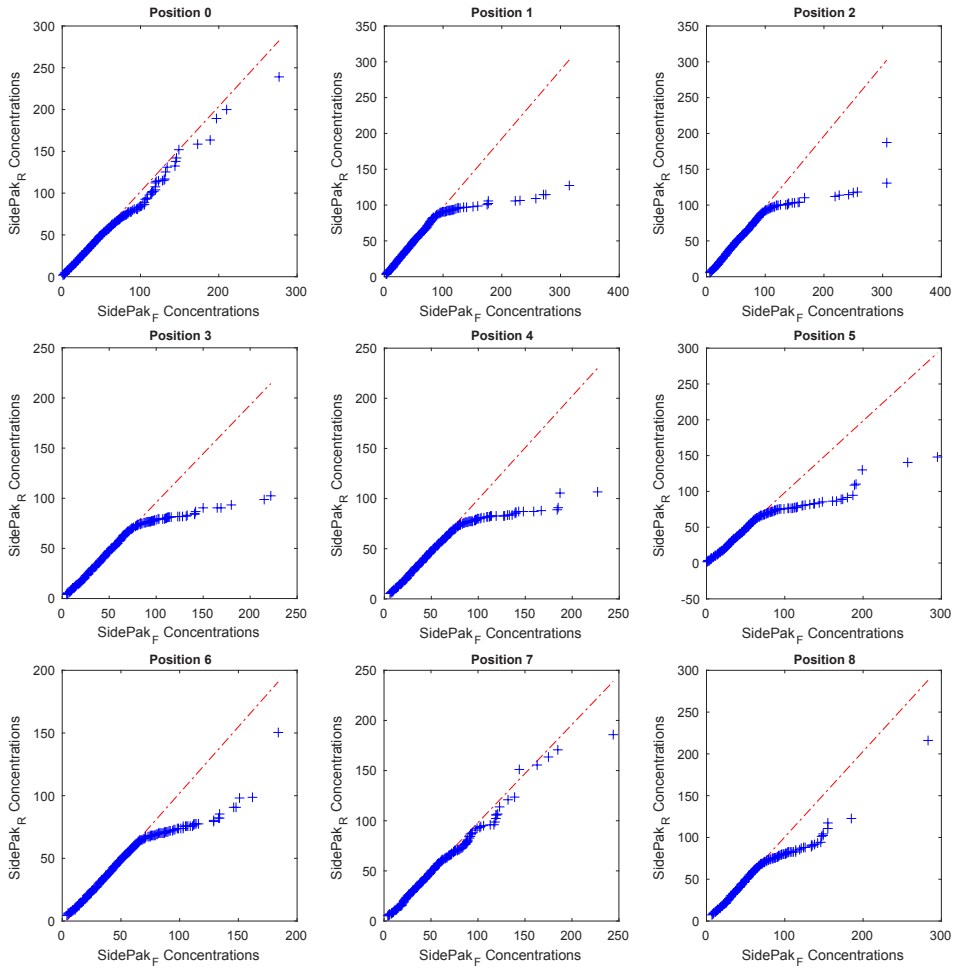


FIGURE 4.8: Phase 2 mixing Q-Q plots

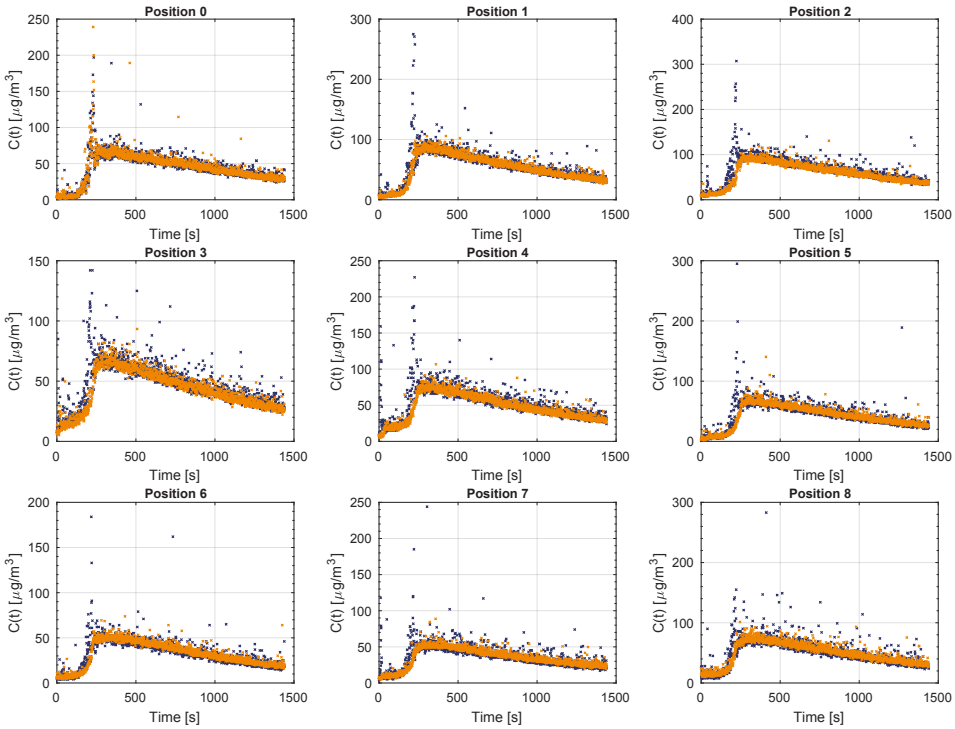


FIGURE 4.9: Phase 2 mixing temporal concentrations
blue = SidePak$_F$; **orange** = SidePak$_R$

TABLE 4.2: Calibration factors

	CF	R^2	SE
SidePak$_F$	0.68	0.999	0.017
SidePak$_R$	0.59	0.999	0.015
Combined	0.63	0.997	0.022

reported in Table 4.2. The high R^2 values indicate the linear regression model is a good fit, even for the combined regression, which has higher associated standard error.

In addition, Figure 4.11 shows the time-resolved $PM_{2.5}$ concentrations during the CF tests, with the α–periods highlighted. These concentrations are from a single SidePak without the custom CF applied. Here the concentration continued to increase between α–periods despite the toaster and toast being sealed off. The test chamber was built to Passivhaus standards so infiltration is considered negligible, and there was no PPV during the CF tests therefore concentrations were not necessarily expected to decay between tests. However it is not clear why

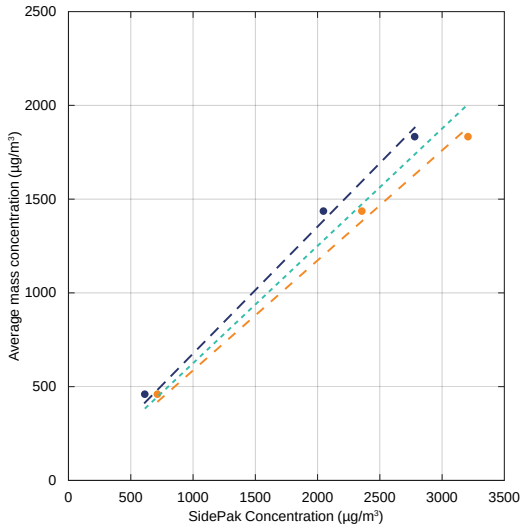


FIGURE 4.10: CF test regression
blue = SidePak$_F$; **orange** = SidePak$_R$; **teal** = Combined

concentrations continued to increase. Possible explanations might include room air mixing over time or secondary particles forming over time.

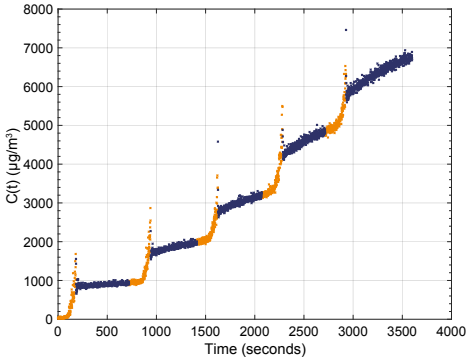
Concentrations reported from this point have the custom CF of 0.63 applied.

4.3.3 Decay Rates

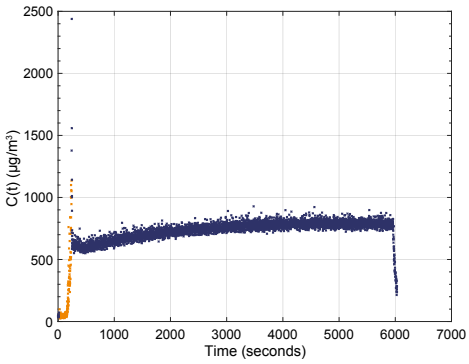
The decay rates ranged between $0.09\,s^{-1}$ and $0.38\,s^{-1}$ with $\mu = 0.23 \pm 0.07\,s^{-1}$. All the predicted decay rates were larger than the estimated air change rate of $0.001\,s^{-1}$. This indicated that deposition or other factors may have contributed more to the decay rate than ventilation. However, the decay still appeared first order logarithmic (see Fig 4.13b). In addition, the decay rates were close to normally distributed, see Figure 4.12.

4.3.3.1 Emission Rates

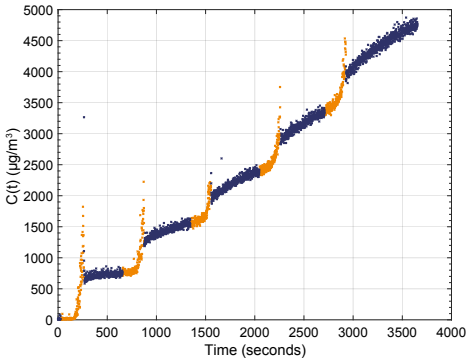
Figure 4.13 shows the concentrations over time for a sample test (Fig 4.13a) and the corresponding decay concentrations with the fitted decay curve (Fig 4.13b). As before, the α–, β– and γ–periods are distinguished by colour. The noisy data that



(A) Test 1



(B) Test 2



(C) Test 3

FIGURE 4.11: Temporal concentrations during calibration tests
orange = α–period

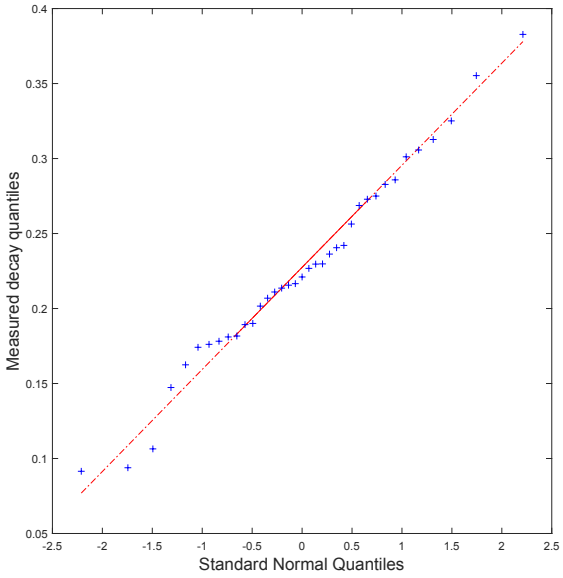


FIGURE 4.12: Q-Q of Phase 2 decay rates (s^{-1})

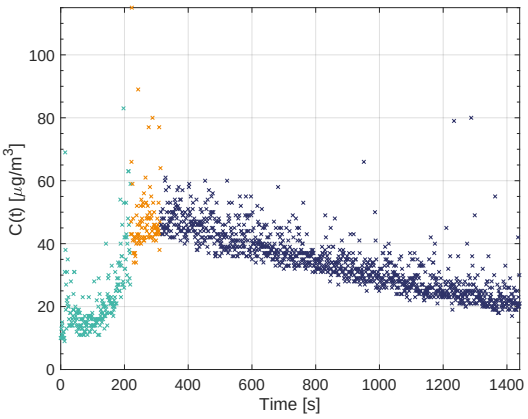
TABLE 4.3: Phase 2 emission rates (mg/min)

CF	Min	LQ	Median	UQ	Max	μ	σ	N
0.63	0.09	0.18	0.22	0.27	0.38	0.23	0.067	37

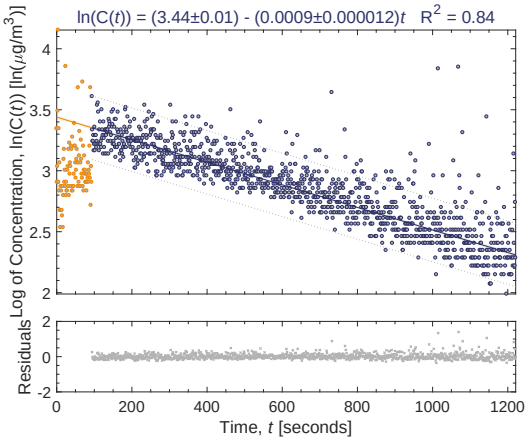
necessitated the updated method to identify the γ–period is evident in Figure 4.13b. Plots for the other tests are included in Appendix D.2.2. 3 of the original 40 tests were rejected due to an $R^2 < 0.7$ and all the remaining tests produced positive emission rates. In addition no tests were rejected by *Chauvenet's Criteria* so $N = 37$.

Unlike in Phase 1, the emission rates were close to normally distributed, see Figure 4.15. The tests produced consistent emission rates, and the mean and median were almost equal, see Table 4.3 and Figure 4.14. Figure 4.16 also includes a direct comparison to the results from Phase 1. The mean g was lower in Phase 2 when compared to the Phase 1 g with either CF, however the Phase 2 median g was higher. This plot also highlights that the Phase 2 g distribution was considerably less spread; the lower variance indicates the Phase 2 test method resulted in more consistent emission rates.

Finally, an empirical cumulative distribution function (eCDF) was produced from



(A) Concentrations over time



(B) Decay concentrations and fitted model

FIGURE 4.13: Example test concentrations
(CF $= 0.63$, wholemeal bread)
teal $= \alpha$–period; **orange** $= \beta$–period; **blue** $= \gamma$–period

the predicted emission rates, see Figure 4.17. This eCDF could itself be used as an input for probabilistic modelling. Alternatively, a normally distributed cumulative distribution function (CDF) with $\mu = 0.23 \pm 0.067$ could be used. This CDF is overlaid in orange in Figure 4.17 and appears to be a reasonably good approximation of the eCDF.

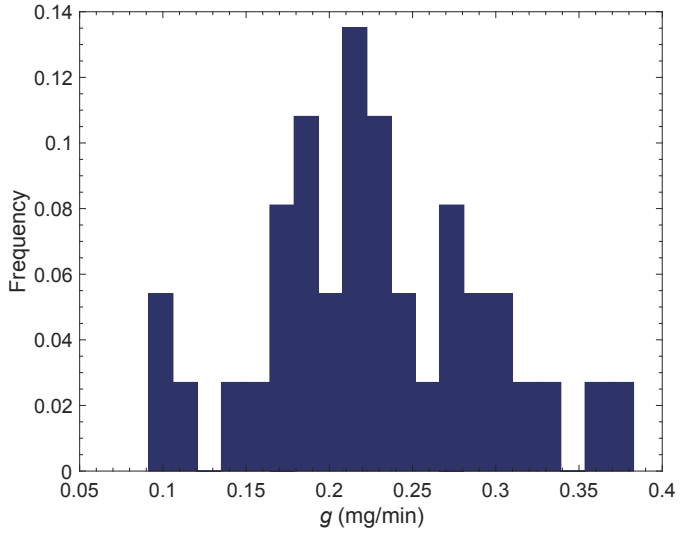


FIGURE 4.14: Histogram of emission rates

4.4 Discussion

The mean emission rates from both Phase 1 and 2 were an order of magnitude lower than those reported by Dacunto *et al.* [40] and Jiang *et al.* [138], see Table 4.4. The degree of charring might explain this difference. Dacunto *et al.* and Jiang *et al.* used a standard char-index to report the degree of charring in their tests, however it was not possible to obtain this index for comparison. The estimated mean emission rate for Phase 2 is closer in magnitude to the median 0.11 mg/min reported by He *et al.* [35].

Other than the changes in the measurement method, a possible further reason for the difference in emission rates between Phase 2 and Phase 1 is the characteristics of the bread itself. The same brand of wholemeal bread was purchased in Phase 1 and Phase 2, however, the manufacturer appeared to have adjusted the properties of the loaf in this time. Photos of slices of toast from each phase are included in Appendix D.3. In Phase 1, the slices of bread were rectangular and the top portion did not fit into the toaster, whereas in Phase 2, they were nearly square and most of the slice was toasted. This change may not itself have influenced g, but may also be

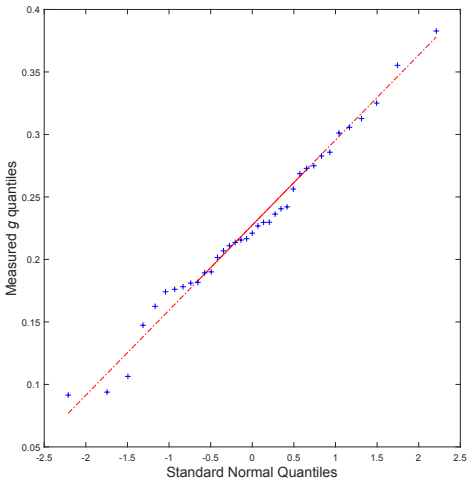


FIGURE 4.15: Q-Q normal comparison

TABLE 4.4: Emission rates comparison (mg/min)

	μ	σ	RSD (%)	N
Phase 1 CF = 0.47	0.9	3.9	433	30
Phase 1 CF = 0.79	1.6	6.6	413	30
Phase 2 CF = 0.63	0.23	0.07	30	39
Toast, 90 − 95% char Dacunto *et al.* [40]	9.5	10.8	114	3
Toast, 70 − 80% char Jiang *et al.* [138]	4.2			
Toasting, He *et al.* [35]	0.11*	0.37	336	18

*Median

indicative of other changes to the bread which could have a confounding effect. This variation could have been corrected if the slices had been weighed and measured before each test.

In Phase 1, the variance of the emission rates, indicated by the relative standard deviation (RSD), remained large despite the high number of repetitions compared to the previous studies. Therefore the test methodology from this phase was not highly repeatable. The improved method in Phase 2 used a smaller room volume with improved mixing to give more resolution in the α–period. This resulted in a more reproducible test indicated by an RSD that was an order of magnitude lower than in Phase 1. The γ–period decay rates were also consistent in Phase 2 and fewer tests were rejected due to a poor model fit.

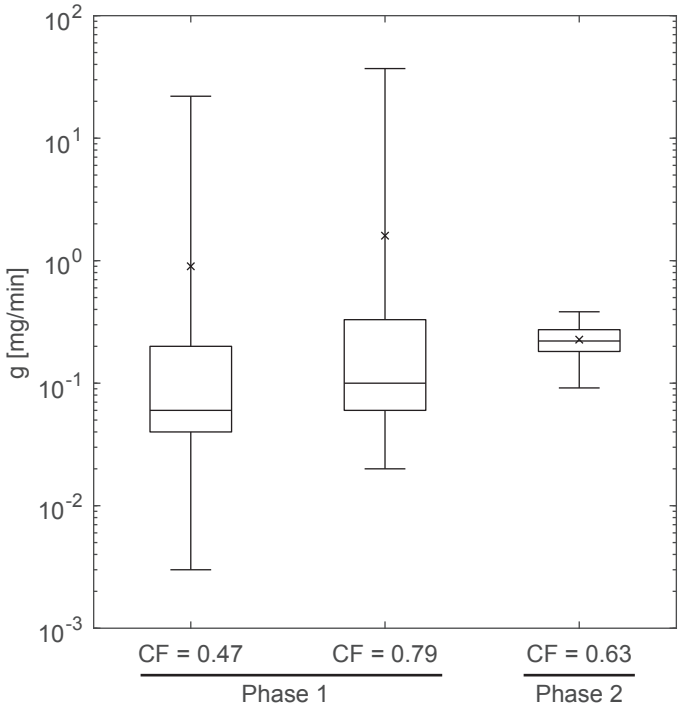


FIGURE 4.16: Comparison of Phase 1 and Phase 2

In Phase 1 the emission rates were not normally distributed, and the histograms suggest a distribution closer to log-normal. The main cause of the skew was a single test with high emission rates. No details were recorded in the test log that might indicate a reason for higher emissions during this test. The more consistent results in Phase 2 might be an indication of a more consistent test method, but the resulting distribution may be less representative of emissions under normal conditions. He *et al.* [35] determined emission rates from residential monitoring data. Their approach results in greater uncertainty derived from: (i) differences between toasting events, (ii) a dependence on occupant records of cooking events, and (iii) less control over the ventilation conditions. However, He *et al.* determined g from monitoring normal occupant behaviour, and so their distribution of g might be more representative of those experienced in a housing stock if the authors used representative sampling techniques to select participating households.

In Phase 1, the calibration factor was the largest source of uncertainty, because

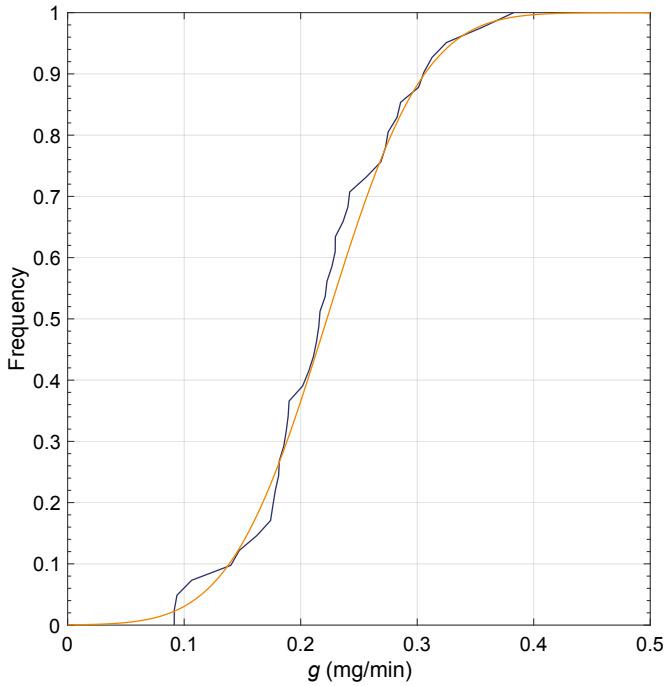


FIGURE 4.17: eCDF (**blue**) and *normal* CDF (**orange**) of g

gravimetric sampling was not used. During Phase 2, this source of uncertainty was reduced, by conducting follow up gravimetric sampling to determine a custom CF. However, this method was not perfect, as the gravimetric sampling was not concurrent. The CFs of 0.59, 0.68 and 0.63 determined in Section 4.3.2.2 are all comparable to the CF $= 0.79$ from Jiang *et al.* [138] and CF $= 0.47$ from Dacunto *et al.* [40] for toasting bread. Jiang *et al.* also found SidePak CFs varied between devices, therefore there is precedent for the differing CFs for the two SidePaks used here. using the combined CF is a source of additional error. The combined CF was selected over the SidePak_F CF to accommodate additional uncertainty as the CFs were not determined during the original test period and were based on just 3 filter samples.

In multiple tests during Phase 2, a high peak $PM_{2.5}$ occured at the end of the α–period, see Figure 4.13 or Appendix D.2.2. These concentrations frequently dropped off rapidly at the start of the β–period. It is possible that the high peaks

were due to the room air initially being poorly mixed. The mixing condition results also suggest that this may be the case, as Figure 4.9 indicates that these high measured peaks were only measured in Point 0 at the centre of the chamber. The predicted C_{Tp} intercepts from the γ regression were typically lower than these high measured peak concentrations, indicating the method to identify the smooth decay period effectively ignored this initial drop off. If mixing was the only cause then this identifies a potential strength of the method. However, an alternative explanation for this initial rapid decay might be that some of the emitted $PM_{2.5}$ dissipated rapidly due to second order decay mechanisms which were not constant over time. If this were the case, then the current method might have underestimated C_{Tp} and hence underestimated g. Nevertheless, the initial rapid decay would also then indicate these particles would not remain in the air for long, and so the risk of them being inhaled is lower. Therefore, this possible underestimation of the peak concentration and resulting emission rate is not problematic for devloping a probability distribution function for use in modelling.

The theoretical peak estimation method may also be a source of uncertainty as this method assumes a constant emission rate, g, over the α–period. The plume tests indicated that g was not constant over time, with no $PM_{2.5}$ emissions in the first 2 minutes of the toasting period. After this time, the emission rate appeared to increase over time. This emission profile is expected for toasting, which relies on radiant heat transfer to first dry out and then brown the bread. As the toast browns, the rate of heat transfer will also increase. One alternative approach to estimate emission rates was suggested by Pagels *et al.* [208], who determined the g iteratively by fitting a curve to the observed data and adjusting the emission rate to achieve the best possible fit. There are two potential problems with this alternative approach: firstly, Pagels *et al.* still assumed a constant g over the α–period, and secondly, the predicted curve was based on the mass balance method which assumes instantaneous full mixing [44]. Neither was true here, therefore this method would not improve the peak estimation method.

A third option would be to use the area-under-the-curve method proposed by Ott *et al.* [44] to estimate the average emission rate $\overline{g(t)}$ over the α–period, see Section 2.5.4.3 for more details. Unlike the peak or theoretical peak estimation method, the area-under-the-curve method is not well established in literature. This approach is theoretically exact, but it assumes instantaneous full-mixing, which was not observed in this case. In addition, the highest concentrations were observed at the end of the α–period, which was relatively short, therefore the theoretical peak estimation method used here appears to be the most appropriate. This method is also best suited to accommodate the non-instantaneous mixing observed in Section 4.3.2.1.

One improvement of the method from Phase 1 to Phase 2 was that the mixing conditions within the chamber were investigated and a desk fan was used to attempt to aid mixing. The results of these tests indicated good mixing within the chamber during the smooth γ–period. However, the mixing conditions were only investigated in two dimensions. Torkmahalleh *et al.* [202] measured the mixing conditions within their small test volume using SF_6 tracer gas and an array of sensors at 10 cm intervals. This method would better assess the mixing conditions throughout the chamber volume. The necessary equipment was not available within this study. In addition, if mixing conditions within the chamber were to be tested in future, it would be best to test the chamber both with and without a mixing fan, to determine whether it is necessary and the best position for it to be located.

Unfortunately a key limitation of this test methodology is that it is time consuming. Each phase of testing took 3 full days despite a short duration cooking source of 3.85 ± 0.9 minutes. However, toasting bread only represents a single cooking source, and so this data is of limited use in modelling. Cooking behaviour is variable and often includes activities of longer duration, therefore using this methodology to measure emission rates from a range of sources would be very

time consuming. The Phase 2 methodology produced consistent emission rates which suggested a normal distribution. If this can be replicated for other cooking sources it might be possible to estimate cumulative distributions suitable for modelling assuming a normal distribution with a mean and standard deviation determined from a smaller sample size of tests.

4.5 Conclusions

To conclude, two methods to measure $PM_{2.5}$ emission rates from a cooking source were developed and tested using toasting bread as a simple repeatable cooking source. Through this process some important considerations for measuring g were identified, namely: (i) the choice time-step for optical devices, (ii) the use of gravimetric sampling to determine custom CFs for optical devices, (iii) choosing an appropriate calculation method based on the shape of the g source function, (iv) the need to measure and control chamber mixing conditions, and (v) the quality of the concentration decay data. However, a key disadvantage of the current test method is that it is time consuming. Phase 2 testing took 3 days despite a short duration of emissions. If the same method were to be repeated for cooking more complex dishes with longer cooking times, the time required to complete the large number of repetitions used here might be prohibitive.

In addition, an empirical CDF and derived normal CDF of $PM_{2.5}$ emission rates were determined from the second phase of tests. Either distribution would be suitable for use in probabilistic stock modelling. However, a question remains over whether these emission rates measured under controlled conditions would be completely representative of those occurring in households. Furthermore, only one cooking source was tested here, using a single cooking method and ingredient, so these distributions were not intended to represent all domestic $PM_{2.5}$ cooking emissions.

Chapter 5

$PM_{2.5}$ Emission Rates from Cooking Full Meals and the Reduction Potential of using a Cooker Hood

This chapter has previously been published in:

O'Leary, C. *et al.* Investigating measurements of fine particle (PM2.5) emissions from the cooking of meals and mitigating exposure using a cooker hood. *Indoor Air* **29,** 423–438. ISSN: 0905-6947 (2019).

This project was a collaborative project with *the Netherlands Organisation for Applied Scientific Research* (TNO). Some of the methodology had already been developed by the team at TNO before I joined the project in early 2017. Specifically, the test chamber and basic ventilation conditions had been decided including the cooker hood, and the Grimm 11-R Mini Laser Aerosol Spectrometer had been selected to measure $PM_{2.5}$ concentrations. However, I was influential in conducting preliminary tests to investigate mixing conditions within the chamber and the decision to include gravimetric sampling within the test protocol. The team had begun to develop the chosen test meals, had chosen the general components and ideas of each meal, and had chosen the variations on Meal 1 to investigate potential

factors that might influence emissions. My contribution began with developing the cooking protocols for each test meal and conducting most of the laboratory tests. During my testing period we attempted to conduct gravimetric sampling to determine custom calibration factors for the Grimm. Unfortunately, the TECORA $PM_{2.5}$ inlet was incorrectly set up so the gravimetric sampling tests had to be repeated at a later date. These follow up tests were conducted by Piet Jacobs from the TNO research team according to the same test procedure. I took the lead with the subsequent data analysis and writing the paper.

5.1 Introduction

There were three main motivations for this research.

Firstly, as introduced in Section 2.5, numerous studies have investigated $PM_{2.5}$ cooking emissions and influencing factors whilst cooking single ingredients. Examples include toasted bread, $9.5 \pm 10.8\,mg/min$, fried chicken breast $15.2\,mg/min$, and deep-fried French fries $0.34 \pm 0.03\,mg/min$ [40]. However, the cooking of individual components, rather than full meals, may not be representative of typical home preparation. The addition of ingredients at various stages of the cooking process may affect $PM_{2.5}$ emission rates; for example, by amending cooking temperatures. However, the need to protect public health is arguably more important than proving this hypothesis, and so there was a need to investigate the effects of repetitively cooking multiple food types concurrently in a controlled and systematic manner, because it had not previously been conducted and the results would be useful in subsequent research to inform policy.

Secondly, there is some evidence that ingredients and non-essential additives, and cooking equipment influence $PM_{2.5}$ emissions, see Section 2.5.2 for more details.

And thirdly, it is important to consider the impact of mitigation strategies on emission rates, as it is unreasonable to expect cooking to be removed from

dwellings *en masse*. Instead, emitted pollutants need to be extracted at their source using a cooker hood, which reduces exposure risks by capturing emitted pollutants before they mix with room air. As introduced in Section 2.3.1, ventilation requirements for kitchens vary around the world. In the UK, kitchens in new dwellings are required to have an intermittent extract rate of 60 L/s, or 30 L/s through a cooker hood, but there is no requirement to modify the ventilation strategy in existing dwellings [16]. However, in the Netherlands, a kitchen ventilation rate of 21 L/s is required in new dwellings. Higher ventilation rates are recommended by ASHRAE 62.2 [179]. The ability of a cooker hood to capture particles is indicated by its capture efficiency (CE), introduced in Section 2.3.1.1. A cooker hood's CE is a function of the airflow rate through the hood, physical features that entrain pollutants towards the device, the installation height and capture volume, and the burner location and coverage by the hood [183]. Lunden *et al.* [181] found particle CEs of $4-39\%$ for stir-frying on the front burner and $70-99\%$ for the back burner. Singer *et al.* [183] reported similar findings.

Therefore, this study aimed to advance the understanding of $PM_{2.5}$ emissions that occur when cooking full main meals. It did this by cooking four hot meals commonly prepared in Northern European countries under controlled conditions in a kitchen laboratory. The cooking methods were then varied to investigate parameters that affect emissions, and the cooker hood was explored as an exposure mitigation strategy.

5.2 Methods and Materials

5.2.1 Laboratory facilities

All of the experiments were performed under controlled ventilation conditions in a test chamber with depth 3.65 m, width 2.66 m, height 2.68 m, and volume, $V = 26.02 \pm 0.08\,\mathrm{m}^3$. The chamber layout, including its dimensions, the placement

of the cooker hood and the size of the cabinets was comparable to the EN 61591 standard [218] for kitchen test facilities. The only addition was the use of a ceiling diffuser to supply air, intended to simultaneously enhance the mixing of the space and minimise stratification and disturbances of the airflow under the cooker hood [219]. Supply air was delivered by an HVAC unit equipped with an AFPRO F7 filter. To minimise uncontrolled infiltration and exfiltration, the supply air flow rate was adjusted so that the pressure difference between the test chamber and its surroundings was $<0.5\,\mathrm{Pa}$ when measured by a Halstrup-Walcher EMA 84 digital pressure gauge.

Tests were performed for two ventilation scenarios that varied the flow rate and location of the ventilation extract. The first was a *low* ventilation scenario with airflow rate of $21\,\mathrm{L/s}$ ($75\,\mathrm{m^3/h}$), the minimum required in domestic kitchens by the Netherlands Building Regulations [220]. It was achieved using a single extract grille located in the middle of the chamber 94 cm below the ceiling; see Position A in Figure 5.1. The second was a *high* ventilation scenario achieved using a cooker hood with a flow rate of $83\,\mathrm{L/s}$ ($300\,\mathrm{m^3/h}$). In both scenarios, the airflow rate was controlled by a centrifugal fan located outside the chamber and set by measuring the pressure drop over an orifice plate following EN ISO 5167-2 [221]. The main tests were conducted using the low ventilation scenario (see Sections 5.2.3.1-5.2.3.3), but the high ventilation scenario was used to support additional analyses (see Section 5.2.3.4).

Mixing conditions were explored during the preliminary test set up period by measuring $PM_{2.5}$ concentrations concurrently in positions $A - D$ (Figure 5.1) under low and high ventilation conditions. The procedure was similar to that set out in Section 4.3.1.1. A SMC TR16 fan was used during the low ventilation scenario to enhance air mixing, and the concentrations appeared similar throughout the space, see Figure E.1. However, mixing tests with both the hood and mixing fan in use indicated $PM_{2.5}$ concentrations were much higher near to position D than position

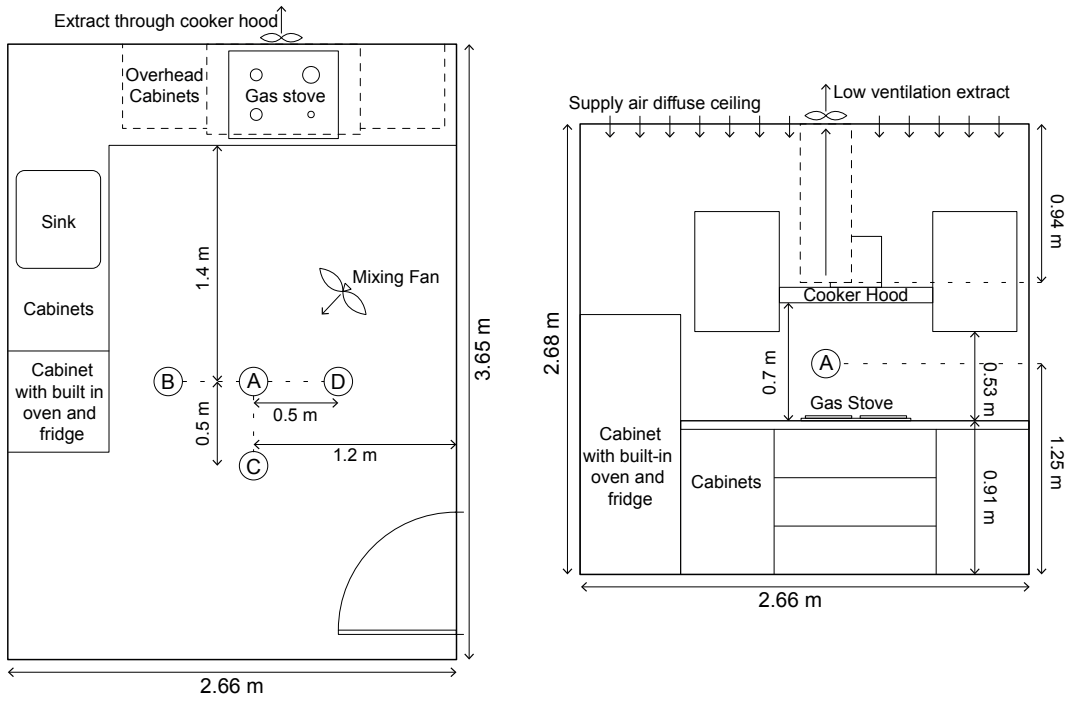


FIGURE 5.1: Test chamber dimensions and layout
A, location of Grimm 11-R Mini-LAS; B, C & D, gravimetric sampling locations

B, see Figure E.2 and smoke tube tests further indicated that the plume above the stove was redirected to the right when the mixing fan was switched on. Therefore, the mixing fan was not used for the *high* scenario to avoid disturbing the airflow under the cooker hood which may reduce the capture efficiency.

A Pelgrim GK 564 gas stove with four burners was used as a heat source; see Figure 5.1. The maximum flow rates, $\dot{Q}$ (L/min), and power, H (kW), per burner were $\dot{Q} = 3.2\,\mathrm{L/min}$ and $H = 1.7\,\mathrm{kW}$ for the back left burner, $\dot{Q} = 4.5\,\mathrm{L/min}$ and $H = 2.4\,\mathrm{kW}$ for the back right burner, $\dot{Q} = 2.5\,\mathrm{L/min}$ and $H = 1.3\,\mathrm{kW}$ for the front left burner, and $\dot{Q} = 1.9\,\mathrm{L/min}$ and $H = 1.0\,\mathrm{kW}$ for the front right burner.

5.2.1.1 Cooker Hood

A consumer standard ATAG WS9011QAM ducted cooker hood was selected to meet the criteria of the high ventilation scenario and installed 70 cm above the

kitchen counter, see Figure 5.1, which approximately agrees with Singer *et al.* [183] and Lunden *et al.* [181]. A *damp buffer*, where the sides that frame the hood protrude below the central horizontal plate, has previously been found to help entrain pollutants towards the hood [185]. The cooker hood was 90 cm wide and 53.6 cm deep, and was equipped with a small damp buffer of height 3 cm (see Figure E.3). The exhaust hood extended over the front burners whose centres were 40 cm from the wall. When frying pans were used on the front burners, the smaller pan ($\varnothing = 24$ cm) was completely covered by the hood, whereas the larger pan ($\varnothing = 28$ cm) protruded by 0.5 cm.

5.2.2 $PM_{2.5}$ Measurement Equipment

The $PM_{2.5}$ concentration and particle size distribution were measured in the test chamber using a Grimm 11-R Mini Laser Aerosol Spectrometer optical particle counter (OPC), factory calibrated using dolomite dust. It measures particles with diameters between 0.25 and 32 µm and classifies them into 31 size bins. It detects concentrations between 0.1 $\mu g/m^3$ and 100 $\mu g/m^3$, and identifies particle counts of up to 2 million particles per litre, at a sampling frequency of 6 seconds. The OPC was placed below the low ventilation extract grille at 1.25 m above the floor, at point A in Figure 5.1. Section 2.2.1.3 introduced a number of known issues with using OPCs to measure $PM_{2.5}$, the consequences of which are discussed in Section 5.3.

OPCs infer mas concentration by measuring the light scattering effect of particles. The resulting need for mass calibration was introduced in Section 2.2.1.3. Mass estimations vary according to the size, shape, and composition of particles [136]. Additionally, the OPC may under-estimate the mass concentrations if a significant proportion of the emitted particles are smaller than the lower detection limit of the device [137]. Accordingly, gravimetric sampling (GS) was used to determine the true mass concentrations using a filter-based GS for each test meal. Due to

equipment availability, GS was conducted during a single repetition of each test meal. Air was drawn in through a TECORA fine air inlet, a low volume sampler head, and a glass-fibre 47 mm filter at 2.3 m^3/h using Gilian Aircon2 electric pumps for a defined period of time, following EN 12341 [133]. The volumetric flow rates were checked with a calibrated Yokogawa RAGL rotameter before and after each collection period. Each filter was weighed before and after use in accordance with MDHS 14/4 [222], and so the mass increase, flow rate, and measurement time were used to calculate the mean concentration, as in Section 4.3.1.2. Unlike Section 4.3.1.2 where the CF was determined using linear regression, here the calibration factor was taken to be the mean average of the ratios of the GS and OPC mean concentrations, following Equation 2.2. Three GS devices were placed at the same height as the OPC and 0.5 m to the left, right, and rear of it; see Positions $B - D$ in Figure 5.1.

5.2.3 Test Meals and Cooking Methods

Data reported for the Dutch National Food Consumption Survey (DNFCS) [223], a periodic survey of food consumed by the Dutch population, was used to derive a typical portion size. The DNFCS weights and aggregates data by age and sex, where each group is designed to be representative of age, region of the country, degree of urbanisation, and education level. The selected portion size was in line with those reported by the DNFCS for the $31 - 50$ age group for men ($n = 348$) and women ($n = 351$) because this age group covers the majority of the adult Dutch population and thus indicates typical adult portion sizes. Four meals were chosen comprising carbohydrates, vegetables, and meat-based protein sources, because the DNFCS indicates that $< 7\%$ of the population has special eating habits that include vegetarianism. The proportions of meat, potatoes, and vegetables were guided by the median mass eaten per consumption day by men and women aged $31 - 50$ years.

These data were used to formulate meal types that could be cooked using a stove with a high degree of repeatability.

5.2.3.1 Test Meal Descriptions

The ingredients of the four meals are given in Table 5.1 and photos of the meals and their components are included in Figure E.4. These meals were selected because they are typically Dutch and broadly European. Meal 1 was a reference meal whose emissions could be compared with the others. The ingredients for Meals 1 and 2 were informed by the DNFCS [223], and the types of meat were consistent with common Western cooking ingredients [34]. The mass of each solid ingredient and volume of each liquid were constant and estimated for a median man and woman. All ingredients were supermarket brand basic ingredients except for the *straight to wok* noodles, which were a popular brand. Fresh ingredients were refrigerated at 4 °C before cooking whereas the canned and dry ingredients, oil, and additives were all stored at room temperature. Solid ingredients were weighed using a Zhongshan Camry EK9210K electronic kitchen scale, and the salt weighed with a Mettler AM50 microbalance. Liquid ingredients were measured using a 25 mL measuring cylinder, and 250 and 1000 mL beakers.

5.2.3.2 Cooking Equipment

A cooking protocol was developed based on gas flow rates and cooking time, following Lunden *et al.* [181]. The gas flow rate was controlled with the cooker controls and monitored using 2 parallel Bronkhorst F-201EV mass flow controllers. It was displayed in real time, summed, and adjusted to meet required flow rates. Pan temperatures were identified during preliminary tests using a ThermaCAM™ SC640 FLIR thermal camera following Buonanno *et al.* [201]. The gas flow rate to the stove along with the time taken to reach the required temperature were recorded, and gas flow rate and time were then used to indirectly control the pan

TABLE 5.1: Cooking ingredients and methods

Meal	Ingredient	Measure	Cooking Instructions
1. Reference meal	Chicken breast fillet	200 g	Shallow fry in olive oil
28 min	Olive oil	10 mL	for the chicken
6 repetitions	Pre-sliced pre-cooked potatoes (5-10 mm thick)	330 g	Fry in olive oil
	Olive oil	50 mL	for the potatoes
	French/green beans	280 g	boil in water
	Water	750 mL	for the beans
2.	Chicken breast fillet	200 g	Shallow fry in olive oil
28 min	Olive oil	10 mL	for the chicken
6 repetitions	Potatoes sliced in half	330 g	Fry in olive oil
	Water	600 mL	for the potatoes
	French/green beans	280 g	boil in water
	Water	750 mL	for the beans
3. Pasta Bolognese	Dried farfalle durum wheat pasta	150 g	Boil in water
28 min	Water	1500 mL	for the pasta
6 repetitions	Smoked lean bacon lardons (24% fat[a])	125 g	Fry in olive oil
	Chopped onion	115 g	Fry in olive oil
	Finely chopped garlic	20 g	Fry in olive oil
	Olive oil	10 mL	for the fried ingredients
	Minced/ground beef ($\leq$ 12% fat[a])	200 g	Fry in own fat
	Tinned.canned chopped tomatoes	400 g	Add to fried ingredients
4. Stir-fry	Pre-sliced chicken breast	200 g	Stir-fry in olive oil
17 min	Olive oil	10 mL	for the chicken pieces
6 repetitions	Pre-chopped fresh vegetables:	330 g	Stir-fry in olive oil
	White cabbage	27%[a]	
	Red pepper/capsicum	20%[a]	
	Leek	20%[a]	
	French/green beans	20%[a]	
	Bean sprouts	13%[a]	
	Straight to wok Noodles	150 g	Stir-fry in olive oil
	Olive oil	20 mL	for the vegetables

All ingredients were fresh unless indicated and had not been frozen and defrosted. No seasoning was used. The olive oil was 95% refined and 5% extra virgin[a]. The prefix "pre" shows ingredients purchased in the described form. Symbol [a] denotes data taken from packaging.

temperature. Four pans were used: (i) a TEFAL Titanium Pro 28 cm non-stick stir-fry pan; (ii) a TEFAL Talent Pro 24 cm non-stick frying pan; (iii) a BK Conical Glass stainless steel 2 L saucepan; and (iv) a BK Conical Glass stainless steel 1.5 L saucepan. Hereon, the frying pans are denoted by their diameter and the saucepans by their volume. The saucepans were covered by a lid during cooking.

Before each test, the pans and cooking materials were cleaned in warm water with standard washing-up liquid (dish soap), rinsed with tap water, and dried. At the end of each cooking period, the burners were turned off and a lid was placed on any frying pan to prevent continued emissions, and to give a clear end to the α–period. The $PM_{2.5}$ concentrations in the test chamber were monitored under constant conditions for a further 30 minutes. Between tests, the chamber was purged of $PM_{2.5}$ by increasing the exhaust ventilation rate and opening the door to the laboratory

TABLE 5.2: Overview of measurements to investigate source strength and exposure reduction potential

Experiment name	N	Cooking duration (min)	Ventilation rate (m^3/h)
Meal 1 (*reference*)[a]	6	28	75
Meal 2	6	28	75
Meal 3	6	28	75
Meal 4[2]	6	17	75
"Blanks"	2	28	75
Meal 1 – Margarine	5	28	75
Meal 1 – Stainless steel pan	5	28	75
Meal 1 – Season meat with salt	5	28	75
Meal 1 (*reference*)[a]	5	28	300
Meal 2	5	28	300
Meal 3	5	28	300
Meal 4	5	17	300
Meal 1 – Frying at backburners	5	28	300

[a]Standard conditions: non-stick pan, frying on front burner, in olive oil gas stove.

until concentrations returned to background levels of $< 1\,\mu g/m^3$.

To investigate the repeatability of emission rates during the cooking period, each meal was cooked 6 times using the low ventilation scenario; see Table 5.2. Gravimetric measurements were made during the final repetition to determine calibration factors; see Section 5.2.2.

5.2.3.3 Cooking Instructions

The steps required to cook each meal are described here but the ingredient measures are only given in Table 5.1 for brevity. In addition, the cooking protocol sheets followed in the laboratory are included in Figure E.5-E.9.

Meal 1 began at minute 0 by heating olive oil in a 28 cm frying pan located on the front left burner with a gas flow rate of 2.5 ± 0.1 L/min. At minute 3, when the pan reached approximately 160 °C, the chicken was added and at minute 4 the gas flow was reduced to 1.3 ± 0.1 L/min. At minute 8, the back left burner was ignited and a 2 L saucepan containing the water an beans was placed over it, giving a total gas flow rate of 4.2 ± 0.1 L/min. At minute 10, the front right burner was ignited and a

24 cm frying pan containing olive oil was placed over it, giving a total gas flow rate 5.3±0.2 L/min. At minute 13, the sliced potatoes were added to the 24 cm frying pan and all ingredients were cooked for a further 15 minutes until the test ended at minute 28. Throughout the cooking period, the chicken was turned every 5 minutes and the potatoes stirred for 30 seconds at 3-minute intervals.

Meal 2 followed a similar method to Meal 1 with one main exception: the potatoes were boiled in water instead of fried. At minute 8, the back left burner was ignited, giving a total gas flow rate of 4.2±0.1 L/min, and a 1.5 L saucepan containing the potatoes and water was placed over it. At minute 13, the front right burner was ignited to boil the French beans, giving a total gas flow rate 5.3±0.2 L/min. In this test, the potatoes were not stirred during cooking. The test ended at minute 28; therefore, the beans were cooked for less time than in Meal 1, but were cooked by the end of the test.

Meal 3 began at minute 0, by heating olive oil in the 28 cm frying pan over the rear right burner with a gas flow rate of 4.5±0.1 L/min. At minute 3, the bacon was added and stirred constantly. At minute 7, the gas flow rate was reduced and the onion and garlic added to the bacon. Simultaneously, the back left burner was ignited under a 2 L saucepan containing water for the pasta, giving a total gas flow rate of 4.1±0.1 L/min. At minute 9, the minced beef was added and the gas flow rate increased to 4.6±0.1 L/min. At minute 13, the tinned tomatoes were added to the mince, mixed thoroughly, and stirred thereafter at 3 minute intervals until the sauce had simmered for 15 minutes in total. At minute 18, the rear right burner was reduced giving a total gas flow rate of 3.9±0.1 L/min, and the pasta was added to the boiling water and cooked for 10 minutes. At minute 21, the back left burner was reduced to give a total gas flow rate of 1.9±0.1 L/min. The test ended at minute 28.

Meal 4 only used the 28 cm frying pan located over the rear right burner with an initial gas flow rate of 4.5±0.1 L/min. After heating the olive oil, at minute 3 the diced chicken was added, turned at minute 4, and the burner reduced to

1.1±0.1 L/min at minute 5. At minute 8, the chicken was removed from the pan, the total gas flow rate was increased to 4.4±0.1 L/min, and the olive oil was added. At minute 10, the vegetables were added, spread thinly, and stirred continuously. At minute 15, the gas flow rate was reduced to 4.4±0.1 L/min, and the chicken and noodles added to the pan and cooked until the test ends at minute 17.

5.2.3.4 Additional Tests

Four additional set of tests were conducted to investigate specific areas of interest, which are reported in Section 5.3.4. Firstly, to investigate $PM_{2.5}$ emission rates from the gas stove, two *blank* tests were conducted; see Table 5.2. These followed the gas flows and timings from the reference meal (see Section 5.2.3.3) and were conducted under the low ventilation conditions (see Section 5.2.1), but neither was food cooked nor pans heated.

Secondly, three tests investigated factors that the literature indicated might affect $PM_{2.5}$ emissions during cooking; see Table 5.2. The reference meal was used with three separate substitutions: (i) any olive oil used for frying was substituted with *Croma* brand "Bakken en Braden" liquid margarine to investigate the findings of Torkmahalleh *et al.* [202] who found that particle emissions varied with oil type; (ii) the non-stick frying pans were replaced by stainless steel pans; and (iii) the chicken was seasoned with 1 g of salt before frying, to investigate whether the findings from Torkmahalleh *et al.* [205] could be applied to realistic cooking methods. Here, Torkmahalleh *et al.* found that adding salt to oil before heating reduced particle emissions under controlled laboratory conditions.

Thirdly, to investigate the reduction potential of extracting the $PM_{2.5}$ at source, the 4 test meals were prepared using the high ventilation scenario with air solely extracted through the cooker hood.

Finally, Lunden *et al.* [181] found higher particle CEs when frying on the back burners of a stove and so this was investigated using the reference meal, and high ventilation scenario, with the frying pans relocated to the back burners; see Table 5.2.

5.2.4 Data Processing and Statistical Analysis

The measurements of $PM_{2.5}$ concentration over time for each test were used to compute a source strength and emission rate. Several methods of calculating an emission rate are described in the literature, see Section 2.5.4. These take the *mass balance* model of Ott *et al.* [44] as their basis, but vary their assumptions about the test conditions or the emission characteristics [44]. The most common method, used in Chapter 4, assumes a constant emission rate and either uses the measured peak concentration [199], or estimates the theoretical peak concentration at the end of the α–period [40], to determine the emission rate. Variations of this *peak estimation* method have been used by He *et al.* [35], Dacunto *et al.* [40], Jiang *et al.* [138], Olson & Burke [199], and Lee & Wang [207].

Trial tests indicated that the emission rate was not constant during the cooking period for Meals $1-4$. Therefore, the *area-under-the-curve* method introduced in Section 2.5.4.3 was selected to determine an average emission rate for the cooking period. It uses the principle that the *area-under-the-curve* of a plot of concentration over time t (s), is equivalent to the total mass emitted, the source strength G (µg). The source strength is divided by the emission period to give an average emission rate [44]. Here, the cooking time and emission period, T (s) were considered identical. The average emission rate, $\overline{g(T)}$ (µg/s), is given by

$$\overline{g(T)} = \phi V \overline{C(T)} + \frac{VC(T)}{T} \tag{5.1}$$

where ϕ (s^{-1}) is the total decay rate (the sum of the ventilation, deposition, agglomeration, and evaporation rates), V (m^3) is the test chamber volume, $\overline{C(T)}$ ($\mu g/m^3$) is the average concentration over the emission period, and $C(T)$ ($\mu g/m^3$) is the concentration at the end of the emission period. Then, $G = \overline{g(T)}T$. Generally, $\overline{g(T)}$ is reported in mg/min and G in mg. The model assumes air homogeneity, zone isolation, and perfect mixing [224].

All parameters were known or obtained from measurements, except for ϕ, which was determined from the log-linear regression of concentrations measured during a 30 minute decay period immediately after the emission period; see Section 5.2.3.2 and Dacunto *et al.* [40]. The precision of estimates in ϕ and the assumptions of the model were determined from the regression where a coefficient of determination (R^2) indicated the proportion of the variance in ϕ that was predictable from the measurements of concentration over time. The standard error (α) in ϕ describes uncertainty in its value. The propogated error in $\overline{g(T)}$ is the root of the sum of the squared of uncertainty in each parameter, obtained by peturbing each one by its standard error, following Hughes & Hase [217]. The calculation of ϕ, $\overline{g(T)}$, and G, and the uncertainty in them, was made using a bespoke MATLAB code[1] [213]. The resulting emission rates were compared using a single factor ANOVA with a 5% significance threshold, and two-sample t-tests with Bonferroni correction, which was used to reduce the probability of a Type 1 error. Here, the 5% significance level was divided by the number of tests to give a revised significance of 0.83%. This analysis was conducted using Excel.

The cooker hood has the effect of reducing $\overline{g(T)}$, by removing contaminants before they are allowed to mix in the space, to give a *net* emission rate. To estimate the potential of the cooker hood to do this, $\overline{g(T)}$ was calculated for each meal (see Section 5.2.3.1) under low and high ventilation conditions and compared to give a percentage reduction in $\overline{g(T)}$ for each meal.

[1]The code is available under a creative commons license from DOI: 10.13140/RG.2.2.35826.66244

5.3 Results and Discussion

$PM_{2.5}$ concentrations were measured over time following the methods given in Section 5.2 for each of the four meals described in Table 5.1. This section presents results and discusses the scaling of the data, the emission characteristics for each meal, meal emission rates, confounding factors, the effectiveness of the cooker hood, and further emissions from the gas burners.

5.3.1 Calibration Factors

In order to interpret the measurements of the OPC, a mean calibration factor, $\overline{C}_n$, for each meal was calculated following the method described in Section 5.2.1, and these were used to scale the mass measurements of the OPC described hereon. There was a marked variation by meal: $\overline{C}_1 = 3.9 \pm 0.15$; $\overline{C}_2 = 5.0 \pm 0.096$; $\overline{C}_3 = 2.7 \pm 0.039$; and $\overline{C}_4 = 1.5 \pm 0.045$. Full results from the gravimetric sampling tests can be found in Table E.1. Gravimetric samples were only collected during a single repetition of each meal, with samples collected on filters in 3 locations within the test chamber. Therefore, the uncertainty in the calibration factors does not account for the variation between repetitions, which may be larger. The variation between meals indicates the composition and optical properties of the emitted $PM_{2.5}$, and proportion of particles below the detection limit of the OPC varied between meals. They also show that the OPC consistently under-estimated particle mass when cooking meals, which agrees with Wang *et al.* [136], whose OPCs were used to measure $PM_{2.5}$ in houses. Wang *et al.* suggested that that is caused by coincidence losses, deviations in the refractive coefficient, or the presence of high concentrations of particles that are smaller than the OPC's detection limit. Each filter was removed from its transport cassette and equalised in a climate chamber for over 4 hours prior to its second weighing. Trapped aqueous aerosols are likely to have evaporated and so are a source of calibration factor bias because the OPC is known to detect them. These calibration factors are higher than others found in

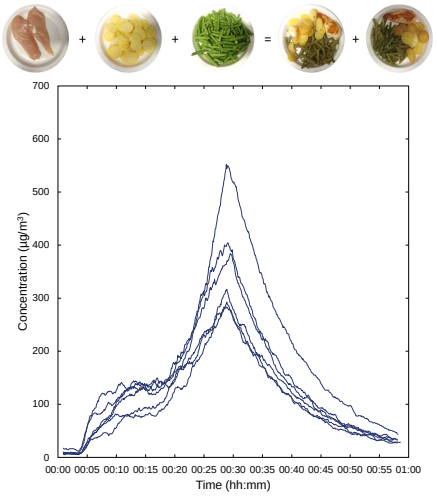
literature [40, 138]. However, the CFs given by Dacunto *et al.* [40] and Jiang *et al.* [138] were not determined using a Grimm calibrated with Dolomite dust and so they are not directly comparable to those given here.

The highest calibration factor was found for Meal 2 whose particle size distribution (see Figure 5.4a) indicates that is emitted the highest proportion of small particles, and so, this supports the theory that the deviation might be partially caused by small particles whose diameters are less than those detected by the Grimm. The differences in $\overline{C}_n$ could also be attributed to changes in the particle composition. Some other affecting factors are discussed in Section 5.3.4.

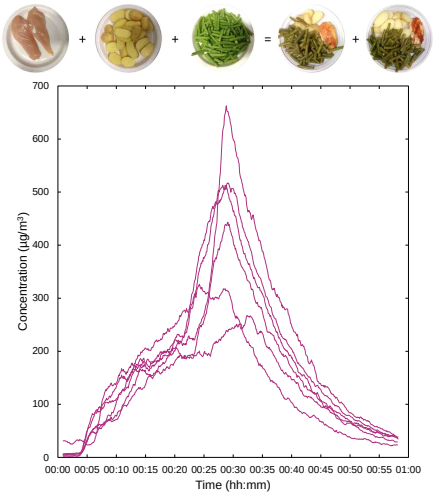
5.3.2 Emission Characteristics

Figure 5.2 shows $PM_{2.5}$ concentrations measured in the test chamber over time during, and after, the cooking period. Consistent gradients indicate steady emission rates and steeper gradients correspond to higher emission rates. The concentrations were logged at 6 second intervals (see Section 5.2.2), however, here they are smoothed over 1 minute intervals for illustrative purposes. Additional figures in Appendix E.2.1 indicate key moments in the cooking process. All meals show repeatable temporal trends in $PM_{2.5}$ concentrations, although there was considerable variance in the magnitude of the concentrations between tests. Cooking involves complex processes, and although the process and ingredients were standardised between repetitions, some level of variation would be expected. In particular, although ingredients were purchased from the same location, their exact composition was not tested.

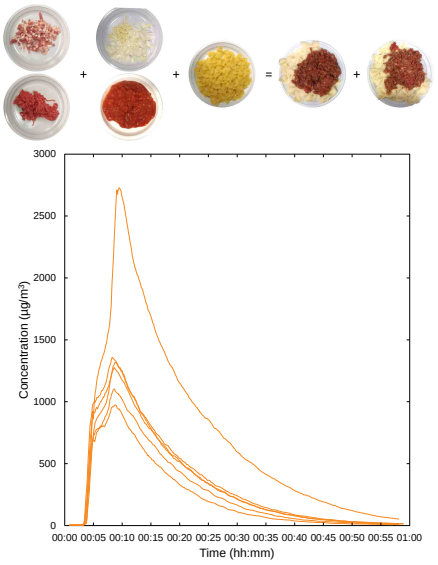
When cooking Meals 1 and 2 (chicken, beans and fried or boiled potatoes respectively), the $PM_{2.5}$ concentrations initially increased steadily for about $20 - 25$ minutes and then increased more rapidly for the remainder of the cooking period. It is not immediately clear what caused this change, but it is possible that over time, the frying pan temperature increased and moisture was removed from the fried



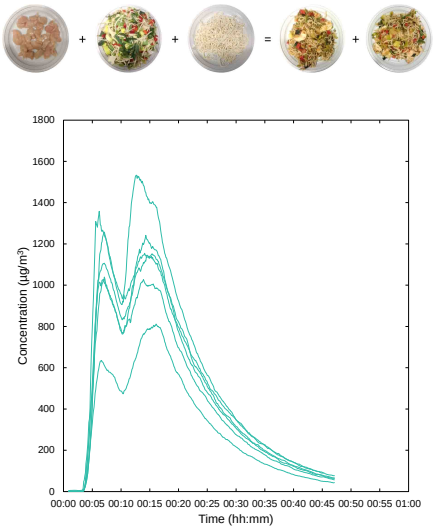
(A) Meal 1



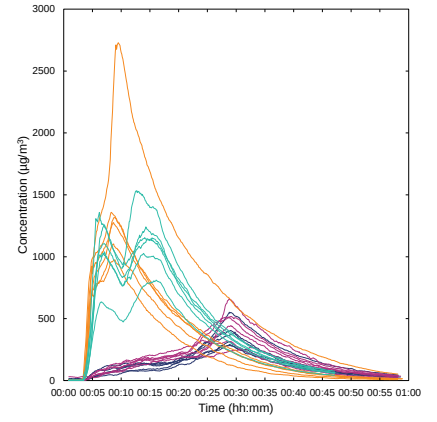
(B) Meal 2



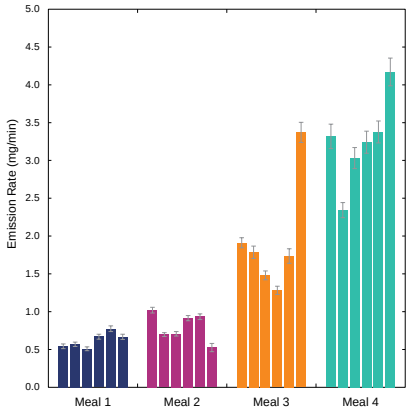
(C) Meal 3



(D) Meal 4



(E) All Meals



(F) $PM_{2.5}$ Emission Rates

FIGURE 5.2: $PM_{2.5}$ concentrations and emission rates for four test meals; for larger copies of the photos see Figure E.4
blue = Meal 1; **pink** = Meal 2; **orange** = Meal 3; **teal** = Meal 4

ingredients leading to Maillard browning. Additionally, it may have been caused by particles below the OPC's detection limit coagulating over time until they reach a detectable size. The consistent changes in gradient exhibited by Figure 5.2a and 5.2b show that Meals 1 and 2 shared similar emission characteristics, and this was expected given their shared ingredients and similar cooking processes. However, Figure 5.2c and 5.2d show that Meals 3 and 4 were distinctly different.

During the cooking of Meal 3 (*pasta bolognese*), the $PM_{2.5}$ concentration initially increased rapidly, correlating with the high temperature frying. The changes between minutes 7 and 13 correspond to the adding of ingredients and changes in gas flow rates. The $PM_{2.5}$ emission rate appears to reduce substantially after the early peak, after the onions and garlic were added and the gas flow turned down. During the final repetition of Meal 3, concentrations were notably higher, as they appeared to increase for longer. The gas supply rate was identical to the other repetitions, and so, the reason for this variation is not clear, but may be related to a variation in the ingredients.

Meal 4 (*stir-fry*) exhibited a high-low-high emission pattern and two distinct peaks. The first peak occurred when frying the chicken, the second when frying the vegetables, and the reduction in concentration occurred immediately after the chicken was removed from the 28 cm frying pan, and prior to the addition of vegetables; see Section 5.2.3.3.

5.3.3 Emission Rates

Figure 5.2f and Table 5.3 show that the mean $PM_{2.5}$ emission rates ($\overline{g(T)}$) and source strengths (G) measured for the four meals described in Section 5.2.3.1 varied between $0.54-3.7\,\mathrm{mg/min}$ and $15-68\,\mathrm{mg}$ with 95% confidence, respectively. Estimated decay rates (see Table E.2) ranged from $4.7\pm0.041\,\mathrm{h}^{-1}$ to $6.1\pm0.042\,\mathrm{h}^{-1}$, although it should be noted that they have little physical meaning in this context because they are a function of particle deposition, agglomeration, evaporation, and

TABLE 5.3: Average emission rates and source strengths for the test meals

	Emission rate, $\overline{g(T)}$ (mg/min)				Source strength, G (mg)			
Test	Meal 1	Meal 2	Meal 3	Meal 4	Meal 1	Meal 2	Meal 3	Meal 4
1	0.54 ± 0.031	1.0 ± 0.038	1.9 ± 0.066^a	3.3 ± 0.016^a	1.5 ± 0.88	29 ± 1.1	55 ± 1.9^a	56 ± 2.8^a
2	0.57 ± 0.029	0.70 ± 0.025	1.8 ± 0.082^a	2.3 ± 0.10^a	16 ± 0.80	20 ± 0.71	50 ± 2.3^a	40 ± 1.7^a
3	0.51 ± 0.025	0.71 ± 0.030	1.5 ± 0.059^a	3.0 ± 0.14^a	14 ± 0.71	20 ± 0.83	41 ± 1.6^a	52 ± 2.4^a
4	0.67 ± 0.033	0.91 ± 0.034	1.3 ± 0.055	3.2 ± 0.15^a	19 ± 0.94	26 ± 0.96	36 ± 1.5	55 ± 2.5^a
5	0.77 ± 0.038	0.93 ± 0.038	1.7 ± 0.10^a	3.4 ± 0.14^a	22 ± 1.1	26 ± 1.1	49 ± 2.7^a	58 ± 2.5^a
6	0.67 ± 0.035	0.52 ± 0.054	3.4 ± 0.13^a	4.2 ± 0.18	19 ± 0.98	16 ± 1.7	95 ± 3.7^a	70 ± 3.1
$\mu \pm \alpha_g$	0.62 ± 0.041	0.80 ± 0.076	1.9 ± 0.30	3.2 ± 0.24	17 ± 1.1	23 ± 2.0	54 ± 8.6	55 ± 3.9
σ	0.10	0.19	0.74	0.59	2.8	4.9	21	9.6

[a] Particle count exceeded 2 000 000 particles/L.
μ = arithmetic mean; α = standard error; σ = standard deviation

other processes, and because the mixed volume of air may not equal the room volume. The volume term in Equation 5.1 is an important source of bias because the room volume applied included cupboards (10% of the room volume), people, and equipment and assumes that the $PM_{2.5}$ is equally mixed within all of these entities. However, it is impossible to determine the validity of this assumption with the measurements made, and so it is possible that the bias in $\overline{g(T)}$ and G could be up to 10% of their values. Volume bias is rarely considered; for example the emission rates reported in Section 2.5.1 [32, 35, 36, 40] all give a volume but do not describe its calculation. Accordingly, future measurements of emission rates and G should seek to minimise the difference between mixed and space volumes. The coefficients of determination for the decay rate were $R^2 > 0.97$ for Meals $1-4$ indicating excellent mixing; see Section 5.2.4 and Sherman [224].

Similar variance in emission rate was found by Fortmann *et al.* [32] who measured 2.92 mg/min when stir-frying using an electrically heated hob, and 3.36 mg/min and 1.54 mg/min when stir-frying ($n = 2$) on a gas burner, which are comparable to the mean $\overline{g(T)} = 3.2 \pm 0.24$ mg/min for Meal 4. Fortmann *et al.* also cooked a full meal using an oven and cooktop and measured the emission rate to be 2.45 mg/min, which lies between the mean emission rates of Meals 3 and 4. Dacunto *et al.* [40] cooked three single dishes in an aluminium frying pan heated on an electric hot plate. First, they measured an emission rate of 0.4 mg/min and $G = 5.7$ mg for chicken, vegetables, and soy sauce stir-fried in olive oil ($n = 2$), which are less than those for all meals cooked here. However, when they pan fried

chicken drumsticks or thighs (on the bone with skin on) for $16-28$ minutes ($n = 6$), the emission rate was $2.5\pm 0.9\,\mathrm{mg/min}$ and G was $62.2\pm 16.9\,\mathrm{mg}$. This emission rate lies between the means for Meals 3 and 4, whereas this G is greater than those for Meals $1-4$. This might suggest they cooked for longer, however, the listed cooking times were shorter. Instead, the higher ratio of G to $\overline{g(T)}$ might be because Dacunto *et al.* used the *peak estimation* method to calculate emission rates, which assumes a constant emission rate. Dacunto *et al.* also panfried chicken breast in olive oil so that it was $25-50\%$ charred, here $g = 15.2\,\mathrm{mg/min}$ and $G = 289\,\mathrm{mg}$. These high values were most likely caused by the charring, whereas Meals $1-4$ only experienced Maillard browning. He *et al.* [35] derived emission rates for a range of cooking events, including complex and simple meals, from measured $PM_{2.5}$ mass concentrations over a 48 hour period in 15 domestic kitchens using an optical device without a calibration factor. General cooking ($n = 37$) gave a median g of $0.11\,\mathrm{mg/min}$ ($\sigma = 0.99\,\mathrm{mg/min}$), frying ($n = 4$) gave a median $2.68\,\mathrm{mg/min}$ ($\sigma = 2.18\,\mathrm{mg/min}$), and stove cooking gave a median of $0.24\,\mathrm{mg/min}$ ($\sigma = 1.29\,\mathrm{mg/min}$). The lack of control over ventilation rates and emission periods, and the lack of a calibration factor, mean there were large uncertainties in these values, but the frying g broadly agree with the emission rates from the meals cooked here whereas the *cooking* and *stove* events were much lower. Nasir & Colbeck [165] followed a similar method to He *et al.* [35] to estimate emission rates from measured concentrations in UK dwellings. Nasir & Colbeck reported emission rates for frying, $1.47\pm 0.82\,\mathrm{mg/min}$ ($n = 10$), and boiling, $1.00\pm 0.21\,\mathrm{mg/min}$ ($n = 15$). Both are between the mean $\overline{g(T)}$ for Meals 2 and 3, although these results are subject to the same limitations as those reported by He *et al.* [35]. Nevertheless, it is reassuring that the values of $\overline{g(T)}$ and G given in Table 5.3 appear plausible given the context provided here.

The measured values of $\overline{g(T)}$ and G can also be compared with those derived from in situ measurements of multiple household activities that include cooking to give a broader understanding of their significance. Chan *et al.* [36] calculated emission

rates for 836 cooking and non-cooking events in 18 dwellings in California, finding a mean emission rate and source strength of 1.72 mg/min and 30 mg, respectively, which are broadly similar to those in Table 5.3. Dacunto *et al.* [40] identified emission rates and source strengths of 0.1 mg/min and 1.4 mg for oven cooked frozen pizza, 9.5 mg/min and 72.5 mg for the toasting of bread ($90-95$% charred), 1.6 mg/min and 18.3 mg for fried salmon, 2.1 mg/min and 24.3 mg for fried pork chop, 3.8 mg/min and 19.9 mg for cigarette smoking, 1.3 mg/min and 16.9 mg for the burning of stick incense and 16.4 mg/min and 215.4 mg for an open fire. For an equivalent release period, the $\overline{g(T)}$ for Meals $1-4$ were greater than those Dacunto *et al.* found for cooking pizza, less than cigarette smoking, and substantially less than an open fire.

Table 5.3 indicates that the particle counts exceeded 2×10^6 particles/L in 10 of the 24 tests. Here, the Grimm OPC may experience *coincidence* errors where multiple particles may be seen as one larger particle. One might expect coincidence errors to affect the regression analysis, as the rate of decay could be expected to change as the concentrations decreased reducing the coincidence rate, yet the values of R^2 were close to unity and $\alpha < 1\%$ of ϕ. For a further discussion of coincidence errors see Section 5.3.6.

The emission rates in Table 5.3 were calculated using the *area-under-the-curve* method to account for a variable emission rate; see Section 5.2.4 and Section 2.5.4.5 for justification. However, several other studies [35, 36, 40] applied the *theoretical peak estimation* method that assumes a constant emission rate, which Figure 5.2 shows to be untrue when cooking these meals. For a direct comparison, this method was applied to the concentration data from these tests, following the method in Section 4.3.1.3, and the constant emission rate g was estimated to be $20-60$% higher than $\overline{g(T)}$ for Meal 1 and 2, up to 70% lower for Meal 3, and between 0% and 12% lower for Meal 4. Clearly, there were significant and non-uniform differences between the two methods. A significant difference relates

to the role of the peak concentration, and this time at which it occurred. Meal 1 and 2 peak concentrations occurred close to the end of the emission period. However, the estimated g value was higher that $\overline{g(T)}$ because the theoretical peak estimation method implicitly assumes a constant emission rate throughout time T, where concentrations rise to $C(T)$ at a consistent rate. Figures 5.2a and 5.2b show that emissions appeared to increase towards the end of the cooking period. C_p occurred early in the 28 minute emission period for Meal 3, and therefore $C(T) < C_p$, which caused the lower estimated g from the peak estimation method. Finally, during Meal 4, the highest concentrations were measured at the end of the test period. However, the theoretical peak method still estimated a lower g as this method effectively ignored the first observed peak concentration observed whilst frying the chicken. The *area-under-the-curve* method was appropriate in this context because it is exact, and also general because it makes no assumptions about the change in the emission rate over time [44]. Accordingly, it is most appropriate to apply this method to the cooking of meals and as such it should also be used in future studies of this type for accuracy and to ensure a fair comparison between tests.

5.3.4 Factors Affecting Emission Rates

The emission rates given in Table 5.3 further highlight the general repeatability of the tests, which is encouraging given the number of unknown or uncontrollable factors involved in the cooking of foods. Table 5.3 also shows there were differences between meals, even when they had similar ingredients. For example, Meals 1 and 2 differed only by the cooking method applied to the potatoes; fried in Meal 1 and boiled in Meal 2. Here, Meal 1 had the lowest emission rates even though frying is known to be a strong source of $PM_{2.5}$ emissions [200]. Boiling the potatoes emitted water vapour, and increased humidity is known to affect the performance of light scattering measurement devices [147]. Additionally, high RH has been linked to the hygroscopic growth of particles [147, 225], and it is possible that boiling the potatoes

created aqueous aerosols that were counted by the OPC. Measurements of RH may have indicated any measurement errors, and this remains a potential confounding factor as RH was not measured. Furthermore, frying is associated with the emission of ultrafine particles, whose diameters are below the detection capability of the OPC [34, 201].

A single factor ANOVA indicated the mean emission rate for all meals was not the same ($p < 0.05$). Two-sample *t*-tests with Bonferroni correction indicated that the emission rates for Meal 1 and Meal 2 were not significantly different ($p > 0.0083$). This suggests that the emissions from frying might have had less influence on the overall emission rate when cooking meals or there are other experimental explanations that were not measured, such as pan temperature. These *t*-tests also found the emission rates from Meals 1 and 3 were significantly different, and that Meal 4 emissions differed from all others ($p < 0.0083$), but those for Meal 2 and 3 were not significantly difference ($p > 0.0083$).

Meal 1 was varied from the base case (see Section 5.2.3.4) in three ways: (i) using liquid margarine instead of olive oil; (ii) using a stainless steel pan instead of a non-stick pan, and; (iii) by adding salt. Figure 5.3 and Table 5.4 show that frying in a stainless steel pan had an immediately obvious effect on the emission rates, with a 940% increase in the mean emission rate. Given that the same volume of oil and mass of the ingredients were used for all tests, the higher emission rates may be a function of the thermal conductivity of the pans, their surface temperatures, and the adhesion between the food and the pan. Here both the chicken and the potatoes were observed sticking to the stainless steel pan in some tests and the surface of the pan charred, see Figure E.15, which could have been reduced by adding more oil, itself a known source of $PM_{2.5}$. This suggests that using a non-stick pan can minimise $PM_{2.5}$ emission during frying. The new stainless steel pan produced the highest emission rate, which then decreased with each subsequent repetition. This indicates there may be an ageing effect that was a function of the changing

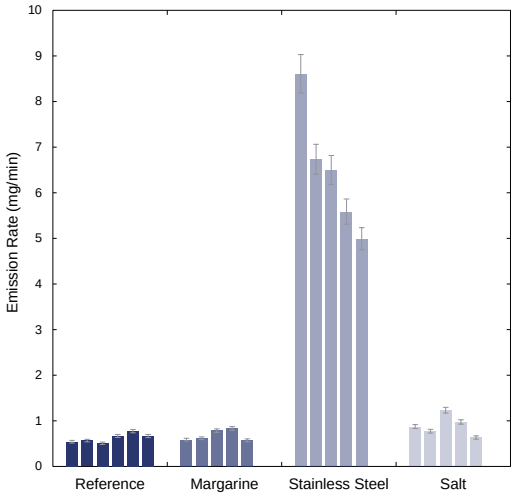


FIGURE 5.3: Influence of factors potentially affecting emissions for Meal 1

properties of the pan's surface, indicated in Figure E.15, which may have continued with further repetitions.

The tests with the liquid margarine and with salt also showed an increase in the mean emission rate of 11% and 47% respectively, when compared with the reference meal. A t-test suggested these changes were non-significant ($p > 0.05$). However, these small differences cannot be ruled out and may be detected by further tests. These results disagree with the findings of Torkmahalleh *et al.* [203, 205] for salt. Torkmahalleh *et al.* [203, 205] added salt to oil before heating, whereas in these tests the salt was added to the chicken before frying. A better comparison may be found with Torkmahalleh *et al.* [206], who found higher particle emissions for grilled salted

TABLE 5.4: Influence of factors potentially affecting emissions

	Meal 1 Emission Rate, $\overline{g(T)}$ (mg/min)				Meal 1 Source Strength, G (mg)			
Test	Reference	Margarine	Steel	Salt	Reference	Margarine	Steel	Salt
1	0.54 ± 0.031	0.59 ± 0.029	8.6 ± 0.42^a	0.87 ± 0.043	15 ± 0.88	17 ± 0.82	240 ± 12^a	24 ± 1.2
2	0.57 ± 0.029	0.62 ± 0.030	6.7 ± 0.33^a	0.77 ± 0.043	16 ± 0.80	17 ± 0.85	190 ± 9.2^a	22 ± 1.2
3	0.51 ± 0.025	0.79 ± 0.038	6.5 ± 0.32^a	1.2 ± 0.062	14 ± 0.71	22 ± 1.1	180 ± 8.9^a	35 ± 1.7
4	0.67 ± 0.033	0.83 ± 0.041	5.6 ± 0.28^a	0.97 ± 0.048	19 ± 0.94	23 ± 1.2	160 ± 7.8^a	27 ± 1.3
5	0.77 ± 0.038	0.57 ± 0.031	5.0 ± 0.24^a	0.63 ± 0.036	22 ± 1.1	16 ± 0.86	140 ± 6.8^a	18 ± 1.0
6	0.67 ± 0.035				19 ± 0.98			
$\mu \pm \alpha_g$	0.62 ± 0.041	0.68 ± 0.054	6.5 ± 0.62	0.90 ± 0.10	17 ± 1.1	19 ± 1.5	180 ± 17	25 ± 2.8
σ	0.10	0.12	1.4	0.23	2.8	3.4	39	6.3
Increase		9%	940%	44%		9%	940%	44%

[a] Particle count exceeded 2 000 000 particles/L.
μ = arithmetic mean; α = standard error; σ = standard deviation

meat than unsalted meat. Table 5.4 shows a small increase in emissions, similar to Torkmahalleh *et al.* [206], however it was not statistically significant. Additionally, the change in emission rate when margarine was also not statistically significant, despite previous suggestions that the oil type is a significant factor [37].

Custom calibration factors were not obtained for each of these variations. In the absence of data $\overline{C}_1 = 3.9$ was used for all three variations, which is a limitation and source of uncertainty.

5.3.5 Cooker Hood Capture Efficiency

The 4 meals were prepared while using an extracting cooker hood located 70 cm above the burners (see Sections 5.2.3.4 and 5.2.4). Table 5.5 gives the resulting emission rates and reductions in the means when compared to those for the main tests in Table 5.3. The percentage reductions in Table 5.5 are an equivalent metric to the CEs defined by Lunden *et al.* [181]. The reductions were $> 90\%$ for all four meals. Additionally, Meal 1 was prepared with the fried components cooked on the back burners, closest to the wall. This resulted in slightly higher reductions that, when tested using a two-sample t-test ($p > 0.05$), were statistically non-significant. This was surprising because Lunden *et al.* [181] found particle CEs of $4 - 39\%$ and $70 - 99\%$ for stir-frying on front and back burners, respectively. Singer *et al.* [183] tested 15 different hoods and reached the same conclusion. Both Lunden *et al.* [181] and Singer *et al.* [183] reported variable the front burner coverage from the hoods. For the two hoods with better burner coverage, Lunden *et al.* measured particle CEs $60 - 80\%$ and $< 60\%$ for stir-frying on the front burner, compared with CEs close to 100% for frying on the rear burner. Of the 15 hoods studied by Singer *et al.*, two had coverage $> 75\%$ during all tests, suggesting good coverage of the front burners. CEs for these hoods measured between 75% and 100%. The exact coverage provided by the hoods was not reported in either study, and both measured some CEs lower than those measured here, even at higher exhaust flow rates. It is

possible that the high emission reduction found here was due to the good coverage of the front burners by the hood and by the presence of a damp buffer [185]. The small volume of the test chamber encourages a closed-loop airflow pattern that allowed particulates to be captured by the hood long after they were emitted having circulated around the chamber, and so a CE determined this way is biased and under-estimates occupant exposure risk. These inherent biases in the technique can be overcome by cancelling out the impact of room concentrations. In theory, one way to do this is to increase the chamber volume and ventilate the chamber far from the hood, or to conduct tests in a full-scale residence [176]; unfortunately, this is impractical to do without introducing new systematic errors, such as poor mixing. A better method is to use a steady state capture efficiency test method [186] that ignores uncaptured pollutants. The method used here may indicate the actual performance of the cooker hood in homes with a kitchen with similar volume to the test chamber, which are common in the English housing stock [226], but it may be less indicative of performance in houses with larger kitchens or in open-plan living spaces where there is less air circulation.

TABLE 5.5: Average emission rates and emission reduction with cooker hood use

	Emission Rate, $\overline{g(T)}$, (mg/min)				Source Strength, G (mg)			
Test	Meal 1	Meal 1 BB	Meal 2	Meal 3	Meal 4	Meal 1	Meal 1 BB	Meal 2	Meal 3	Meal 4
1	0.027 ± 0.0043	0.040 ± 0.0020	0.023 ± 0.0036	–[a]	0.093 ± 0.038	0.75 ± 0.12	1.1 ± 0.056	0.64 ± 0.10	–[a]	1.6 ± 0.65
2	0.064 ± 0.017	0.029 ± 0.0022	0.053 ± 0.0056	0.0068 ± 0.0010	0.072 ± 0.022	1.8 ± 0.48	0.80 ± 0.062	1.5 ± 0.16	0.19 ± 0.029	1.2 ± 0.38
3	0.031 ± 0.0039	0.053 ± 0.011	0.071 ± 0.017	0.0080 ± 0.00094	0.038 ± 0.017	0.87 ± 0.11	1.5 ± 0.31	2.0 ± 0.47	0.22 ± 0.026	0.65 ± 0.030
4	0.024 ± 0.0045	0.024 ± 0.0056	0.015 ± 0.0021	–[a]	0.027 ± 0.006	0.68 ± 0.13	0.68 ± 0.16	0.42 ± 0.060	–[a]	0.45 ± 0.10
5	0.062 ± 0.012	0.026 ± 0.0039	0.037 ± 0.0039	–[a]	0.23 ± 0.075	1.8 ± 0.25	0.73 ± 0.11	1.0 ± 0.11	–[a]	3.9 ± 1.3
$\mu \pm \alpha_{\overline{g}}$	0.042 ± 0.0089	0.034 ± 0.0054	0.040 ± 0.010	0.0074 ± 0.00060	0.091 ± 0.036	1.2 ± 0.25	0.96 ± 0.15	1.1 ± 0.29	0.15 ± 0.04	1.6 ± 0.61
σ	0.020	0.012	0.023	0.00084	0.081	0.56	0.34	0.64	0.024	1.4
Reduction	93%	94%	95%	99.6%	97%	93%	94%	95%	99.7%	97%

[a]Concentrations too low to estimate decay rate. BB = back burners; μ = arithmetic mean; α = standard error; σ = standard deviation

Meals 3 and 4 both resulted in higher reductions than Meals 1 and 2. However, it was only possible to calculated emission rates for two of the five repetitions of Meal 3, because the chamber $PM_{2.5}$ concentrations were too low in other tests. In these tests, the log-linear regression indicated the concentration increased during the decay period, which may be due to incomplete mixing, and so these tests were discounted. The total decay rates (ϕ) ranged from $0.076 \pm 0.043\,h^{-1}$ to $5.6 \pm 0.15\,h^{-1}$ (see Table E.4). Here R^2 values decreased substantially ($R^2 < 0.83$ for accepted tests) and the α of the decay rate increased, indicating a decrease in the mixing quality when compared with the initial tests (see Section 5.3.2). These are limitations of the method, as the chamber concentrations are likely to be low when using a functioning cooker hood with a high capture efficiency. Particles will have deposited on surfaces during all tests, but the cooker hood may have changed the velocity profile around the cooker, and the resulting deposition rates. The method used here accounts for this change by identifying how much the emission rate was effectively reduced, and so is measuring an *apparent* capture efficiency, rather than a true capture efficiency. This metric is useful because it can be used to estimate indoor $PM_{2.5}$ concentrations from a known source and used to inform regulations, but there is significant uncertainty in the measurements.

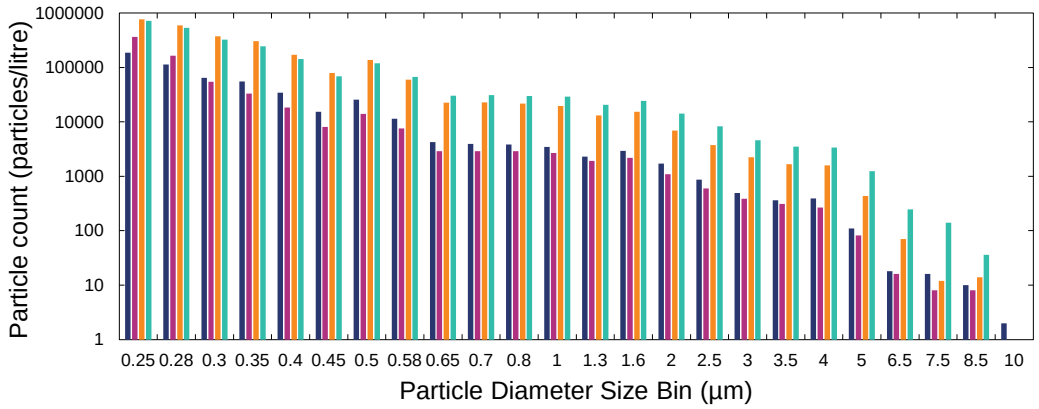
Kim *et al.* [186] have developed a standard method to derive capture efficiencies using ideal gases. Although it does not fully account for particle behaviour, there is lower measurement uncertainty in this test method. Singer *et al.* [183] calculated their hood CEs by using CO_2 as a tracer and measuring its concentration in the exhaust duct. However, Lunden *et al.* [181] followed the method given in Section 5.2.3.4 arguing that particle losses in the hood and ductwork would bias both the measured concentrations and the particle CE. This highlights issues with all methods of measuring cooker hood CE for different pollutants and is an area of ongoing work. The airflow rate through the cooker hood of 83 L/s was high especially when compared with the 50, 30 and 21 L/s required by ASHRAE, in the UK and in the Netherlands, respectively. This flow rate was clearly effective but it

has associated noise and energy penalties. The airflow rate was not varied but the relationship between it and the apparent capture efficiency may be non-linear, and so, it could be possible to reduce the flow rate without significantly affecting the performance. Further work is required to investigate this.

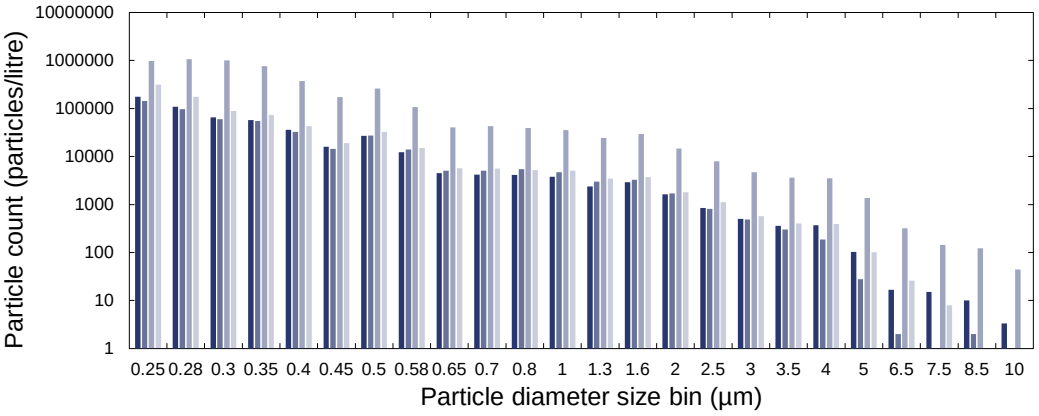
5.3.6 Particle Size Distributions

The distribution of the optical diameter of particles varied over the cooking and decay periods. Figure 5.2 shows that the peak concentration occurred towards the end of the cooking and emission period for Meals 1 and 2. Therefore, at this point the vast majority of particles discharged during the cooking period had already been emitted and so the moment of peak concentration was chosen as a suitable moment to analyse the variance in particle diameter for all meals. Figure 5.4a shows the distribution of particle diameters between 0.25 and 10 µm at the peak concentration time, averaged over the 6 tests for each meal type. It shows that they were similar for all four meals, with more particles emitted in the smaller size fractions. Figure 5.4a shows that when compared with Meal 1, Meal 2 emitted more particles in the smallest size fractions. Figure 5.2 shows that Meals 3 and 4 had higher peak concentrations then Meal 1 and 2, and Figure 5.4a confirms that they also had higher particle counts. The distribution for Meal 4 is weighted more toward the larger particle sizes. This agrees with previous findings that particle diameter increased at higher frying temperatures [34]. Table 5.3 indicates that the OPC may have experienced coincidence errors in some repetitions of Meals 3 and 4. These increase uncertainty in Figure 5.4 where large size bins may be over populated and smaller bins under populated in affected meals.

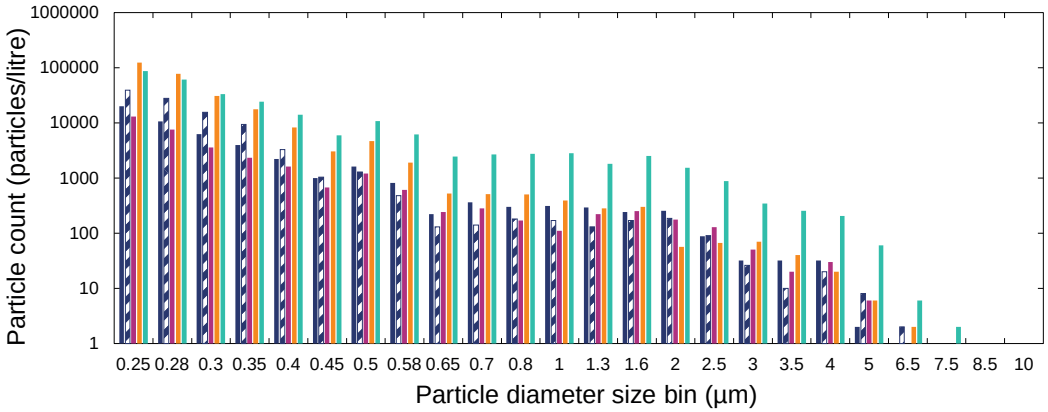
The three variations in the preparation of the Meal 1 base case altered the emission rates and the particle size distributions, see Figures 5.3 and 5.4b respectively. Using liquid margarine resulted in lower particle counts in the larger size fractions ($>$ 2.5 µm in diameter). Frying in stainless steel resulted in higher emissions



(A) Base test meals, mean of 6 tests;
blue = Meal 1; **pink** = Meal 2; **orange** = Meal 3; **teal** = Meal 4



(B) Factors potentially affecting emissions, mean of 5 tests;
blue = Meal 1; **dark** = Margarine; **mid** = Steel; **light** = Salt



(C) Reduction potential, mean of 5 tests;
blue = Meal 1; **blue stripe** = Meal 1 Back Burners; **pink** = Meal 2; **orange** = Meal 3; **teal** = Meal 4

FIGURE 5.4: Peak concentration particle size distributions

overall, but the particle size distribution was similar to the base case meal. With the addition of salt, the distribution was weighted toward the smaller size fractions ($< 2.5\,\mu m$).

Figure 5.4c illustrates the particle size distributions when the cooker hood was used. Cooking Meal 1 on the back burners rather than the front burners also changed the particle size distributions within the chamber. Table 5.5 shows the cooker hood captured a slightly greater mass of particles when cooking on the back burners. However, the differences between Figures 5.4a and 5.4c suggest that the cooker hood captured a greater number of smaller particles ($< 0.4\,\mu m$) when frying on the front burners. It is not clear why this occurred, but the particle size distributions are compared at a single moment in time and so may change at other times. Also, more of the larger particles ($> 0.65\,\mu m$) appear to have been captured in Meal 3 than Meal 4, although it is not clear why this occurred.

5.3.7 Gas Burners

$PM_{2.5}$ concentrations were measured during two *blank* tests (see Section 5.2.3.4) where the combustion elements of the Meal 1 preparation were followed for the low ventilation scenario, but no food was cooked. The concentrations were generally $< 1\,\mu g/m^3$ (see Figure E.10), and so, it was impossible to identify any decay at the end of the test once the stove burners were switched off. This shows that the emissions of $PM_{2.5}$ from the gas burners can be considered negligible and so did not contribute to the temporal variation in $PM_{2.5}$ concentration shown in Figure 5.2 or the emission rates given in Table 5.3. However, gas burners are a known emitter of ultrafine particles and nitrogen oxides [32, 201], and both pollutants are associated with negative health effects [38].

5.3.8 Impacts

The $PM_{2.5}$ source strengths and emission rates of the four meals suggest that cooking for a prolonged period in a house without adequate ventilation could lead to indoor $PM_{2.5}$ concentrations that exceed those found outside and could negatively affect the health of the occupants; see the health risks discussed in Section 2.1. This is especially likely in airtight dwellings where ventilation may be inadequate, especially during the heating season when occupants may seek to reduce ventilation rates to minimise heating fuel costs. Section 5.3.5 shows that a cooker hood can be used to reduce the $PM_{2.5}$ emissions during cooking, although it is not yet clear what combination of airflow rates and capture efficiencies should be prescribed by standards or norms. Here, the emission rates in Table 5.3 could be used to derive appropriate ventilation rates and capture efficiencies for cooker hoods following the statistical method described by Salthammer [10] and introduced in Section 2.4.3. The emission rates and their standard errors could also be used as stochastic inputs to stock-scale models of housing used to estimate exposure and predict the chronic health impacts from exposure to $PM_{2.5}$ from cooking and the positive changes that may arise from mitigation measures, such as the installation and use of a cooker hood.

5.4 Conclusions

This work provides further evidence that the cooking of meals emits $PM_{2.5}$. The emission rate varied over time as a particular meal was cooked and this variation was likely caused by a range of factors, many of which are not quantifiable. However, frying, the browning of food, the presence of oil or fat, the pan temperature, and the pan type all contributed.

It is possible to reduce $PM_{2.5}$ emissions by using cooking methods that do not brown or char the food and by using a non-stick pan when frying. Other methods

were tested that have been shown elsewhere to affect $PM_{2.5}$ emission rates, such as replacing oil with liquid margarine and adding salt, but were found to only have a minimal effect. A limitation of these tests was the small number of replicates, hence, more repetitions might help identify small variations in emissions.

The apparent capture efficiency of a cooker hood at a particular airflow rate is an indication of the proportion by which it reduces an emission rate to the room. This *net* emission rate of $PM_{2.5}$ from the cooking of meals was reduced substantially by using a cooker hood with good coverage of all burners at a high airflow rate. Although the apparent capture efficiency has the advantage of being derived from the measured $PM_{2.5}$ concentrations, there is significant uncertainty in its measurement caused by systematic biases.

Measuring capture efficiencies using ideal gases under steady state conditions is an ideal test method because it is independent of room dynamics and contaminant interactions. However, in real-world environments cooking is rarely conducted under steady state conditions and its pollutants infrequently act as ideal gases. In particular, these methods do not account for particle behaviour, but they are less uncertain and only measure the ability of a hood to capture pollutants at their source. This first-order approximation is acceptable for rating cooker hoods, but a detailed estimation of occupant exposure inside a dwelling may need to consider these extra factors.

The calibration factors obtained for each meal varied away from unity, and so, the optical properties of the $PM_{2.5}$ emitted by cooking differed from those of the calibration source, here dolomite dust. Therefore, when measuring $PM_{2.5}$ concentrations in domestic kitchens using an optical measurement device calibrated using dolomite dust, the measured concentrations must be corrected using an appropriate calibration factor before negative health effects can be estimated from them with any accuracy. Furthermore, the calibration factors are shown to vary by meal and so it is not possible to use a single factor for all meals

without introducing significant uncertainty. Calibration factors can be obtained either from concurrent gravimetric sampling, or values from the literature can be used with significant uncertainty. It is likely that devices calibrated by sources other than dolomite dust will also require calibration factors.

Chapter 6

Predicted $PM_{2.5}$ Concentrations in the English Housing Stock

The first part of this chapter has previously been presented as a poster and paper at the AIVC conference in 2018:

O'Leary, C. *et al. Estimated distributions of $PM_{2.5}$ concentrations in the kitchens of the English housing stock for infiltration and mechanical ventilation scenarios* in *39th Air Infiltration and Ventilation Centre Conference* (Antibes Juan-Les-Pins, France, 2018), 1090–1099.

In addition, the second part of this chapter has been accepted for publication in:

O'Leary, C. *et al.* Setting the standard: The acceptability of kitchen ventilation for the English housing stock. *Building and Environment*, 106417. ISSN: 0360-1323 (2019)

In both cases, the model inputs have since been updated slightly which has resulted in small changes to the predicted values, however these changes did not significantly effect the main findings.

6.1 Introduction

To this point, the work in this thesis has focused on measurement; including measuring $PM_{2.5}$ concentrations in domestic kitchens, measuring $PM_{2.5}$ emission rates from cooking events, and investigating ventilation based mitigation strategies both in situ and under laboratory conditions. Cooking has been established as a key indoor source of $PM_{2.5}$, and there is some evidence both presented here and elsewhere in literature (see Section 2.3) to suggest that appropriate ventilation methods can be used to reduce indoor concentrations, and, subsequent personal exposure and dose.

Stock scale models and simulation techniques, introduced in Section 2.4, can be used to predict $PM_{2.5}$ concentrations in entire building stocks. In addition, input parameters can be varied to predict the effect of changes to the housing stock, for example ventilation conditions. Many of the modelling approaches introduced in Section 2.4 are computationally expensive, which limits their utility for finding the ideal conditions. Alternatively, Salthammer [10] proposed a statistical approach to generate a probability density function (PDF) of pollutant concentrations using a simple mass-balance model and Monte-Carlo sampling, see Section 2.4.3 for more details. The resulting PDF can then be compared to an appropriate health based pollutant guideline concentrations to predict the percentage of the stock where concentrations remain below the guideline.

Therefore, the aims of this chapter were to develop a statistical model which could be used to provide a rudimentary assessment of the English housing stock, and to assess the suitability of the current English Building regulations given in ADF for removing $PM_{2.5}$ emitted during cooking. The mathematical model, based on Salthammer's method, was developed to predict kitchen $PM_{2.5}$ concentrations for different ventilation strategies using Monte-Carlo sampling techniques. A final aim was to consider the ventilation strategies required to keep $PM_{2.5}$ concentrations

below guideline values in 50%, 75% and 98% of English kitchens, and thus minimise population exposure.

6.2 Steady State Approach

This first approach closely followed the approach outlined by Salthammer [10] to predict steady state $PM_{2.5}$ concentrations in a representative sample of English domestic kitchens emitted by a single cooking source.

6.2.1 Method

In a well-mixed space, where the emission and ventilation rates are constant over time, concentrations tend towards an asymptotic $PM_{2.5}$ concentration, namely the steady state concentration, C_{ss}. As such, C_{ss} is a theoretical concentration that is never reached. The rate at which the indoor concentration approaches steady state concentration is a function of the ventilation and emission rates. In a small space with a low ventilation rate and high emission rate, near steady state concentrations may be reached in only a few minutes, but in larger, well ventilated spaces it can take much longer. Accordingly, three distributions were predicted: (i) kitchen steady state $PM_{2.5}$ concentrations, C_{ss} ($\mu g/m^3$); (ii) the time taken to reach 95% of C_{ss}, T (minutes), and; (iii) the time (minutes) taken to reach the WHO [128] 24-hour mean guideline of $25\,\mu g/m^3$.

A bespoke MATLAB code[1] [213] was used to mathematically predict these concentrations using Monte-Carlo sampling techniques.

[1]The code is available under a creative commons license from DOI: 10.13140/RG.2.2.19049.44645

6.2.1.1 Model

The widely used mass balance model introduced in Section 2.5.4 formed the basis of the calculations, where the concentration at time t, $C(t)$ was defined in Equation 2.6. Under steady state conditions the temporal component of 2.6 tends to $0\,\mu g/m^3$, and as such the $C_{ss}$is defined in Equation 6.1, and $C(t)$ can be defined as a function of C_{ss} as in Equation 6.2. The steady state condition was previously described in more detail in Section 2.5.4.1.

$$C_{ss} = \frac{\lambda_T P C_a}{(\lambda_T + k)} + \frac{(1-\eta)g}{(\lambda_T + k)V} \tag{6.1}$$

$$C(t) = C_{ss} + (C(0) - C_{ss})\, e^{-(\lambda_T + k)t} \tag{6.2}$$

Along with $C_{ss}(\mu g/m^3)$, $C(t)$ ($\mu g/m^3$) is a function of the initial concentration $C(0)$ ($\mu g/m^3$), total ventilation rate, λ_T (s^{-1}), and deposition rate, k (s^{-1}). C_{ss} is itself a function of the ambient concentration, C_a ($\mu g/m^3$), penetration coefficient, P (–), source emission rate, g ($\mu g/s$), capture efficiency, η (–), zone volume, V (m^3), ventilation and deposition rates. In addition, Equation 6.2 can be used to find the time, T, to reach a specific concentration, $C(T)$, as in Equation 6.3.

$$T = \frac{1}{(\lambda_T + k)} \ln\left(\frac{C(0) - C_{ss}}{C(T) - C_{ss}}\right) \tag{6.3}$$

6.2.1.2 Model Inputs

This first model only considered $PM_{2.5}$ from cooking events, and assumed that $PM_{2.5}$ from any previous events has successfully been removed. Therefore, C_a and $C(0)$ were assumed to equal $0\,\mu g/m^3$. As such, the penetration coefficient, P was redundant and was not included as a model input. All other inputs were modelled

probabilistically, each sampled from an appropriate distribution, and were assumed to be independent variables.

Emission Rate, g This first model was the first stage of the model development process, and so a single cooking source of toasting bread was modelled. Emission rates were uniformly sampled from the eCDF defined in Phase 2 of Chapter 4, with $\mu = 0.23\,\mathrm{mg/min}$ and $\sigma = 0.067\,\mathrm{mg/min}$.

Deposition Rate, k A deposition rate of $0.39 \pm 0.16\,\mathrm{h}^{-1}$ was, taken from Ozkaynak *et al.* [170]. This value is based on 1780 personal monitoring measurements in non-smoking residences, and has previously been used in $PM_{2.5}$ modelling studies (see Das *et al.* [37], Milner *et al.* [192], and Hamilton *et al.* [193]). Deposition rates were assumed to be normally distributed, but with a minimum deposition rate of $0\,\mathrm{h}^{-1}$.

Volume, V The English housing stock comprises 22.3 million dwellings, of which a statistically representative sample of 16 150 dwellings is documented in the 2009 English Housing Survey (EHS) [226]. The 2009 data set was not the most recent EHS data available, however, it was selected to match the data set used to derive the infiltration rate distributions (see below). Entries with no recorded kitchen dimension data were ignored, and the remaining data set was considered representative of 99.75% of the housing stock. This data set was used to create an eCDF in MATLAB[2], from which volumes were sampled.

Ventilation Rate, λ_T Three ventilation strategies were considered based on the requirements set out in ADF. These are set out in Table 6.1.

Infiltration was assumed to occur in all scenarios, and was considered to be a function of a measurement of *air permeability*, the airflow rate through adventitious openings at a pressure differential of 50 Pa normalised by dwelling surface area.

[2]The code is available under a creative commons license from DOI: 10.13140/RG.2.2.15694.00326

TABLE 6.1: Ventilation strategies

Strategy		Fan Airflow Rate	Details
A		0 L/s	Infiltration only
B		60 L/s	General extract ventilation
C	20%	30 L/s	Extract through a cooker hood, $\eta = 20\%$
	40%	30 L/s	Extract through a cooker hood, $\eta = 40\%$
	60%	30 L/s	Extract through a cooker hood, $\eta = 60\%$
	80%	30 L/s	Extract through a cooker hood, $\eta = 80\%$

Distributions of hourly infiltration rates in English houses were used in order to explore the maximum possible variability in infiltration. The distributions were generated by DOMVENT3D, an open source[3] implicit model of infiltration and exfiltration through any number of façades [228–232]. It assumes that all façades are uniformly porous and that the pressure distribution over a vertical surface is linear. Further assumptions, merits, limitations, and the corrobotation of its predictions are discussed widely by Jones & Lowe [230] and Jones *et al.* [231], and an analysis of its predictions to its inputs has been undertaken [232]. The infiltration distributions were generated using DOMVENT3D with identical distributions to those described by Jones *et al.* [232].

The majority of English houses share a party wall with another house and, because their permeability is unknown, they were considered to be either impermeable or equally permeable as other walls with equal probability of either assumption.

Under all ventilation scenarios, all ventilation was assumed to occur via infiltration or the mechanical ventilation rates describes in Table 6.1. All other purpose provided openings were assumed to be closed, to model the heating season condition. When the mechanical system was switched off (strategy A), the total airflow rate, λ_T, was equal to the infiltration rate λ_N (h^{-1}). When a mechanical system was switched on, as for strategy B or C, all make-up air was assumed to occur via infiltration or exfiltration. Generally, in a *leaky* house with a high value of air permeability, a small mechanical extract flow rate, λ_M (h^{-1}), creates a small reduction in the external air pressure that increased the infiltration rate and decreases the exfiltration rate by the

[3]The code is available under a creative commons license from DOI: 10.13140/RG.2.2.30311.39844

same magnitude. Therefore, the change to the total airflow rate attributable to the mechanical system is equal to $\lambda_M/2$, so that $\lambda_T = \lambda_N + \lambda_M/2$. In an *airtight* house, a small extract flow rate creates a much larger reduction in indoor air pressure that eliminates exfiltration so that $\lambda_T = \lambda_M$. These limits have been discussed previously [230, 233, 234] and the transition between them in UK houses has been shown to be well modelled by superposition using a power law [230]. However, the exponent is unique to every house and so, in the absence of knowledge, a high exponent is advised. Then, the model of total airflow rate reduces to

$$\lambda_T = max\left\{\lambda_N + \frac{\lambda_M}{2}, \lambda_M\right\} \tag{6.4}$$

which is known as the *half-fan* model. This model was used to determine the total ventilation rate from the probabilistically sampled infiltration rate, λ_N, and the deterministic mechanical ventilation rate, λ_M.

When comparing the differences in hourly infiltration rates in US houses predicted by a physically correct numerical airflow mass-balance model and the half-fan model, the root mean square error was found to be $< 5\%$ [234].

The half-fan model has been shown to be less accurate than an exponential relationship between λ_M and λ_N. However, the exponential model has not been evaluated for the English stock whereas the half-fan model has. In addition, the half-fan model is explicit and, therefore, computationally inexpensive so it can be used to explore a range of ventilation scenarios in a short period of time.

For both strategy A and B, it was assumed the emitted $PM_{2.5}$ instantaneously mixed fully within the space, meaning the capture efficiency, $\eta = 0\%$. In strategy C, to model the additional benefit of extracting at source via a cooker hood, the probabilistically sampled emission rates were reduced by η, which has a value between 0 and 1 where 1 is 100% efficient. Section 2.3.1.1 showed that CEs vary considerably. There is also no information in the literature about typical CEs for

cooker hoods in the English housing stock. Accordingly, four different CEs were modelled deterministically: 20%, 40%, 60% and 80%.

6.2.1.3 Sampling Method

The sampling method followed that described by Das *et al.* [37] and Jones *et al.* [232]. There were four direct input variables to find C_{ss}, which were assumed to be independent of one another. These variables were sampled in sets of 100 using a Latin Hypercube, and each set was applied to predict C_{ss} using Equation 6.1, and the time to reach 95% of C_{ss} and the WHO 24-hour guideline using Equation 6.3. The total sample size increased incrementally by the set size. After each set of predictions was made, the overall arithmetic mean (μ) and standard deviation (σ) were calculated. When the change in μ and σ from one set of samples to the next was $\leq$ 0.01%, the stopping criterion was met, and the total number of samples deemed adequate to have produced a representative PDF.

6.2.1.4 Sensitivity Analysis

A sensitivity analysis was used to test the dependence of steady state $PM_{2.5}$ concentrations on the model inputs. The method followed that of Jones *et al.* [232] and a full description is found in the reference. The method tests for linear (Kendall's tau, Pearson's product moment, linear regression), monotonic (Spearman's rank correlation coefficient, rank-transformed standardised variables), and non-monotonic (Kolmogorov-Smirnov, Kruskal-Wallis) relationships between inputs and outputs. All inputs were ranked according to the magnitude of the regression coefficient. A fundamental requirement of this sensitivity analysis is that all tested inputs are independent of each other, which was also an assumption of the model.

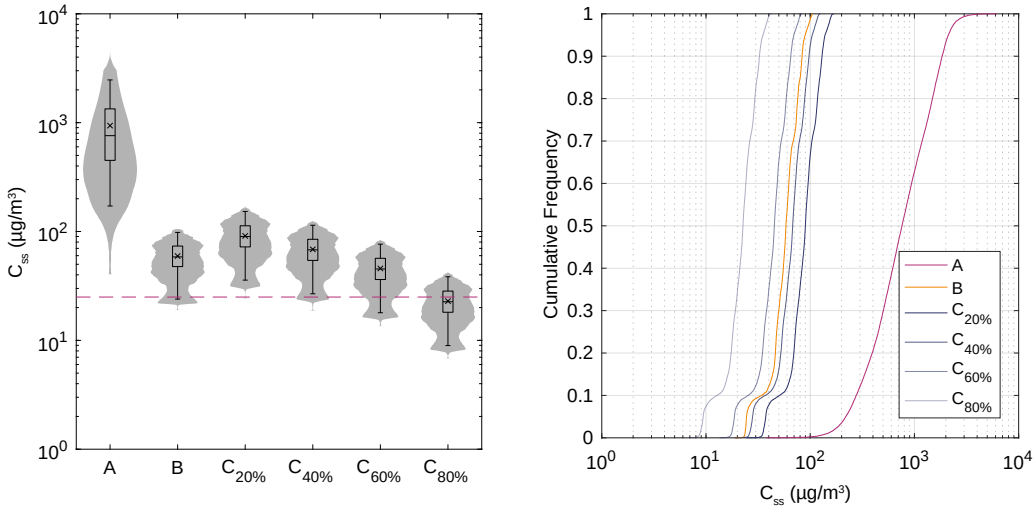
TABLE 6.2: Predicted steady state $PM_{2.5}$ concentrations ($\mu g/m^3$) in English kitchens for ventilation strategies A – C, see Table 6.1

Centile/ Statistic	A	B	C			
			$\eta = 20\%$	$\eta = 40\%$	$\eta = 60\%$	$\eta = 80\%$
2%	171	23.9	35.8	26.8	17.9	8.96
25%	451	47.6	72.2	54.3	36.2	18.1
50%	760	58.5	89.9	67.9	45.3	22.6
75%	1340	73.5	113	84.8	56.7	28.4
98%	2470	98.1	153	114	76.5	38.4
μ	940	59.5	91.1	68.6	45.8	22.9
σ	634	18.3	28.9	21.8	14.5	7.27
GM	743	56.3	85.9	64.7	43.2	21.6
GSD	2.0	1.4	1.4	1.4	1.4	1.4

6.2.2 Results

Figure 6.1 and Table 6.2 show the results for C_{ss} for the three ventilation strategies. For strategy A, where infiltration was the only form of ventilation, C_{ss} concentrations were predicted to exceed the WHO 24-hour guideline $25\,\mu g/m^3$ in all kitchens. With ventilation strategy B, C_{ss} was predicted to exceed the guideline in 94% of kitchens. The best case scenario was for strategy C with $\eta = 80\%$, where C_{ss} was predicted to exceed the guideline in 35% of kitchens. The distributions, indicated by the shapes of the *violins* in Figure 6.1a, appear non-normal for all ventilation strategies. Furthermore, for strategy B and C, the C_{ss} distribution appears multi-modal.

Figures 6.2 and 6.3 show distributions of the predicted time taken to reach 95% of the steady state concentration and the WHO guideline of $25\,\mu g/m^3$, respectively. Increasing the ventilation rate simultaneously reduced C_{ss} and the time required to reach 95% of C_{ss}, and increased the predicted time required to reach $25\,\mu g/m^3$. Figure 6.2 also shows that the capture efficiency η was predicted to have no effect of the time taken to reach a fraction of C_{ss}. This is also defined mathematically as η cancels in the quotient of Equation 6.3. However, this was not the case when predicting the time taken to reach a specific concentration, as in Figure 6.3. Here, increasing η also increased the time taken to reach the guideline concentration as the capture efficiency reduces the fraction of all emitted particles which mix through the



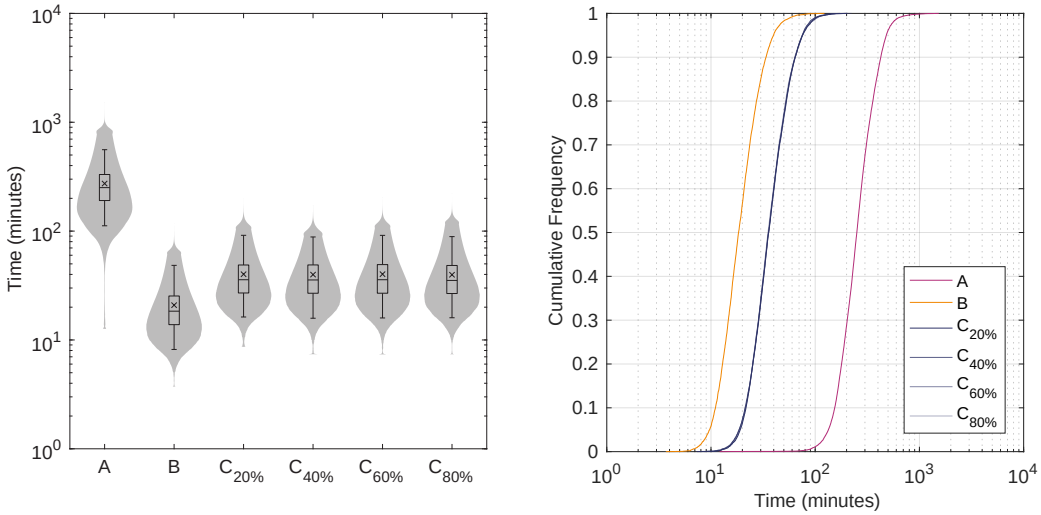
(A) Violin plot (B) Cumulative frequency

FIGURE 6.1: Predicted steady state concentrations in English kitchens for ventilation strategies A – C, see Table 6.1

kitchen volume.

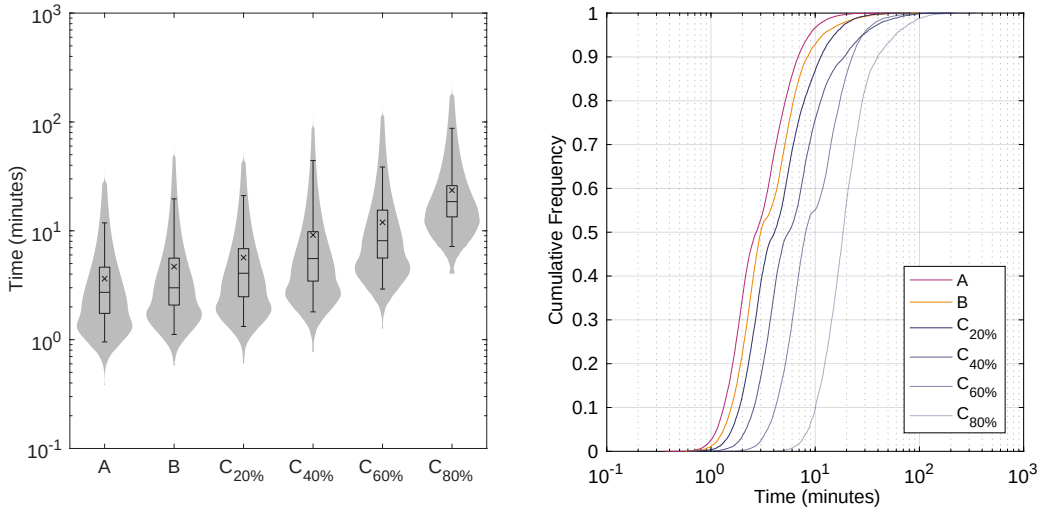
6.2.2.1 Sensitivity Analysis

The sensitivity analysis in Section 6.2.1.4 was used to determine the relative importance of the input variables on the C_{ss} predictions. All inputs were obtained simultaneously by the Latin Hypercube sampling method and so any interactions between them were accounted for. The ranks of each model input are given in Table 6.3, where 1 indicated the most important input, assessed as having highest correlation or regression coefficient. Emission rate, g, was ranked the most influential by all tests, with the deposition rate the least important, a finding that is mathematically sound from Equation 6.1. This further emphasises the importance of $PM_{2.5}$ cooking emission rates in modelling. Almost all the relationships between these inputs and C_{ss} were statistically significant, where $p \leq 0.05$ with one exception, indicated in Table 6.3.



(A) Violin plot (B) Cumulative frequency

FIGURE 6.2: Predicted time (minutes) to reach 95% of C_{ss}



(A) Violin plot (B) Cumulative frequency

FIGURE 6.3: Predicted time (minutes) to reach $25\,\mu g/m^3$

TABLE 6.3: Sensitivity of C_{ss} to model inputs. 1 is the highest rank.

Input\Test	S_{Kend}	S_{Pear}	S_{Spear}	$S_{regress}$	$S_{rankgreg}$	S_{Kolm}	S_{Krusk2}	S_{Krusk5}	$S_{Krusk10}$	$S_{Krusk20}$
g	1	1	1	1	1	1	1	1	1	1
V	2	2	2	2	2	2*	2	2	2	2
λ_N	3	3	3	3	3	3	3	3	3	3
k	4	4	4	4	4	4	4	4	4	4

*indicates tests with non-significant association where $p > 0.05$

6.2.3 Implications and Limitations

The predictions in Section 6.2.2 highlight that the current ADF ventilation requirements for kitchen extract are inconsistent. ADF does not specify a minimum cooker hood capture efficiency that will provide equivalent protection the the general extract flow rate of 60 L/s. Of the capture efficiencies considered, equivalent concentrations were only predicted for $\eta = 60\%$ and above. Existing literature on cooker hood capture efficiencies has reported a wide variation; for example Rim *et al.* [182] measured ultrafine particle CEs ranging from 30% to $> 90\%$. However, the higher CEs were measured at flow rates over 180 L/s, which is six times higher the 30 L/s specified in ADF. Lunden *et al.* [181] measured a similar range of CEs but with a minimum flow rate of 51 L/s. Further investigation and empirical tests are required to investigate whether existing cooker hoods installed in English dwellings meet ADF requirements, or what flow rates and CEs are currently achieved. In addition, according to the 2009 EHS data, only 47% kitchens were fitted with any form of extractor fan [226].

In the best strategy considered, $C_{80\%}$, C_{ss} was predicted to exceed the WHO guideline in 30% of kitchens. However, a considerable limitation of this approach is the use of steady state concentrations, as these are unlikely to be reached for cooking source $PM_{2.5}$ due to the relatively short cooking duration. Adams & White [235] suggested a minimum cooking period of 30 minutes to cook a main meal. With no PPV, a 30 minute source emission period would cause concentrations to approach steady state in $< 1\%$ kitchens, from strategy A in Figure 6.2a, although concentrations would exceed 25 $\mu g/m^3$ in $> 99\%$ kitchens. With strategy B or $C_{80\%}$, a thirty minute emission period was predicted to result in near steady state concentrations in 85% and 34% kitchens respectively, see Figure 6.2. The length of the cooking event is an unknown and was not an input to the model, but is clearly an important parameter.

In addition, the comparisons to the WHO 24-hour guideline are not entirely

appropriate. These comparison do help to put the predicted concentrations into context. However, this guideline is for a 24-hour mean concentration rather than either the theoretical C_{ss}, which is unlikely to be reached, or a peak concentration occurring at the end of a defined cooking period. In addition, external or background concentrations were ignored, whereas these concentrations also contribute to real indoor exposures.

This approach was deemed unsuitable for assessing the ADF requirements in detail because: (i) it did not allow comparison to metrics as it did not predict average concentrations; (ii) only one cooking emission source was considered; (iii) C_a was not included as a model input; and (iv) it did not allow for comparison between the constant and intermittent ventilation rates prescribed in ADF. Therefore, a second model was developed using similar techniques to improve on this first iteration.

6.3 Average Concentrations Approach

The first modelling approach highlighted multiple issues with the Salthammer [10] methodology when applied to cooking source $PM_{2.5}$, primarily due to the short duration of cooking emissions meaning steady state concentrations are unlikely to be reached. In addition, the model over-simplified the problem, considering only a single cooking source where emission rates have previously been found to vary considerably, see Section 2.5 and Chapter 5. Therefore, this second approach sought to improve the model by predicting 24-hour mean concentrations in English domestic kitchens. As well as providing a rudimentary assessment of the English housing stock and the suitability of the English Building Regulations given in ADF for removing $PM_{2.5}$ emitted during cooking, this approach sought to consider the required ventilation strategies, cooker hood capture efficiencies, and ventilation rates to keep $PM_{2.5}$ concentrations below health-based guidelines in 50%, 75% and 98% of English kitchens, and thus minimise population exposure to them.

6.3.1 Methods

The method used a bespoke MATLAB code[4] to run Monte-Carlo simulations to predict 24-hour mean concentrations in a representative sample of English kitchens, based initially on the method outlined by Salthammer [10].

6.3.1.1 Model

Once again, the basis of the model was the widely used *mass balance* model of pollutant concentrations in a single zone from Ott *et al.* [44]. The base equations were the same as those used in Section 6.2.1.1, repeated here for ease. The equation parameters have previously been defined.

$$C_{ss} = \frac{\lambda_T P C_a}{(\lambda_T + k)} + \frac{(1-\eta)g}{(\lambda_T + k)V} \tag{6.5}$$

$$C(t) = C_{ss} + (C(0) - C_{ss})\, e^{-(\lambda_T + k)t} \tag{6.6}$$

The mean concentration, $\overline{C(t)}$ (µg/m^3), between times $t = 0$ and $t = t$, can be found by integrating Equation 6.6 between these limits and dividing by the time period of interest, t. This results in Equation 6.6.

$$\overline{C(t)} = C_{ss} - \frac{C(t) - C(0)}{(\lambda_T + k)\, t} \tag{6.7}$$

All three equations assume constant conditions over time [44]. Therefore, to predict the mean average concentration over a significant period of time, T (seconds), such as a day, it was divided into j distinct sub-periods where the total time is equal to the sum of the durations of each sub-period, so $T = \sum_{i=1}^{j} t_i$. Each sub-period corresponded to a change in the emission or ventilation rates so that the final

[4]The code is available under a creative commons license from DOI: 10.13140/RG.2.2.22404.88963

concentration of each sub-period was also the initial concentration of the next. The overall mean concentration over time T was then calculated as the weighted mean, $\overline{C(t_i)}$, for each sub-period where

$$\overline{C(T)} = \frac{\sum_{i=1}^{j} \left(\overline{C(t_i)} \cdot t_i \right)}{T} \tag{6.8}$$

6.3.1.2 Stocks, Strategies, and Scenarios

Two main housing stocks were considered: a stock of new build houses, and the existing housing stock that includes new houses. This was to reflect the separation in ADF of requirements for work to existing dwellings and new houses, and allowed the differences in kitchen volume and air permeability between the two stocks to be assessed. In addition, a *future* stock was considered. The inclusion of this stock was an attempt to future-proof the work, by considering the total ventilation rate. The future stock was a modified version of the new housing stock.

Six ventilation strategies were tested within the contexts of two scenarios. The strategies were chosen because they are either specified in ADF, or because ADF implicitly allows them.

The first scenario considered the existing requirements of the building regulations to assess their suitability. The requirements for new kitchens were assessed for all three stocks, because existing dwellings have to maintain an existing ventilation strategy whose details were unknown. The six ventilation strategies, outlined in Table 6.4 and Figure 6.4, were simulated in each stock. There is currently no capture efficiency requirement in ADF, however the required ventilation rate through a cooker hood is 50% lower than for a room extract fan, which implies a capture efficiency of 50%, hence $\eta = 50\%$ was used for the initial assessment. In addition, two strategies were included to investigate the potential benefits of continuing intermittent ventilation for an additional 10 minutes after the end of the cooking period. The appropriateness of this time is discussed in Section 6.4.3. Predicted

TABLE 6.4: Ventilation strategies

Strategy	Fan Airflow Rate	Details
A	0 l/s	Infiltration only
B	13 l/s	Constant general extract ventilation at the *high* rate from ADF
C	60 l/s	Intermittent general extract ventilation just during cooking
D	60 l/s	Same as *C* but for an additional 10 minutes after cooking
E	30 l/s	Intermittent extract through a cooker hood, η=50% just during cooking
F	30 l/s	Same as *E* but for an additional 10 minutes after cooking

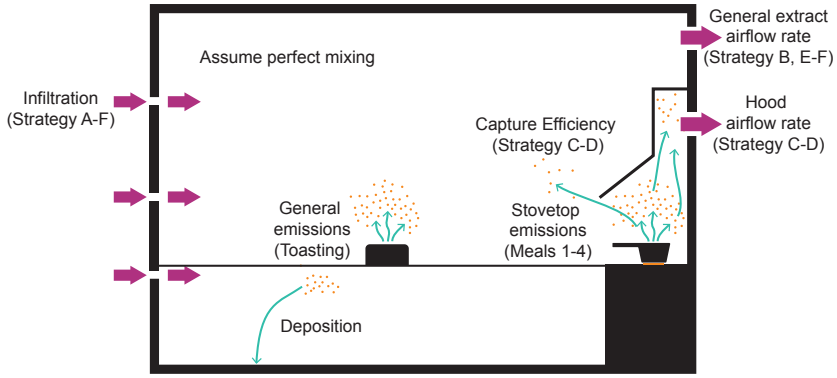


FIGURE 6.4: Kitchen ventilation strategies and model inputs.

concentrations were aggregated to a daily mean concentration to allow direct comparison with the WHO 24-hour mean guideline of 25 $\mu g/m^3$ [128], and the peak predicted concentration was recorded to allow further comparison between the strategies.

The second scenario considered the fan airflow rates required to minimise occupant exposure to $PM_{2.5}$. Here, the model was run for a range of capture efficiencies ($0 - 100\%$) and ventilation rates ($0 - 1200$ L/s). For each combination of capture efficiency and fan airflow rate, the model was used to predict the percentage of kitchens in which the daily mean concentration remained below the WHO 24-hour mean guideline. These percentages were then interpolated to develop curves to show the combination of ventilation rate and capture efficiency required for the daily mean concentration to remain below the guideline value in 2%, 25%, 50%,

75% and 98% kitchens.

6.3.2 Model Inputs

The flow rate, capture efficiency and duration of ventilation use for the modelled ventilation strategy were independent variables, and so these inputs were modelled deterministically. All other inputs were modelled probabilistically, sampled from appropriate distributions. Empirical distributions were fitted using piecewise cubic hermite interpolating polynomials [232].

Kitchen Volume, V The 2009 English Housing Survey was used to derive kitchen volumes, as in Section 6.2.1.2. The kitchen volume of each surveyed house is the product of the reported height, width, and depth. These volumes, and the corresponding weight of each sampled house, were used to create two empirical CDFs, from which volumes were sampled[5]. The first CDF was for the entire housing stock, used in the whole stock scenario, and the second was for houses built posts 2002, used in the new and future stock scenarios. *Post 2002* is the newest age bin given by the 2009 EHS. Entries with no recorded kitchen dimension data were ignored, the remaining data sets represented 99.75% of the entire English housing stock, and 99.65% of the post 2002 housing stock. Figure 6.5 shows volume distributions for both stocks. Neither distribution is normally distributed, and both are positively skewed. The mean (standard deviation) are 26.5 m^3 (13.8 m^3) for the existing stock, and 30.6 m^3 (18.6 m^3) for new houses. This indicates a trend towards larger kitchens in new English houses.

Ventilation Rate, λ_T Airflow in houses comprises ventilation through purpose-provided openings, infiltration and exfiltration through adventitious openings, and airflow through mechanical systems. Six ventilation strategies were

[5]The code is available under a creative commons license from DOI: 10.13140/RG.2.2.15694.00326

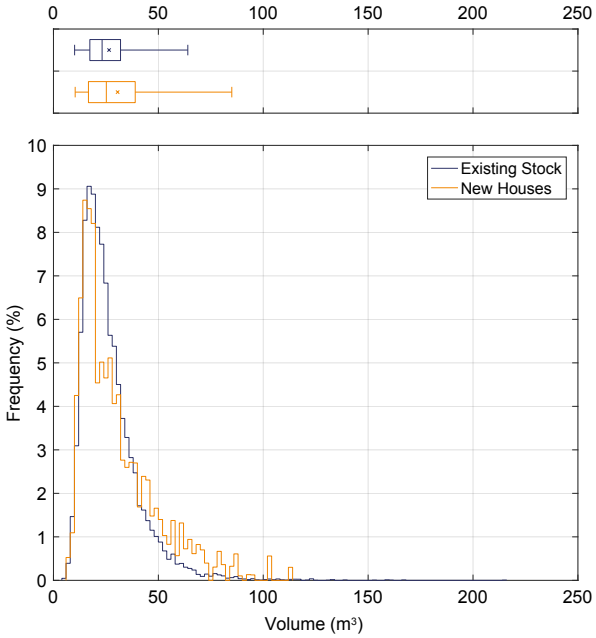


FIGURE 6.5: Distributions of English kitchen volumes [226]

considered intially, see Table 6.4. All assumed that purpose-provided openings were sealed, representing limiting conditions during the heating season. Infiltration was assumed to occur in all scenarios, and was considered to be a function of a measurement of air permeability, see Section 6.2.1.2 for more information. As for the steady state model, λ_N was sampled from distributions of hourly infiltration rates in English houses. These distributions were generated using DOMVENT3D [228–232]. As before, party walls were considered to be either impermeable or as permeable as other walls with equal probability.

All inputs to DOMVENT3D were identical to those described by Jones *et al.* [232], except for the distributions of air permeability. For pre-2002 houses, the Stephen distribution [236] was used, comprising measurements made in 384 UK dwellings built before 2000. For post-2002 houses, a distribution was applied derived from measurements tested under the Air Tightness Testing and Measurement Association (ATTMA) scheme in 144024 new UK houses between 2015 and 2016, which has been evaluated previously [237, 238].

The DOMVENT3D infiltration rate distributions described above were sampled for the existing and new stocks. For the future stock, the infiltration rate was set to always equal $0\,h^{-1}$. This was not to assume no infiltration would occur in the future stock, but instead to eliminate infiltration as a variable to use the model and to assess required total ventilation rates (λ_T) where the existing and new stocks consider the mechanical ventilation rate (λ_M).

The half-fan model introduced in Section 6.2.1.2 was once again applied to find λ_T from the infiltration rate, λ_N, and mechanical ventilation rate λ_M according to Equation 6.4. The basis and merits of this model were explained previously.

Penetration Coefficient, P The *penetration coefficient* is a non-dimensional parameter between 0 and 1 that represents the filtering effect of the building envelope [44]. The value of P depends on the size distribution of the aerosol and the flow characteristics through an airflow path [239], where $P = 1$ for airflow through large openings, such as open windows, and $P < 1$ for other airflow paths [44]. P has been found to vary between 0.7 and 0.9 with particle size [44], but previous models have treated $PM_{2.5}$ as an homogeneous pollutant and modelled $P = 0.8$ for the heating season conditions [240] considered here. Therefore, P was considered uniformly distributed between 0.7 and 0.9.

Capture Efficiency, η Two types of cooking were considered: foods cooked on a stove and those cooked elsewhere. For the general extract ventilation scenarios (B–D), the cooking location was unimportant because it was assumed that all emitted $PM_{2.5}$ fully mix within the space so that $\eta = 0$. When meals were not cooked on a hob the same mixing and capture efficiency assumptions were made. However, when a cooker hood was used and the meal cooked on a hob, the proportion of emitted $PM_{2.5}$ captured by the cooker hood before they can mix in the kitchen was given by η; see Equation 6.1. There are no typical values of η for English devices given in the literature. Accordingly, an initial value of 50% was assumed for the

assessment of the existing building regulations; later values between $0 - 100\%$ were tested deterministically.

Deposition Rate, k A deposition rate of $0.39 \pm 0.16\,\mathrm{h}^{-1}$ was used, taken from from Ozkaynak *et al.* [170] as in Section 6.2.1.2. It is derived from 1780 personal monitoring measurements in non-smoking residences, and has been used by other $PM_{2.5}$ modelling studies [37, 192, 193, 196]. Deposition rates were assumed to be normally distributed with the distribution truncated between $0\,\mathrm{h}^{-1}$ and $0.78\,\mathrm{h}^{-1}$, for a positive distribution with the same mean.

Ambient Concentration, C_a Unlike Section 6.2.1.2, the ambient, or external, $PM_{2.5}$ concentration was assumed to be uniformly distributed between $8.6\,\mu\mathrm{g/m}^3$ and $14.6\,\mu\mathrm{g/m}^3$. These concentrations were reported for London [240], however there is a precedent of applying this data to the entire housing stock [39].

Initial Concentration, $C(0)$ The initial concentration was assumed to equal the background indoor concentration, which is also the steady state concentration in the absence of internal sources

$$C(0) = C_b = C_{ss} = \frac{\lambda_T P C_a}{(\lambda_T + k)} \tag{6.9}$$

$C(0)$ was calculated using Equation 6.9 for λ_T at the start of the day. If the concentration at the end of the total time period, $C(T)$, was significantly different from that at the beginning, $C(0)$, the calculations were repeated with $C(0) = C(T)$ and the process continued until $[C(T) - C(0)] \leq 1\%$. The final value of $C(0)$ was then recorded as an input.

Cooking Frequency and Emission Rate, g The total time period was one day, so $T = 24\,\mathrm{hours}$, and 3 meals were cooked per day. This was to simulate a significant,

TABLE 6.5: Mean (μ) and standard deviation (σ) of main meal and toast emission rates and durations

	Emission rate (mg/min)		Duration (min)	
Meal Type	μ	σ	μ	σ
Meal 1 – *fried chicken, fried potatoes, boiled green beans*	0.62	0.10	28	5
Meal 2 – *fried chicken, boiled potatoes, boiled green beans*	0.80	0.19	28	5
Meal 3 – *pasta Bolognese*	1.9	0.74	28	5
Meal 4 – *chicken and vegetable stir-fry*	3.2	0.59	17	5
Toast	0.22	0.065	3.85	0.9

but not extreme, emissions profile, such as a weekend where occupants are at home all day and cook multiple times. Similar emission profiles were considered in other analyses of IAQ [37, 193].

The first meal was always the toasting of bread, representing breakfast. Emission rates were sampled from the empirical CDF derived in Section 4.3 with $\mu = 0.23\,\text{mg/min}$ and $\sigma = 0.067\,\text{mg/min}$. The meal duration was assumed to be normally distributed with $\mu = 3.8\,\text{minutes}$ and $\sigma = 0.9\,\text{minutes}$, to match the duration of the α–periods recorded in Section 4.3.

The other meals were considered typical of those cooked on a stove top in Northern Europe and their emission rates and cooking durations were those reported for Meals $1 - 4$ in Chapter 5, see Table 6.5.

Each of the meals was assumed equally likely to be cooked. First, the meal type was sampled, and then the emission rate and cooking duration were sampled from their corresponding normal distributions in Table 6.5.

The cooking schedule was made highly variable to account for a lack of data describing cooking behaviour in the UK. The relative importance of this input was interrogated by a sensitivity analysis; see Section 6.3.2.3. The start time of each meal was assumed to be uniformly distributed between 0600–1000 for breakfast, 1000–1400 for lunch, and 1600–2000 for dinner.

TABLE 6.6: Example scenario general inputs

Input	Value		
Volume (m^3)	22.6		
Infiltration Rate (h^{-1})	0.40		
Deposition Rate (h^{-1})	0.25		
Penetration Coefficient (-)	0.70		
External Concentration ($\mu g/m^3$)	12.8		
Initial Concentration ($\mu g/m^3$)			
Number of cooking periods	3		
Emission number	1	2	3
Meal Type	Toast	Meal 1	Meal 3
Emission Rate (mg/min)	1.22	2.70	12.4
Duration (minutes)	3.5	28.1	20.8
Post cooking interval (hours)	4.6	9.1	

To maximise the decay period after the final cooking event, the simulation start time ($t = 0$) always coincided with the beginning of the breakfast cooking period.

6.3.2.1 Example Strategies

To help explain how the model works, Figure 6.6 shows predicted concentrations over time for ventilation strategies A, C, and F (see Table 6.4) in the same kitchen. All other inputs were identical and are given in Table 6.6. All of the inputs are arbitrary and are only for illustrative purposes. Table 6.7 shows the breakdown of the calculation process. The daily mean $PM_{2.5}$ concentrations, $\overline{C(T)}$, calculated using Equations 6.2-6.8, were 214 $\mu g/m^3$, 73.3 $\mu g/m^3$ and 38.1 $\mu g/m^3$ for strategies A, C, and F, respectively.

6.3.2.2 Sampling Method

Again, the sampling method followed that described by Das *et al.* [37] and Jones *et al.* [232]. There were 17 input variables required to calculate $\overline{C(T)}$, which were sampled in sets of 100 using a Latin Hypercube. Each set of samples was applied to predict $\overline{C(T)}$; see Section 6.3.1.1. In addition, the peak concentration C_{max} was recorded for each sampled kitchen. The total sample size increased incrementally by the set size.

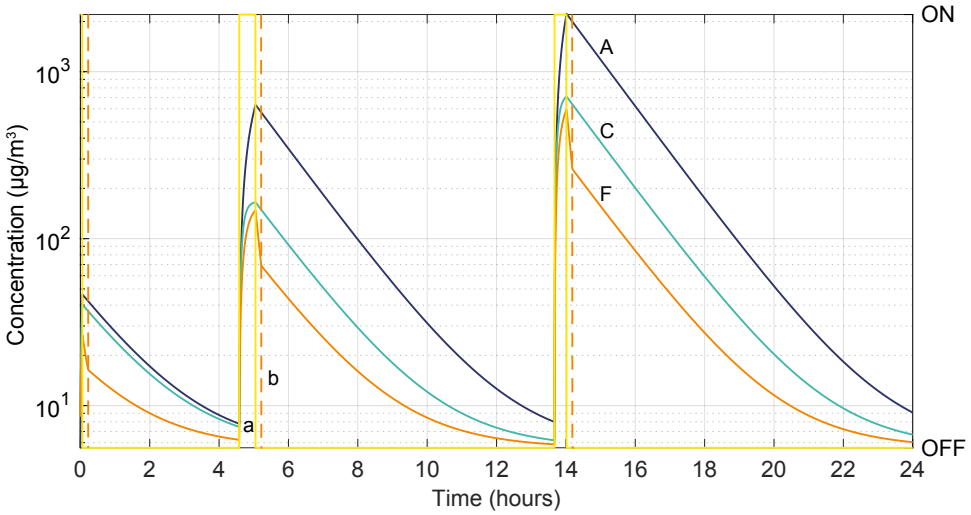


FIGURE 6.6: Example scenarios; A (**blue**), infiltration only; C (**teal**), 60 l/s general extract during cooking; F (**orange**), 30 l/s extract through cooker hood with $\eta = 50\%$ during cooking plus 10 minutes after; a (**yellow**), source emission function; b (**orange dashed**), marks 10 minutes after the end of cooking.

TABLE 6.7: Example scenarios calculation breakdown

Step	Δt s	g µg/s	λ_T s^{-1}	C_{ss} µg/m³	$C(0)$ µg/m³	$C(t)$ µg/m³	$\overline{C(t)}$ µg/m³	$t\overline{C(t)}$ µg s/m³
Strategy A								
1	214	4.4	0.00011	1087	9	50	30	6323
2	16284	0.0	0.00011	6	50	8	20	323703
3	1687	9.7	0.00011	2404	8	634	337	568389
4	31037	0.0	0.00011	6	634	8	118	3661039
5	1251	44.6	0.00011	11017	8	2222	1156	1446121
6	35928	0.0	0.00011	6	2222	9	349	12525044
Total	86400						214	18530620
Strategy C								
1	214	4.4	0.00266	80	7	39	24	5239
2	16284	0.0	0.00011	6	39	7	16	267743
3	1687	9.7	0.00266	167	7	165	132	223424
4	31037	0.0	0.00011	6	165	6	34	1058410
5	1251	44.6	0.00266	734	6	710	527	659561
6	35928	0.0	0.00011	6	710	7	115	4115634
Total	86400						73.3	6330011
Strategy F								
1	214	4.4	0.00133	148	6	43	25	5403
2	600	0.0	0.00133	9	43	23	32	18976
3	15684	0.0	0.00011	6	23	7	12	180417
4	1687	9.7	0.00133	163	7	148	103	173351
5	600	0.0	0.00133	9	148	69	103	61717
6	30437	0.0	0.00011	6	69	6	17	520653
7	1251	44.6	0.00133	716	6	592	380	475653
8	600	0.0	0.00133	9	592	261	404	242155
9	35328	0.0	0.00011	6	261	6	46	1616430
Total	86400						38.1	3294755

After each set of predictions was made, the overall mean (μ) and standard deviation (σ) of $\overline{C(T)}$ for all sets of samples were calculated. When the change in μ and σ from the addition of one set of samples to the next was $\leq$0.01% the total number of samples was deemed adequate.

6.3.2.3 Sensitivity Analysis

Again, a sensitivity analysis was used to test the dependence of $\overline{C(T)}$ on the model inputs, following the method of Jones *et al.* [232] and a full description is found in the reference. The method tests for linear (Kendell's tau, Pearson's product moment, linear regression), monotonic (Spearman's rank correlation coefficient, rank-transformed standardised variables) and non-monotonic (Kolmogorov-Smirnov, Kruskal-Wallis) relationships between inputs and outputs. All inputs were ranked according to the magnitude of the regression coefficient.

A fundamental requirement is that all tested inputs are independent of one another. Therefore, the emission rates, g, and source durations were combined as a product to form a single input variable, the total *source strength*, ΣG, which is the total mass of $PM_{2.5}$ emitted per day. The tested input variables were V, λ_N, k, C_a, P, $C(0)$, ΣG, and the mean average time between cooking events, $\overline{\Delta t}$. The sensitivity of $\overline{C(T)}$ to the number of emission periods was not tested because it was assumed to be constant.

6.3.3 Results

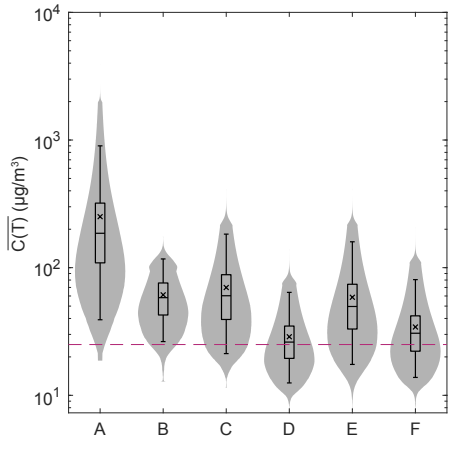
6.3.3.1 Existing Regulations

Figure 6.7 and Table 6.8 show the predicted $PM_{2.5}$ concentrations for ventilation strategies A $-$ F (see Table 6.4) for the existing stock (Figure 6.7a), new houses (Figure 6.7b), and the future stock (Figure 6.7c). When infiltration was the only source of fresh air (strategy A), the predicted daily mean $PM_{2.5}$ concentrations were at, or below, the WHO guideline of 25 $\mu g/m^3$ in 0.3% of existing kitchens, 0.2% in

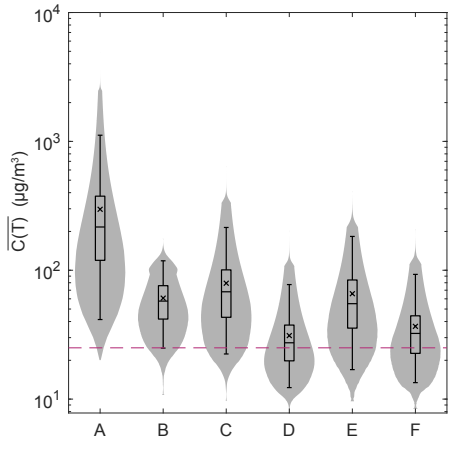
new kitchens, and 0.04% in future kitchens. This improved when 13 L/s of continuous mechanical ventilation was used (strategy B) to 1.5% of existing kitchens, 2.1% of new kitchens, and 1.9% of future kitchens. When 60 L/s general extract was used during cooking (strategy C), 5.4% of existing kitchens, 3.8% of new kitchens, and 1.5% of future kitchens met the WHO guideline. The best performance occurred when using a cooker hood with an airflow rate of 30 L/s and a capture efficiency of 50% (strategy E), where 11% of existing, 10% of new, and 6% of future kitchens were below the WHO guideline. These predictions highlight that halving a general extract airflow rate and assuming a 50% capture efficiency will not guarantee the same IAQ. Overall, the existing requirements perform poorly, especially in new houses, where $\leq$ 10% of kitchens were below the WHO guideline. This is because new houses are more airtight. The future stock presumed an infiltration rate of $0\,h^{-1}$, so it was expected to perform worst of the three stocks as the mechanical ventilation equalled the total ventilation.

However, continuing an intermittent ventilation strategy for 10 minutes after cooking ends was predicted to increase the percentage of kitchens below the WHO guideline. For 60 L/s general extract (strategy D), 46% of existing kitchens, 42% of new kitchens or 37% of future kitchens were predicted to be below the WHO guideline. And when using 30 L/s extract through a cooker hood with 50% capture efficiency (strategy F), 34% of existing, 31% of new, and 23% of future kitchens were below the WHO guideline. Here, the general extract strategy achieved lower $PM_{2.5}$ concentrations because the ventilation rate was higher and the capture efficiency did not apply after cooking has ended.

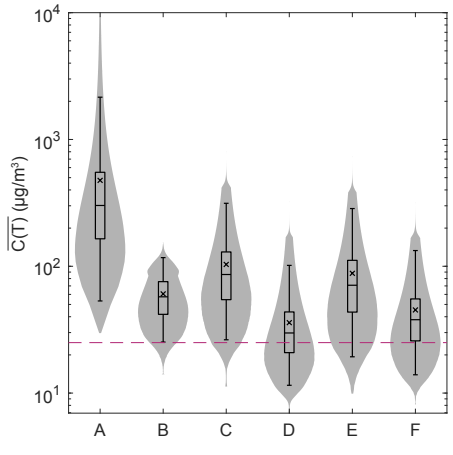
The distributions of data are shown by the shaded *violins* in Figure 6.7, and were similar in shape for all of the intermittent ventilation scenarios (strategies C $-$ D). There was greater variability, particularly for higher concentrations, in the predictions for existing kitchens than new. Strategy A (infiltration only) has the greatest variance, whereas Strategy B (constant extract) has the least and is



(A) Existing houses

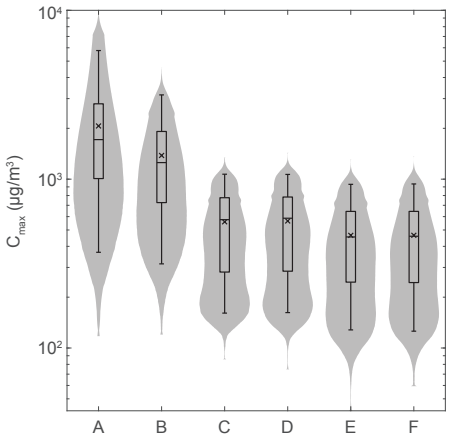


(B) New houses

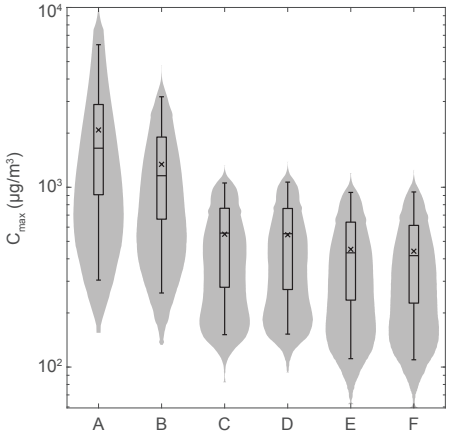


(C) Future houses

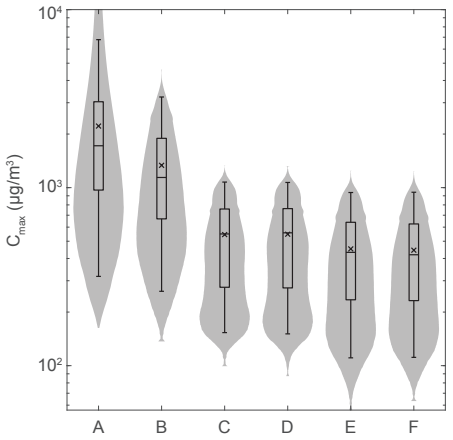
FIGURE 6.7: Predicted average $PM_{2.5}$ concentrations; WHO 24-hour in **pink**



(A) Existing houses



(B) New houses



(C) Future houses

FIGURE 6.8: Predicted maximum $PM_{2.5}$ concentrations

TABLE 6.8: Predicted 24-hour mean average concentrations ($\mu g/m^3$) in English kitchens for all ventilation strategies

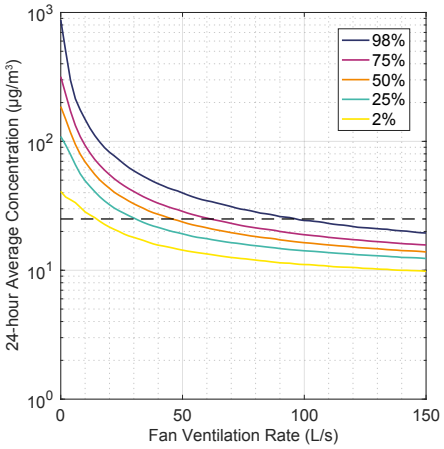
Centile/	Strategy (see Table 6.4 for details)					
Statistic	A	B	C	D	E	F
Existing Stock						
2%	39.1	26.4	21.2	12.5	17.5	13.8
25%	109	42.6	39.3	19.5	33.1	22.2
50%	186	58.4	60.3	26.1	49.7	30.6
75%	320	75.9	88.1	34.9	74.1	41.9
98%	900	117	183	64.0	160	80.6
μ	251	61.3	69.9	28.8	58.6	34.3
σ	222	23.5	41.6	12.9	36.1	17.1
GM	186	56.9	59.8	26.3	50.0	30.9
GSD	2.2	1.5	1.7	1.5	1.8	1.6
New Houses						
2%	41.4	24.9	22.4	12.3	16.9	13.4
25%	119	41.8	43.1	19.8	35.5	22.6
50%	217	57.6	68.0	27.4	55.0	32.3
75%	376	75.9	101	37.5	84.0	44.3
98%	1120	118	215	77.4	183	92.9
μ	297	60.8	79.3	31.1	65.8	36.7
σ	287	23.8	49.9	16.5	42.4	20.2
GM	213	56.2	67.0	27.8	55.0	32.4
GSD	2.3	1.5	1.8	1.6	1.8	1.6
Future Stock						
2%	53.3	25.3	26.3	11.5	19.3	13.9
25%	165	41.8	54.5	20.8	43.5	25.8
50%	302	57.4	86.1	29.8	70.9	37.9
75%	551	75.6	130	43.6	111.4	55.3
98%	2155	117	314	101.6	285	133.1
μ	476	60.6	103.4	35.9	88.0	45.3
σ	705	23.5	70.9	23.1	65.5	29.3
GM	306	56.1	85.3	30.7	70.5	38.5
GSD	2.5	1.5	1.9	1.7	1.9	1.7

bi-modal.

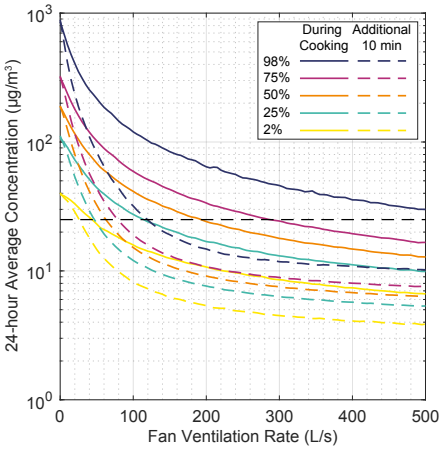
For comparison, Figure 6.8 shows the predicted daily maximum concentrations for each scenario. Here there was no benefit to continued ventilation after cooking for either scenario C/D or E/F, as the key factor here is the rate of removal during the cooking period. In addition, higher peak concentrations were predicted for the costant ventilation scenario than any intermittent ventilation options.

6.3.3.2 Required Ventilation Rates

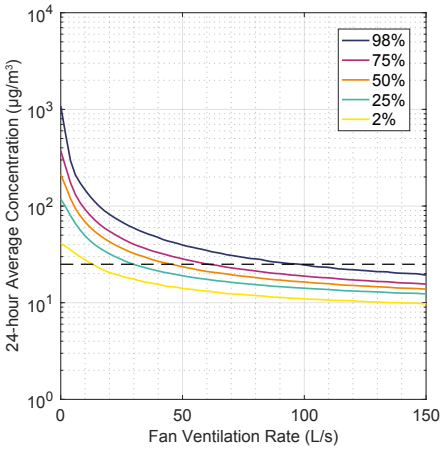
Constant General Extract Figure 6.9 shows the predicted concentrations at a range of airflow rates for constant mechanical extract ventilation. For 98% of kitchens to remain below the $25\,\mu g/m^3$ 24-hour mean guideline, the predicted a required fan extract rate was $97\,L/s$ (or $25\,h^{-1}$) for the existing housing stock or



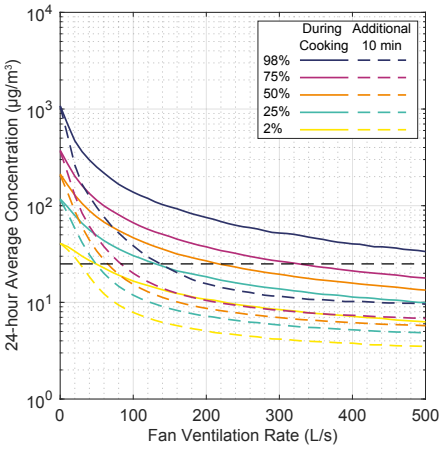
(A) Existing houses



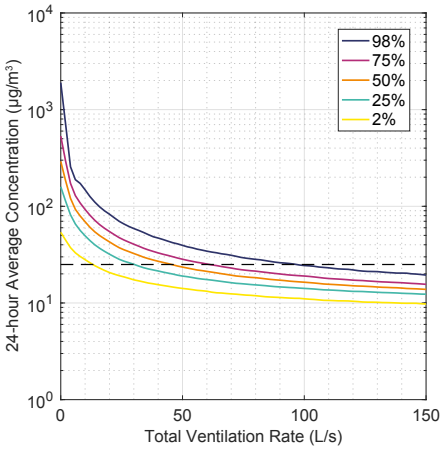
(A) Existing houses



(B) New houses

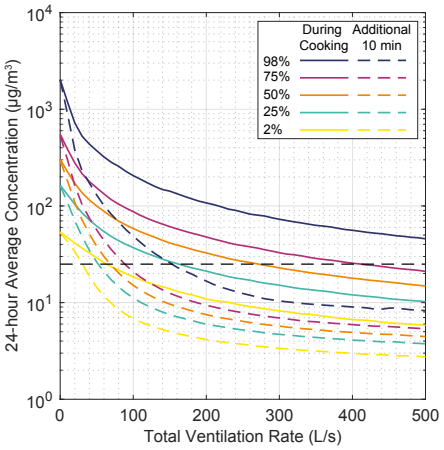


(B) New houses



(C) Future houses

FIGURE 6.9: Constant General Extract; WHO 24-hour in **black**



(C) Future houses

FIGURE 6.10: Intermittent general extract ventilation; WHO 24-hour in **black**

TABLE 6.9: Required general extract ventilation rates

	Constant Ventilation		Intermittent Ventilation			
			During Cooking		Plus 10 min	
Centile	L/s	h^{-1}	L/s	h^{-1}	L/s	h^{-1}
Existing Stock						
2%	15	1.1	43	4.1	26	2.4
25%	31	4.1	115	16	47	6.7
50%	45	6.8	193	29	62	9.7
75%	61	11	285	50	79	13
98%	97	25	619	131	119	22
New Houses						
2%	13	0.81	49	3.5	27	2.0
25%	30	3.4	131	16	49	6.4
50%	46	6.3	216	30	65	9.4
75%	61	11	326	56	83	13
98%	99	25	723	154	137	22
Future Stock						
2%	13	0.86	63	4.9	30	2.4
25%	30	3.4	162	20	52	7.0
50%	46	6.3	272	38	69	10
75%	61	11	419	69	88	14
98%	97	25	1025	202	153	22

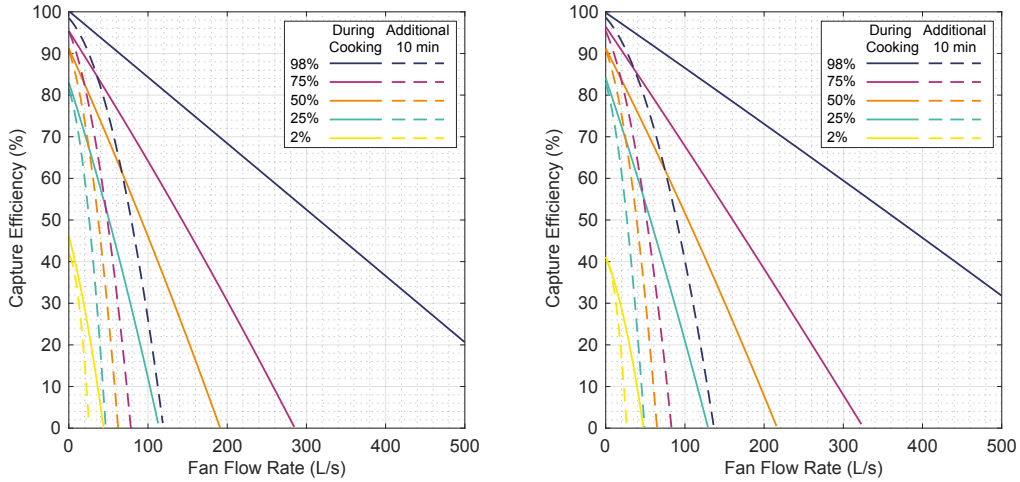
99 L/s (or 25 h^{-1}) for new houses. New houses are more airtight and so they generally require a higher fan airflow rate. In the future stock, Figure 6.9c predicts a required total ventilation rate of 97 L/s (25 h^{-1}). The ventilation rates required for other percentages of kitchens to remain below the guideline concentration are included in Table 6.9. The predicted constant ventilation rate for 75% houses (existing, new or future) to remain below the guideline, at 61 L/s, was almost five times the current required constant ventilation rate required by ADF.

Intermittent General Extract When using intermittent extract ventilation away from a stove (not through a cooker hood), the exhaust ventilation rates required for 98% of kitchens to remain below the WHO guideline were predicted to be 619 L/s (131 h^{-1}) for existing kitchens and 723 L/s (154 h^{-1}) for new, and a total ventilation rate of 1025 L/s (202 h^{-1}) was predicted for the future stock. See Table 6.9 for other centiles. The predicted ventilation rates for both 75% and 98% kitchens to comply were considerably higher than the existing requirements, and high air change rates

can cause additional problems, such as increased energy demand and pressure differentials [230].

Continuing ventilation for 10 minutes after cooking was also beneficial here. Figure 6.10 shows the predicted concentrations for a range of ventilation rates. When ventilation continued after cooking, the mean concentrations dropped considerably for the same flow rate. Here, for 98% kitchens to remain below the WHO guideline, the predicted fan exhaust rate was 119 L/s ($22\,h^{-1}$) for existing houses, and 137 L/s ($22\,h^{-1}$) for new houses, and a total ventilation rate of 153 L/s ($22\,h^{-1}$). These ventilation rates are high when compared to those specified by ADF. However, the 75th centile exhaust ventilation rates of 79 L/s for existing kitchens, and 83 L/s for new, are closer to the ADF required 60 L/s; see Table 6.9.

Cooker Hood When air is extracted through a cooker hood, its capture efficiency must also be considered. This is where the percentage of kitchens below the guideline is useful, and can be used to produce a three-dimensional relationship between flow rate, capture efficiency and the percentage of kitchens below the threshold, see Figure F.1. Whilst the three-dimensional plot helps to visualise the relationship it is more useful to produce a plot relating the required capture efficiency and flow rate for each centile of kitchens, effectively taking horizontal slices through Figure F.1. Figure 6.11 shows the relationships between fan airflow rate and capture efficiency for the existing housing stock and new dwellings.The capture efficiency of cooker hoods installed in UK dwellings is unknown, and cooker hood capture efficiencies have been shown to increase with the hood airflow rate [181, 183]. For this reason the future stock was not considered here as that scenario predicts total rather than fan airflow rates. For the 50% capture efficiency implicitly assumed by ADF, the predicted ventilation rates were at least 92 L/s, 144 L/s or 317 L/s for 50%, 75%, or 98% of existing kitchens to comply, respectively, if the hood is only switched on during cooking; see Figure 6.11a. In new houses, the



(A) Existing houses (B) New houses

FIGURE 6.11: Required combinations of airflow rate and capture efficiency

corresponding minimum ventilation rates required were 104 L/s, 161 L/s and 371 L/s; see Figure 6.11b.

Continuing to run the cooker hood after cooking ends lead to lower predicted flow rates and/or capture efficiencies, indicated by the dashed lines in Figure 6.11. Assuming the same 50% capture efficiency, the minimum required flow rates for 50%, 75% or 98% kitchens to comply were predicted to be 37 L/s, 49 L/s or 80 L/s in existing houses (Figure 6.11a) and 39 L/s, 51 L/s or 90 L/s in new houses (Figure 6.11b).

These theoretical combinations of capture efficiency and flow rate have limited use unless they are compared to the measured performance of installed cooker hoods. Figure 6.11 could be used as a design tool by overlaying empirical curves that relate measured capture efficiency, $\eta_{measured}$, and airflow rate for real cooker hoods. An example is given in Figure 6.12. The two curves labelled *Example A* and *Example B* were produced by fitting empirical data to the equation $\eta = \eta_{max}(1 - e^{-\alpha\lambda_M})$, where $\eta_{max} = 100\%$ and α is a constant equal to the magnitude of the gradient from the log–linear regression of $\eta_{max} - \eta_{measured}$. The data for *Example A* was obtained from

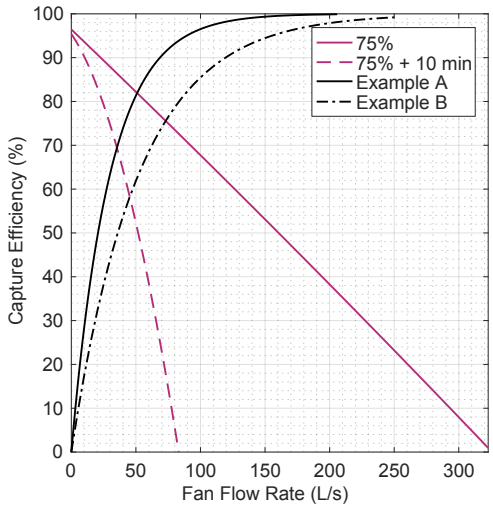


FIGURE 6.12: Measured cooker hood performance with 75th centile airflow rate-capture efficiency curves for new houses

measurements of the performance of a single hood [241] similar to the one used in Chapter 5, whereas *Example B* comprises the combined data for 7 hoods located at multiple mounting heights [188]. These examples show how their intersections with the efficiency–airflow rate curves could be used to set the hood's airflow rate. For example, if a cooker hood with the same properties as *Example A* was installed with an airflow rate of 51 L/s, its corresponding capture efficiency of $\geq$82% would keep $PM_{2.5}$ concentrations below the WHO guideline in 75% of new kitchens when used just during cooking.

6.3.3.3 Sensitivity Analysis

Table 6.10 shows the results for the sensitivity analysis described in Section 6.3.2.3. In addition, Figure F.3 shows plots of $\overline{C(T)}$ against each model input. The sensitivity analysis was used to determine the relative importance of independent input variables, where a rank of 1 indicates the most important input variable. The total source strength, ΣG, was ranked the most influential by all tests, also indicated in Figure F.3. The least important input varied between tests, but the ambient concentration, penetration coefficient and average time between meals

were consistently ranked lowest. This is important because these inputs involved the most assumptions. The relationships between inputs and outputs were statistically significant ($p \leq 0.05$) for most high ranked inputs across all tests. Lower ranked inputs were often non–significant; see Table 6.10. The lower ranking of the time between meals means the model could be simplified by fixing this input with the penalty of removing the capability to test the effect of continuing ventilation after cooking ends.

TABLE 6.10: Sensitivity of $\overline{C(T)}$ to model inputs. 1 is the highest rank

Input\Test	S_{Kend}	S_{Pear}	S_{Spear}	$S_{regress}$	$S_{rankreg}$	S_{Kolm}	S_{Krusk2}	S_{Krusk5}	$S_{Krusk10}$	$S_{Krusk20}$
ΣG	1	1	1	1	1	1	1	1	1	1
V	2	3	2	3	2	2	2	2	2	2
λ_N	3	5	3	5	3	3	3	3	3	3
k	4	4*	4	4	4	4	4	4	5	5
$C(0)$	5	2	5	2	5	5	5	5	4	4
C_a	6*	7*	6*	7	6	7*	6*	6*	7*	7*
$(\overline{\Delta t})$	7*	6*	7*	6	7	8*	8*	8*	8*	8*
P	8*	8*	8*	8	8	6*	7*	7*	6*	6*

* indicates tests which were non–significant at the $p > 0.05$ significance level

One assumption of the sensitivity analysis is that the model inputs are independent of one another. Most inputs included in Table 6.10 were sampled independently, and Figure F.4 does not appear to show correlations between input variables. The initial concentration, $C(0)$, was not sampled independently. However, it was included in the sensitivity analysis to check whether the step to repeat the calculations for each sampled "day" were necessary for model predictions. Visual inspection of Figure F.4e indicates some weak correlation between $C(0)$ and other input variables. However, as $C(0)$ ranked in the middle of the inputs in the sensitivity analysis, it was still important to include this parameter in the sensitivity analysis and the convergence loop in the calculation method.

6.4 Discussion

6.4.1 Drawbacks

Section 6.2.3 explored the limitations of the initial, simplified steady state model. The method introduced in Section 6.3.1 attempted to address these limitations by instead predicting average 24-hour concentrations with multiple cooking emission events and non-zero ambient $PM_{2.5}$ concentration. However, there are sill limitations to the updated method.

In Section 6.2.3 and Section 6.3.3, predicted steady state and 24-hour mean $PM_{2.5}$ concentrations, respectively, were compared to the WHO guideline. This comparison assumed that occupants spend all of their time in the kitchen. This is generally not the case, and so the focus was on $PM_{2.5}$ exposures experienced by cooks and others who may consistently occupy a kitchen, such as young children. In dwellings with open plan kitchens, both non-cooks and cooks are likely to be exposed to elevated concentrations for longer. Open plan kitchens are included in the volume distributions (see Figure 6.5), however the model did not distinguish between open plan and enclosed kitchens. A multi-zone model coupled with occupancy patterns, similar to the approaches undertaken by [191, 193, 194], may give a more realistic estimation of exposure for different types of occupant. However, this would require the use of appropriate archetypes and occupancy patterns, and would take more computational time, although it should be investigated in the future. This study was not intended to investigate personal exposures in that way. Instead, the intention was to perform an investigation into the $PM_{2.5}$ concentrations that might be found in domestic kitchens, and the appropriate ventilation mechanisms to reduce them. Comparing $\overline{C(T)}$ to the WHO's 24-hour guideline in Section 6.3.3 does provide a rudimentary assessment of acute exposure.

In order to consider the chronic impact of indoor $PM_{2.5}$ concentrations from

cooking sources a model would be required whose predictions could be compared to the WHO annual mean guideline of $10\,\mu g/m^3$. This would require a more complex model that considers multiple cooking and occupancy patterns, similar to [39, 193, 194], and should also consider seasonal variations in ventilation use. The current model only considered worst-case heating season conditions where infiltration was the only source of natural driven airflow. However, occupants are more likely to open windows in summer months as air temperatures increase [242], thereby increasing the total ventilation rate. In those circumstances, it is important to understand how naturally driven ventilation through purpose-provided openings and kitchen fans affect one-another.

The current average concentration model did not consider the energy consequences of the different ventilation strategies. Any mechanical ventilation system will result in increased energy demand, both to power the ventilation itself, and the heating load associated with the increased ventilation rate. Future work should consider the comparative energy consequences of a lower air exchange rate over an extended period versus a high air exchange rate over a shorter period. This could be achieved by using a coupled dynamic model, see Shrubsole *et al.* [39]. However, this modelling approach is more computationally expensive and so would be more suited to testing a smaller number of model iterations than included in Section 6.3.3.2.

6.4.2 Context

The predicted 24-hour mean concentrations given in Table 6.8 can be compared to *in-situ* measurements. For example, the 24-hour average $PM_{2.5}$ concentrations were measured in 11 residential kitchens located in Colchester, England [165]. The reported averages were grouped by housing type, and were found to be $56\,\mu g/m^3$ (Coefficient of variation, $C_v = 71\%$) in student accommodation, $46\,\mu g/m^3$ ($C_v = 71\%$) in single bedroom flats, and $10\,\mu g/m^3$ ($C_v = 70\%$) in 2 and 3 bedroom

houses. All kitchens were equipped with an extractor fan, and so the measured concentrations are best compared to Ventilation Strategy C (see Table 6.8), although the fan type and frequency of use were not reported. The measured 24-hour mean concentrations in the student accommodation and single bedroom flats sit between the 25th centile and median, although the measured concentrations in the houses were lower. The predicted $\overline{C(t)}$ can also be compared to the measured concentrations from Chapter 3. Daily mean concentrations were not reported in Section 3.3.2, instead the mean concentration was given for each 7 day monitoring period. During the first week all airflow was assumed to occur solely via infiltration (Strategy A), and the 7-day mean concentrations range between $14.1\,\mu g/m^3$ ($C_v = 310\%$) and $70.3\,\mu g/m^3$ ($C_v = 566\%$). The 7-day mean concentrations in 4 of the 5 houses were near or below the 2nd centile for Strategy A. These measured $PM_{2.5}$ concentrations suggest that the predictions for Strategy A may overestimate those found in domestic kitchens. It should be noted that the sample sizes of both studies were small ($N = 11$ and $N = 5$) so they cannot be considered representative of the entire English housing stock. A full validation of the model would require $PM_{2.5}$ measurements in a representative sample of English residential kitchens, and should be the subject of future work.

Another consideration is the equivalence of constant and intermittent ventilation solutions. The 24-hour mean concentrations were predicted here to allow direct comparison to the WHO 24-hour guideline concentration. This is currently the only guideline for acute exposures. However, it may be worth considering the peak concentrations predicted for each system, see Figure 6.7a. Where strategy B predicted slightly lower average concentrations than strategy C, and similar $\overline{C(T)}$ to strategy E, the peak concentrations were considerably higher for strategy B. This would be a consideration if future health research were to establish links between acute health effects and instantaneous or average concentrations over a shorter averaging period. Continuing intermittent ventilation was also predicted to have no effect on the maximum concentrations, as here the only consideration is the flow

rate and capture efficiency during cooking.

The predictions from the average concentration model can also be compared to those of previous modelling studies. Here it is difficult to make direct comparisons because they typically report annual, rather than 24-hour, mean concentrations, and the reported concentrations are not specifically for kitchens. For example, the annual mean concentration that a member of an average non-smoking household is exposed to was predicted to be $28.4\,\mu g/m^3$ [191]. Furthermore, cooks in domestic kitchens were predicted to be exposed to annual average concentrations of $30-40\mu g/m^3$ by Shrubsole *et al.* [39]. Both studies included kitchen extract fans in some dwellings, and also modelled window opening behaviour [39, 191]. Their predicted concentrations were closest to the 2nd centile for Strategy A (see Table 6.8) and the 25th centile for Strategy C. This also suggests the current model may overestimate concentrations. However, the annual averages reported in these studies include different cooking schedules for weekends and weekdays, and use a single $PM_{2.5}$ emission rate of 1.6 mg/min that accounts for less variability than those used here. Table 6.5 shows that this value lies within the range applied here, but neither this study, nor those discussed herein, accounted for extreme emission events, such as those where charring or burning may occur from dirty equipment or as part of the cooking process.

6.4.3 Policy, Standards and Guidelines

Figure 6.7a predicted that over 98% of English houses are too airtight to provide sufficient infiltration to dilute $PM_{2.5}$ emissions from cooking sufficiently so that daily mean concentrations in kitchens are below the WHO guideline of $25\,\mu g/m^3$. The EHS [226] recorded that 47% of English houses have a fan in their kitchen, and so the majority will solely rely on infiltration for dilution during the heating season when windows and vents are closed. In these circumstances, their occupants are likely to be exposed to short term $PM_{2.5}$ concentrations that exceed WHO daily guideline.

It is not desirable to increase infiltration because it is positively correlated with heating energy demand. As current policies seek to improve the energy performance of housing stocks, infiltration reduction is a primary method [232]. Therefore, controlled ventilation is required in all kitchens to mitigate against negative impacts on occupant health from cooking. The EHS does not record if fans are wall-mounted or cooker hoods. The difference is important because many cooker hoods recirculate air without extracting it directly outside and do not contain $PM_{2.5}$ filtration. In future versions of the EHS it would be helpful to record the fan type and whether it exhausts externally. It is also unclear what ventilation rate these fans might provide and so field measurements are required.

Figure 6.7 shows that the existing domestic kitchen ventilation strategies and airflow rates prescribed in ADF were predicted to be inadequate in over 89% of houses when used only during cooking. Continuing to ventilate after cooking ends reduced the daily mean $PM_{2.5}$ concentration significantly, but $\overline{C(T)}$ was still above the WHO guideline in 54% of kitchens. Dobbin *et al.* [176] found similar benefits of continuing ventilation after emissions end. They measured similar reductions in mean $PM_{2.5}$ concentration over the monitoring period for keeping the cooker hood on for 15 minutes after cooking as for an increase of almost 50 L/s. This is a similar effect to the reductions seen in Figure 6.10b, although they are not directly comparable as Dobbin *et al.* only took the average concentration over a single cooking period and the following 15 minutes, rather than a full day with multiple emission periods [176].

The choice of continuing to ventilate for exactly 10 minutes after cooking ends was somewhat arbitrary. However, the optimum duration is a balance between maximising the rate of concentration reduction and psycho-social factors, such as memory and noise, especially when people also eat in the kitchen after cooking. Therefore, additional tests were run for strategy C/D and E/F where the extended ventilation period varied between 0 and 40 minutes. The results, see Figure F.2,

show a law of diminishing returns where an initially steep decline tends towards an asymptote. For 60 L/s general extract, in 98% kitchens, the daily average concentration reduced by 70% for 10 minutes extra ventilation after cooking ended, but increasing the additional ventilation period to 20 minutes only reduced concentrations by a further 6%, to a total reduction of 76%. A similar pattern was observed for 30 L/s through a cooker hood with 50% capture efficiency, although the benefit of additional ventilation after cooking was reduced, see Figure F.2. In Figure 6.7a, strategy E predicted lower $\overline{C(T)}$ than strategy C, comparing general extract and a cooker hood just during cooking. However, the $\overline{C(T)}$ was predicted to be higher for strategy F than strategy D. This finding is repeated and further explained in Figure F.2, where the higher ventilation rate in strategy C/D means that for less than 5 minutes extra ventilation after cooking, predicted $\overline{C(T)}$ was lower than strategy E/F. It might be possible to identify the ideal combination of fan airflow rate, capture efficiency, and duration of ventilation after cooking using a 4-dimensional approach similar to the 3-dimensional approach used to find the relationship between airflow rate and capture efficiency in Section 6.3.3.2, however, this approach would not account for human behaviour. Therefore, ventilating for an additional 10 minutes appears appropriate and may also be memorable because it is simple, making it a suitable public health guideline.

The predictions can also be compared against other standards and guidelines for domestic kitchens. For example, ASHRAE 62.2 recommends minimum extract airflow rates of 150 L/s or 5 h^{-1} for enclosed kitchens, or 150 L/s for non-enclosed kitchens [179]. The current model did not differentiate between enclosed and open plan kitchens, although both were included in the volume distributions given in Figure 6.5. Figure 6.10b shows that an extract ventilation rate of 150 L/s was predicted to keep concentrations below the WHO guideline in <50% of new kitchens, when used just during cooking, or >98% if they continue to operate for 10 minutes after cooking ends. Alternatively, ASHRAE 62.2 requires an airflow rate of 50 L/s through a cooker hood. Figure 6.12 shows that the *Example A* fan would

provide sufficient ventilation in 75% kitchens, and the *Example B* fan would provide enough ventilation in more than 75% kitchens if it operates for an additional 10 minutes after cooking. Both example fan performance curves predict required capture efficiencies of over 50% for an airflow rate of 50 L/s: 81% for *Example A* and 62% for *Example B*.

European ventilation requirements for residential kitchens vary between countries and are often less specific. For example, the Netherlands Building Decree [220] specifies a capacity of 21 L/s. Dutch ventilation systems typically have 3 settings: 7 L/s when the dwelling is unoccupied, 14 L/s when occupied, and 21 L/s during cooking. The current model cannot test this combination because constant and intermittent ventilation strategies are treated as separate scenarios.

European standard EN 16798-1:2019 provides recommended ventilation rates for use in the absence of national standards [178]. Its kitchen extract airflow rate is dependent on the number of main rooms in the dwelling and occupant expectations. For example, for a normal dwelling with 4 main rooms, the specified kitchen extract is 35 L/s. However, the standard is unclear whether this is an intermittent or a constant airflow rate, although it does state that the ventilation rate can be halved if there is also a cooker hood present. The prescribed ventilation rate varies with dwelling size and so it is not directly comparable to the predictions made here, which assume the same extract rate for the entire stock. It may be valuable to test the approach given in EN 16798-1 as a method of reducing ventilation rates in smaller dwellings, thereby simultaneously lowering the energy demand and maintaining the level of protection found in larger dwellings. This will be the subject of future work.

The capture efficiency of installed cooker hoods is predicted to play a key role in reducing kitchen $PM_{2.5}$ concentrations. Measured capture efficiencies have been found to increase with increased flow rate [181, 183, 243], but other factors also affect capture efficiency. They include stove-top coverage, installation height, and

kitchen configuration [181, 183]. ASTM standard E3087-18 gives a procedure to measure capture efficiency for wall mounted cooker hoods [187], but the same procedure is not yet applicable to island hoods or downdraft devices, where the airflow patterns are different [186]. The ASTM procedure also measures with the hood mounted between wall-mounted cabinets [187], which has previously been found to increase the capture efficiency [183]. Additionally, Dobbin *et al.* [176] measured flow rates through installed cooker hoods that were $12 - 31\%$ lower than the manufacturer specified rates. For these reasons, manufacturers need to provide installation instructions, and details of how the hood performance may be affected if they are not followed, and to ensure the installed performance matches testing.

ASTM standard E3087-18 should be cited in standards as the appropriate method for measuring capture efficiency. In the absence of a manufacturer's empirical value, standards should also recommend a punitive values to be used in calculations. A large surveys of the capture efficiencies of *everyday* cooker hoods is required and could be used to determine appropriate punitive values in the medium term. In the short term, values will require an educated guess.

6.5 Conclusions

Through two stages of model development, the work included in this chapter predicted uncertainty in mean average daily concentrations of $PM_{2.5}$ emitted by cooking in English kitchens. The initial steady state model proposed by Salthammer [10] was trialled first but later rejected in favour of an average concentration model better suited to the transient nature of cooking emissions. It is predicted that over 98% of English houses are too airtight to provide sufficient infiltration to dilute $PM_{2.5}$ emissions from cooking sufficiently so that daily mean concentrations in kitchens are below the WHO guideline of $25\,\mu g/m^3$. Therefore, controlled ventilation is required in all kitchens to mitigate against cooking related negative health impacts.

The application any of the mechanical ventilation strategies prescribed by England's ADF was predicted to be adequate in less than 11% of houses when used only during cooking. When the requirements of ASHRAE 62.2 were applied, it was predicted that concentrations are below the WHO guideline in more than 25% of new kitchens.

When using an intermittent ventilation strategy, continuing to ventilate for a period of time after cooking has finished had a significant effect on the daily mean $PM_{2.5}$ concentration. However, there were diminishing returns as time increases. A 10 minute period was found to be appropriate. When using 10 minutes of additional ventilation after cooking, $PM_{2.5}$ concentrations were below the WHO guideline in 46% and >98% of kitchens when ADF and ASHRAE 62.2 were applied, respectively. Incorporating the additional 10 minutes of ventilation into the assumptions used to define ventilation strategies and airflow rates prescribed by standards means it must also be a public health guideline. Then, for 75% of houses to have a daily mean $PM_{2.5}$ concentration below the WHO guideline, a constant ventilation rate of 61 L/s or an intermittent ventilation rates 79 L/s is required. However, the comparative energy and carbon emission implications of the different ventilation strategies remain an area for future work.

The most effective ventilation strategy uses a cooker hood located over the hob that captures a proportion of emitted particles and extracts them directly outside. Its ability to do this is a function of its capture efficiency and the airflow rate and so both must be specified in standards. Standards should cite ASTM standard E3087-18 as an appropriate capture efficiency test method that cooker hood manufacturers must follow. In the absence of empirical data, punitive values should be specified. For 75% of houses to have a daily mean $PM_{2.5}$ concentration below the WHO guideline, a cooker hood with a 50% capture efficiency requires an airflow rate of 51 L/s when used during cooking and for 10 minutes afterwards.

Chapter 7

Discussion and Recommendations for Future Work

The work presented in this thesis consists of a series of empirical and theoretical investigations into $PM_{2.5}$ concentrations in UK domestic kitchens. Therefore, the aim of this chapter is to draw together the work presented previously and to expand the discussion included in Chapter 3 – 6.

Firstly, the average concentration model introduced in Chapter 6 predicted that using ventilation compliant with current ADF recommendations would substantially reduce $PM_{2.5}$ concentrations, though not below guideline concentrations in the majority of kitchens. However, the intervention study presented in Chapter 3 only found additional ventilation to significantly reduce temporal $PM_{2.5}$ concentrations in 2 of the 5 houses.

The lognormality of pollutant concentrations is established in the literature [156]. Therefore the median or geometric mean concentration are likely to be more representative summary statistics than the arithmetic mean, which is more effected by the positive skew of the data. Many of the previous residential and personal monitoring studies included in Table 2.7 and Table 2.8 recognised this and included summary statistics other than the arithmetic mean and standard deviation. The

concentrations measured in the kitchens of Houses A – E were not normally or lognormally distributed, see Figure C.9, which informed the decision to compare the Week 1 and Week 2 concentration distributions using the non-parametric Mann-Whitney U or Wilcoxon's Rank-sum test.

However, the existing epidemiological data (see Tables 2.1 – 2.4) and health guidelines rely on 24 hour or annual mean concentrations, as they rely on concentrations measured using the standard gravimetric test method. For example, the WHO [128] cite the American Cancer Society [55] and the Harvard Six-Cities [49] studies which both used gravimetric data. The gravimetric method is only able to provide mean average concentrations over the measurement time. Where measured temporal concentrations are not summarised with arithmetic means over the same averaging period as the guidelines, it is not possible to compare the measured concentrations to the guideline. There is no real benefit to finding the number of hours that concentrations were above $25\,\mu g/m^3$, as this is not a fair comparison and the health effects of exposures for time periods less than 24 hours have not been established.

The large size of the data sets in Chapter 3 ($n \approx 10081$) may have contributed to the statistically significant results. Also, in House D, the MW test indicated a small but significant decrease in concentrations in Week 2 despite an increase in the arithmetic mean. The opposite was true in House B. An alternative approach might have been to calculate the 24-hour mean concentrations for each consecutive 24-hour period, and use an appropriate statistical test to compare the 7 resulting concentrations for each week. The smaller data sets would be less likely to result in a statistically significant result, and it is important to avoid cherry picking data. In addition, this test method would be more comparable to the distributions included in Figure 6.7.

Additionally, the performance of the existing ventilation systems installed in the intervention test kitchens was not measured. Instead, they were assumed to perform

to existing ADF requirements. Here, cooking behaviour was a confounding factor, as the total amount of time spent cooking increased in Week 2 in House B, and the total time spent frying increased in Houses A, D and E, see Figure 3.6.

A second confounding factor was the existing ventilation mechanisms and the way in which occupants elected to use them. The average concentration model assumed the only form of additional ventilation was mechanical extract, whether constant or intermittent, and that intermittent ventilation was used for the entirety of all cooking events. Extracting air from the kitchen in this way is designed to depressurise the kitchen, so air is drawn into the kitchen from other parts of the house, which prevents the spread of pollutants from the kitchen to other parts of the house [244]. In contrast, the students frequently used the cooker hood to provide additional extract ventilation in conjunction with opening external windows or doors. Furthermore, the occupants did not use ventilation during all recorded cooking events in Week 2. As the use of ventilation was recorded using a simple tick box method, it was not possible to investigate whether the indicated ventilation mechanism was used for all or part of the corresponding cooking event. Participants were not given specific instructions on which ventilation mechanisms to choose, therefore it would be beneficial to issue more specific instructions in future, to reflect the findings from Chapter 6. This would include recommending occupants only use only mechanical extract ventilation such as a cooker hood or general extractor fan, and using it throughout cooking and for 10 minutes after. In addition, participants could be better informed of which cooking processes are associated with high $PM_{2.5}$ emissions, so as to better target their ventilation use.

The average concentration model assumed that intermittent ventilation was used during, and after, all emission events, including the toasting of bread. However, within the intervention study, occupants did not record using ventilation during all cooking events in Week 2. From Table 3.6, additional ventilation was used during $55 - 75\%$ of cooking events. Occupants most frequently used the cooker hood

during cooking events using the stovetop. It was used less frequently during cooking events using appliances elsewhere in the kitchen, such as the microwave or the toaster. This suggests that in order for the model predicted reductions to be achieved in the stock, people may need to be advised to use ventilation during all cooking events, not just when using the stove.

Finally, the relationship between concentrations, exposure and dose should be considered. Total daily or annual exposure is a function of the concentration and time spent in each microenvironment. This was demonstrated in the yearlong personal monitoring study conducted by Braniš & Kolomazníková [153]. There, the location which contributed most to annual exposure was the primary home. However, the third lowest average $PM_{2.5}$ concentrations were recorded in this location. In contrast, the second highest contributor to annual average exposure was from time spent in restaurants and bars, where the participant spent just 1.2% of her time but was exposed to concentrations of nearly $300\,\mu g/m^3$. Shrubsole *et al.* [191] used a simple ratio to predict annual exposures for a household average, cook and non-cook from living, bedroom and kitchen concentrations. Concentrations were weighted by 0.45, 0.45 and 0.1 (living room, bedroom and kitchen) for the overall household average. Applying this same weighting to the predicted average concentrations in the existing stock in Table 6.8 suggests time spent in the kitchen might contribute a median $18.6\,\mu g/m^3$ to 24-hour mean personal exposure for Strategy A. This is below the WHO 24-hour guideline concentration, however, it does not account for the exposures elsewhere. In addition, this adjustment method does not account for the fact that individuals are more likely to be in the kitchen during cooking or immediately after cooking, when concentrations are highest. Furthermore, it does not account for open plan kitchens, where occupants are more likely to remain in the kitchen for more of the day. A future model could be adapted to report the average predicted concentration during cooking periods as well as the 24-hour average, however, without predicted concentrations in other zones and the corresponding occupancy behaviour, it would not be possible to

predict overall average exposures.

7.1 Model Limitations and Future Development

The $\overline{C(T)}$ model developed in Chapter 6 has a number of limitations, some of which were discussed in the same chapter, but these are explored in more detail here.

The first and most pressing issue is the need for model validation. There is little gravimetrically calibrated $PM_{2.5}$ concentration data for the English housing stock. A future study is required to sample concentrations in kitchens of a representative sample of English houses to allow for future model validation. If concurrent measurements were made in other rooms within the dwelling, following a similar method to Wigzell *et al.* [163] and Hill *et al.* [169], the same data set could also be used to investigate pollutant transport within the dwellings. Similarly, the same sample of houses could be used to implement a similar intervention study to the one presented in Chapter 3 on a larger scale. In this situation, the key improvements to make would be (i) to include gravimetric sampling, preferably with one filter for each 24 hour period, so the measured concentrations can be compared to guidelines; (ii) to include a standardised survey of available ventilation methods and normal use of this ventilation, conducted between Week 1 and Week 2; and (iii) to improve the ventilation instructions to occupants to match the recommendations made in Chapter 6.

A second considerable development is to develop an annual model suitable for predicting average daily and annual concentrations for comparison to guidelines. This model would preferably include pollutant transport between zones, and occupancy schedules for a true comparison. The current model only predicts concentrations for a single day, and assumes a three cooking events per day. An annual model would require multiple cooking schedules, at the very least distinct

weekday and weekend schedules as adopted by Das *et al.* [37] and Shrubsole *et al.* [39, 191].

7.1.1 Cooking Behaviour

The current average concentration model assumed three cooking events per day to represent a reasonable worst case scenario. However, this behaviour was assumed due to the limited data available. This approach is reasonable for predicting 24-hour concentrations, and the probabilistic cooking schedule was similar to the deterministic weekend schedule used previously by Shrubsole *et al.* [39, 191]. The 2005 Time Use Survey aggregated cooking and washing up into a single statistic, where respondents spent and average of 40 minutes per day cooking and washing up at home [7]. The *Time Use Survey* is a large scale household survey of how people in the UK aged over 8 years spend their time, and is uses participant responses in daily time activity diaries [245]. As such, the Time Use Survey results in a large data set, which could be used to derive occupancy and behaviour schedules. The 2014-15 Time Use Survey did include separate categories for "Unspecified food management", "Food preparation and baking", "Dish washing", "Preserving" and "Other specified food management" [245]. As such, the results from this survey could be used to develop new probabilistic cooking and occupancy schedules suitable for use in $PM_{2.5}$ concentration models. However, this was deemed outside the scope of the current work as the large quantity of data involved would make determining the schedules an independent project.

Adams & White [235] used data from the 2005 Time Use Survey to investigate relationships between time spent cooking and population demographics. They found that 60.4% men and 85.0% women spent some time cooking or washing up in one 24 hour period. In contrast, only 33.0% men and 59.5% women spent at least 30 continuous minutes cooking or washing up in the same period. The authors deemed 30 minutes to be the shortest amount of time required to prepare a main

meal. The cooking event duration modelled in Section 6.3 was assumed to equal the α–period from the corresponding emission rate tests in Chapter 4 and Chapter 5. In contrast, the Time Use Survey and Adams & White [235] include all types of food preparation activity within their definition of cooking, which will include chopping and preparing food before heat is applied. In fact, preparing a sandwich entirely from ready-to-eat ingredients would be classified as time spent cooking in the latter definition, despite it not including processes known to emit $PM_{2.5}$.

For comparison, across the total ten monitoring weeks included in Chapter 3, the mean time spent cooking ranged between 23 minutes/day (House C Week 2) and 261 minutes/day (House E Week 1). This time cooking was divided across multiple events, ranging between mean 2.6 (House C Week 2) to 11.9 (House E Week 1) cooking events per day. However, this behaviour is only representative of a very small percentage of the English housing stock, as multiple occupancy households such as student housing compose less than 1% of the English housing stock [226].

News headlines in 2014 reported a trend towards *"Can't cook won't cook Britain"*, stating that the time spent cooking in Britain had halved since the 1980s [246]. The *Daily Mail* is not the most reputable source of information, and the news report does not clearly declare the data source. However, the National Food and You Survey from the Food Standards Agency recently reported the popularity of eating pre-made food [247]. 21% of respondents reported eating pre-packaged sandwiches and 25% reported eating ready meals at least once per week [247]. Eating out is also popular, with 38% of respondents reporting eating out or buying takeaway for breakfast, 70% eating or buying lunch outside the home, and 85% eating out for dinner [247]. However, 71% women and 46% men did report cooking at least once per day. Compared to the first wave of the survey from 2010 [248], the percentage of men cooking at least once per day had increased, and the percentage of women had remained the same.

Firstly, these datasets offer some evidence to suggest that the assumption of

cooking three meals per day in Section 6.3 might be an overestimation in many UK houses. However, these sources contain insufficient data to create an alternative cooking schedule. Dimitroulopoulou *et al.* [194] determined time-activity profiles from the BBC Survey for Daily Life, but they appear to be based on survey data from 1998. The recent changes to cooking habits discussed previously suggest the Dimitroulopoulou *et al.* cooking and occupancy schedules might not be an accurate representation of contemporary Britain.

The existing surveys discussed here were not created with the intention of creating cooking schedules for probabilistic models of cooking source $PM_{2.5}$ concentrations in dwellings. Only the Time Use Survey includes details of time spent conducting a particular activity, but here the participants report activity based on a activity diary divided into 10 minute intervals [7]. This method might miss shorter cooking activity, such as toasting bread or frying eggs.

7.1.2 Representative Cooking Emission Rates

In Chapter 6, cooking emissions were modelled using the emission rates measured in Chapters 4.3 and 5, however, multiple factors influence emission rates from cooking, see Section 2.5.2. Figure 7.1 outlines key parameters identified in literature and explored in empirical tests within this work, and some not yet investigated in detail.

The test meals designed and tested in Chapter 5 were derived from quantities of ingredients eaten in the Dutch National Food Consumption Survey (DNFCS) [220], but the cooking methods were entirely constructed by the research team as the DNFCS does not include details on the corresponding cooking behaviour. Therefore, it is reasonable to consider whether these test meals are representative of meals cooked in the UK, and methods by which representative cooking emission rates might be obtained.

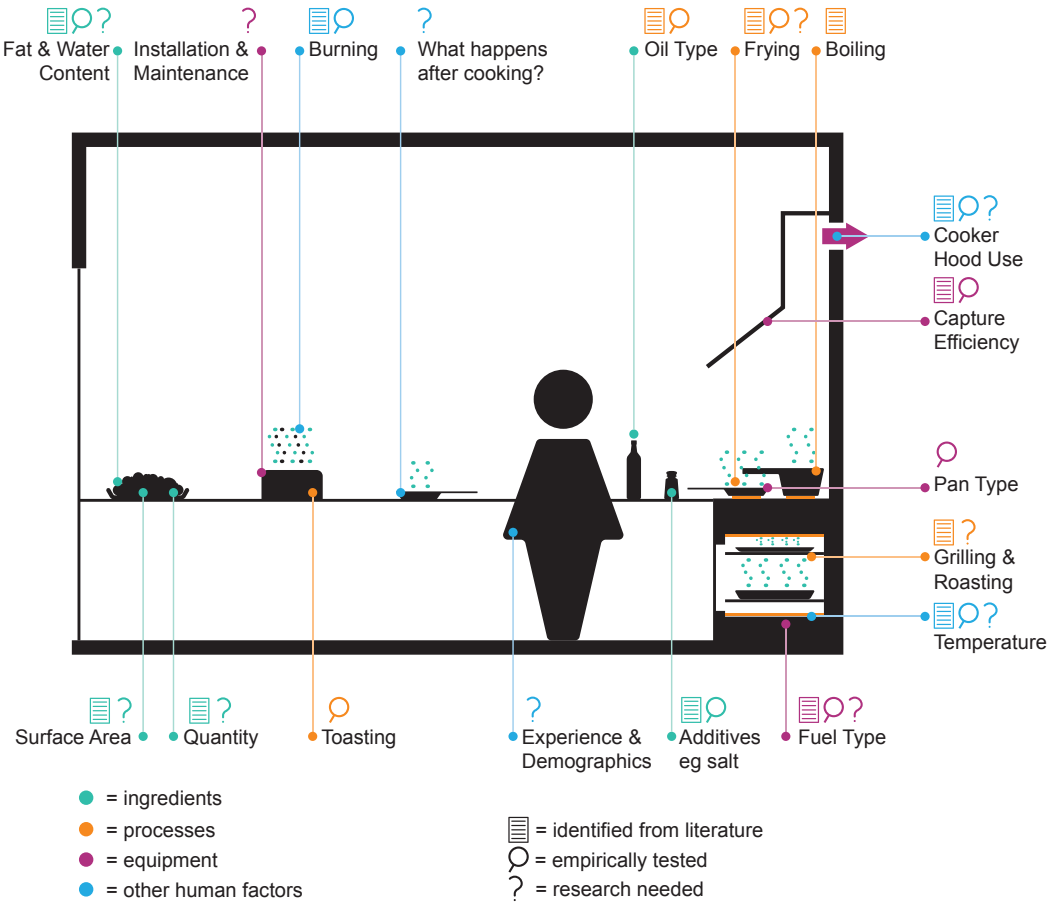


FIGURE 7.1: Factors that influence $PM_{2.5}$ emissions to the room

In the UK, the National Diet and Nutrition Survey (NDNS) is a rolling survey conducted on behalf of Public Health England and the Food Standards Agency to assess diets, nutrient intake and nutritional status in the UK population [249]. The data is similar to that produced by the DNFCS, however there is less resolution in the reported data. For comparison, the mean mass of meat and meat products, potatoes and other tubers, and vegetables consumed by adults aged $31-50$ were given in the DNFCS as 125 g/day (men) and 88 g/day (women), 194 g/day (men) and 140 g/day (women), and 143 g/day (men) and 135 g/day (women) respectively [220]. However, in the NDNS, the results for all adults aged $19-64$ are aggregated. In addition, there is no category equivalent to potatoes and other tubers. However, the average total meat consumption is comparable to the DNFCS, with mean

131 g/day for men and 86 g/day for women [249]. The NDNS also does not report separate statistics for vegetable consumption, instead grouping fruit and vegetables together [249].

An alternative source of information might be the *Food and You* Survey, also conducted by the Food Standards Agency. Where the NDNS aims to investigate the population nutritional intake [249], the Food and You survey is more interested in the public's reported behaviours, attitudes and knowledge of food safety with regards to purchasing, storage, preparation, and consumption [247]. 81% of respondents reported eating chicken or turkey at least once a week, and 55% ate beef, lamb or pork once a week, with men more likely to eat meat than women [247]. The reverse was true for fruit and vegetables, where women were more likely to eat raw fruit, and raw or cooked vegetables every day. However, the Food and You survey data does not include quantities of food consumed, so may be less informative for constructing representative meal portions.

A common flaw with these surveys when trying to construct representative meal types is that they include details of the key food types eaten, but not the cooking methods used to prepare them. Both food type and cooking method have been found to influence $PM_{2.5}$ emissions, see Section 2.5.2 and Figure 7.1. Chapter 4 considered a single dry cooking process, namely toasting, and Chapter 5 included multiple stovetop cooking processes. Most significantly, the concentrations measured whilst preparing Meal 3 and Meal 4 in Chapter 5 suggest that high temperature frying emits large quantities of $PM_{2.5}$, see Figures E.13 and E.14. Previous studies introduced in Section 2.5.2 suggested that cooking foods with high fat content causes higher $PM_{2.5}$ emissions than lower fat [116, 123, 201]. In Meal 3, the highest concentrations were measured whilst frying the bacon (see Figure E.13), the ingredient with the highest fat content (see Table 5.1). However, in Meal 4, stir-frying the vegetables at a high temperature only appears to have resulted in slightly lower $PM_{2.5}$ emissions than stir-frying the chicken, indicated by the

gradient in Figure E.14, despite the lower fat content of the vegetables. This indicated process may be as important, if not more important, as the components of meals being prepared.

In Chapter 3, the cooking methods used most varied between houses (see Figure 3.6). Cooking events which used water based processes such as boiling or steaming, the oven, and multiple cooking methods contributed most to the overall cooking time. Comparing the frequency of different cooking methods by time rather than number of occurrences weights the finding towards slower cooking methods. For example, it typically takes longer to a cook single food type, such as sausages, in the oven than grilling it, which in turn is slower than frying. Many of the cooking events recorded as using multiple cooking methods in Section 3.3.4 were similar to Meals $1-4$ in Chapter 5 in that they were composed of multiple stovetop cooking processes, and most other multiple method cooking events included a combination of oven based and stovetop cooking events. Again the data collected in Chapter 3 is not representative of cooking behaviour across the UK as it was based on a small sample size and because multiple occupancy households including student households represent less than 1% of the English housing stock [226]. Figure 7.1 also identified that occupant demographics and experience might influence cooking behaviour, with less experienced 'cooks' more likely to be dependent on heating preprepared components or blindly following recipes without knowing what to expect. The resulting impact on emissions has not been investigated.

Few studies included in Table 2.10 reported emission rates from cooking known complete meals. Chapter 5 aimed to fill this gap by measuring emissions from cooking a series of full meals, however, these emissions were not compared to those from cooking each of the elements separately. Here, the aim was to produce emission rates suitable for use in probabilistic modelling. However, as part of a future process to develop representative meals, it may be appropriate to undertake

a *component based* analysis of cooking emission rates. If the emission rates from each component have an additive effect when combined, then values of g measured from cooking individual ingredients could be combined to produce meals. It is possible that the relationship is more complex.

Meals $1-4$ were all composed of similar stovetop cooking processes including frying and boiling, yet the $PM_{2.5}$ concentration profiles differed between meals, see Figure 5.2. One possible cause for the discrepancy is the difference in the ingredients, however, the combination of processes might also be significant. With these test meals as a base, series of tests could be used, to investigate the effect of factors including changing the ingredients, their surface area or mass, the speed and temperature at which they are cooked, and the cooking processes used. For instance, how do the emission rate and source strength change if Meal 1 is prepared with sausages instead of chicken, or if the chicken is grilled or baked instead of fried. The effect of substituting the boiled ingredients might also warrant investigation, but is likely to have less effect.

Figures E.11 to E.14 indicate that the structure of the cooking process is also important. Meal 3 involved initial high temperature frying, then frying at a lower temperature before adding liquid after which the contents of the large frying pan was left to simmer. In this case, the resulting meal was a pasta bolognese, but other meals, perhaps risotto [250] or curry [251], may also follow a similar sequence of processes. A systematic component based approach to determining representative meal types and corresponding g and G in this manner would require interdisciplinary research, most likely including food or nutritional scientists. Initial investigations to identify key mechanisms and ingredients could be used to inform the development of a representative survey to investigate cooking behaviour in the UK.

A key consideration is that Meals $1-4$ in Chapter 5 only used stovetop cooking processes, however, cooking events using the oven were popular, see Figure 3.6.

Furthermore, some of the highest peak concentrations observed in House B coincided with times the occupants used the oven, see Figure C.10. The same was not true in other participating houses. These high concentrations were observed in House B regardless of the food type being prepared, which might suggest the oven itself contributed to the high emissions. One possible explanation is that the oven was dirty; Torkmahalleh *et al.* [200] considered the cleaning conditions of appliances and utensils a possible influencing factor for emission rates. If a future test revealed similar results, an intervention might be to clean the oven and repeat the measurement period.

House B was also the only house included in the intervention fitted with a gas oven. Dennekamp *et al.* [31] found UFPs were emitted whilst from stovetop gas burners whereas the *blank* tests in Chapter 5 found that the gas burners did not emit $PM_{2.5}$, therefore there is mixed evidence to suggest whether the burner in the gas oven could have itself been the $PM_{2.5}$ source. Similarly, $PM_{2.5}$ can be a product of incomplete combustion [40], as can carbon monoxide (CO) [104]. CO was measured in the participating kitchens but the measured concentrations were not presented in Chapter 3. Figure C.10 shows the temporal $PM_{2.5}$ and CO concentrations measured in House B. Here, the concentrations corresponding to cooking events using the oven are highlighted in orange. Some of the peak CO concentrations coincided with the oven being used, which may suggest the oven was incorrectly installed, poorly maintained or poorly ventilated. Further tests were not conducted as it was outside the scope of the intervention test and specific to this house. As an aside, the peak CO concentrations from House B were $1.864\,mg/m^3$ in Week 1 and $0.8155\,mg/m^3$ in Week 2, both of which were comfortably below the lowest CO guideline set out by the WHO of $7\,mg/m^3$ average over 24 hours [104].

There are two reasonable approaches to investigating representative emission rates. The first, explored thus far, would use a representative survey of time spent cooking, food cooked and cooking methods to construct truly representative meal

types. The emission rates from cooking these meals would then be measured under laboratory conditions. Chapter 5 aimed to take this approach, however, due to lack of available data, the cooking methods and times were largely assumed. An additional disadvantage of this method is that the high complexity and subsequent variation of cooking makes it difficult to define representative meals. For modelling purposes, probability distributions would then be required to define the frequency of each meal type, and the corresponding emission rate and duration of emissions, as in Chapter 6.

An alternative, more direct method might be to produce two probability distribution functions, one to represent emission rates from all cooking sources, and one for the duration of all cooking events. This could be achieved through residential monitoring similar to the methods used by He *et al.* [35], Chan *et al.* [36], and Nasir & Colbeck [165]. For distributions suitable to estimate $PM_{2.5}$ concentrations in a housing stock such as the English housing stock, monitoring would need to be conducted in a representative sample of houses from the same stock. Here, it might be important to ensure households demographics within the sample are also representative of the population, as this may influence cooking behaviour [235]. Ideally, the selection process would align with that of either or both the EHS and the Time Use Survey to allow for analysis of sections of the stock as well as considering the stock as a whole.

A measurement protocol including records of cooking activity similar to He *et al.* [35] and Nasir & Colbeck [165] would be preferred over the method used by Chan *et al.* [36]. All cooking events are identified with this method, not just those that result in identifiable concentration peaks, and concentration peaks not related to cooking events can be excluded. In addition, using a time activity diary allows the total duration of the cooking period to be identified. Chan *et al.* [36] defined the emission period as being from the time concentrations began to increase above background levels to the time of peak concentration. In the case of cooking

emissions, this method only includes the parts of the event which produce $PM_{2.5}$. In order to produce emission rates suitable to use coupled with population cooking behaviour, it is more useful to define an emission rate to represent the entirety of the cooking period. The concentration profiles presented in Figure 4.13 and Figure 5.2 show that the definition of the emission period adopted by Chan *et al.* might not include the entire emission period. The resulting emission rates and source strengths might be high compared to those from a method that includes the total duration of the cooking period. Neither method is intrinsically wrong, but it is important to understand the definition of the cooking period in the population behaviour data and the measured emission rate data when selecting or collecting data sets suitable for use in modelling, to ensure their compatibility.

He *et al.* [35], Chan *et al.* [36], and Nasir & Colbeck [165] all used optical measurement devices, and their methods depend on time resolved $PM_{2.5}$ concentrations to determine emission rates. The calibration method is a source of error in the estimated emission rates. Only Nasir & Colbeck [165] used concurrent gravimetric sampling, employing the in-built filter on their OPCs. Chan *et al.* [36] also attempted to adjust their concentrations using a general calibration factor. The variation in CFs found between meals in Chapter 5 and between SidePaks in Chapter 4 shows that custom calibration is necessary for optical measurement devices to provide accurate mass measurements. In addition, as calibration factors have been found to vary between sources, both in Chapter 5 and previously by Dacunto *et al.* [40] and Jiang *et al.* [138], any attempt to determine emission rates from residential monitoring will have additional associated calibration errors if a single CF is used.

As discussed in Section 3.4, the standard CF method aims to correct the mean mass concentration measured by the optical device over a given time period to match the mean mass concentration from the gravimetric method. This method provides sufficient resolution to compare the measured concentrations to guidelines, as

current guidelines are also set as mean concentrations. However, using a single CF over the measurement period means the magnitude of elevated mass concentrations during cooking may not be accurate when the CF varies between meals. Ideally, separate gravimetric samples should be collected for each cooking period. Current gravimetric sampling techniques require occupants of investigators to manually change the filters to achieve multiple CFs, which is an impractical monitoring protocol. Therefore any emission rates determined from residential monitoring would be subject to calibration error.

Both methods of determining representative emission rates are time intensive, and the second method utilising a representative monitoring programme is also an invasive approach. However, this second method has a distinct advantage that it would also produce indoor $PM_{2.5}$ concentrations for a representative sample of the housing stock which could then be used to aid model validation.

7.2 Aggregated vs Granular Stock Modelling

The models developed and used to predict concentrations in Chapter 6 considered the entire English housing stock in a single, aggregated model. Infiltration, kitchen volume and cooking behaviour were all treated and sampled as independent inputs. However, there may be some correlation between these properties related to factors including the housing type, occupant demographics, and dwelling age. For the aggregated model approach used here, it is reasonable to sample these inputs independently, and assume that the number of samples is sufficient to produce a representative output distribution.

In contrast, Shrubsole *et al.* [39] separately considered the effect of energy retrofit measures on predicted $PM_{2.5}$ concentrations in dwellings above and below the low income threshold. They used relevant variables from the EHS to identify the built form, permeability, current and potential retrofit level, tenure, indoor pollutant

regime, and household income level. The authors were then able to separately consider the impact of retrofits of four different sub-stocks, defined by household income level. Shrubsole *et al.* used a deterministic model however, probabilistic methods could also be used to create granulated models suitable for comparing the performance of different subsections of the entire stock. Here, many parameters could be used to define the sub-stocks, so it would be important to clearly establish the research question before developing model. As discussed above, Shrubsole *et al.* [39] compared different income groups.

Occupant demographics are likely linked to occupancy and cooking patterns. Aragon *et al.* [252] developed probabilistic English domestic occupancy profiles, to present an alternative to the two deterministic occupancy profiles frequently used in literature. These two typical profiles are: (i) a family with dependent children where the parents work full time; and (ii) a retired elderly couple who spend the majority of their time indoors. Das *et al.* [37] and Shrubsole *et al.* [39, 191] used $PM_{2.5}$ emission schedules intended to represent a similar household to the first of these typical household types. However, Aragon *et al.* [252] identified that these household types represented just 19% of English households between them, and as such, these occupancy and behaviour profiles might not be representative of the population. Instead, Aragon *et al.* defined 33 household types using employment status and household composition data from the EHS. These types were then matched with householders in the Time Use Survey, and the resulting data used to develop occupancy profiles with 3 states: (i) at home and sleeping; (ii) at home and not sleeping; and (iii) away from home. A similar process could be followed to develop an improved model of cooking behaviour, or to include occupancy patterns for different rooms within the dwelling, both parameters which impact exposure. However, the middle stage of matching data from the EHS and Time Use Survey was complicated by the fact that both surveys do not report the same parameters to identify household information. Instead the researchers matched the household composition and individual work status by analysing the Time Use

Survey interviews and re-categorising the data accordingly.

7.3 Measuring and Calculating $PM_{2.5}$ Emission Rates

Two different methods have been used to calculate $PM_{2.5}$ emission rates within this thesis, and a third option was also presented in Section 2.5.4.1. All three methods are based on the mass balance model, but each makes slightly different assumptions about the conditions within the test chamber, and the duration and nature of emissions.

The steady state method assumes constant $PM_{2.5}$ emissions until concentrations reach a near constant level. This behaviour was not observed in any of the $PM_{2.5}$ measurements in this thesis, therefore this model was rejected as it appeared to be unsuitable. Torkmahalleh *et al.* [202, 205] used this method to measure emissions from heating different types of cooking oil. They conducted the experiments in a small volume and measured for an extended time period under constant conditions. Therefore, concentrations were more likely to be near the steady state concentration.

The *peak* or *theoretical peak* estimation method used in Chapter 4 is most frequently used in literature, and assumes constant emissions during the α–period, so that the peak concentration occurs at the end of the α–period and can be used to estimate the emission rates. If this assumption is not met, as in Chapter 5, then the resulting emission rate and source strength from the peak method might be higher or lower than that predicted by the area-under-the-curve method. Here, the use of time-resolved optical measurement devices can help select the most appropriate method, but the intended purpose of the predicted emission rates is also a consideration. For example, Figure 2.5 showed a comparison of the predicted concentration curves from emission rates determined with each method in turn. If the purpose of the emission rates is as input to a model of average concentrations,

as in Chapter 6, then the most appropriate calculation method is the one that predicts the average $PM_{2.5}$ emission rate over the total emission period. Therefore the area-under-the-curve method was most appropriate for Chapter 5.

The concentration profiles for toast did not suggest a constant emission rate, however, as the peak concentrations occurred after the end of the emission period, the theoretical peak estimation method was deemed to best approximate the emission rate. In addition, this method was able to account for the lag while the emitted particles mixed throughout the space.

Chapter 4 also highlighted the important of the measurement interval for time-resolved data. In phase 1, the 1 minute logging interval resulted in too few data points within the α–period to clearly understand the emission profile. The 1 second interval in phase 2 provided greater resolution, but there was also considerable noise in the data. This noise created additional challenges, especially with regards to identifying the end of the β–period. However, visual inspection of the decay plots indicates the method was generally successful, see Figure D.8.

For non-constant source emissions, such as the test meals in Chapter 5, there might sometimes be value to using the peak estimation methods. The current $PM_{2.5}$ health guidelines focus only on average concentrations, however, if a metric were established based on instantaneous exposures it might also be beneficial to predict peak $PM_{2.5}$ concentrations. When the peak concentration occurs near to the end of the emission period, Figure 2.5 shows that the peak estimation method is best suited to predicting the final, or end, concentration. Therefore, here it would be more appropriate to define the emission period following Chan *et al.* [36], and only consider the time up to the peak concentration. However, it may be difficult to source corresponding cooking behaviour data.

Dacunto *et al.* [40] reported their measured $PM_{2.5}$ emissions in three forms, as a source strength (mg), an emission rate (mg/min), and as an *emission factor* (mg/g). This latter metric was the emitted mass per mass burned with combustion sources,

or per mass of raw ingredients in the cooking tests. Similarly, Torkmahalleh *et al.* [206] also reported the *emission flux* ($\mathrm{mg}/(\mathrm{min\,m^2})$), where they normalised the measured mass emission rate by the surface area of the pan or the meat cooked. The emissions measured in Chapter 4 and Chapter 5 were reported as emission rates and source strengths as these are the most useful for the current modelling approach. In Chapter 4, the slices of bread were not weighed before or after cooking, and their surface area was not measured. During the second phase of tests, they appeared to be smaller in size, see Figure D.9c, but they were not measured so it was not possible to conduct further investigations.

In Chapter 5, the solid ingredients were weighed before cooking, but not after. In addition, the liquid ingredients were not weighed. The second Dacunto *et al.* [40] definition of emission factor might be obtained from the recorded data, which could form another means to determine emission distributions from distributions of mass of ingredients used. However, oil is a known source of $PM_{2.5}$ during cooking and the mass of the oil was not measured directly. Furthermore, the ingredients were weighed or measured out within the tolerances established in the cooking protocol in Table 5.1 to maintain consistency between repetitions, however the exact mass used in each repetition was not recorded, as the investigation did not aim to estimate emission factors.

With regards to cooking processes, perhaps the first definition of emission factor set out by Dacunto *et al.* [40] for combustion processes might be of more interest. From the basis of the mass balance principle, mass cannot be created nor destroyed. Not all loss of mass from ingredients during cooking will be to form $PM_{2.5}$. Here toasting is a good example, where the bread is first dried out causing emissions of water vapour before Maillard browning begins to occur. $PM_{2.5}$ is also unlikely to be the only pollutant emitted. However, comparing the estimated mass lost, obtained by weighing the bread before and after cooking, to the $PM_{2.5}$ source strength would provide a means to investigate what proportion of total emissions are attributed to

$PM_{2.5}$. Naturally, this would require an accurate source strength, so the area-under-the-curve calculation method may be preferable.

In more complex cooking processes, such as those included in Chapter 5, this definition of emission factor would require the combined mass of all solid and liquid ingredients to be measured before and after cooking. The reason for including wet ingredients in the mass measurement is that, depending on the cooking process, wet ingredients are often absorbed by the solid ingredients causing the mass of the solid ingredients to increase. One such example is boiling pasta in water. However, after cooking, it might be difficult to accurately measure the mass of the ingredients if, for example, the cooking process has caused them to stick to the pan.

The Torkmahalleh *et al.* [206] definition of emission flux may also have offered insight into the variation in emission rates both between meals, and tests, in Chapter 5 and between measurement phases in Chapter 4. A larger surface area per unit mass causes the food to dehydrate and cook faster, and there is a larger exposed surface on which Maillard browning can occur. Meal 1 and Meal 2 both specified chicken breast with mass within a specific range. Where possible, this was achieved using two chicken breast fillets of roughly equal size. Sometimes this was not possible and smaller pieces of chicken were also fried, resulting in a higher ratio of surface area to mass. Meal 4 also used pre-diced chicken breast which had a higher surface area to mass ratio than the chicken fillets in Meal 1. Combined with the higher surface area, this may have contributed to the higher emission rates observed in Meal 4. The relationship between food surface area and particulate emissions has not been widely considered in literature.

Torkmahalleh *et al.* [253] measured $PM_{2.5}$ emissions when deep frying tofu in corn oil. They compared $PM_{2.5}$ concentrations for heating just corn oil, and then deep frying tofu with mass 12.5 g and surface area 32 cm^2, mass 30 g and surface area 60 cm^2, and mass 75 g and surface area 108 cm^2. There was no major increase in

particle concentrations from corn oil alone and with the two smaller pieces of tofu added, however concentrations were 10 times higher with the highest piece of tofu added. However, this investigation was not a true test of the effect of surface area, as the mass also changed between tests. Additionally, while Torkmahalleh *et al.* [206] normalised their measured emission rates by food surface area to determine emission flux, they did not investigate the relationship between food surface area an resulting emissions. Perhaps one reason the effect of surface area has not been widely investigated is because the irregular shape of ingredients makes it difficult to determine their surface area, however, this relationship warrants systematic investigation as part of a component based analysis of emissions.

7.4 Assumptions of Homogeneity and Pollutant Spread

Chapters 3 to 6 all made the same assumption that emitted $PM_{2.5}$ was uniformly mixed within the space. Within Chapter 5 and Chapter 6, this complete mixing was assumed to be instantaneous, whereas accommodation was made for a delay in the the mixing conditions in Chapter 4. This assumption is common throughout the existing literature, but there are some limitations to this approach.

In the intervention test, the consequence of this assumption was that the concentrations measured at a single point were assumed to be representative of those throughout the space. However, the measured concentrations were not used to estimate health impact or dose, and so this assumption is not important to the results of this particular test.

As first mentioned in Section 5.3.3, the assumed volume of the space may not equal the mixing volume when measuring emission rates. In both the tests measuring emissions from toasting bread and those measuring emissions from cooking full meals, the volume of the test chamber was taken to equal the volume of the room. However, the volume of furniture, equipment and people within the space was not

subtracted. As such, the volume used to estimate g was likely larger than the volume occupied by fully mixed air.

Other published estimates of emission rates included in Table 2.10 also assumed full mixing within the volume, though it is not always clear how the mixing volume is defined. Furthermore, one personal monitoring study assumed the air was fully mixed throughout the entire dwelling volume [199]. Full mixing is unlikely to occur throughout the dwelling, as demonstrated by Wigzell *et al.* [163], Nasir & Colbeck [165], and Hill *et al.* [169] who measured lower particle concentrations in rooms other than the kitchen than in the kitchen in most cases.

The modelled approach in Chapter 6 also assumed the mixing volume of the kitchens was equal to the full volume of the kitchens as reported in the EHS. An advantage of using the whole, empty kitchen volume when measuring emission rates is that there is no information on furniture for use in modelling, and so predicted concentrations will be correct. This is true in kitchens with a similar size and configuration to the TNO test kitchen, where the proportion of the total volume occupied by furniture is likely to be similar. Under these circumstances, the same assumptions were made in both the emission rate calculation process and the subsequent model. However, the EHS also includes kitchens with much larger volumes, see Figure 6.5, where furniture and other items may occupy a smaller percentage of the total volume. This survey does not include data on the volume of the contents of the space, so any estimation of the mixing volume has to make similar assumptions. Other previous studies of pollutant concentrations in the English housing stock have not accounted for items in the space reducing the mixing volume [39, 191, 194].

With regards to estimating total or indoor $PM_{2.5}$ exposure, the average concentration model used in Section 6.3 has one distinct disadvantage, in that it considers the kitchen as a single zone in isolation from the rest of the dwelling. Shrubsole *et al.* [191] could to estimate overall occupant exposures using a simple

ratio because their approach employed internal pollutant transport software to predict concentrations in multiple zones of the house. Shrubsole *et al.* used stock archetypes and deterministic modelling, which helped limit the number of simulations run. Alternatively, Logue *et al.* [254] developed a physics-based mathematical model to investigate residential $PM_{2.5}$ infiltration in the US housing stock. In this model, the entire house was modelled as a single zone, taking the opposite extreme approach to Section 6.3. Logue *et al.* did not include $PM_{2.5}$ from indoor sources, therefore it may have been more appropriate to represent the house with a single mixing volume.

Where indoor $PM_{2.5}$ sources are modelled, proximity of the occupant to the source is also important to exposure. Most mathematical models or simulation tools, whether producing a single or multizone model, consider the air to be fully mixed within the zone. However, even when the room air is reasonably well mixed, individuals in close proximity to the source might be exposed to higher concentrations. Wallace *et al.* [145], Adgate *et al.* [154], Lai *et al.* [155], Mohammadyan & Ashmore [158], Williams *et al.* [160], and Ozkaynak *et al.* [170] all measured higher personal exposure concentrations than fixed residential particulate concentrations and fixed outdoor concentrations. This difference can be attributed to time spent and concentrations in different microenvironments, and proximity to sources and incomplete mixing. Ozkaynak *et al.* [170] suggested the presence of a *personal cloud* to explain the higher personal concentrations. This personal cloud has been attributed to a combination of the *proximity effect*, where individuals are closer to the source, and a *Pigpen effect*, where particles fall off clothes and are resuspended from surfaces due to individual movement [145]. This phenomenon has since been explored elsewhere in literature [145, 154], and may offer a means to scale predicted room concentrations from models to estimate personal exposures. However, the magnitude of measured personal clouds varies between and within studies, and includes negative personal clouds where personal exposures were lower than the concentrations in microenvironments. For example,

Adgate *et al.* [154] found personal clouds in the range $-64.5 - 198.3\,\mu g/m^3$, with mean $11.5\,\mu g/m^3$ and median $5.7\,\mu g/m^3$, and Wallace *et al.* [145] reported individual personal clouds of $-15 - 20\,\mu g/m^3$. Adgate *et al.* also found some correlation between time spent outdoors and individual personal clouds. The factors affecting the personal cloud are not currently well understood, and as such more data would be needed to include this parameter in model predictions.

7.5 Ventilation and Kitchen Design

Some parameters which influence emission rates, mitigation methods and pollutant spread throughout the dwelling might be used to inform kitchen design to promote healthy dwellings.

The first parameter to consider is ventilation. Ventilation rates for constant and intermittent were recommended in Section 6.3.3.2, assuming mechanical extract only. UK dwellings are typically naturally ventilated with local mechanical extract ventilation in the "wet" zones, primarily the kitchen and bathrooms. This helps to create negative pressure in these zones to help prevent the pollutants spreading to other occupied rooms. Therefore, these conditions were the only options considered. However, the combined effect of constant mechanical extract with intermittent boost ventilation during cooking, as is common in Dutch dwellings, was not investigated. Furthermore, only the winter ventilation conditions were considered. The effect of opening windows to provide additional natural ventilation was not considered. Here it would be important to consider the effect of natural ventilation on the airflow patterns both within the kitchen and the entire dwelling.

With regards kitchen ventilation, regulators the choice of specifying ventilation rates as airflow rates (L/s or similar), air change rates (h^{-1}), or both. ASHRAE 62.2 gives options for both and air change rate and volume flow rate in enclosed

kitchens, where the smaller volume means the required air change rate might be met with a lower airflow rate. For larger, open plan kitchens, the same option is not available, with only a minimum airflow rate being specified. Specifying air exchange rates might provide more IAQ consistency between dwellings. However, as the air change rate depends on the volume of the space in which the fan is installed, specified volume flow rates are more easily implemented and tested by installing a fan specified to provide the correct airflow. In cooker hoods, where capture efficiency is a function of the flow rate, manufacturers could not test and produce curves such as those in Figure 6.11 for air change rates, again because this parameter depends on the room volume.

In the current circumstances of a National Climate Emergency [255] it is critical to minimise the energy demand of the housing stock. Here the additional energy demand associated with driving the fan and compensating for the heat loss of additional ventilation is critical. Section 6.3.3.2 did not model energy demand. From the model predictions, using a cooker hood rather than general kitchen extract to provide intermittent exhaust ventilation can mean a lower fan airflow rate is required. Using a high performance hood would reduce the energy demand of the kitchen ventilation system. The energy requirements of running the fan itself would be relatively easy to predict; it is a function of the airflow rate, pressure losses across the systems, fan efficiency and the operating time, values for which can be assumed. Heat loss is slightly more complex, as it is a function of the temperature difference from internal to external. The Chapter 6 models do not model the thermal environment and do not include external weather conditions. An updated model using either CONTAM or EnergyPlus would have to include these conditions, and so would then be able to predict the energy demand associated with the proposed ventilation strategies. Here, the time at which cooking occurs would be more important, as it would relate to the external temperature. Within the energy analysis, the energy demand of using a fan with higher flow rate just during cooking should be compared with that of a lower flow

rate during cooking and for an additional 10 minutes. The high and low flow rate could be set using Figure 6.10 to provide the same daily average concentrations.

Another consideration is that local extract ventilation can be noisy, and the noise of a system often deters occupants from using such ventilation methods [183]. Singer *et al.* [183] found many of the domestic hoods they measured increased the sound levels above 60 dB even when used on low or medium settings, which they state would interfere with conversation. There was also a correlation between hood flow rate and sound levels, and so devices with a high capture efficiency at low flow rate may be used more, and therefore perform better as a mitigation tool.

Overall the optimum ventilation system would be a cooker hood that can achieve near 100% capture efficiency at low airflow rates, thereby reducing the noise and associated energy requirements. The feasibility of such a hood is unlikely, but this ideal hood could be used to focus cooker hood design. Of course, to be popular, this ideal cooker hood would also be aesthetically pleasing, ergonomic and affordable. The challenge for manufacturers, therefore, is to use current knowledge to produce and test hoods with high capture efficiencies at low airflow rates.

The design and position of cooker hoods is also crucial to capture efficiency and resulting dwelling concentrations. The example hood curves in Figure 6.12 show that increasing the airflow rate through a cooker hood increases its capture efficiency. However, other factors that affect the capture efficiency have also been identified, including mounting height, hob coverage, and design features to entrain pollutants towards the device, see Section 2.3.1.1 and Section 5.3.5 for more information. Most studies of capture efficiency, including the ASTM test method [187], focus on wall mounted hoods positioned between wall cupboards. This configuration was used in the TNO test kitchen, see Figure E.3, and was observed in House A and House E, see Figure C.3 and Figure C.2. A different test method, not yet developed, is required for island hoods and downdraft devices [186], which display different airflow characteristics to wall mounted hoods. Jacobs *et al.* [185]

found increased capture efficiencies when side plates were fitted to the hood, and Sjaastad & Svendsen [184] found the cooker hood was most effective when positioned in the corner of the room. In wall mounted hoods, the wall itself helps to restrict the plume and direct it towards the hood. This benefit is lost in island hoods, located away from the walls [184].

There is also evidence to suggest that good burner coverage is important to increase hood capture efficiency, as discussed in Sections 2.3.1.1 and 5.3.5. The primary means to achieve this is of course part of cooker hood design, however, hob design might also offer an indirect mitigation method by influencing cooking behaviour. Lunden *et al.* [181], Singer *et al.* [183], and Walker *et al.* [188] found that the hoods they used provided better coverage and higher capture efficiency when cooking on the back burners. This was not observed in Section 5.3.5, however this hood was large and provided good coverage when cooking on the front burners. The cooker hoods from House A and House E (see Figure C.3 and Figure C.2 respectively) did not appear to be particularly high performance, although the burner coverage was not investigated in any of the intervention test kitchens. If smaller cooker hoods are common practice, locating the hotter burners at the back of the stove, closer to the wall, might encourage users to use the rear burners for higher emitting cooking events such as high temperature frying. The hottest burner in the TNO test kitchen was located was located on the back right, used to cook Meal 4, but this is not necessarily the standard in kitchen design.

In open plan dwellings, it is likely even more beneficial to extract emitted pollutants at source through a cooker hood, although this was not tested in this current models. Firstly, if $PM_{2.5}$ is not extracted at source and general ventilation is used to dilute the concentration in the room air, the larger room volume means it would take longer for concentrations to decay back to background level, unless a higher extract flow rate were used. Of course, for the same source strength, the larger mixing volume would result in lower room concentrations. Any benefits of

the dilution in the larger mixing volume might be reduced by the longer occupancy times in these spaces, especially for occupants who are not the cook. Some of these characteristics might also be observed in houses with separate kitchens if, as in House B and D, the internal doors to the kitchen are normally left open. Here, cooker hoods with high capture efficiencies might be more beneficial.

The current $\overline{C(T)}$ model proposed in Chapter 6 assumed that occupants switch on any intermittent mechanical extract ventilation at the exact moment that they start cooking. A public health message could be used to encourage this behaviour, but there is currently no data on normal usage of kitchen exhaust ventilation. One possibility is that occupants forget to use any ventilation until there is a noticeable difference in the IAQ. The model could be modified to test the effect of delaying the ventilation start time by a defined number of minutes or until concentrations reach a specific value above background levels. Similar to how using ventilation after cooking was predicted to have more effect for general extract ventilation than for a cooker hood, see Figure F.2, delaying the ventilation start time is likely to have a greater impact on concentrations when using a cooker hood. This is because the capture efficiency of the hood is only beneficial when it is used to reduce the emissions to the room during cooking.

There has been some suggestion that consumer grade particle or $PM_{2.5}$ monitors could be integrated into smart home systems to help control IAQ and ventilation [186]. However, as discussed previously, designing a cooker hood to switch once a cooker hood has already been reached might limit the effectiveness of the hood. A simpler solution might simple be to link the cooker hood to the cooker itself, so it automatically switches on when the cooker is in use, and runs for 10 minutes after it is switched off to match the recommendations in Chapter 6. Sensors located in the exhaust flue could then be used to detect concentrations of $PM_{2.5}$ and other pollutants, and used to reduce the airflow rate if these concentrations are low so as to avoid over ventilating during low emission events. This area of research is not yet

established and therefore requires further work.

7.6 Health Effects and Guidelines

A final consideration for interdisciplinary research is the need for further research into the health effects of $PM_{2.5}$ from cooking sources. Current guidelines [104] and papers (for example Logue *et al.* [12]) assume equal toxicity of $PM_{2.5}$ from all sources. However, literature included in Section 2.1.3 suggested that the physical and chemical composition of $PM_{2.5}$ might have an impact of the resulting health effects, and that $PM_{2.5}$ composition varies with source. In effect, $PM_{2.5}$ might be better described as a pollutant class rather that a single pollutant. It is not feasible to suggest investigating the health effects of residential source $PM_{2.5}$ using epidemiological methods, due to the measurement constraints. Alternatively, the toxicity of $PM_{2.5}$ collected using gravimetric sampling could be investigated using in-vitro and in-vivo methods similar to those used for ambient $PM_{2.5}$, see Section 2.1.2 for more information.

Secondly, the current $PM_{2.5}$ guidelines are set based on 24-hour or annual average concentrations [104, 128, 129]. This is related to how $PM_{2.5}$ has historically been measured using the gravimetric method, which remains the reference method [133]. The gravimetric method can only be used to measure average concentrations and so the current understanding of concentration-response relationships (see Section 2.1.1.6) relies on time averaged $PM_{2.5}$ concentrations. However, the measured data in Chapters 3 to 5 demonstrates clear peaks and troughs in the temporal $PM_{2.5}$ concentrations. There is currently no evidence to suggest whether exposure to instantaneous elevated peak $PM_{2.5}$ concentrations is associated with health effects; hence, there is a need to identify the presence of appropriate biomarkers in response to short term peak exposures. These questions are not for engineers to answer, but the answers could be critical to guiding future research in the field.

Finally, around 40 000 excess deaths each year are attributed to ambient pollution [1]. In the UK, as in most industrialised countries, the health effects of indoor exposures remain understudied. There will also be a trade off between indoor and outdoor exposure to $PM_{2.5}$ originating indoors and outdoors. Ventilation mechanisms that do not filter incoming air ought to be used strategically, to remove indoor source $PM_{2.5}$ and other pollutants without encouraging pollutant ingress during outdoor pollution events. The kitchen ventilation strategies considered herin so not normally filter extracted air before it is exhausted to outside, thus it might be prudent to consider the effect of the proposed ventilation strategies on ambient $PM_{2.5}$ concentrations, which also impact total exposure and health.

7.7 Recommendations for Future Work

A number of recommendations for future work have been suggested throughout this thesis, and so the key ideas are summarised here for clarity.

1. The addition of energy demand for the proposed mechanical ventilation systems in Chapter 6 to allow comparison between the different ventilation strategies and to assess the energy implications of these recommendations.

2. Develop the average concentration model to estimate total exposures and consequential QALYs or DALYs using a method similar to Hamilton *et al.* [193] or Logue *et al.* [12] in order to conduct a cost benefit analysis of the ventilation system changes recommended in Chapter 6.

3. $PM_{2.5}$ measurement with gravimetric sampling in the kitchens of a representative sample of English houses suitable for use in model validation. This same study could include measurements of multiple pollutants, sampling locations in different rooms of the house to investigate pollutant transport, and an intervention study similar to the one presented in Chapter 3

but with more specific ventilation instructions to match the recommendations from Chapter 6.

4. Measurement of cooker hood capture efficiencies of hoods typically found in the English housing stock, following a method similar to Lunden *et al.* [181] and Singer *et al.* [183] or the ASTM test method [187].

5. An interdisciplinary project to systematically investigate $PM_{2.5}$ emissions from cooking, possibly using a component based analysis. The primary aim of this project would be to develop representative cooking emission probability density functions.

6. Develop probabilistic cooking schedules and occupancy types for different household demographics for use in future modelling.

7. Toxicology research into the toxicity of cooking source $PM_{2.5}$. This could follow similar in-vitro and in-vivo methods to those used by studies introduced in Section 2.1.2 but for $PM_{2.5}$ samples collected in Western kitchens.

Chapter 8

Conclusions

The primary aims for this thesis were to investigate $PM_{2.5}$ emissions from cooking meals typical to a European or western diet, to investigate the associated $PM_{2.5}$ concentrations within the UK housing stock, and to assess the suitability of the current ventilation requirement as set out within Approved Document F of the UK Building Regulations (ADF) [16]. These aims were achieved through a series of empirical and theoretical investigations.

8.1 Empirical Findings

In line with findings from existing literature, cooking activity was found to elevate measured temporal $PM_{2.5}$ concentrations above background levels in the kitchens of multiple occupancy student houses in Nottingham, UK. $PM_{2.5}$ has previously been identified as a priority indoor air pollutant with long established associations between ambient $PM_{2.5}$ and multiple health effects. However, the concentrations measured within this thesis could not be compared to the current World Health Organisation guidelines for $PM_{2.5}$ as the SidePak photometer was not gravimetrically calibrated. Furthermore, the literature review also found a need for research to investigate the health effects of $PM_{2.5}$ from cooking sources. Current

guidelines assume equal $PM_{2.5}$ toxicity regardless of source, and so the guidelines themselves may need revisiting.

A behavioural intervention where the occupants were encouraged to employ existing ventilation during cooking resulted in a statistically significant, small reduction in $PM_{2.5}$ concentrations in 2 of the 5 participating houses. A small yet significant increase in $PM_{2.5}$ concentrations was observed in the remaining houses. However, this study was limited, primarily because cooking behaviour was not consistent between the two weeks, because the performance of the existing cooker hoods was not directly investigated, and because the calibration factor was not measured. Additionally, comparing the 1 minute concentrations from Week 1 with those from Week 2 was more likely to find a statistically significant result due to the large number of data points. Ventilation was used during most cooking events following the intervention, though future interventions may benefits from providing occupants with more precise instructions to use a mechanical extractor fan or externally exhausting cooker hood only.

$PM_{2.5}$ emission rates from cooking were found to vary widely in current literature, with five identifiable methods used to measure them. Therefore, a method was developed to measure $PM_{2.5}$ emission rates with a high number of repetitions suitable for producing probability density functions of emissions suitable for use in probabilistic modelling. Important parameters identified were understanding the room mixing conditions and controlling conditions to aid good mixing, controlling the ventilation conditions to maintain a constant air exchange rate, ensuring an appropriate time step for measuring concentrations with an optical device, and using gravimetric sampling to obtain custom calibration factors.

Following this method, toasting bread was found to emit $PM_{2.5}$ at a rate of $\mu = 0.23\,\mathrm{mg/min}$ ($\sigma = 0.07\,\mathrm{mg/min}$) and an empirical cumulative distribution was produced suitable for use in modelling. These measured emission rates were lower some of than those found elsewhere for the same source. The difference was likely

due to a difference in the degree of charring, although the high repeatability of the final tests might also have produced emission rates less representative of normal behaviour.

Emission rates from cooking full meals using a multiple stovetop processes were measured to range between $\mu = 0.62\,\mathrm{mg/min}$ ($\sigma = 0.10\,\mathrm{mg/min}$) for fried chicken breast, fried potatoes and boiled green beans and $\mu = 3.2\,\mathrm{mg/min}$ ($\sigma = 0.24\,\mathrm{mg/min}$) for chicken and vegetable stir-fry. Custom calibration factors varied between test meals, therefore it is imperative that concurrent gravimetric sampling is used to adjust $PM_{2.5}$ concentrations measured using optical devices. The browning of food, presence of oil or fat and pan temperature all contributed to the magnitude of cooking emissions. Frying in a non-stick pan resulted in significantly lower emissions than using a stainless steel pan, so individuals are recommended to cook using non-stick pans where possible. Using a high performance cooker hood to extract air at a rate of $83\,\mathrm{L/s}$ reduced room $PM_{2.5}$ emissions by $\geq$ 93%, though the test method used here resulted in increased uncertainty in the estimated cooker hood capture efficiency. Instead, the ASTM test method [187] is recommended for rating the performance of cooker hoods.

8.2 Theoretical Predictions

The measured emission rates were used to develop a probabilistic model of 24-hour average kitchen concentrations in the English housing stock informed by the English Housing Survey. Over 98% kitchens were found to be too airtight to provide sufficient ventilation through infiltration to dilute $PM_{2.5}$ concentrations from cooking to concentrations below the current WHO guideline concentrations. The current ADF mechanical extract ventilation strategies were predicted to provide adequate ventilation in $<$ 11% kitchens if only used during cooking. When intermittent ventilation was considered to run for 10 minutes after cooking 46% kitchens were predicted to comply. Continuing ventilation after cooking followed a

law of diminishing returns, and as such individuals are recommended to use existing mechanical extract ventilation in the form of a kitchen extractor fan or exhausting cooker hood throughout the cooking period and for 10 minutes after cooking ends. Recirculating cooker hoods were not considered here.

The model was then used to predict required ventilation rates for 24-hour average concentrations in 75% kitchens to remain below WHO guidelines. For general kitchen extract, a constant airflow rate of 61 L/s ($11\,h^{-1}$) is recommended. Alternatively, 79 L/s ($13\,h^{-1}$) intermittent general extract is recommended when used for the entire cooking period and 10 minutes after cooking ends.

A cooker hood that exhausts air to outside the building is recommended as the most effective form of kitchen ventilation, and its performance is defined by the capture efficiency. Probabilistic modelling was used to produce a tool to relate the required flow rate and capture efficiency for 75% kitchens to remain below guideline concentrations. In absence of empirical capture efficiency data, punitive values should be specified. Assuming a capture efficiency of 50%, an airflow rate of 51 L/s was recommended to maintain 24-hour mean concentrations below the WHO guideline in 75% kitchens of the English housing stock. An additional, as yet unquantified benefit of installing a cooker hood is that the lower required airflow rates result in reduced associated energy demand and lower noise levels.

List of Publications

Included here is a list of my publications produced during the course of this PhD. Any content reproduced within the thesis has been indicated at the start of the appropriate chapter.

Journal Articles

O'Leary, C. *et al.* Investigating measurements of fine particle (PM2.5) emissions from the cooking of meals and mitigating exposure using a cooker hood. *Indoor Air* **29,** 423–438. ISSN: 0905-6947 (2019)

O'Leary, C. *et al.* Setting the standard: The acceptability of kitchen ventilation for the English housing stock. *Building and Environment,* 106417. ISSN: 0360-1323 (2019)

Conference Papers

Long Presentation

O'Leary, C. *et al. An intervention study of $PM_{2.5}$ concentrations measured in domestic kitchens* in *39th Air Infiltration and Ventilation Centre Conference* (Antibes Juan-Les-Pins, France, 2018), 1063–1072

Poster Presentation

O'Leary, C. & Jones, B. *A Method to Measure Emission Rates of $PM_{2.5}$s from Cooking* in *38th Air Infiltration and Ventilation Centre Conference* (Nottingham, UK, 2017), 463–472

O'Leary, C. *et al. Estimated distributions of $PM_{2.5}$ concentrations in the kitchens of the English housing stock for infiltration and mechanical ventilation scenarios* in *39th Air Infiltration and Ventilation Centre Conference* (Antibes Juan-Les-Pins, France, 2018), 1090–1099

Appendix A

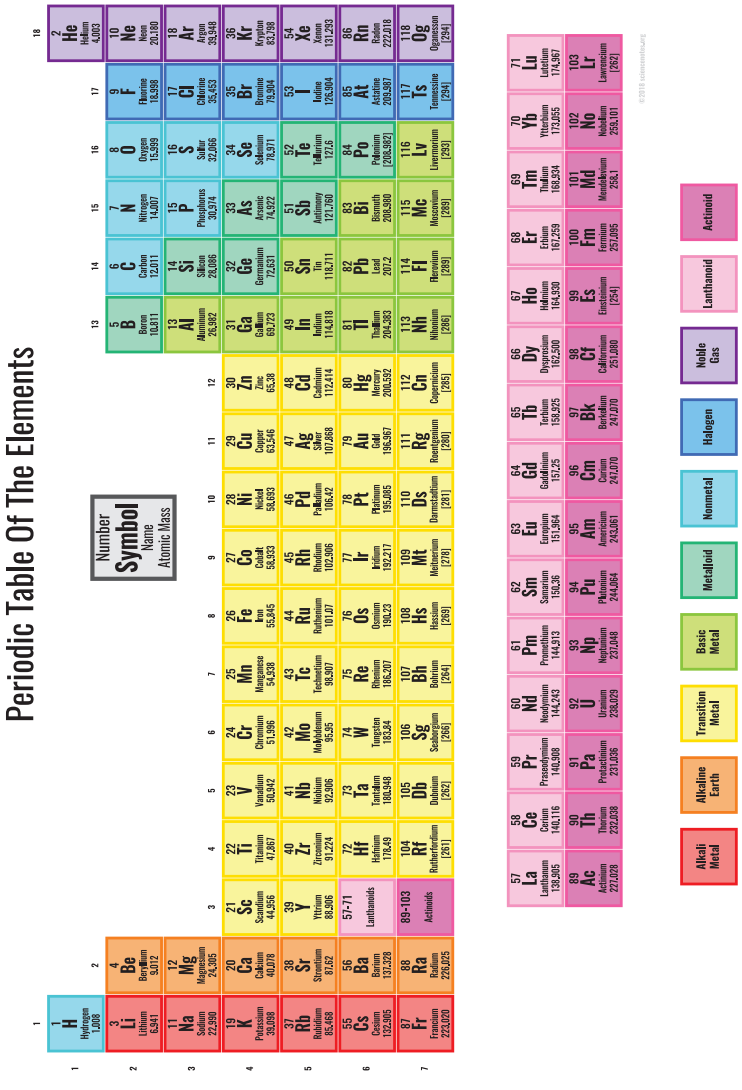


FIGURE A.1: Periodic Table of Elements
from sciencenotes.org/periodic-table-pdf-2

Appendix B

Derivation of the Mass Balance Equation

Included in this appendix is the mathematical derivation of the mass balance equations used throughout the thesis. This derivation is based on that provided by Ott *et al.* [44], but updated to incorporate non-ventilation decay mechanisms, the penetration factor and cooker hood capture efficiencies. The starting point is Equation B.1.

$$\left(\begin{array}{c}\text{Mass generated}\\ \text{between Time}\\ t=0 \text{ and } t=T\end{array}\right) - \left(\begin{array}{c}\text{Mass lost}\\ \text{between Time}\\ t=0 \text{ and } t=T\end{array}\right) = \left(\begin{array}{c}\text{Mass inside}\\ \text{chamber at}\\ \text{Time } t=T\end{array}\right) \tag{B.1}$$

Or alternatively, considering the key mechanisms from Figure 2.2:

$$\left(\begin{array}{c}\text{The change}\\ \text{in concentration}\\ \text{over time } t\end{array}\right) = \left(\begin{array}{c}\text{Increases from}\\ \text{pollutant}\\ \text{ingress}\end{array}\right) + \left(\begin{array}{c}\text{Increases}\\ \text{from source}\\ \text{emissions}\end{array}\right) - \left(\begin{array}{c}\text{Ventilation}\\ \text{losses}\end{array}\right) - \left(\begin{array}{c}\text{Other}\\ \text{losses}\end{array}\right) \tag{B.2}$$

Mathematically, this can be expressed as Equation B.3, which is similar to the equation used by Olson & Burke [199].

$$\frac{dC(t)}{dt} = \lambda P C_{ext} + \frac{(1-\eta)g}{V} - \lambda C(t) - kC(t) \tag{B.3}$$

[H] $C(t)$	= Pollutant concentration at time t	[M/L^3]
C_{ext}	= External concentration	[M/L^3]
P	= Penetration coefficient	[-]
V	= Volume	[L^3]
g	= Emission rate, considered constant	[M/T]
η	= Cooker hood capture efficiency	[-]
t	= Time	[T]
λ	= Decay due to ventilation and	[T^{-1}]
k	= Decay due to all other mechanisms	[T^{-1}]
	(L length, M mass, T time)	

Dimensional analysis of Equation B.3 gives:

$$\frac{\text{kg}}{\text{m}^3} \cdot \frac{1}{\text{s}} = \frac{1}{\text{s}} \cdot \frac{\text{kg}}{\text{m}^3} + \frac{\text{kg}}{\text{s}} \cdot \frac{1}{\text{m}^3} - \frac{1}{\text{s}} \cdot \frac{\text{kg}}{\text{m}^3} - \frac{1}{\text{s}} \cdot \frac{\text{kg}}{\text{m}^3}$$

Substituting $\phi = \lambda + k$ into Equation B.3 gives Equation B.4 which is then rearranged to group $C(t)$.

$$\frac{dC(t)}{dt} = \lambda P C_{ext} + \frac{(1-\eta)g}{V} - \phi C(t) \tag{B.4}$$

$$\frac{dC(t)}{dt} + \phi C(t) = \lambda P C_{ext} + \frac{(1-\eta)g}{V} \tag{B.5}$$

This is integrated using Integrating factor IF $= e^{\int \phi \, dt} = e^{\phi t}$ to give:

$$e^{\phi t} \cdot \frac{dC(t)}{dt} + \phi C(t) e^{\phi t} = \lambda P C_{ext} e^{\phi t} + \frac{(1-\eta)g}{V} e^{\phi t} \tag{B.6}$$

$$\int \left(\frac{d}{dt} C(t) e^{\phi t} + \phi C(t) e^{\phi t} \right) dt = \int \left(\lambda P C_{ext} e^{\phi t} + \frac{(1-\eta)g}{V} e^{\phi t} \right) dt \tag{B.7}$$

$$C(t) e^{\phi t} = \frac{\lambda P C_{ext}}{\phi} e^{\phi t} + \frac{(1-\eta)g}{\phi V} e^{\phi t} + a \tag{B.8}$$

$$C(t) = \frac{\lambda P C_{ext}}{\phi} + \frac{(1-\eta)g}{\phi V} + a e^{-\phi t} \tag{B.9}$$

Here a is a constant of integration, and can be determined using the initial condition that $C(0)$ occurs at time $t = 0$.

$$C(0) = \frac{\lambda P C_{ext}}{\phi} + \frac{(1-\eta)g}{\phi V} + a e^{0} \tag{B.10}$$

$$a = C(0) - \frac{\lambda P C_{ext}}{\phi} - \frac{(1-\eta)g}{\phi V} \tag{B.11}$$

Therefore a can be substituted into Equation B.9 to give Equation B.12 or Equation 2.6.

$$C(t) = \frac{\lambda P C_{ext}}{\phi} + \frac{(1-\eta)g}{\phi V} + \left(C(0) - \frac{\lambda P C_{ext}}{\phi} - \frac{(1-\eta)g}{\phi V} \right) e^{-\phi t} \tag{B.12}$$

Dimensional analysis of Equation B.12:

$$\frac{\mathrm{kg}}{\mathrm{m}^3} = \frac{1}{\mathrm{s}} \cdot \frac{\mathrm{kg}}{\mathrm{m}^3} \cdot \mathrm{s} + \frac{\mathrm{kg}}{\mathrm{s}} \cdot \frac{\mathrm{s}}{\mathrm{m}^3} + \left(\frac{\mathrm{kg}}{\mathrm{m}^3} - \frac{1}{\mathrm{s}} \cdot \frac{\mathrm{kg}}{\mathrm{m}^3} \cdot \mathrm{s} - \frac{\mathrm{kg}}{\mathrm{s}} \cdot \frac{\mathrm{s}}{\mathrm{m}^3} \right)$$

Under steady state conditions, $e^{-\phi t} \rightarrow 0$ as as $t \rightarrow \infty$, so from Equation B.12, the steady state concentration C_{ss} is given by:

$$C_{ss} = C(\infty) = \frac{\lambda P C_{ext}}{\phi} + \frac{(1-\eta)g}{\phi V} + \left(C(0) - \frac{\lambda P C_{ext}}{\phi} - \frac{(1-\eta)g}{\phi V} \right) \cdot 0 \tag{B.13}$$

$$C_{ss} = \frac{\lambda P C_{ext}}{\phi} + \frac{(1-\eta)g}{\phi V} \tag{B.14}$$

B.1 Emission Rate Calculations

In studies measuring emission rates using the *peak estimation* or *theoretical peak estimation* methods introduced in Section 2.5.4 equations resembling Equations B.3 and B.12 are used. Olson & Burke [199] assume no capture efficiency ($\eta = 0$) and background internal concentration C_b which is a function of the external concentration and equals the initial concentration such that:

$$C_b = \frac{\lambda P C_{ext}}{\phi} \tag{B.15}$$

$$C_b = C(0) \tag{B.16}$$

Substituting $C_b = C(0)$ into Equation B.12:

$$C(t) = C_b + \frac{g}{\phi V} + \left(C_b - C_b - \frac{g}{\phi V} \right) e^{-\phi t} \tag{B.17}$$

$$C(t) = C_b + \frac{g}{\phi V} + \left(-\frac{g}{\phi V} \right) e^{-\phi t} \tag{B.18}$$

$$C(t) = C_b + \frac{g}{\phi V} \left(1 - e^{-\phi t} \right) \tag{B.19}$$

Appendix C

Kitchen Intervention Study

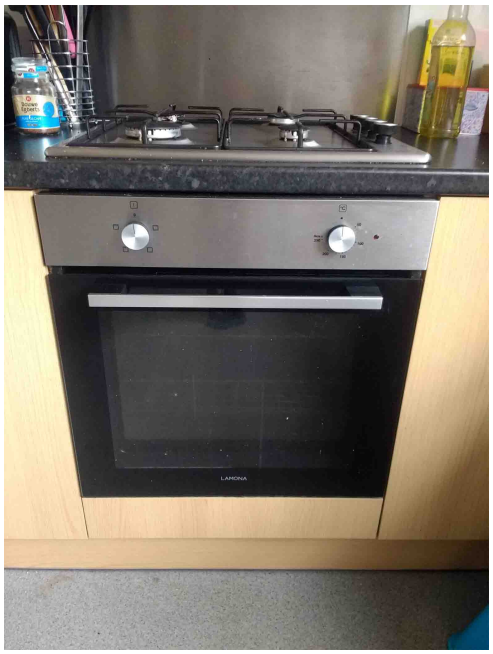
This appendix contains further figures and tables relevant to Chapter 3.

C.1 Example Appliances

Included in this section are example photos of appliances found in the kitchens included in the intervention study in Chapter 3. Mostly these examples are from House E, however also included are photos of the cooker hood in House A. The coffee machine present in Figure C.1a was not present during the intervention study. Figure C.2 shows further photos of the cooker hood installed in House E to illustrate the investigations undertaken in all houses, and Figure C.3 shows the damaged ductwork found in House A.



(A) Stove, oven and cooker hood



(B) Electric oven



(C) Microwave



(D) Kettle (left) and toaster (right)

FIGURE C.1: Example appliances from House E



(A) Cooker hood



(B) Speed controls



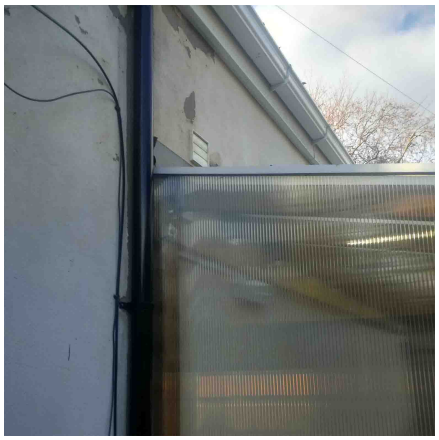
(C) Hood with grease screens removed



(D) Fan



(E) Duct work



(F) External vent location

FIGURE C.2: House E cooker hood



(A) Hood location



(B) Damaged duct work

FIGURE C.3: House A cooker hood
©Stewart Somerville

C.2 Blank Cooking Logs

Included here are blank copies of the cooking log issued to the lead occupants for the household to record cooking events in Week 1 (Fig C.4) and also ventilation use in Week 2 (Fig C.5).

COOKING LOG

Date	Time (hh:mm)		Food cooked, methods used, notes.
	Start	Finish	

FIGURE C.4: Blank *Household Cooking Log* Week 1

COOKING LOG

Date	Time (hh:mm)		Food cooked, methods used, notes.	Win-dows	Door	Fan
	Start	Finish				

FIGURE C.5: Blank *Household Cooking Log* Week 2

C.3 Lead Occupant Instructions

Figure C.6 and Figure C.7 show email text and a Week 1 concentration plot. Both were shared with the lead occupant to aid them in briefing the other occupants before commencing Week 2. The plot is produced directly with the TrakPro™software provided by TSI to download the SidePak data. It reports concentrations in mg/m^3 rather than $\mu g/m^3$.

Key points to make are:

- You've been measuring $PM_{2.5}$ (fine particulate matter) a type of air pollution
- Long term and short term exposure to raised concentrations have been linked to increased risk of various health effects
- The World Health Organisation (WHO) set daily and annual guidelines for maximum average concentrations, these are 25µg/m³ and 10µg/m³ respectively
- Show the plot
- These are the concentrations measured in your kitchen
- Explain how they compare to the WHO thresholds
- Peaks coincide with cooking activity
- Suggest ways to reduce concentrations in this coming week such as
  - Open the window
  - Open the external door
  - Use the extractor fan
- Ask them to make a note of if they use ventilation during cooking

FIGURE C.6: Week 2 instructions for lead occupant

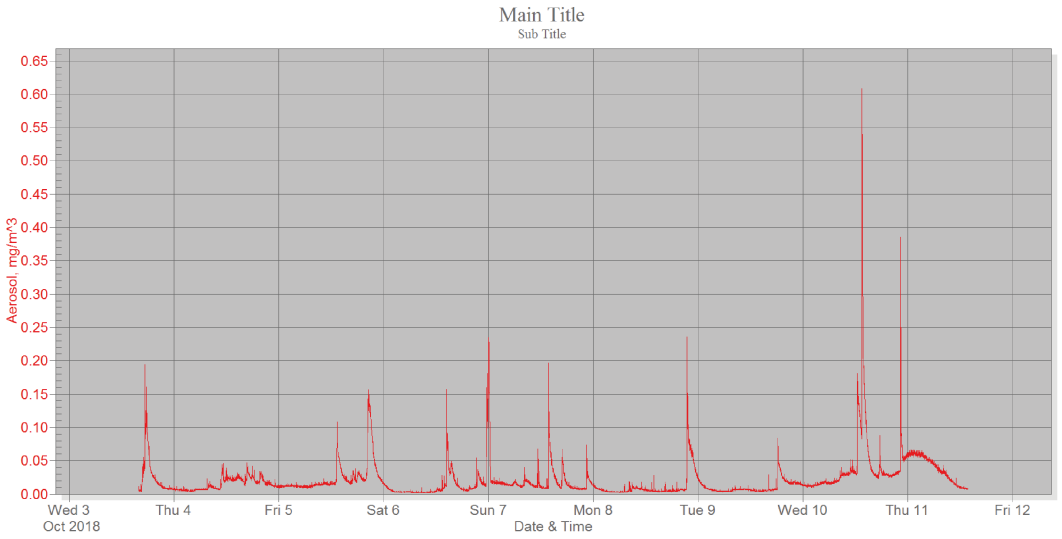
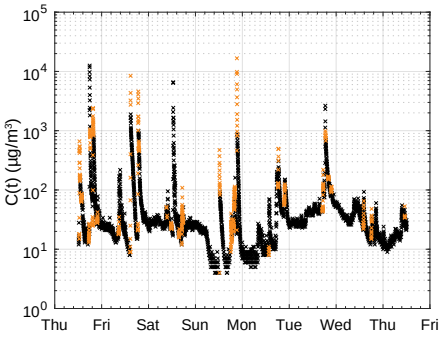


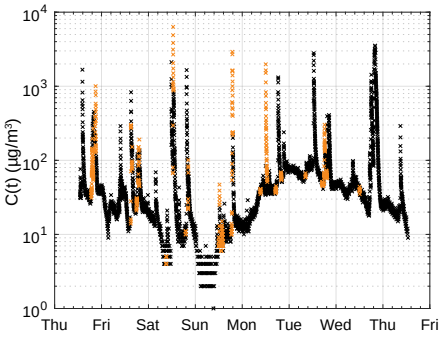
FIGURE C.7: Example Week 1 plot

C.4 Additional Plots

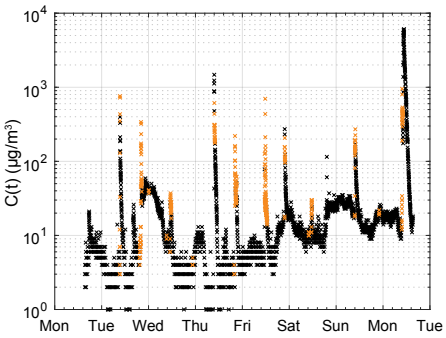
Figure C.8 shows the $PM_{2.5}$ concentrations over time for Week 1 and Week 2 in all five houses. The points highlighted in **orange** indicate the concentrations measured during occupant recorded cooking events. Figure C.9 shows the corresponding Q-Q plots comparing the natural log of the concentrations to a normal distribution.



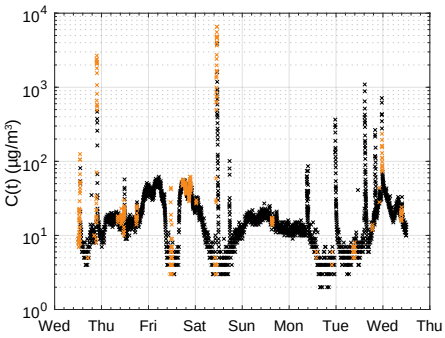
(A) House A Week 1



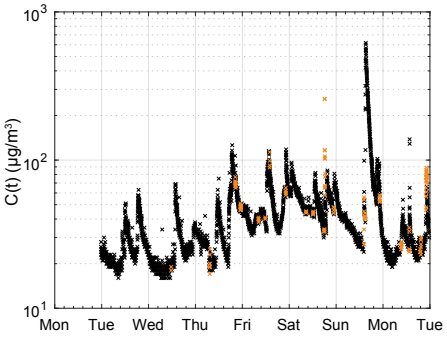
(B) House A Week 2



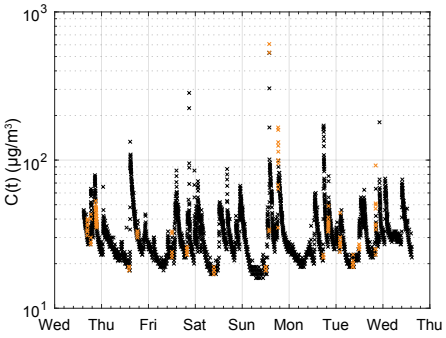
(C) House B Week 1



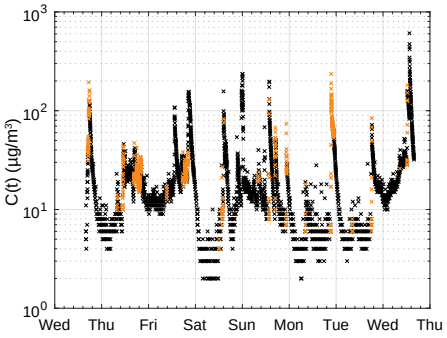
(D) House B Week 2



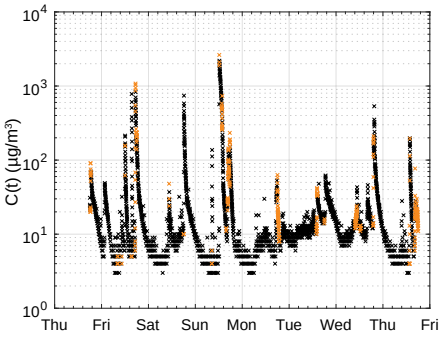
(E) House C Week 1



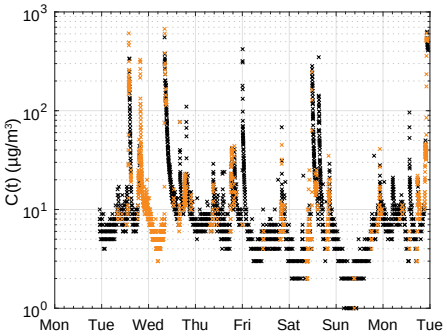
(F) House C Week 2



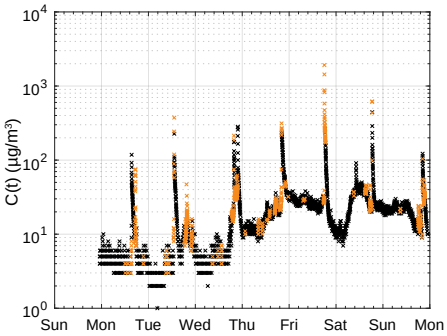
(G) House D Week 1



(H) House D Week 2

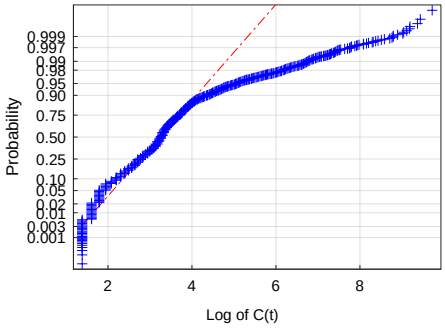


(I) House E Week 1

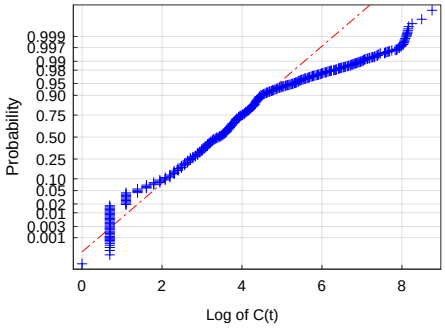


(J) House E Week 2

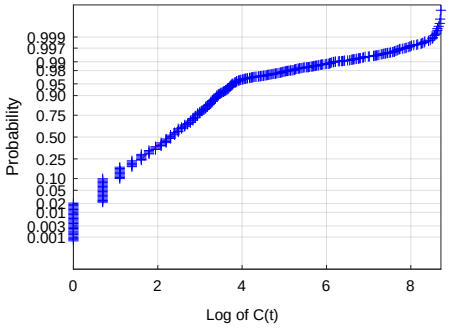
FIGURE C.8: All temporal concentrations
orange indicates concentrations during recorded cooking events



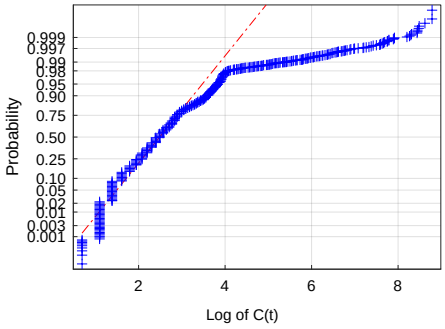
(A) House A Week 1



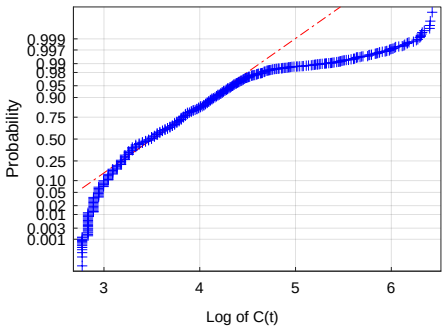
(B) House A Week 2



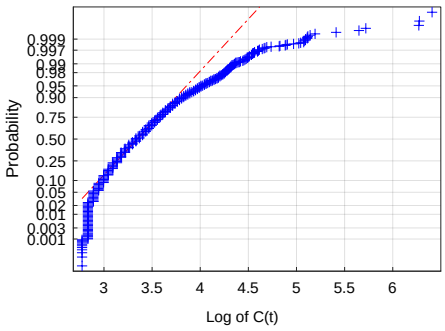
(C) House B Week 1



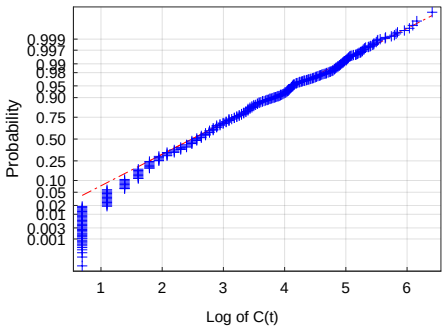
(D) House B Week 2



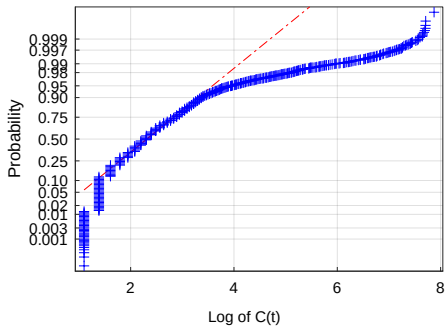
(E) House C Week 1



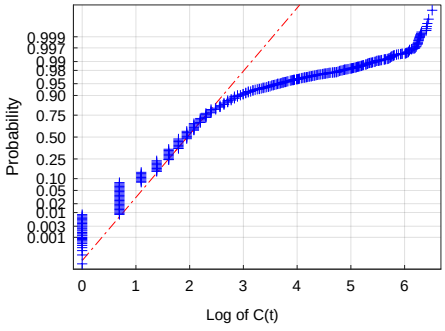
(F) House C Week 2



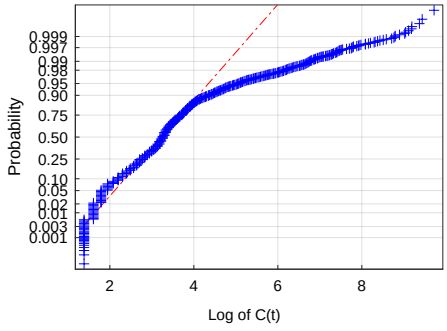
(G) House D Week 1



(H) House D Week 2

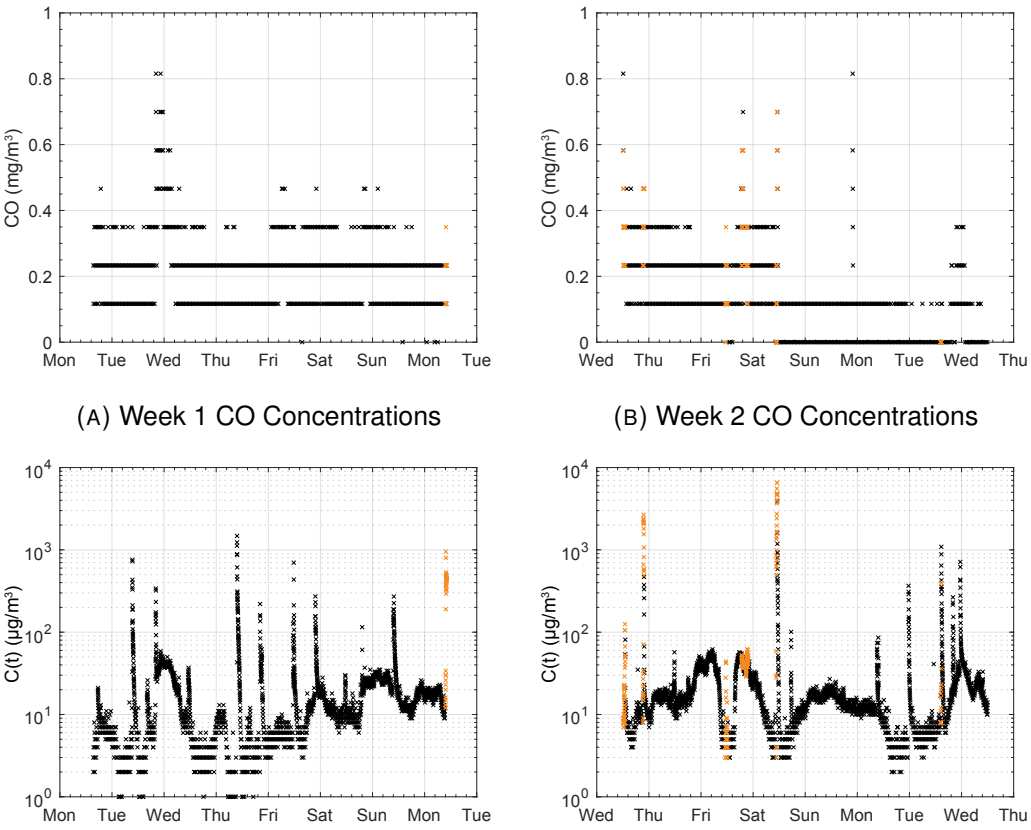


(I) House E Week 1



(J) House E Week 2

FIGURE C.9: All Q-Q Plots



(A) Week 1 CO Concentrations

(B) Week 2 CO Concentrations

(C) Week 1 $PM_{2.5}$ Concentrations

(D) Week 2 $PM_{2.5}$ Concentrations

FIGURE C.10: Temporal CO and $PM_{2.5}$ concentrations measured in House B
orange indicates concentrations during recorded cooking events using the oven

Appendix D

Toast

This appendix contains additional supporting figures and information for Chapter 4.

D.1 Phase 1



FIGURE D.1: The toaster

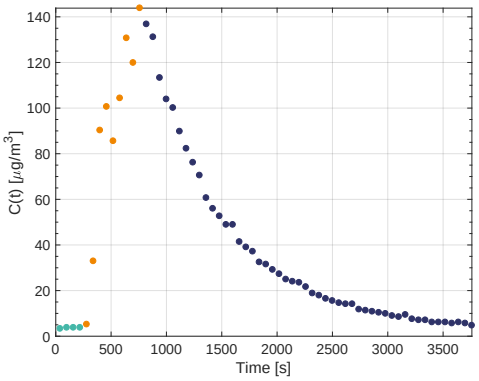


FIGURE D.2: The set up during a plume test

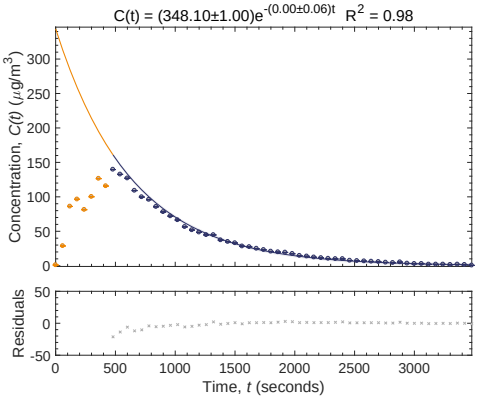
D.1.1 All Decay Plots

This section contains plots for all the tests in Phase 1. All concentrations here have been adjusted with CF = 0.47. Throughout this section **teal** = α–period; **orange** =

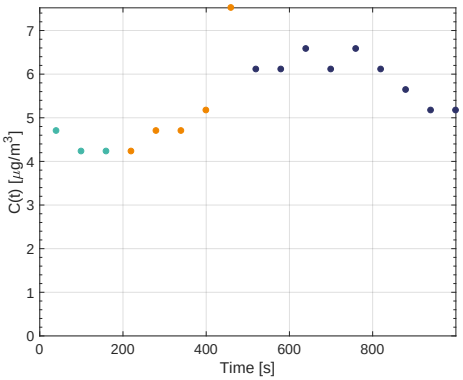
β–period; **blue** $= \gamma$–period. tests which were eliminated from the final data set are also indicated.



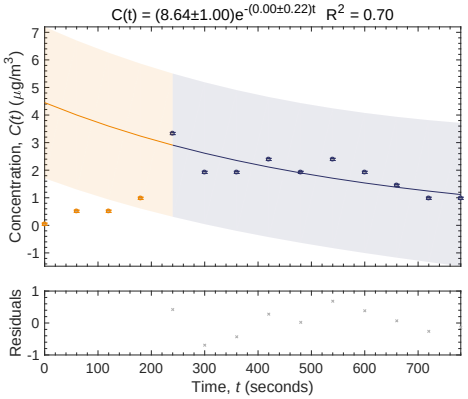
(A) All - test 1



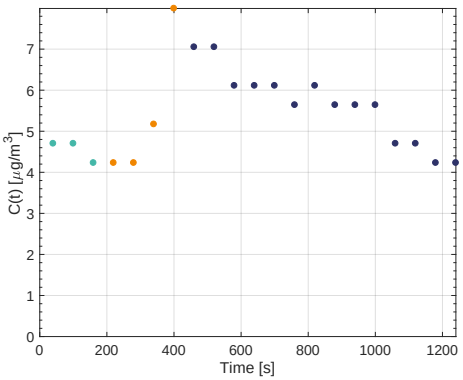
(B) Decay - test 1



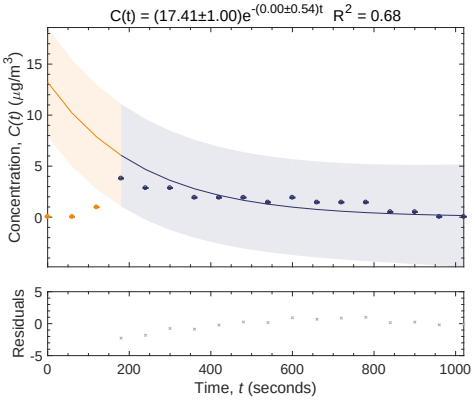
(C) All - test 2



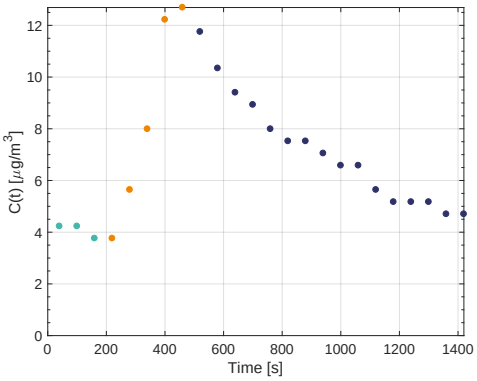
(D) Decay - test 2
removed as $R^2 < 0.7$



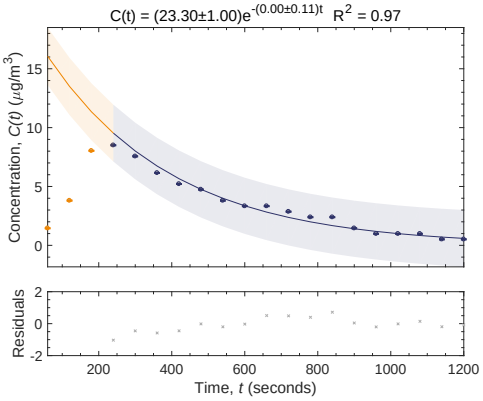
(E) All - test 3



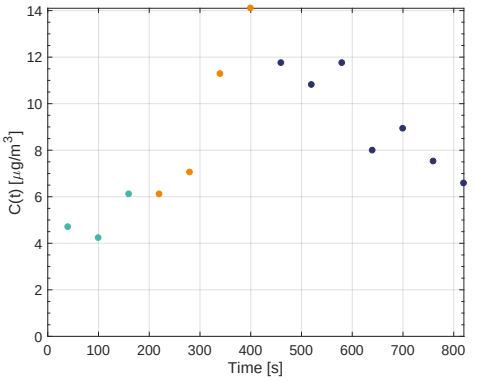
(F) Decay - test 3
removed as $R^2 < 0.7$



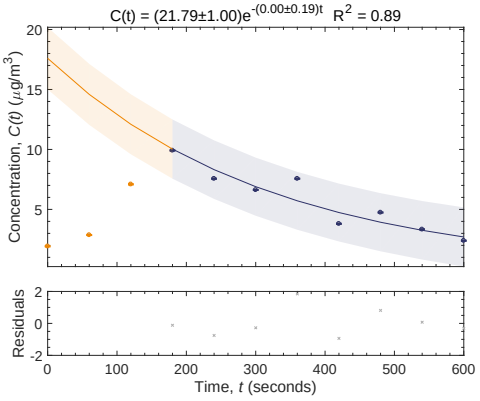
(G) All - test 4



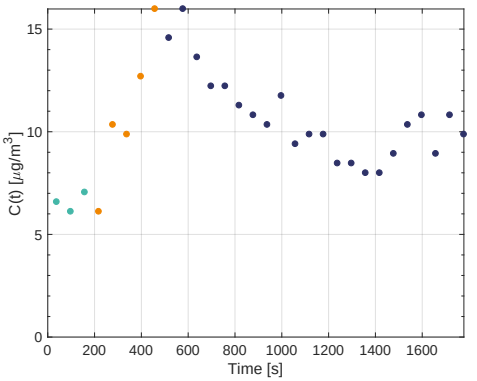
(H) Decay - test 4



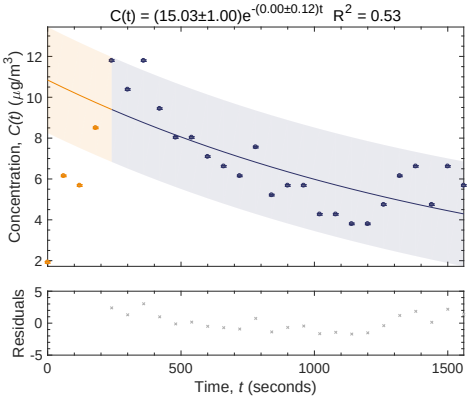
(I) All - test 5



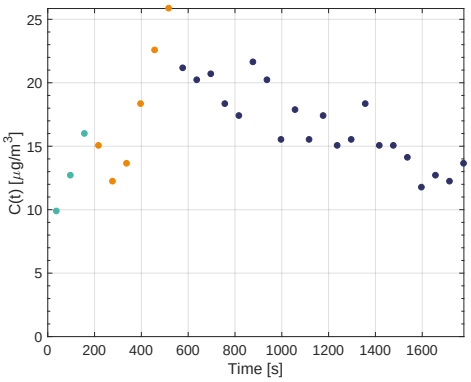
(J) Decay - test 5



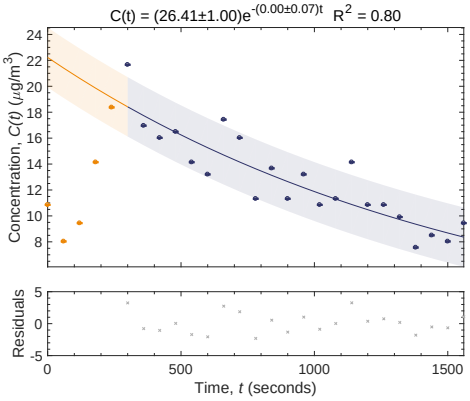
(K) All - test 6



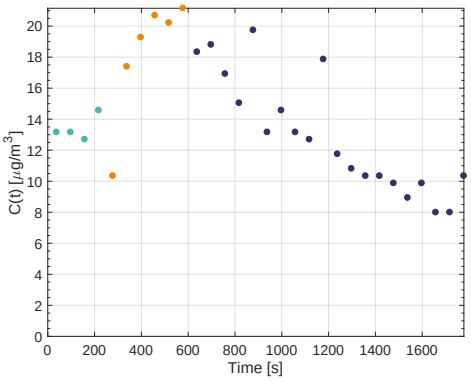
(L) Decay - test 6
removed as $R^2 < 0.7$



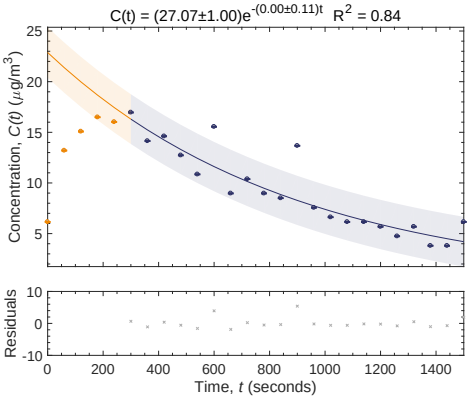
(M) All - test 7



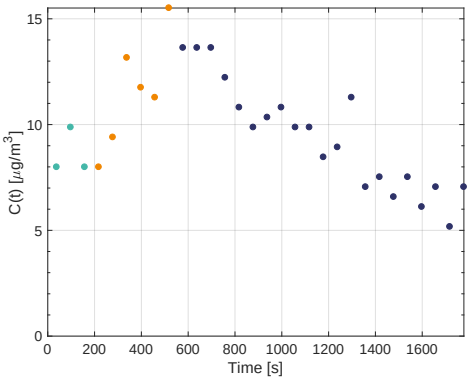
(N) Decay - test 7



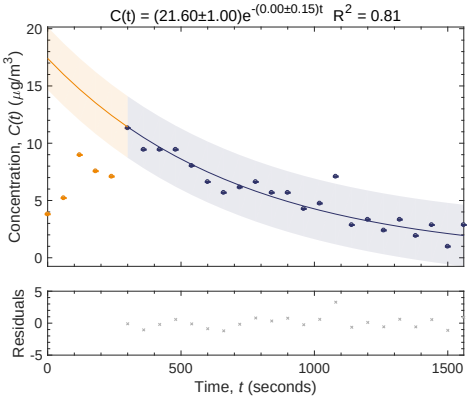
(O) All - test 8



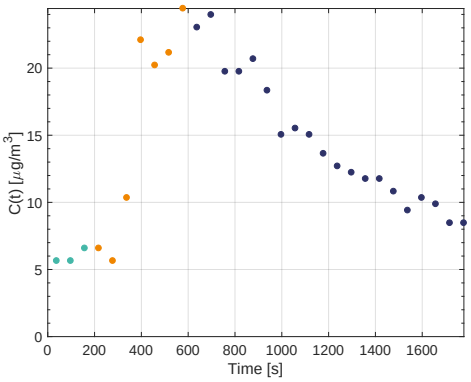
(P) Decay - test 8



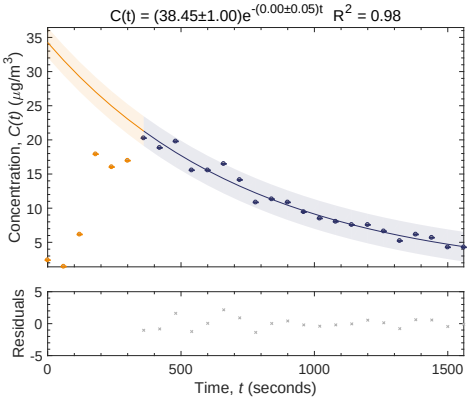
(Q) All - test 9



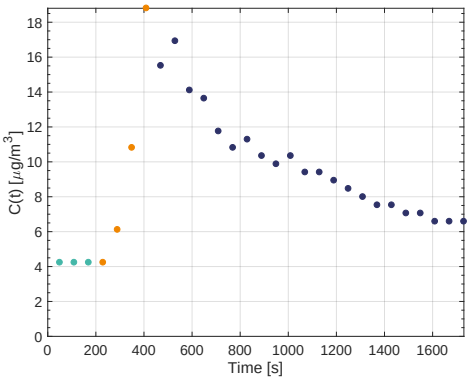
(R) Decay - test 9



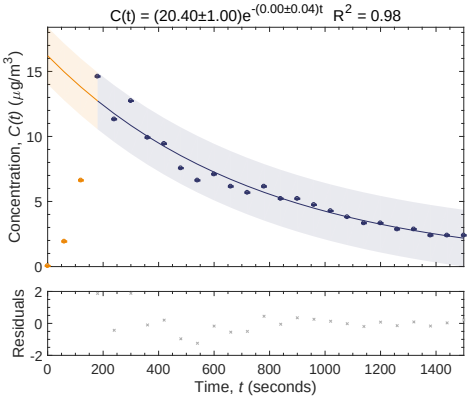
(S) All - test 10



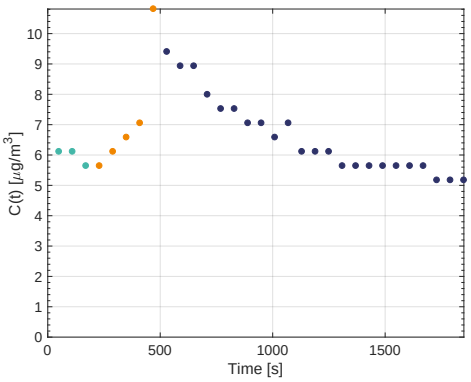
(T) Decay - test 10



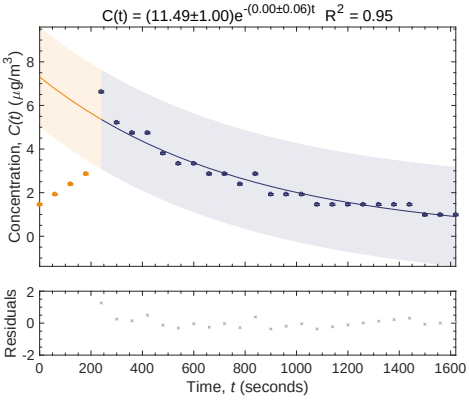
(U) All - test 11



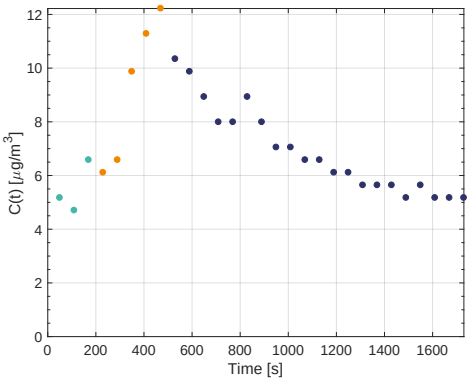
(V) Decay - test 11



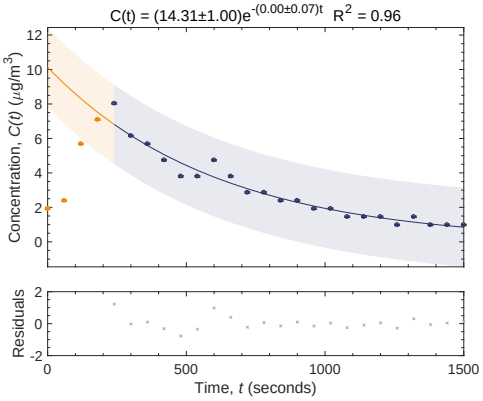
(W) All - test 12



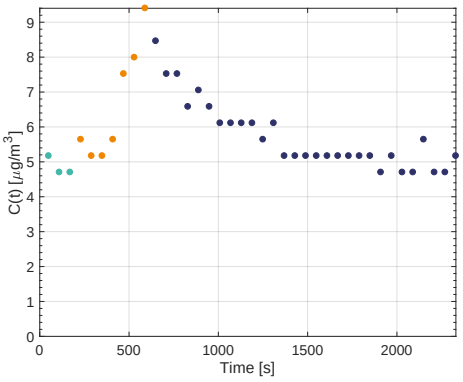
(X) Decay - test 12



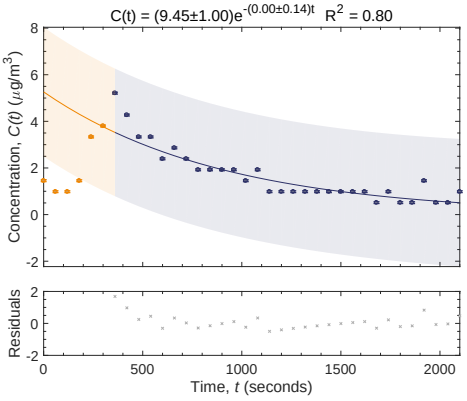
(Y) All - test 13



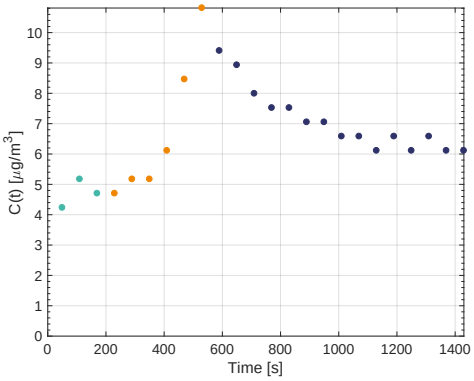
(Z) Decay - test 13



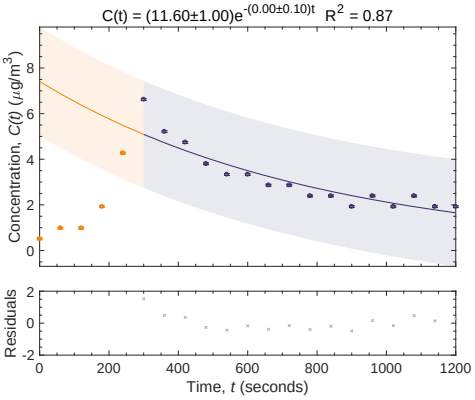
(AA) All - test 14



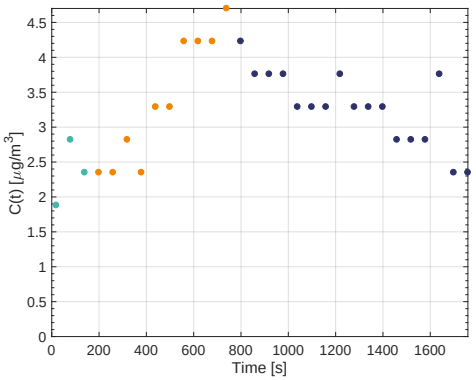
(AB) Decay - test 14



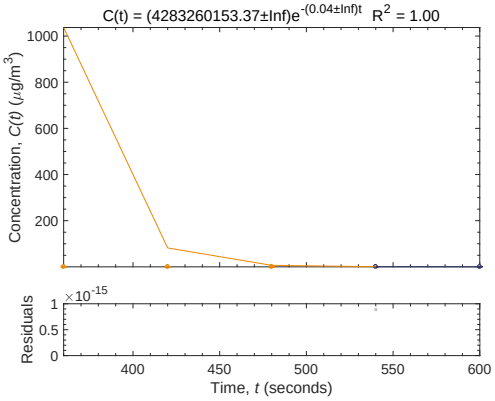
(AC) All - test 15



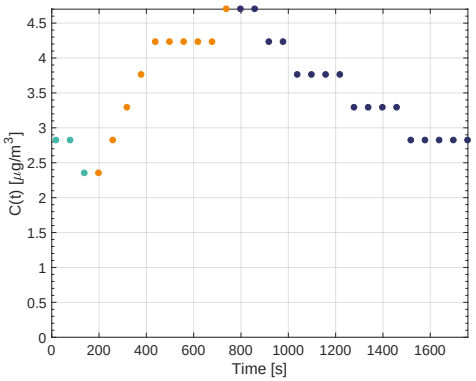
(AD) Decay - test 15



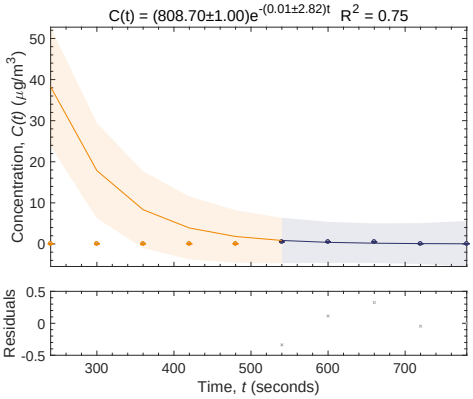
(AE) All - test 16



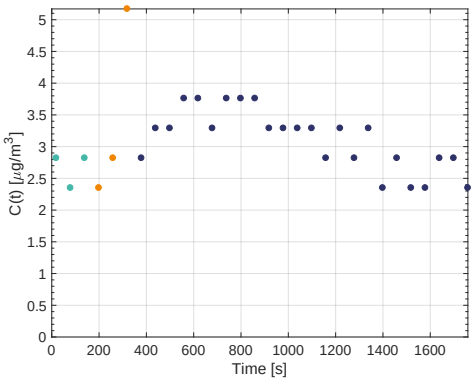
(AF) Decay - test 16 removed as an outlier



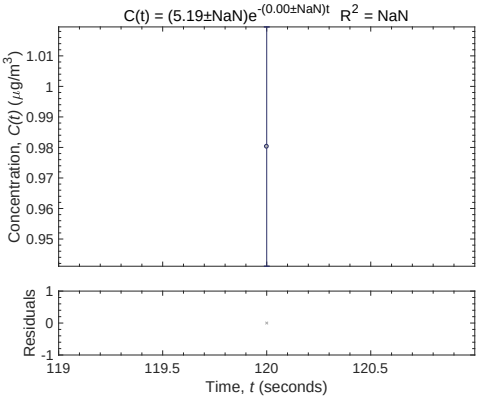
(AG) All - test 17



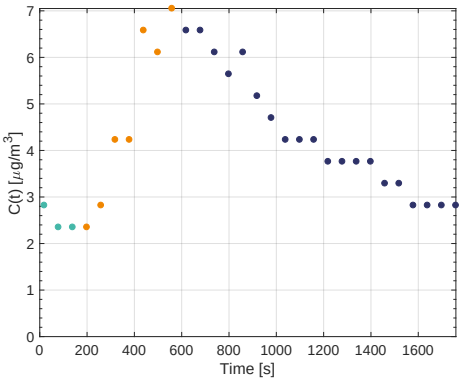
(AH) Decay - test 17



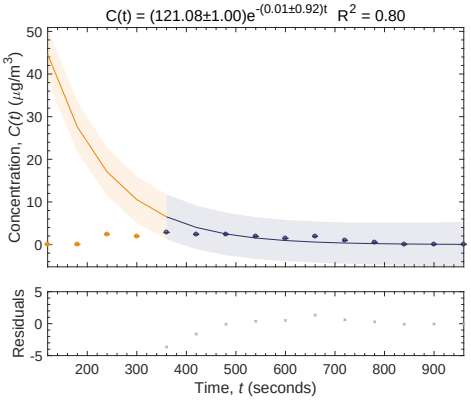
(AI) All - test 18



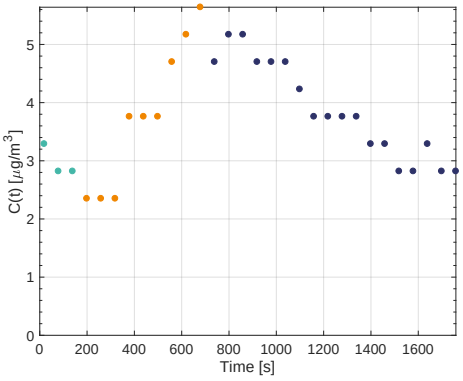
(AJ) Decay - test 18



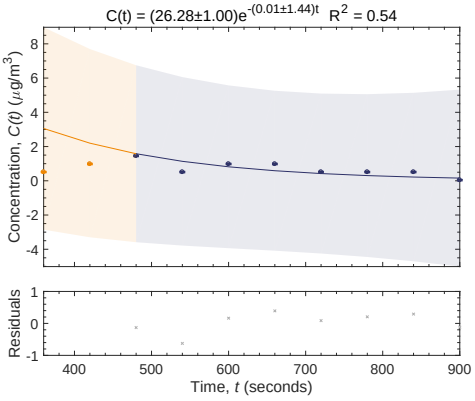
(AK) All - test 19



(AL) Decay - test 19

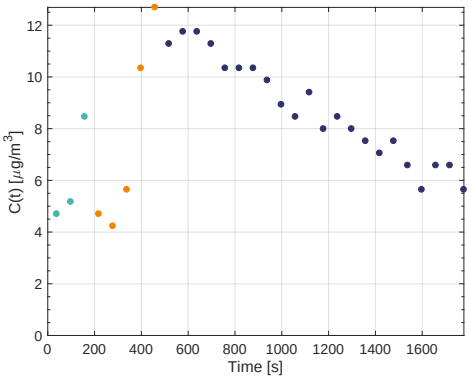


(AM) All - test 20

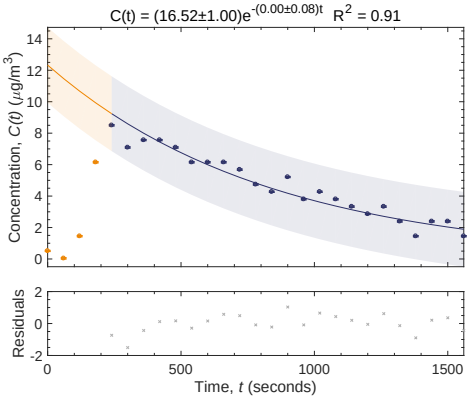


(AN) Decay - test 20 removed as $R^2 < 0.7$

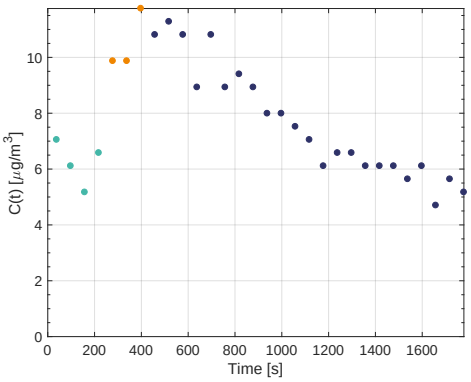
FIGURE D.3: Wholemeal bread tests, concentrations and decay



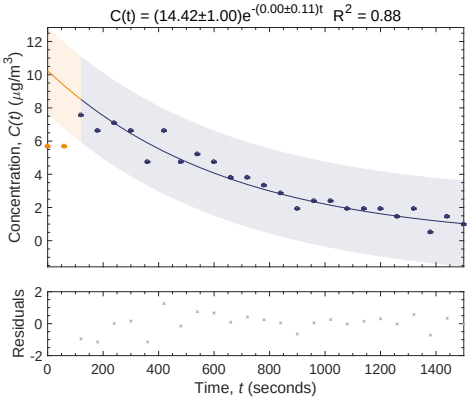
(A) All Data - test 1



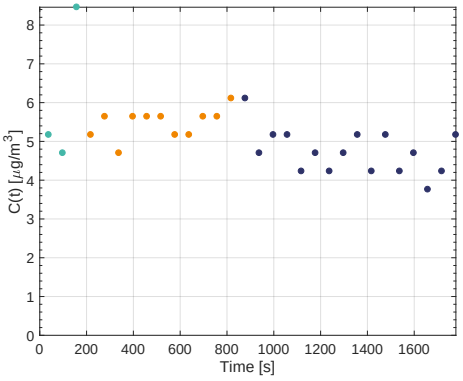
(B) Decay - test 1



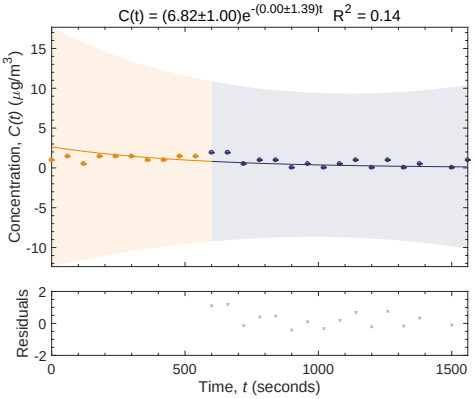
(C) All - test 2



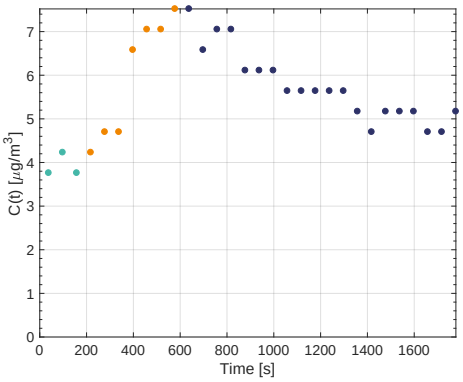
(D) Decay - test 2



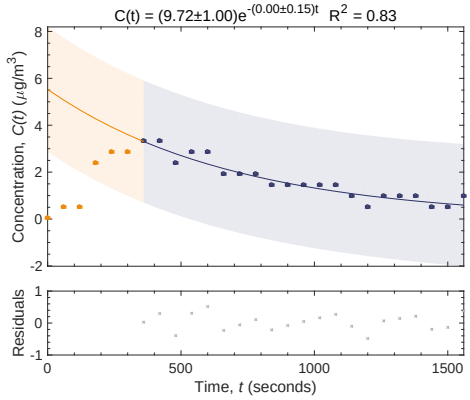
(E) All - test 3



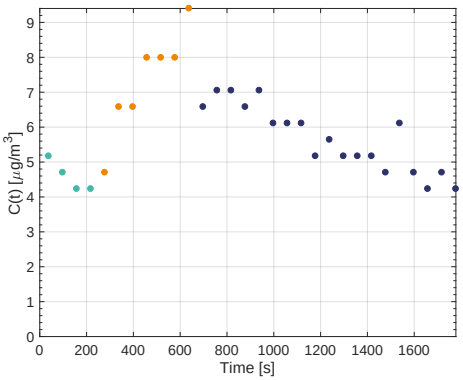
(F) Decay - test 3
removed as $R^2 < 0.7$



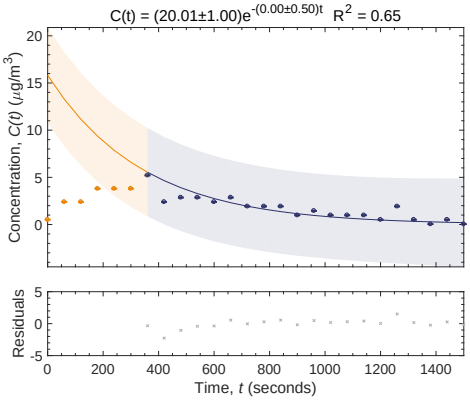
(G) All - test 4



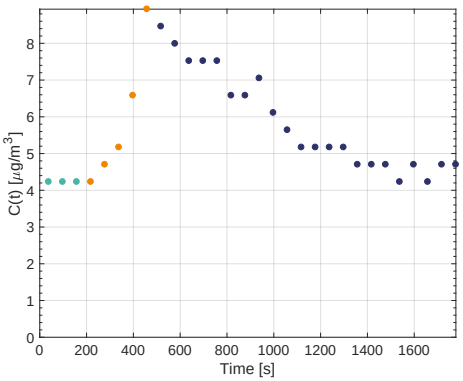
(H) Decay - test 4



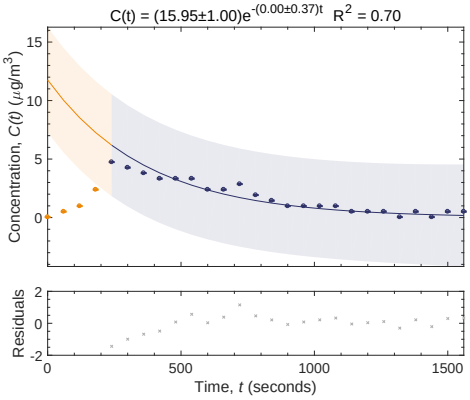
(I) All - test 5



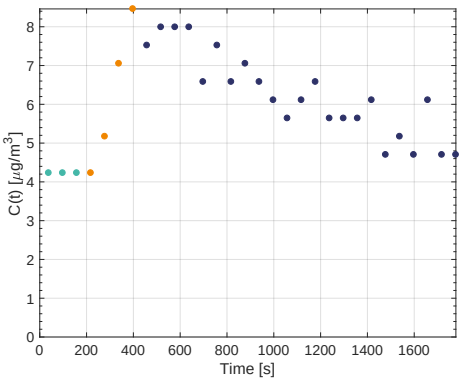
(J) Decay - test 5
removed as $R^2 < 0.7$



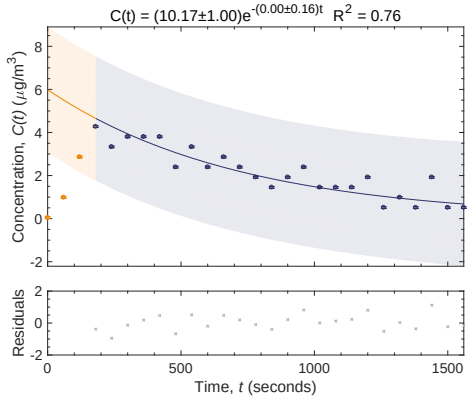
(K) All - test 6



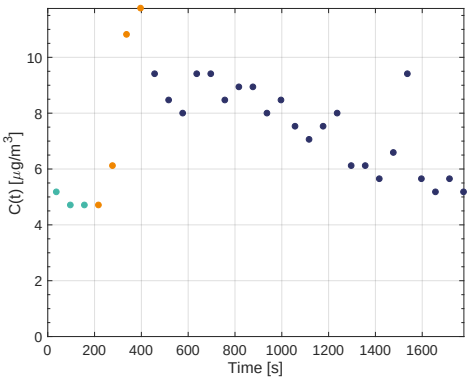
(L) Decay - test 6



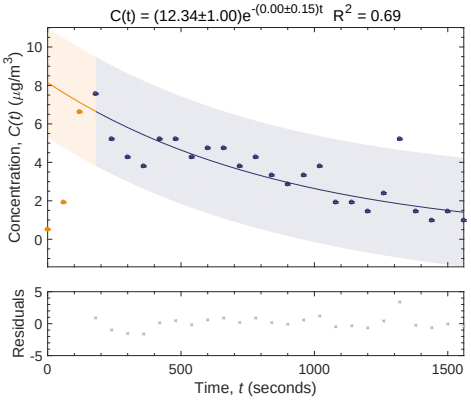
(M) All - test 7



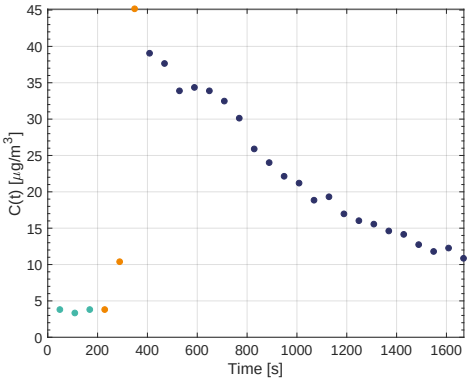
(N) Decay - test 7



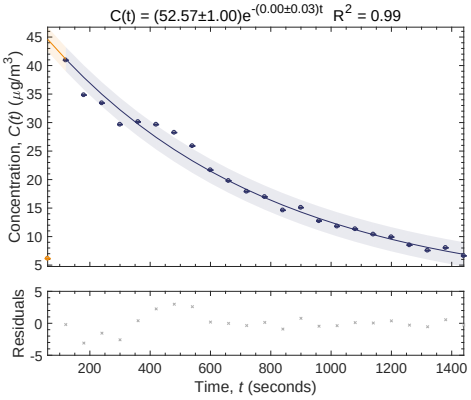
(O) All - test 8



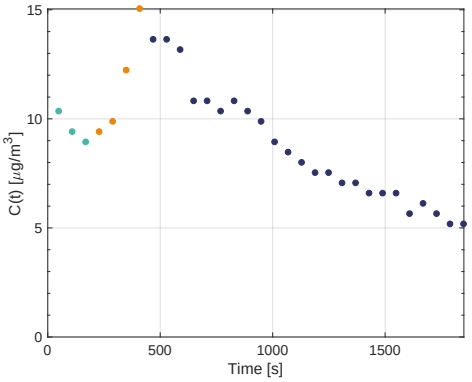
(P) Decay - test 8
removed as $R^2 < 0.7$



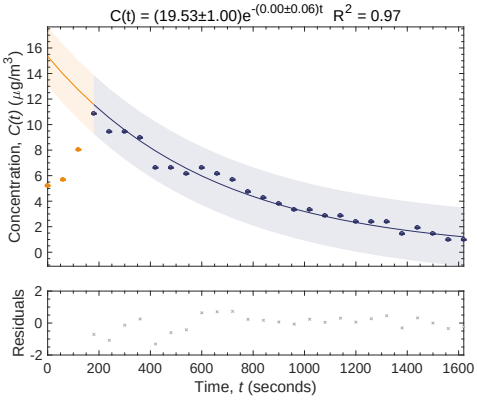
(Q) All - test 9



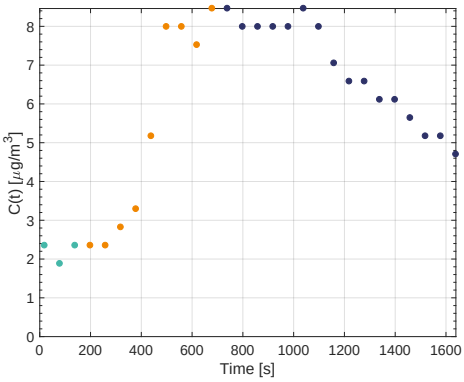
(R) Decay - test 9



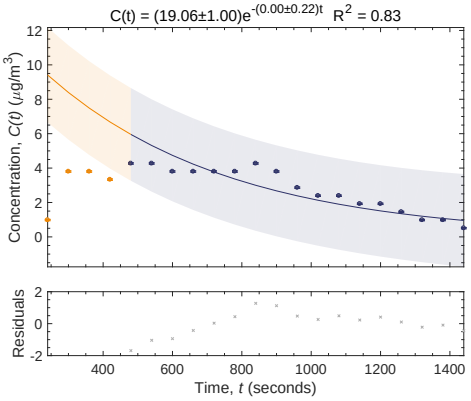
(S) All - test 10



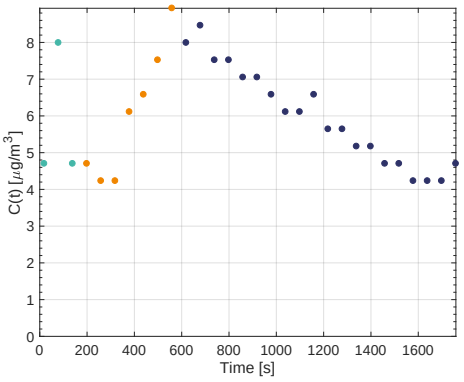
(T) Decay - test 10



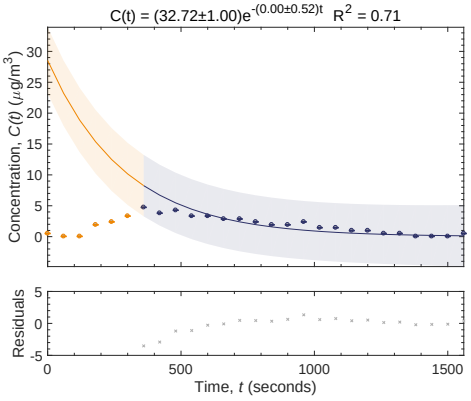
(U) All - test 11



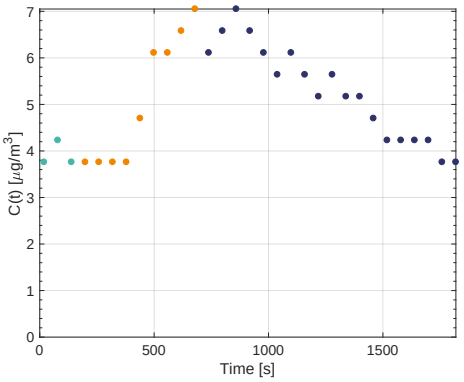
(V) Decay - test 11



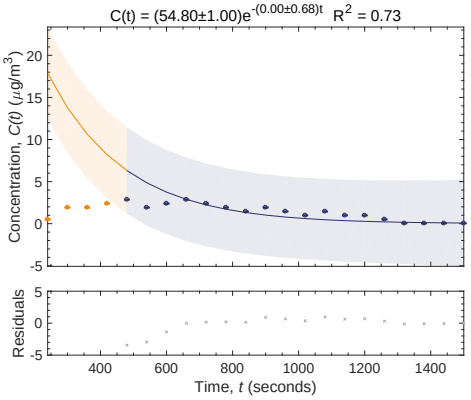
(W) All - test 12



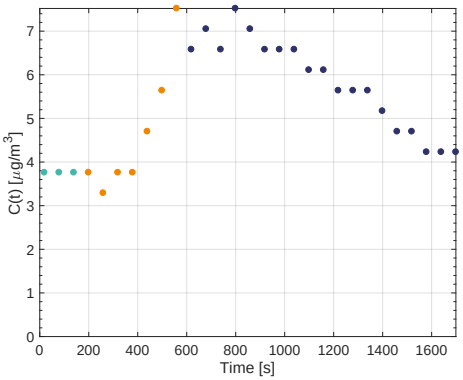
(X) Decay - test 12



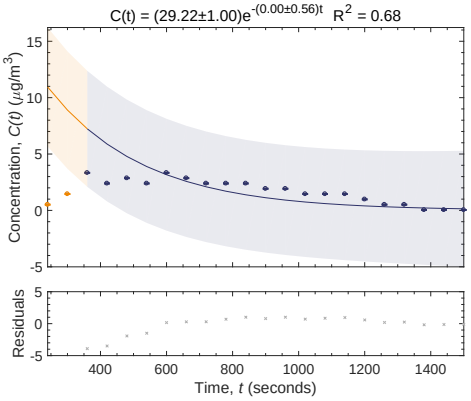
(Y) All - test 13



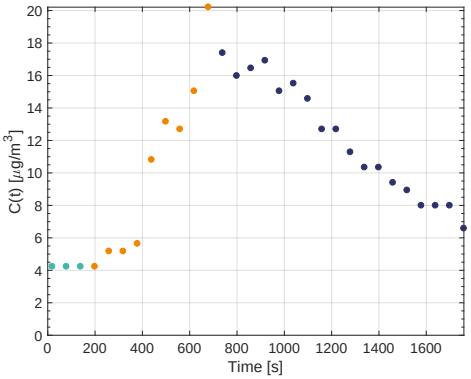
(Z) Decay - test 13



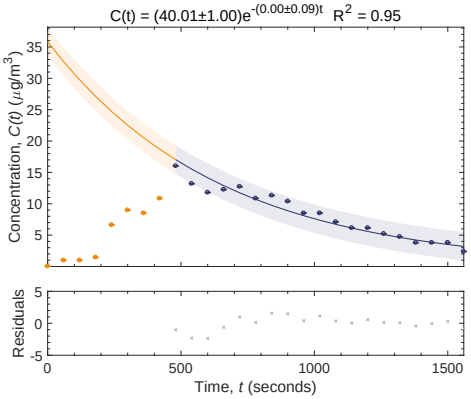
(AA) All - test 14



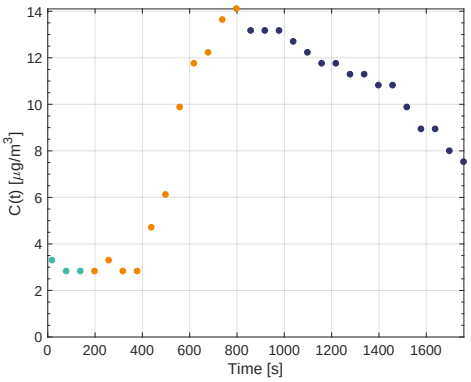
(AB) Decay - test 14
removed as $R^2 < 0.7$



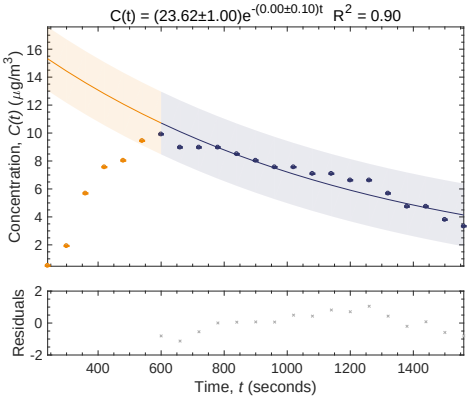
(AC) All - test 15



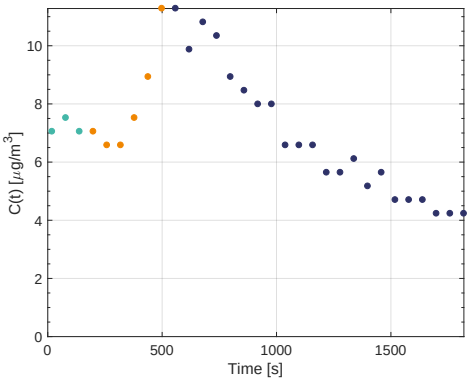
(AD) Decay - test 15



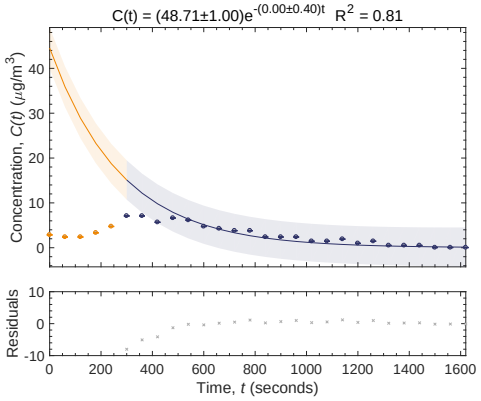
(AE) All - test 16



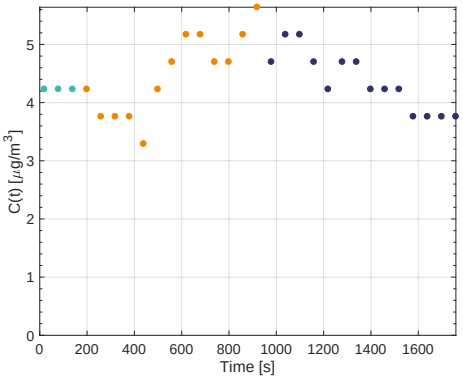
(AF) Decay - test 16



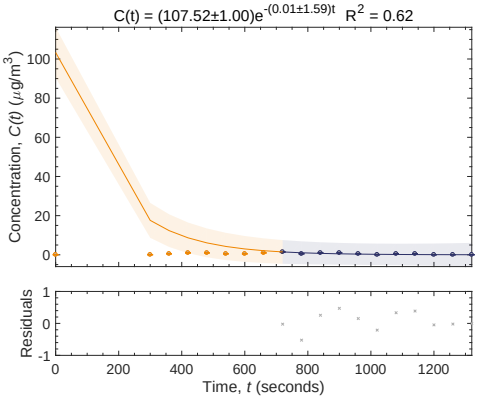
(AG) All - test 17



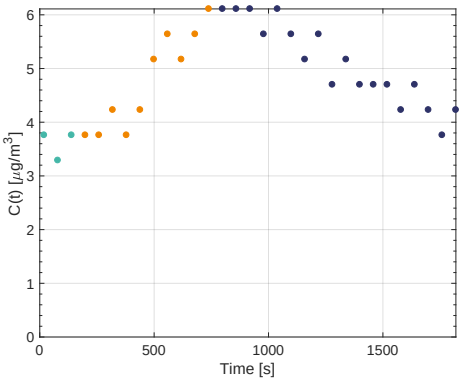
(AH) Decay - test 17



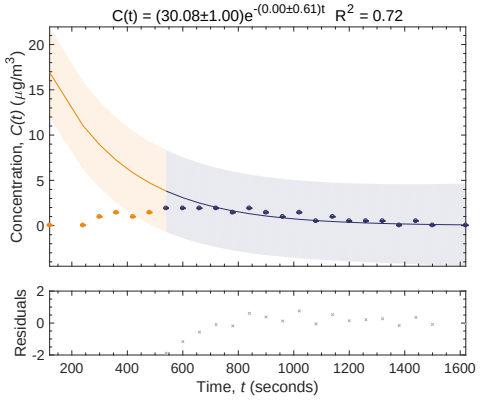
(AI) All - test 18



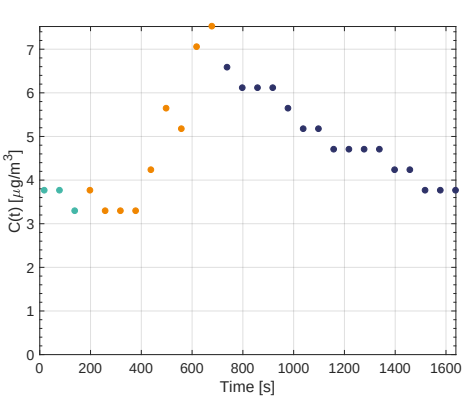
(AJ) Decay - test 18
removed as $R^2 < 0.7$



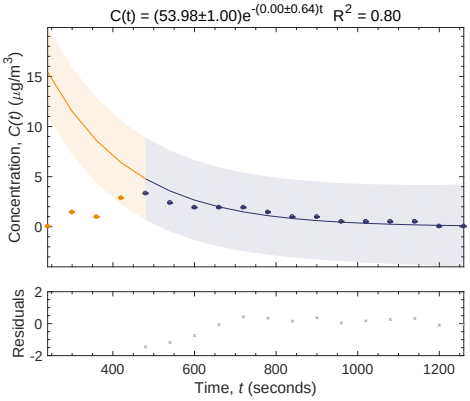
(AK) All - test 19



(AL) Decay - test 19



(AM) All - test 20



(AN) Decay - test 20

FIGURE D.4: White bread tests, concentrations and decay

D.1.2 Additional Results Plots

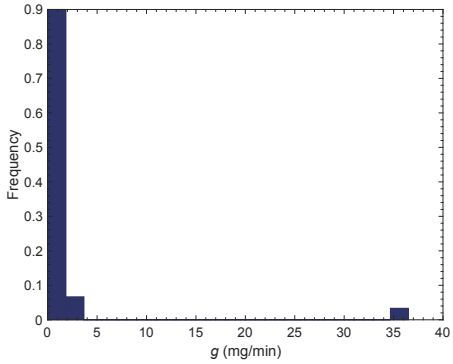


FIGURE D.5: Phase 1 emission rates histogram CF $= 0.79$

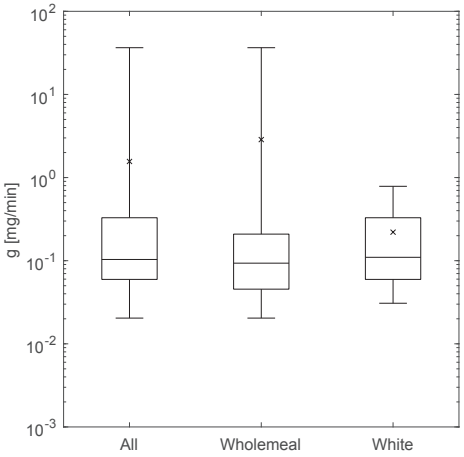


FIGURE D.6: Phase 1 emission rates box plots comparison CF $= 0.79$

D.2 Phase 2

D.2.1 Chamber Set Up



(A) Doorway and installed board



(B) Window and extract fan



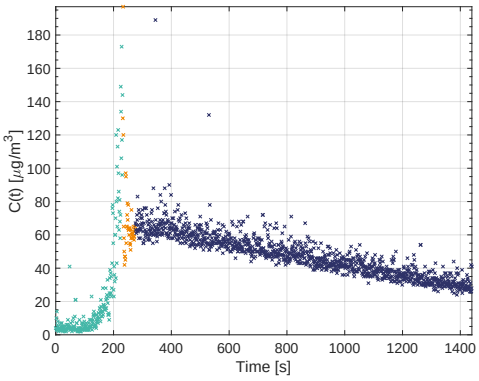
(C) Mixing fan

FIGURE D.7: Phase 2 test chamber ventilation set up

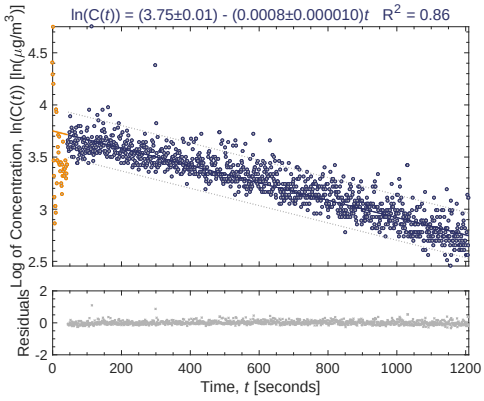
D.2.2 All Decay Plots

The concentrations over time and decay plots with fitted regression are included here. All concentrations have had the custom CF = 0.63 applied. It is indicated in the caption if a test was removed during post processing, and the reason for this

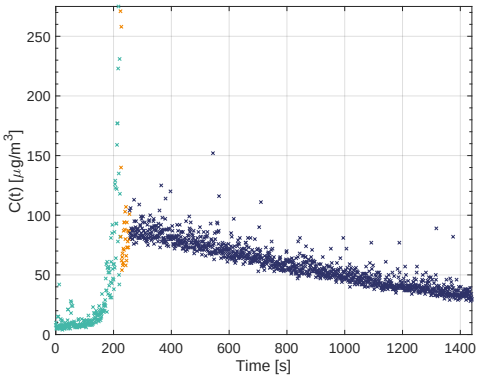
removal is also given. Throughout this section **teal** $= \alpha$–period; **orange** $= \beta$–period; **blue** $= \gamma$–period.



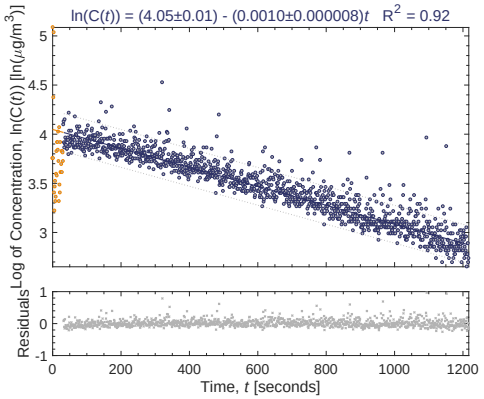
(A) All - test 1



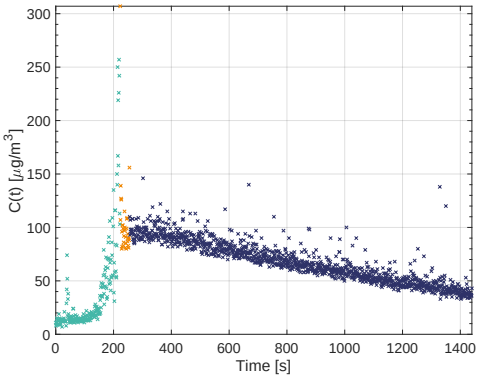
(B) Decay - test 1



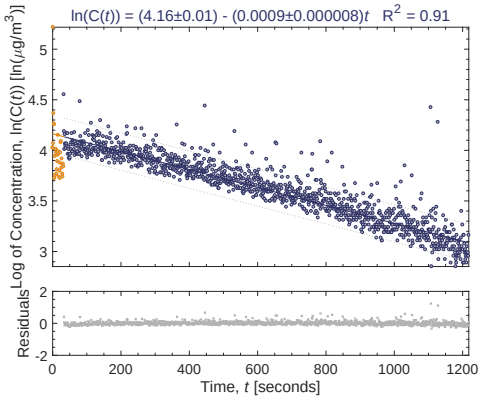
(C) All - test 2



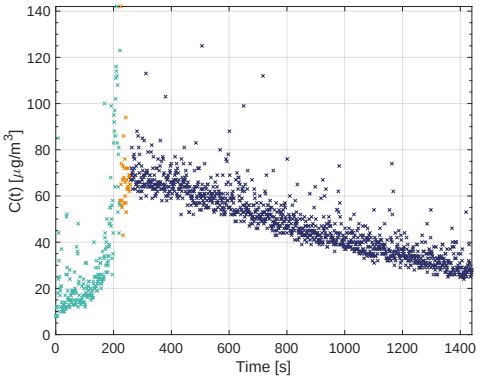
(D) Decay - test 2



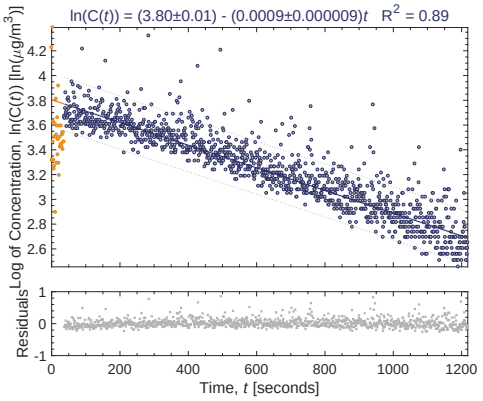
(E) All - test 3



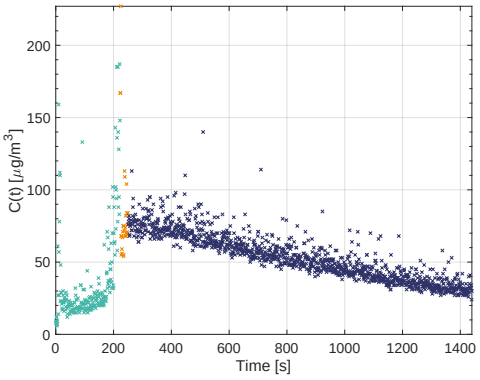
(F) Decay - test 3



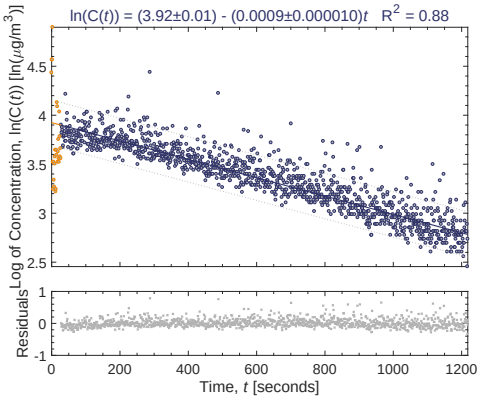
(G) All - test 4



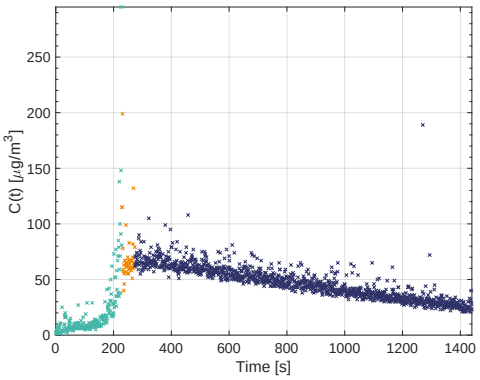
(H) Decay - test 4



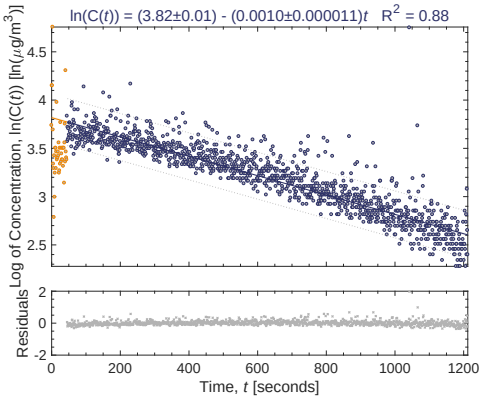
(I) All - test 5



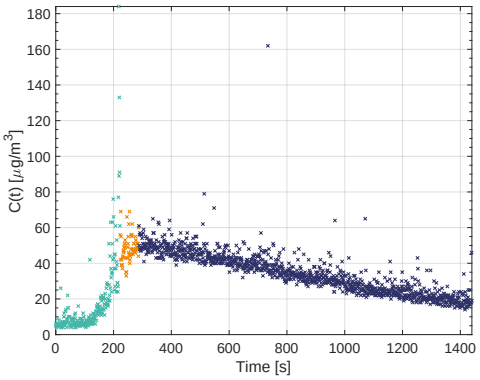
(J) Decay - test 5



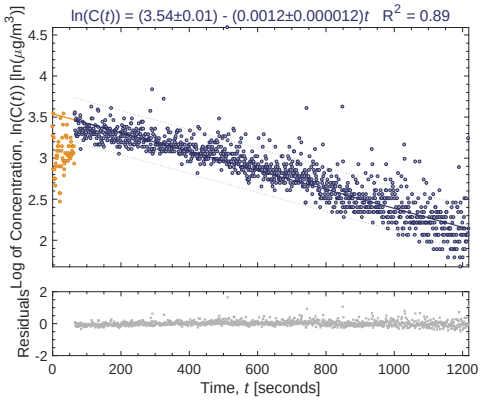
(K) All - test 6



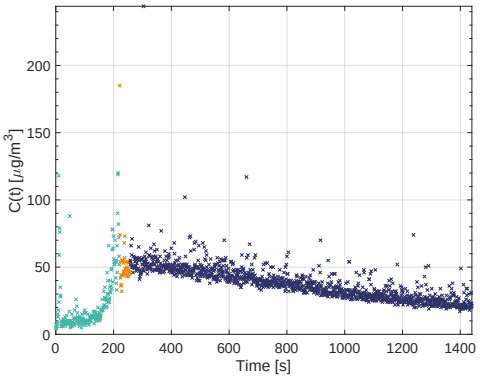
(L) Decay - test 6



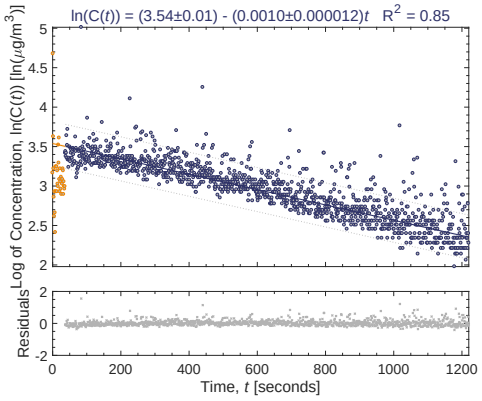
(M) All - test 7



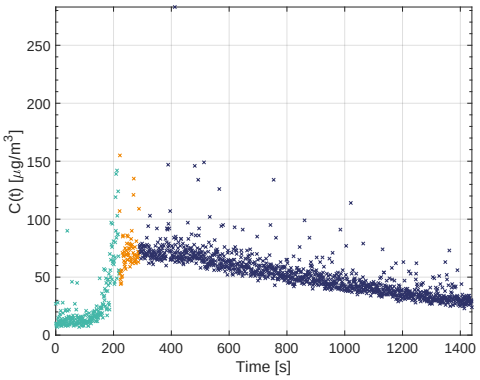
(N) Decay - test 7



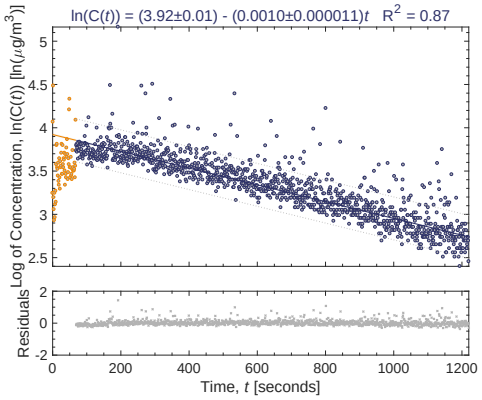
(O) All - test 8



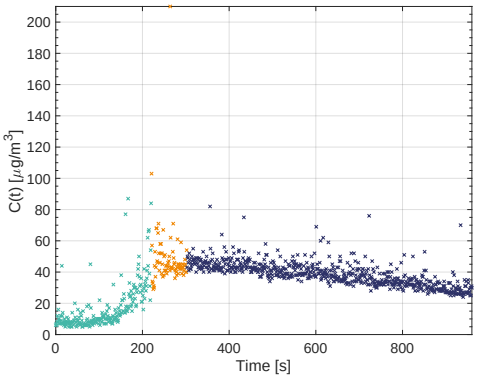
(P) Decay - test 8



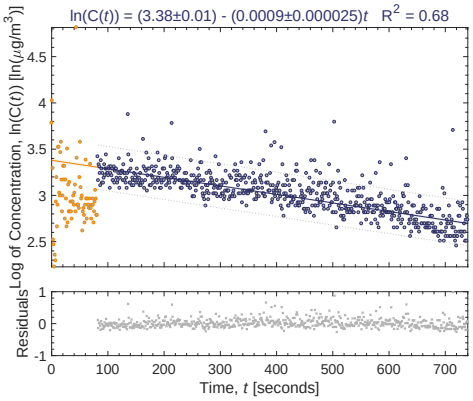
(Q) All - test 9



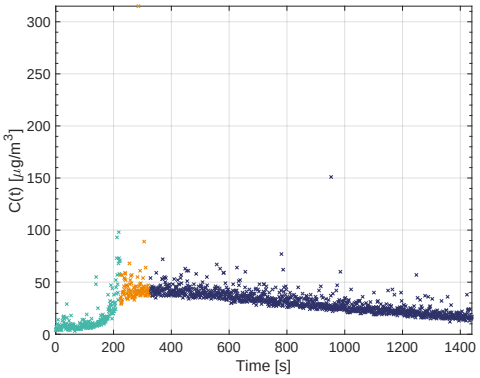
(R) Decay - test 9



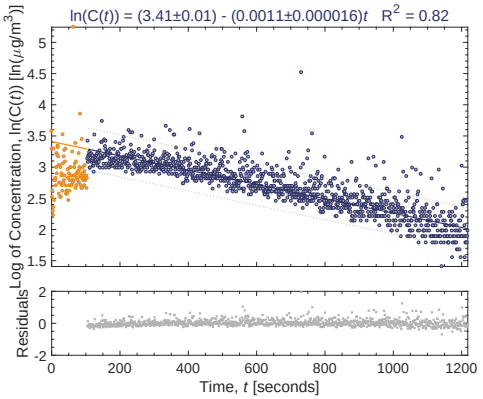
(S) All - test 10



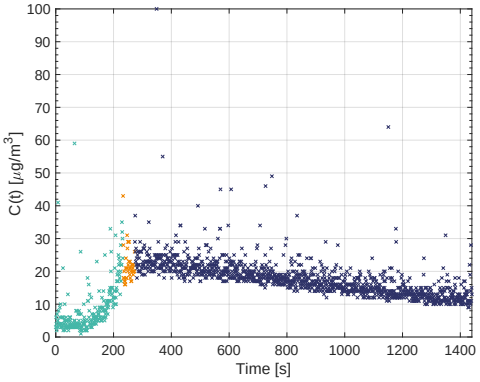
(T) Decay - test 10
removed as $R^2 < 0.7$



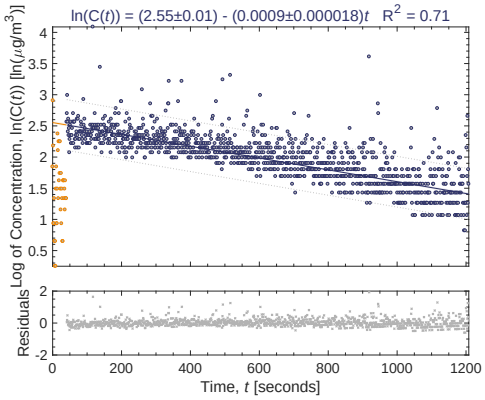
(U) All - test 11



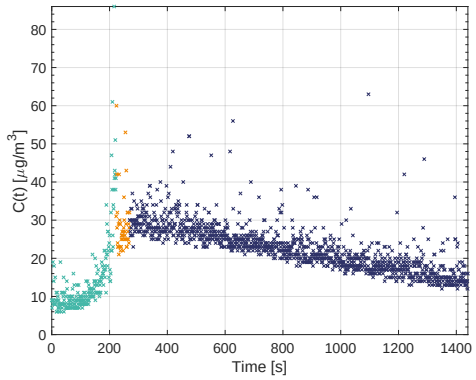
(V) Decay - test 11



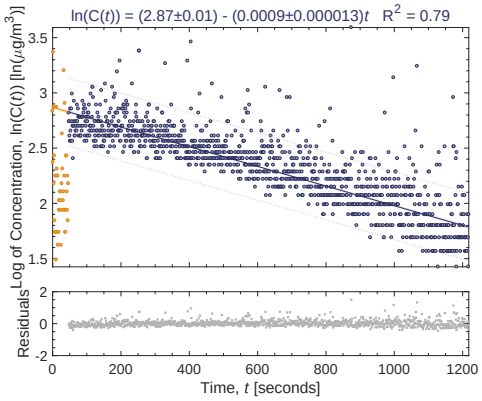
(W) All - test 12



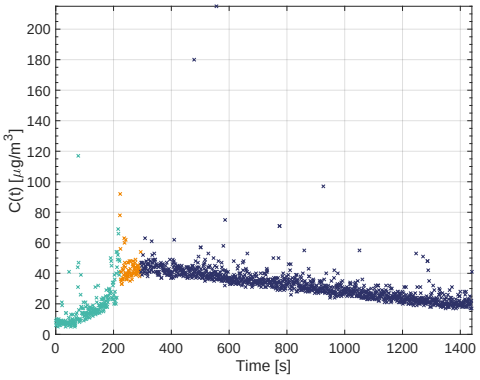
(X) Decay - test 12



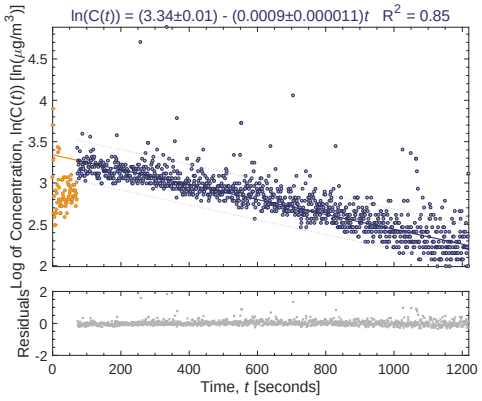
(Y) All - test 13



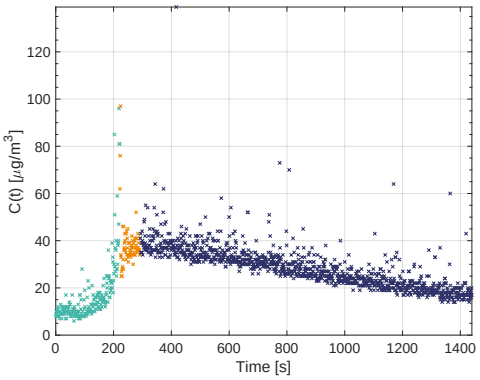
(Z) Decay - test 13



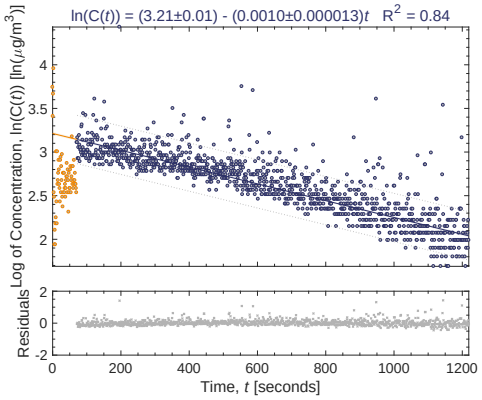
(AA) All - test 14



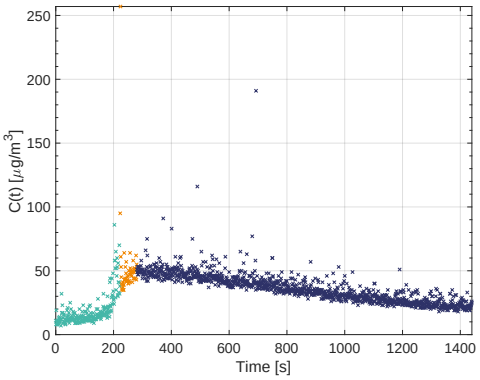
(AB) Decay - test 14



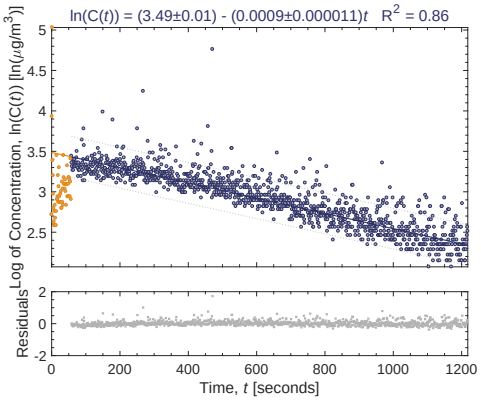
(AC) All - test 15



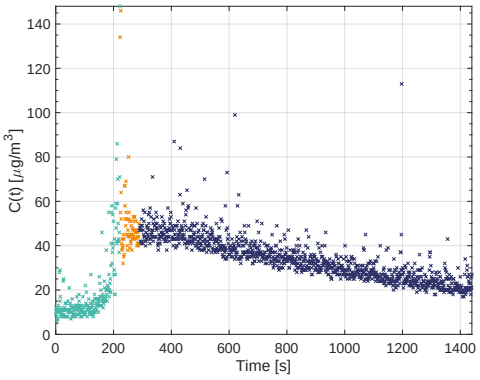
(AD) Decay - test 15



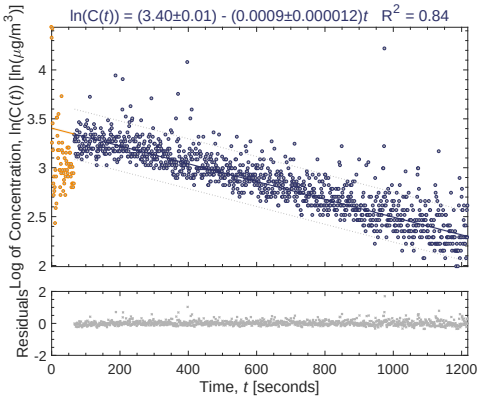
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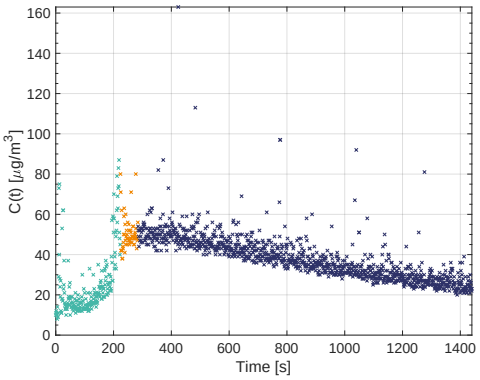
(AF) Decay - test 16



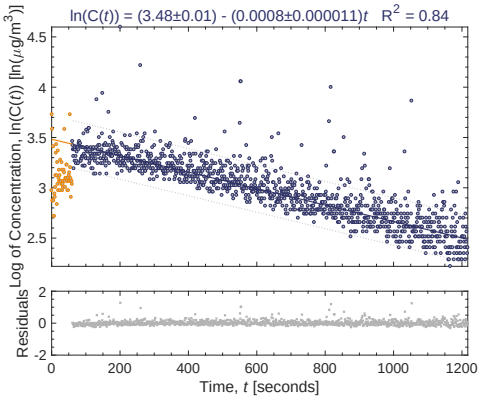
(AG) All - test 17



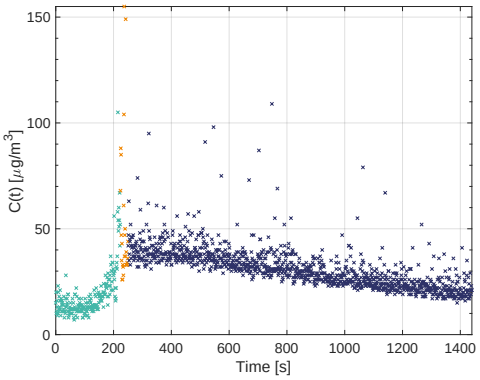
(AH) Decay - test 17



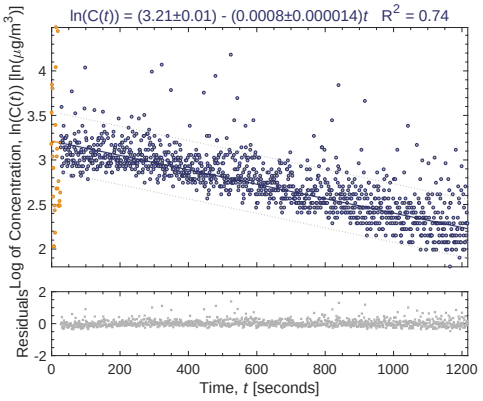
(AI) All - test 18



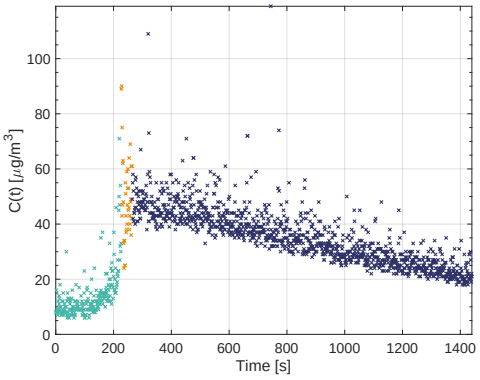
(AJ) Decay - test 18



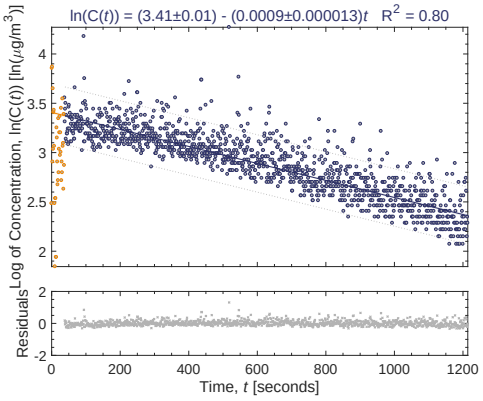
(AK) All - test 19



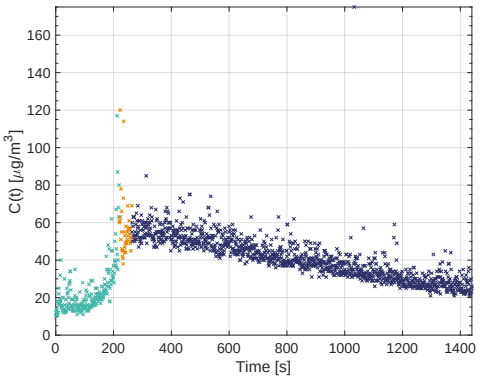
(AL) Decay - test 19



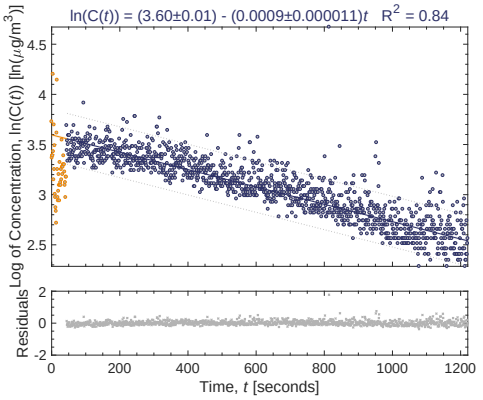
(AM) All - test 20



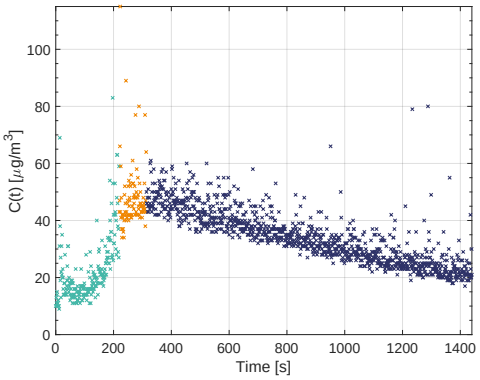
(AN) Decay - test 20



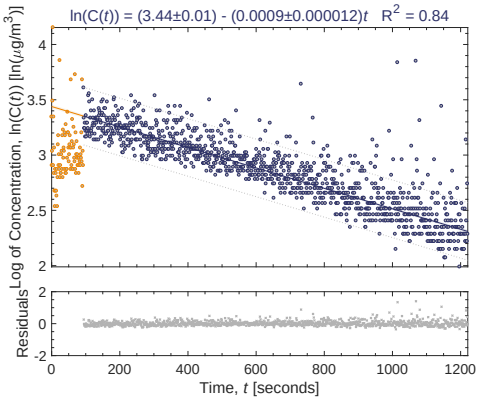
(AO) All - test 21



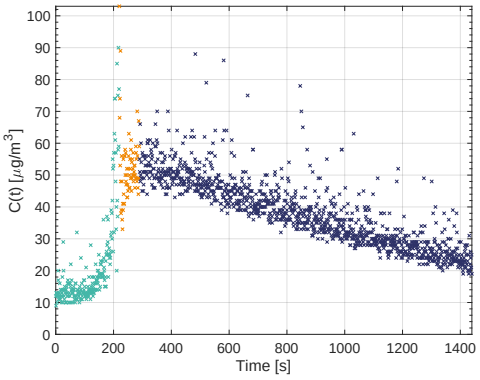
(AP) Decay - test 21



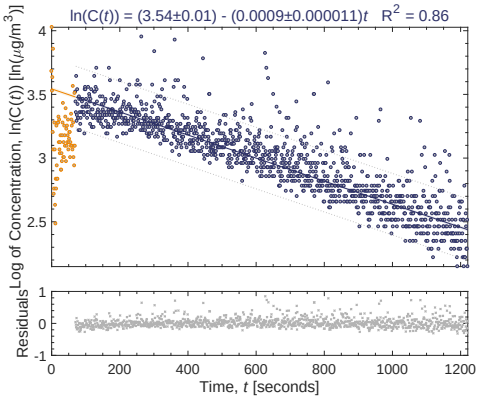
(AQ) All - test 22



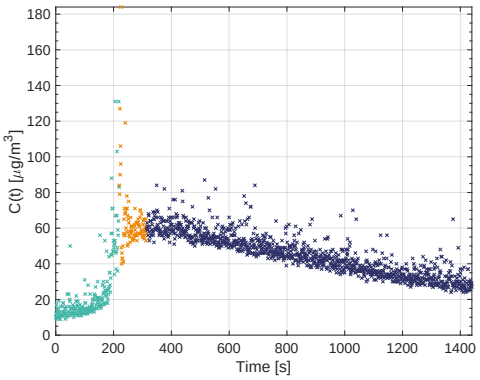
(AR) Decay - test 22



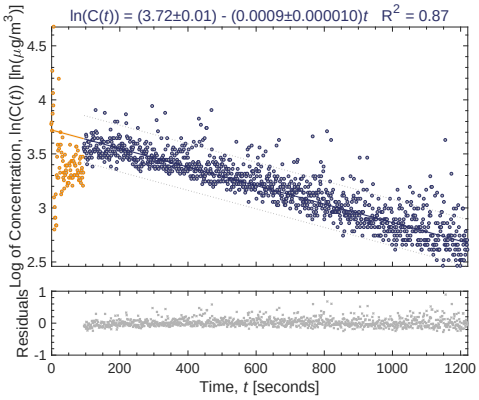
(AS) All - test 23



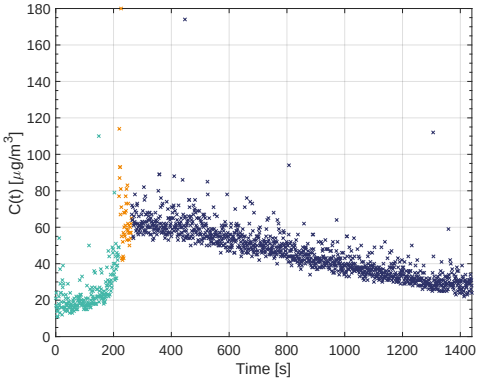
(AT) Decay - test 24



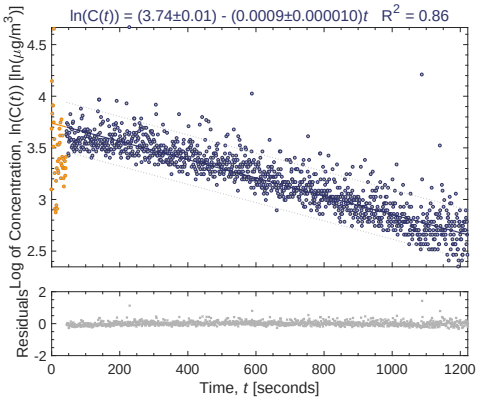
(AU) All - test 24



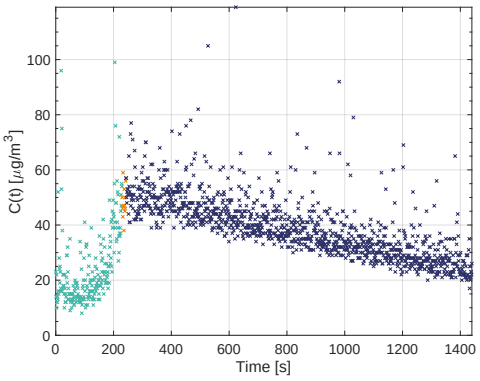
(AV) Decay - test 24



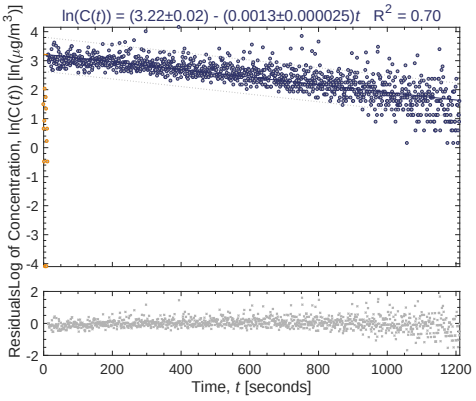
(AW) All - test 25



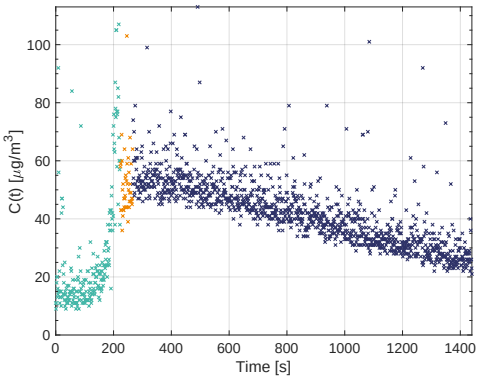
(AX) Decay - test 25



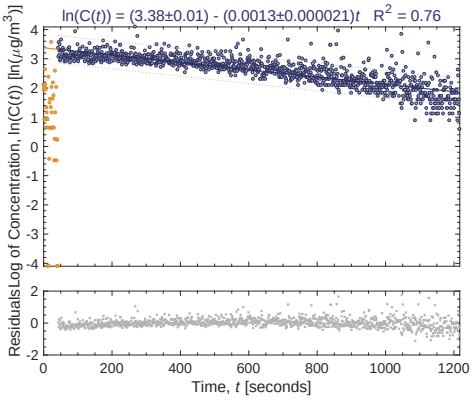
(AY) All - test 26



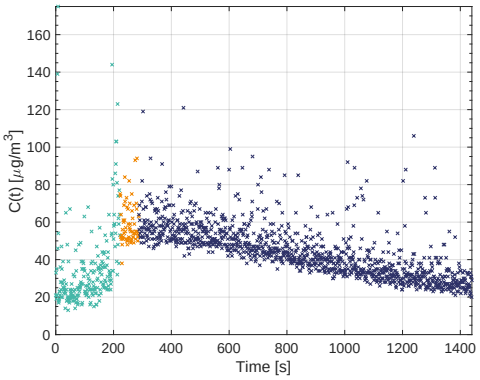
(AZ) Decay - test 26
removed as $R^2 < 0.7$



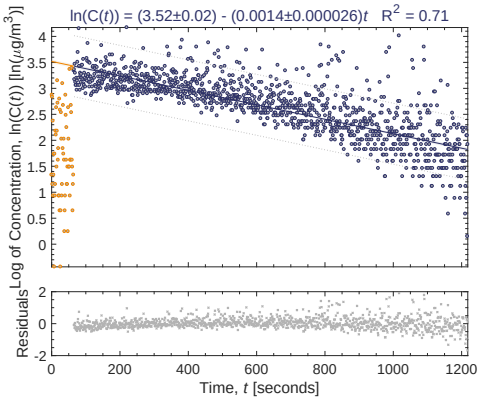
(BA) All - test 27



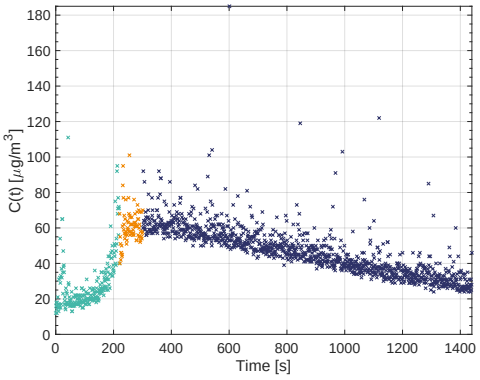
(BB) Decay - test 27



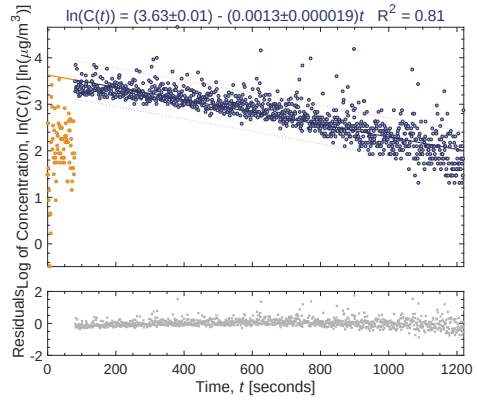
(BC) All - test 28



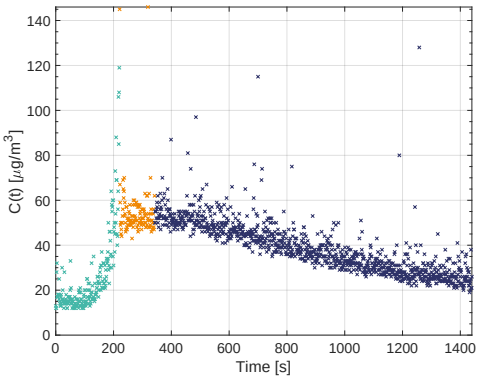
(BD) Decay - test 28



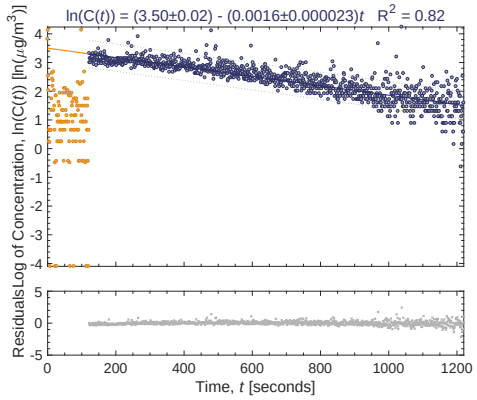
(BE) All - test 29



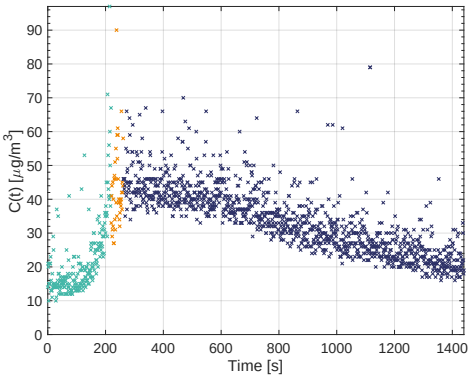
(BF) Decay - test 29



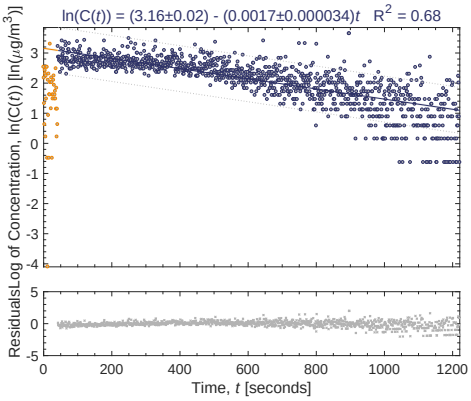
(BG) All - test 30



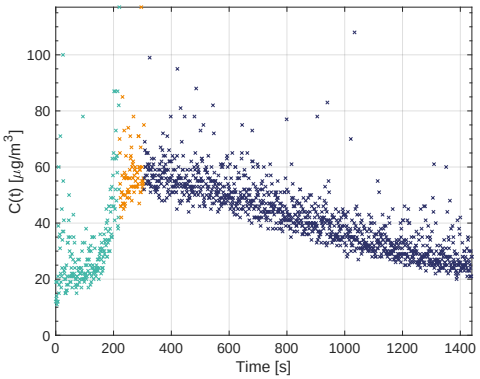
(BH) Decay - test 30



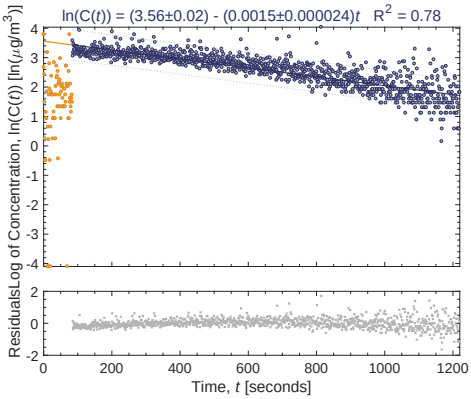
(BI) All - test 31



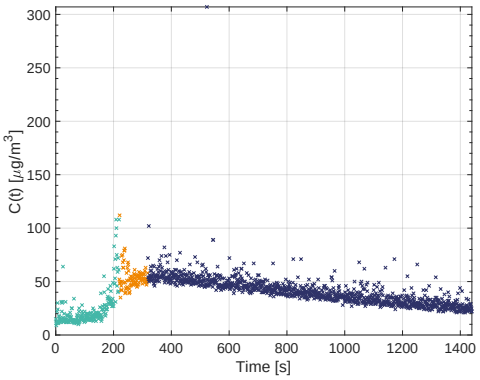
(BJ) Decay - test 31
removed as $R^2 < 0.7$



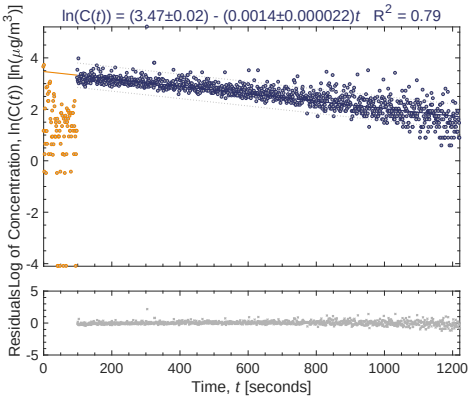
(BK) All - test 32



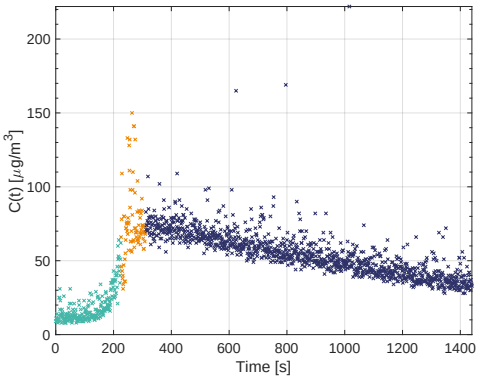
(BL) Decay - test 32



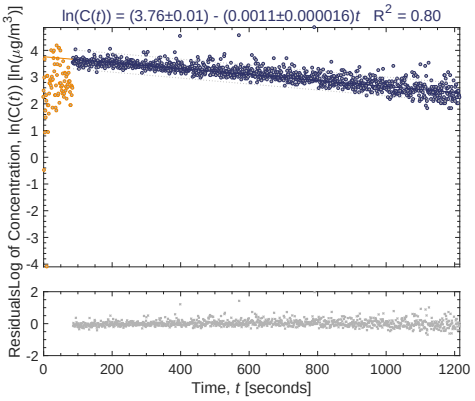
(BM) All - test 33



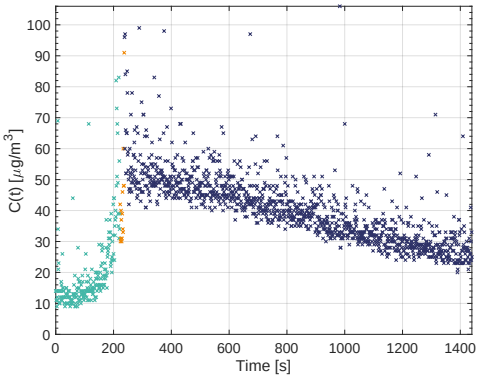
(BN) Decay - test 33



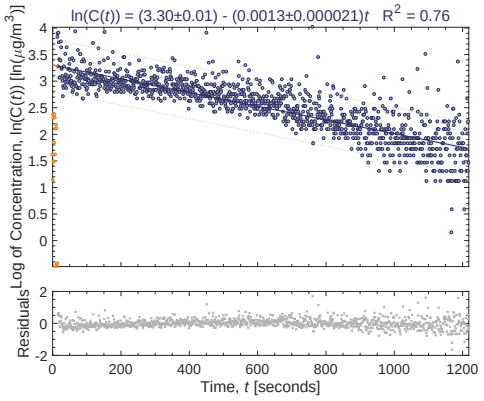
(BO) All - test 34



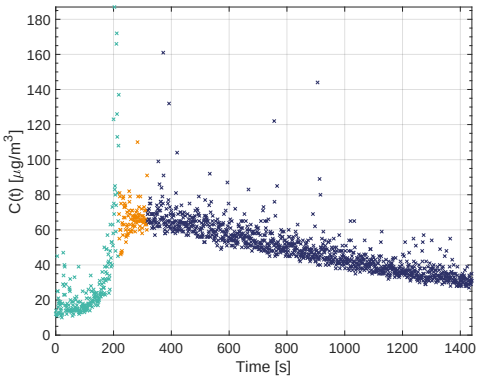
(BP) Decay - test 34



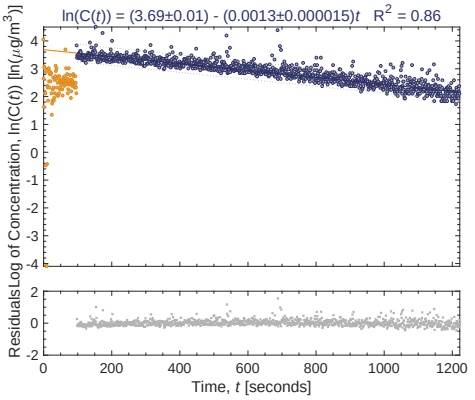
(BQ) All - test 35



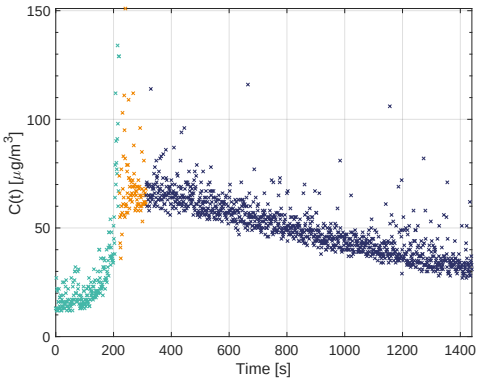
(BR) Decay - test 35



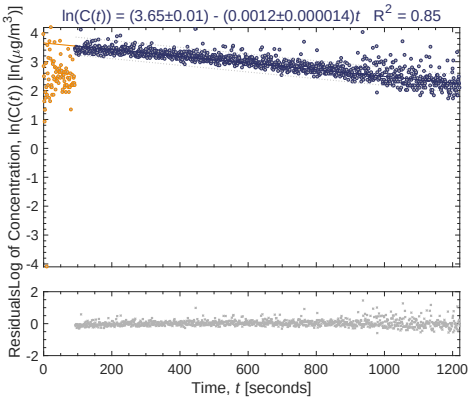
(BS) All - test 36



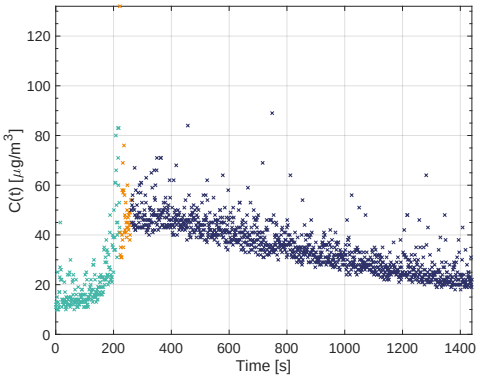
(BT) Decay - test 36



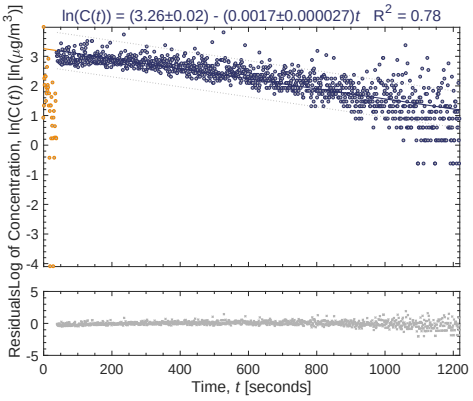
(BU) All - test 37



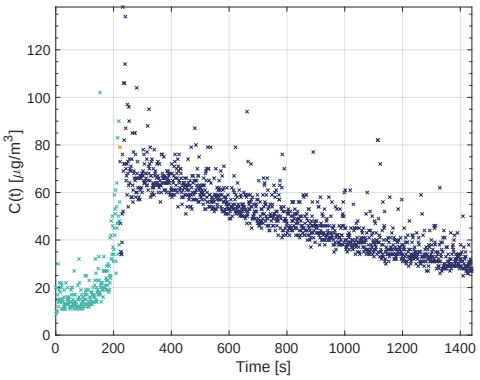
(BV) Decay - test 37



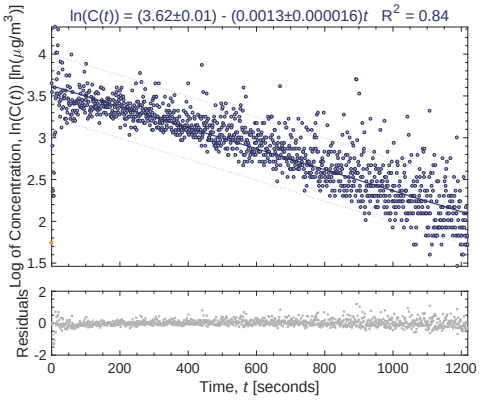
(BW) All - test 38



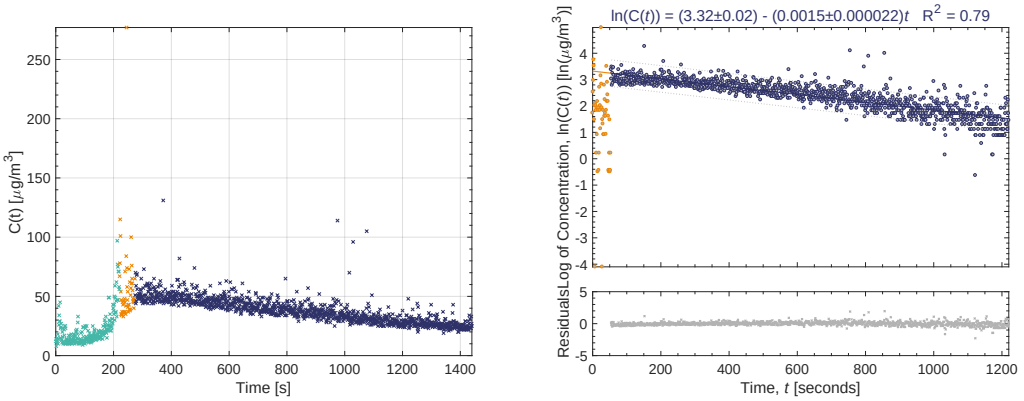
(BX) Decay - test 38



(BY) All - test 39



(BZ) Decay - test 39



(CA) All - test 40 (CB) Decay - test 40

FIGURE D.8: Phase 2 tests, concentrations and decays

D.3 Toast Comparison



(A) Phase 1 wholemeal toast (B) Phase 1 white toast (C) Phase 2 toast

FIGURE D.9: Toast photos

Appendix E

Full Meal $PM_{2.5}$ Emissions

This appendix contains additional supporting information for Chapter 5.

E.1 Method

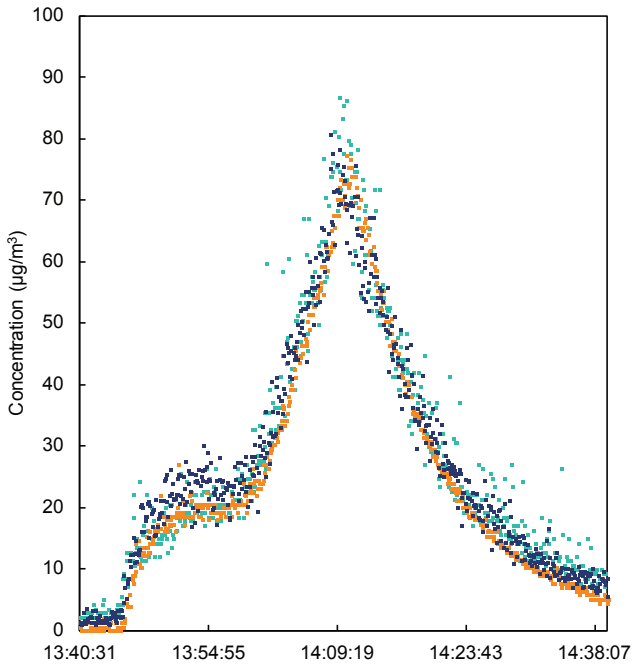


FIGURE E.1: Low ventilation mixing test
blue = Position A; **orange** = Position B; **teal** = Position D

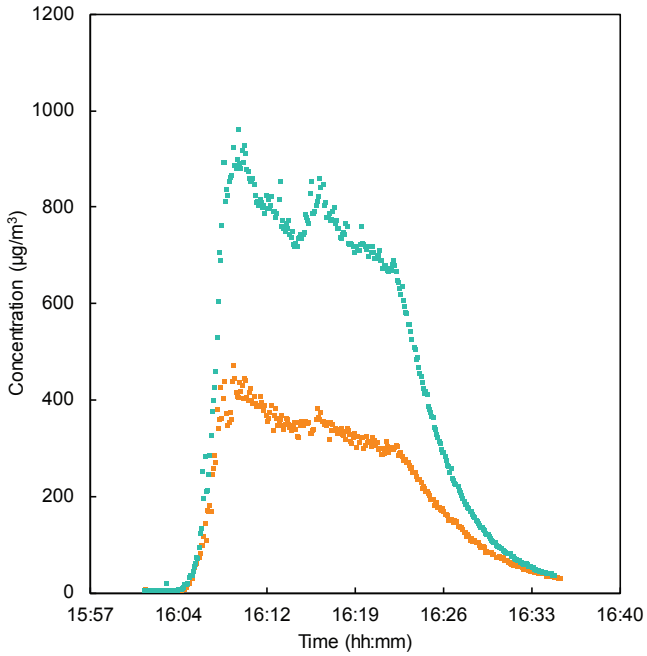
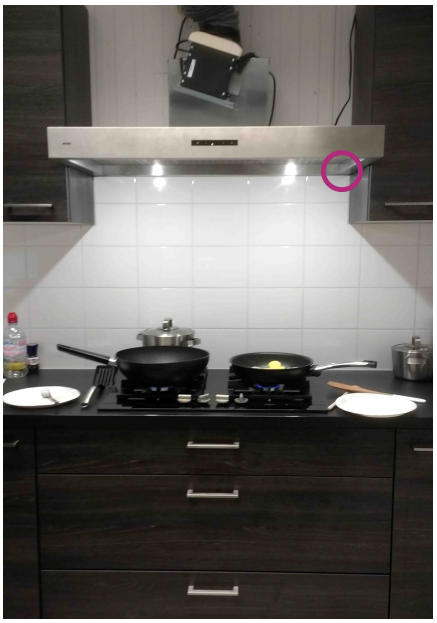
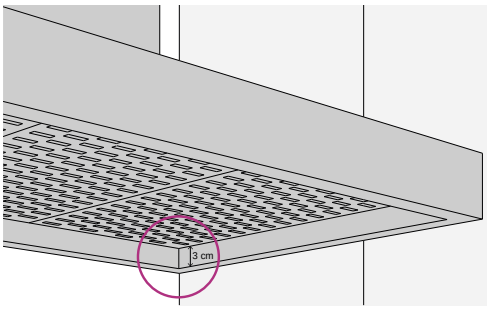


FIGURE E.2: High ventilation mixing test
orange = Position B; **teal** = Position D

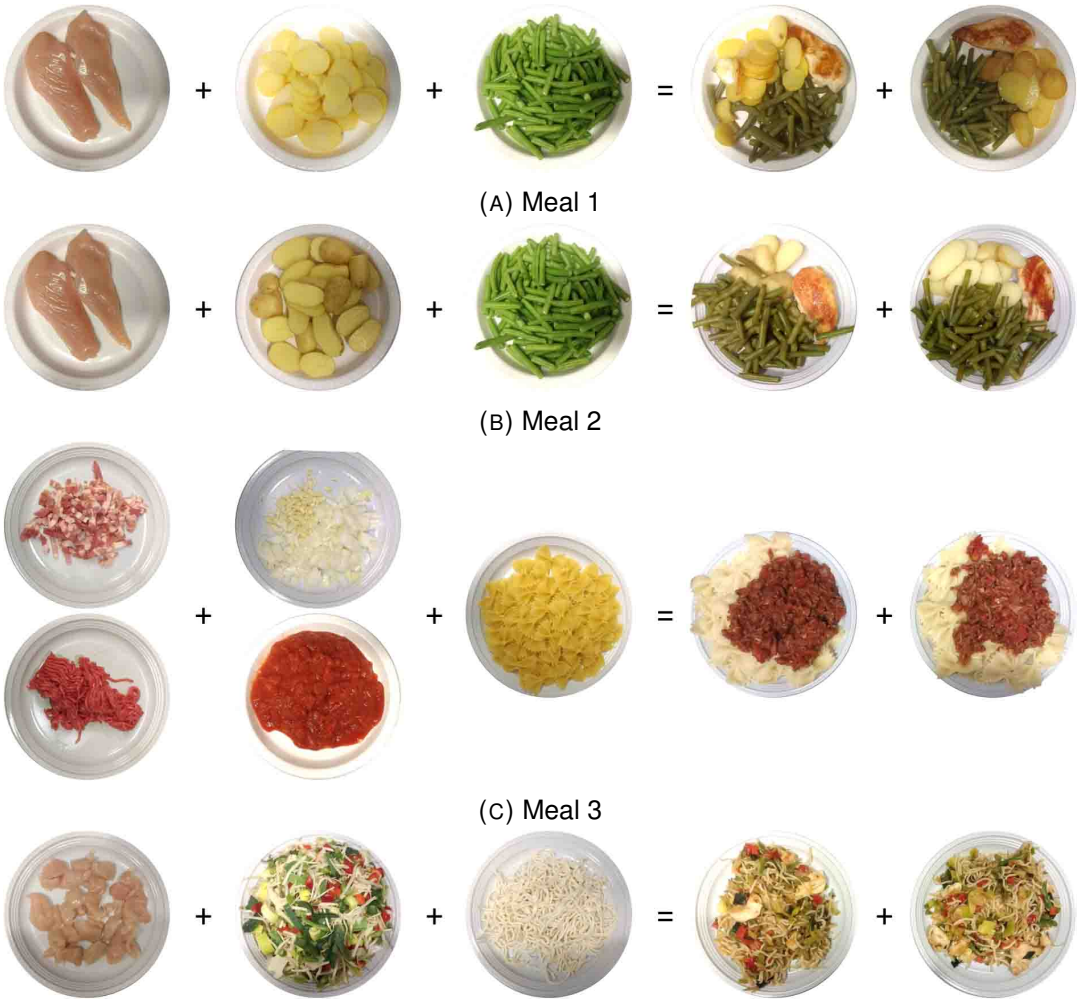


(A) Photo of the test rig with the *damp buffer* circled



(B) 3D diagram of the hood showing the *damp buffer*

FIGURE E.3: The test rig



(A) Meal 1

(B) Meal 2

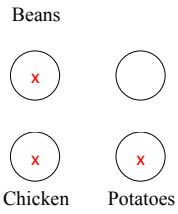
(C) Meal 3

(D) Meal 4

FIGURE E.4: Components of each test meal

Meal 1 - Chicken, Fried Potatoes and French Beans

Chicken	Beans	Potatoes
in 2 pieces	*as purchased*	*peeled and sliced – as purchased*
25 minutes	20 minutes	15 minutes
200 g	280 g	330 g
10 ml oil	750 ml water	50 ml oil

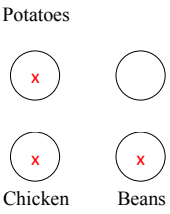


Time (min)	Chicken	Beans	Potatoes	Total gas flow (lpm)
-3	Heat pan and oil			2.5 ± 0.1
0	Add chicken to pan			
1	Turn down			1.3 ± 0.1
2				
3				
4				
5	Turn chicken	Turn on ring		4.2 ± 0.1
6				
7			Heat pan and oil	5.3 ± 0.2
8				
9				
10	Turn chicken		Add potatoes to pan	
11				
12				
13			Stir potatoes (30 sec)	
13				
15	Turn chicken			
16			Stir potatoes (30 sec)	
17				
18				
19			Stir potatoes (30 sec)	
20	Turn chicken			
21				
22			Stir potatoes (30 sec)	
23				
24				
25		Turn off all rings and put on lids – end of test		

FIGURE E.5: Meal 1 cooking protocol

Meal 2 - Chicken, Boiled Potatoes and French Beans

Chicken	Beans	Potatoes
in 2 pieces	*as purchased*	*chopped new potatoes*
25 minutes	20 minutes	15 minutes
200 g	280 g	330 g
10 ml oil	750 ml water	600 ml water

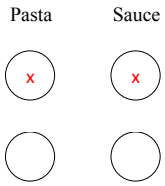


Time (min)	Chicken	Potatoes	Beans	Total gas flow (lpm)
-3	Heat pan and oil			2.5 ± 0.1
0	Add chicken to pan			
1	Turn down			1.3 ± 0.1
2				
3				
4				
5	Turn chicken	Turn on ring		4.2 ± 0.1
6				
7				
8				
9				
10	Turn chicken		Turn on ring	5.3 ± 0.2
11				
12				
13				
13				
15	Turn chicken			
16				
17				
18				
19				
20	Turn chicken			
21				
22				
23				
24				
25	Turn off all rings and put on lids – end of test			

FIGURE E.6: Meal 2 cooking protocol

Meal 3 - Pasta Bolognese

Sauce		Pasta
25 minutes		*as purchased*
10 ml oil	2 cloves garlic	21 minutes
125 g smoked bacon lardons	200g minced beef	150 g
1 onion (chopped)	400g can chopped tomatoes	1500 ml water

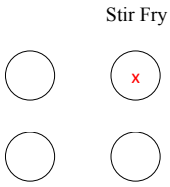


Time (min)	Sauce	Pasta	Total gas flow (lpm)
-3	Heat pan and oil		4.5 ± 0.1
0	Add bacon to pan		
1			
2			
3			
4	Add onion and garlic to pan, and turn down	Turn on pan	4.1 ± 0.1
5			
6	Add beef mince and turn up		4.6 ± 0.1
7			
8			
9			
10	Add tomatoes and mix through for 1 minute		
11			
12			
13	Stir		
14			
15	Turn down	Add pasta to pan	3.9 ± 0.1
16	Stir		
17			
18		Turn down	1.9 ± 0.1
19	Stir		
20			
21			
22	Stir		
23			
24			
25	Turn off all rings and put on lids – end of test		

FIGURE E.7: Meal 3 cooking protocol

Meal 4 - Stir Fry

Stir Fry	
14 minutes	
200 g chicken	10 ml oil
330 g stir fry vegetables	20 ml oil
150 g straight to wok noodles	

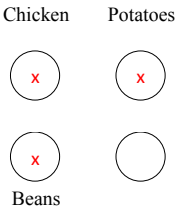


Time (min)	Stir Fry	Total gas flow (lpm)
-3	Heat pan and 10 ml oil	4.5 ± 0.1
0	Add chicken to pan	
1	Turn chicken	
2	Turn down	1.1 ± 0.1
3		
4		
5	Remove chicken, add 20 ml oil to pan and turn up	4.4 ± 0.1
6		
7	Add vegetables, stir continuously	
8		
9		
10		
11		
12	Add chicken and noodles (broken up) and turn down	1.1 ± 0.1
13		
14	Turn off all rings and put on lids – end of test	

FIGURE E.8: Meal 4 cooking protocol

Meal 1 - Chicken, Fried Potatoes and French Beans

Chicken	Beans	Potatoes
in 2 pieces	*as purchased*	*peeled and sliced – as purchased*
25 minutes	20 minutes	15 minutes
200 g	280 g	330 g
10 ml oil	750 ml water	50 ml oil



Time (min)	Chicken	Beans	Potatoes	Total gas flow (lpm)
-3	Heat pan and oil			2.5 ± 0.1
0	Add chicken to pan			
1	Turn down			1.3 ± 0.1
2				
3				
4				
5	Turn chicken	Turn on ring		3.7 ± 0.1
6				
7			Heat pan and oil	5.0 ± 0.2
8				
9				
10	Turn chicken		Add potatoes to pan	
11				
12				
13			Stir potatoes (30 sec)	
13				
15	Turn chicken			
16			Stir potatoes (30 sec)	
17				
18				
19			Stir potatoes (30 sec)	
20	Turn chicken			
21				
22			Stir potatoes (30 sec)	
23				
24				
25		Turn off all rings and put on lids – end of test		

FIGURE E.9: Meal 1 backburners cooking protocol

E.2 Results

TABLE E.1: Gravimetric sampling results

	Sampling	Total Mass (µg)			Average Concentration (µg/m^3)				CF ±
	Times (s)	Filter 1	Filter 2	Filter 3	Filter 1	Filter 2	Filter 3	OPC	Standard Error
Meal 1	3480	310	340	300	139	153	135	36.3	3.92 ± 0.15
Meal 2	1680	180	170	170	168	158	158	32.3	4.99 ± 0.096
Meal 3	3240	1620	1570	1650	783	758	797	290*	2.69 ± 0.039
Meal 4	2760	1020	1030	1120	578	584	635	401	1.50 ± 0.045
Pump flow rate was 2.3 m^3/h (0.64 L/s) during all tests									

* peak particle count exceeded 2 000 000 particles/litre

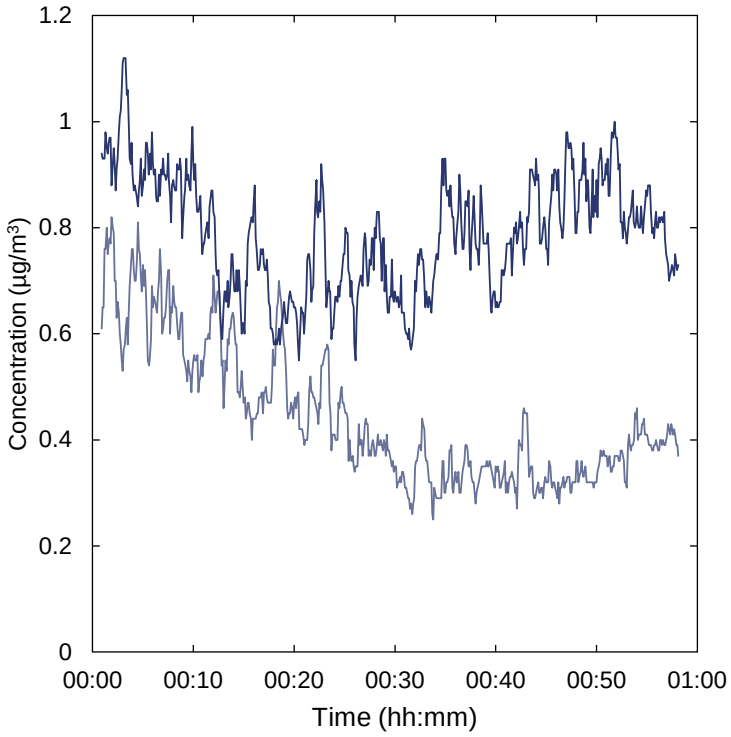


FIGURE E.10: $PM_{2.5}$ concentrations during *blank* tests

E.2.1 Key Moments

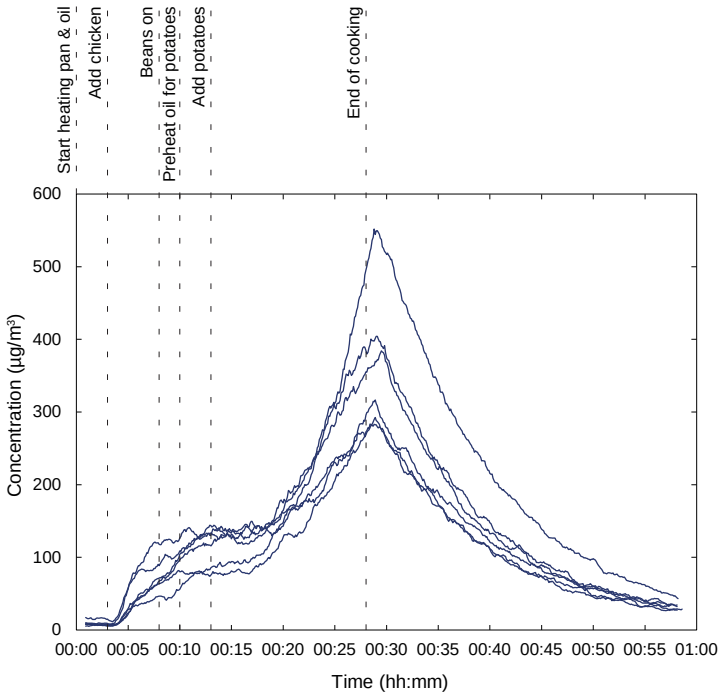


FIGURE E.11: Meal 1 - key moments

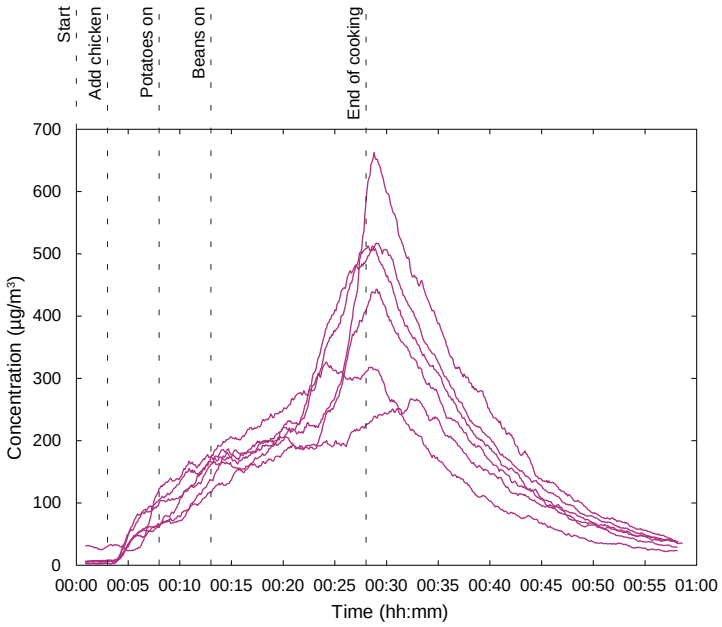


FIGURE E.12: Meal 2 - key moments

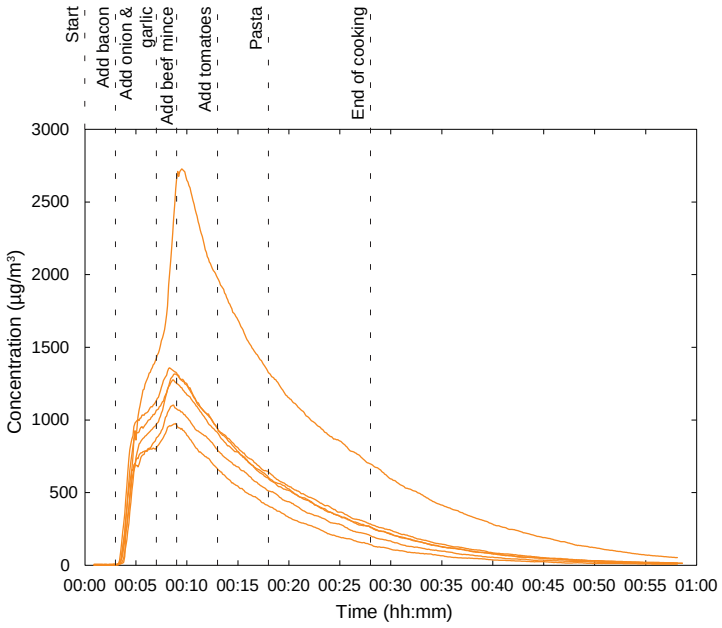


FIGURE E.13: Meal 3 - key moments

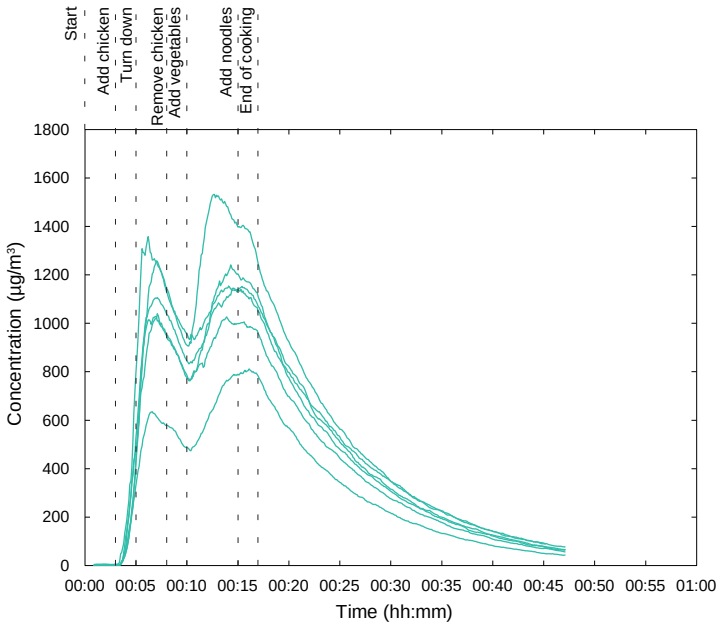
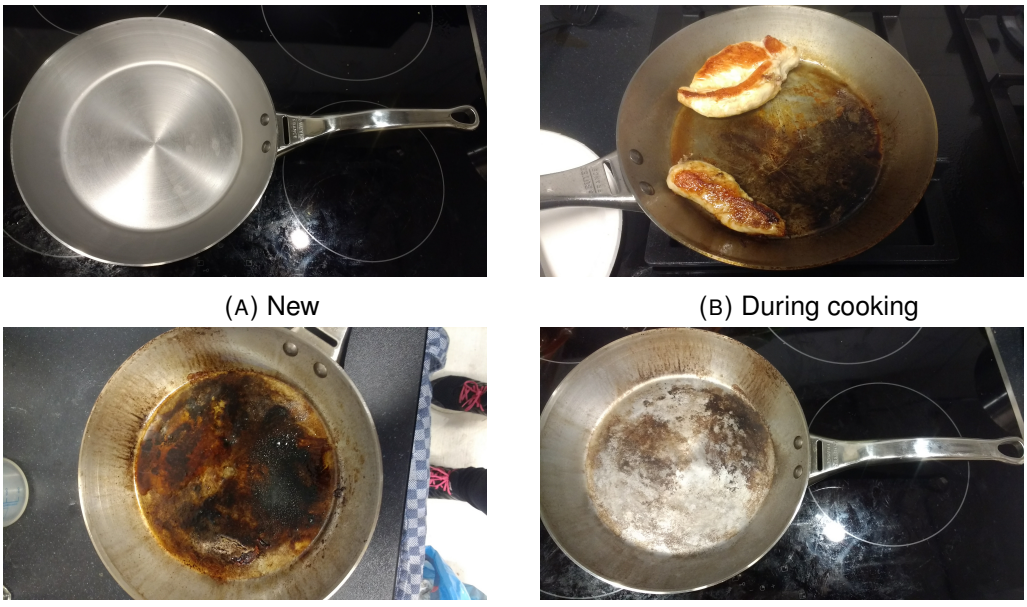


FIGURE E.14: Meal 4 - key moments



(A) New (B) During cooking

(C) Unwashed, after use (D) Washed, after final test

FIGURE E.15: Stainless steel pan ageing

E.2.2 Decay Rates

TABLE E.2: Measured decay Rates, base test meals, low ventilation

	Decay Rate (h^{-1})							
	Meal 1		Meal 2		Meal 3		Meal 4	
Test	Decay	R^2	Decay	R^2	Decay	R^2	Decay	R^2
1	4.8 ± 0.044	0.98	5.9 ± 0.039	0.99	5.9 ± 0.035^a	0.99	5.3 ± 0.016^a	1.00
2	4.7 ± 0.039	0.98	5.3 ± 0.037	0.99	5.8 ± 0.038^a	0.99	5.7 ± 0.019^a	1.00
3	5.0 ± 0.046	0.98	5.6 ± 0.053	0.97	5.8 ± 0.055^a	0.97	5.5 ± 0.016^a	1.00
4	5.3 ± 0.040	0.98	5.6 ± 0.027	0.99	6.0 ± 0.062	0.97	5.5 ± 0.017^a	1.00
5	5.2 ± 0.028	0.99	6.1 ± 0.042	0.99	5.9 ± 0.034^a	0.99	5.6 ± 0.017^a	1.00
6	5.1 ± 0.049	0.97	4.7 ± 0.041	0.98	5.2 ± 0.025^a	0.99	5.6 ± 0.016^a	1.00
$\mu \pm \alpha_\phi$	5.0 ± 0.10		5.5 ± 0.21		5.7 ± 0.12		5.5 ± 0.06^a	

[a] particle count exceeded 2 000 000 particles/L

μ = arithmetic mean; α = standard error

TABLE E.3: Measured decay rates, Meal 1 variations, low ventilation

	Decay Rate (h^{-1})					
	Margarine		Steel		Salt	
Test	Decay	R^2	Decay	R^2	Decay	R^2
1	5.4 ± 0.045	0.98	4.5 ± 0.014^a	1.00	5.7 ± 0.035	0.99
2	5.5 ± 0.041	0.98	3.9 ± 0.012^a	1.00	5.5 ± 0.037	0.99
3	5.6 ± 0.034	0.99	4.5 ± 0.013^a	1.00	5.8 ± 0.032	0.99
4	5.4 ± 0.031	0.99	4.4 ± 0.012^a	1.00	5.7 ± 0.031	0.99
5	5.0 ± 0.044	0.98	3.7 ± 0.010^a	1.00	5.3 ± 0.039	0.99
$\mu \pm \alpha_\phi$	5.4 ± 0.11		4.2 ± 0.16		5.9 ± 0.09	

[a] particle count exceeded 2 000 000 particles/L

μ = arithmetic mean; α = standard error

TABLE E.4: Measured decay rates, high ventilation

	Decay Rate (h^{-1})									
	Meal 1		Meal 1 BB		Meal 2		Meal 3		Meal 4	
Test	Decay	R^2	Decay	R^2	Decay	R^2	Decay	R^2	Decay	R^2
1	2.7 ± 0.11	0.66	1.2 ± 0.062	0.55	1.8 ± 0.13	0.39	$-^a$	-	3.2 ± 0.12	0.71
2	3.9 ± 0.12	0.77	0.70 ± 0.029	0.65	4.7 ± 0.17	0.72	0.076 ± 0.043	0.010	3.0 ± 0.11	0.73
3	2.3 ± 0.11	0.60	2.0 ± 0.059	0.79	4.5 ± 0.17	0.71	0.36 ± 0.047	0.16	2.3 ± 0.073^a	0.76
4	1.9 ± 0.12	0.45	1.1 ± 0.059	0.54	1.3 ± 0.14	0.24	$-^a$	-	2.1 ± 0.088	0.66
5	3.8 ± 0.13	0.75	1.7 ± 0.067	0.67	3.6 ± 0.14	0.70	$-^a$	-	5.6 ± 0.15	0.83
$\mu \pm \alpha_\phi$	2.9 ± 0.40		1.3 ± 0.22		3.2 ± 0.14		0.22 ± 0.14		3.3 ± 0.68	

[a] Concentrations too low to estimate decay rate.

BB = back burners; μ = arithmetic mean; α = standard error

Appendix F

Predicted Kitchen $PM_{2.5}$ Concentrations

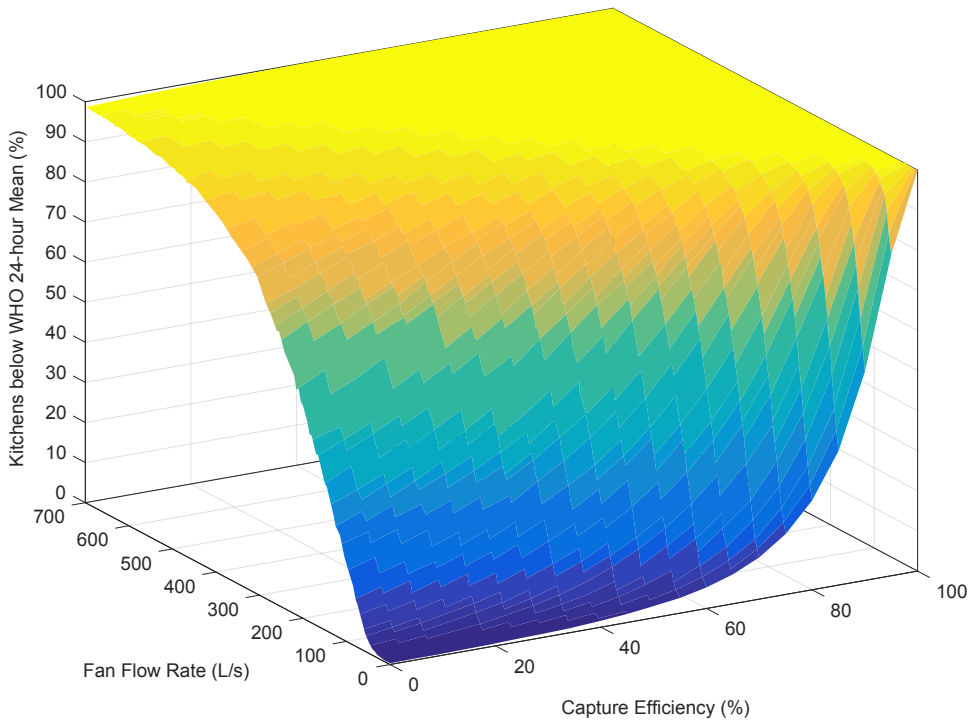


FIGURE F.1: 3-dimensional plot, existing houses

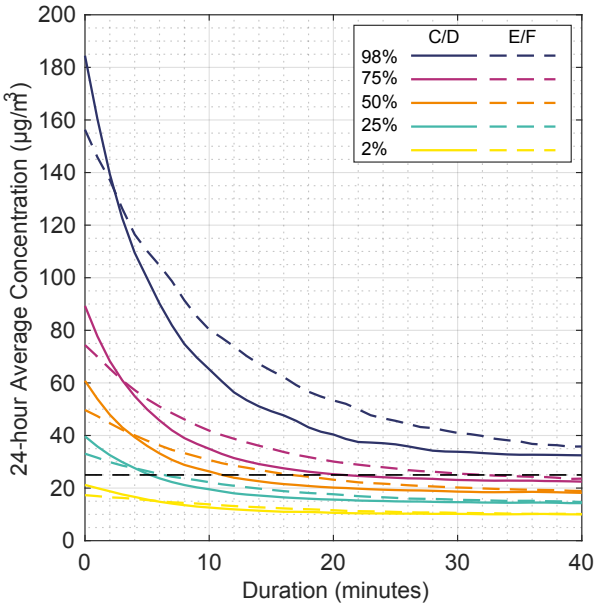


FIGURE F.2: Variation of additional ventilation, existing stock

F.1 Sensitivity Analysis Plots

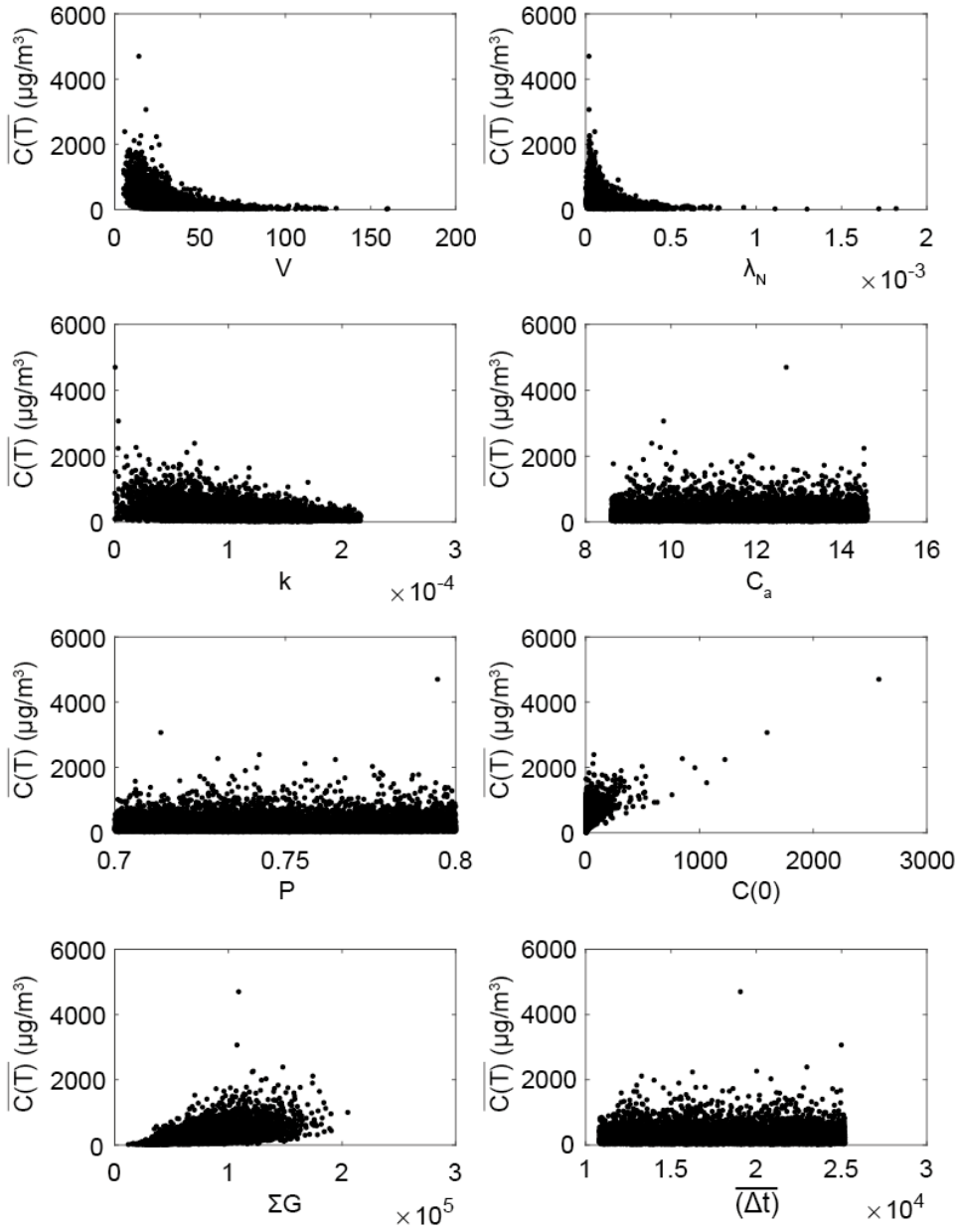
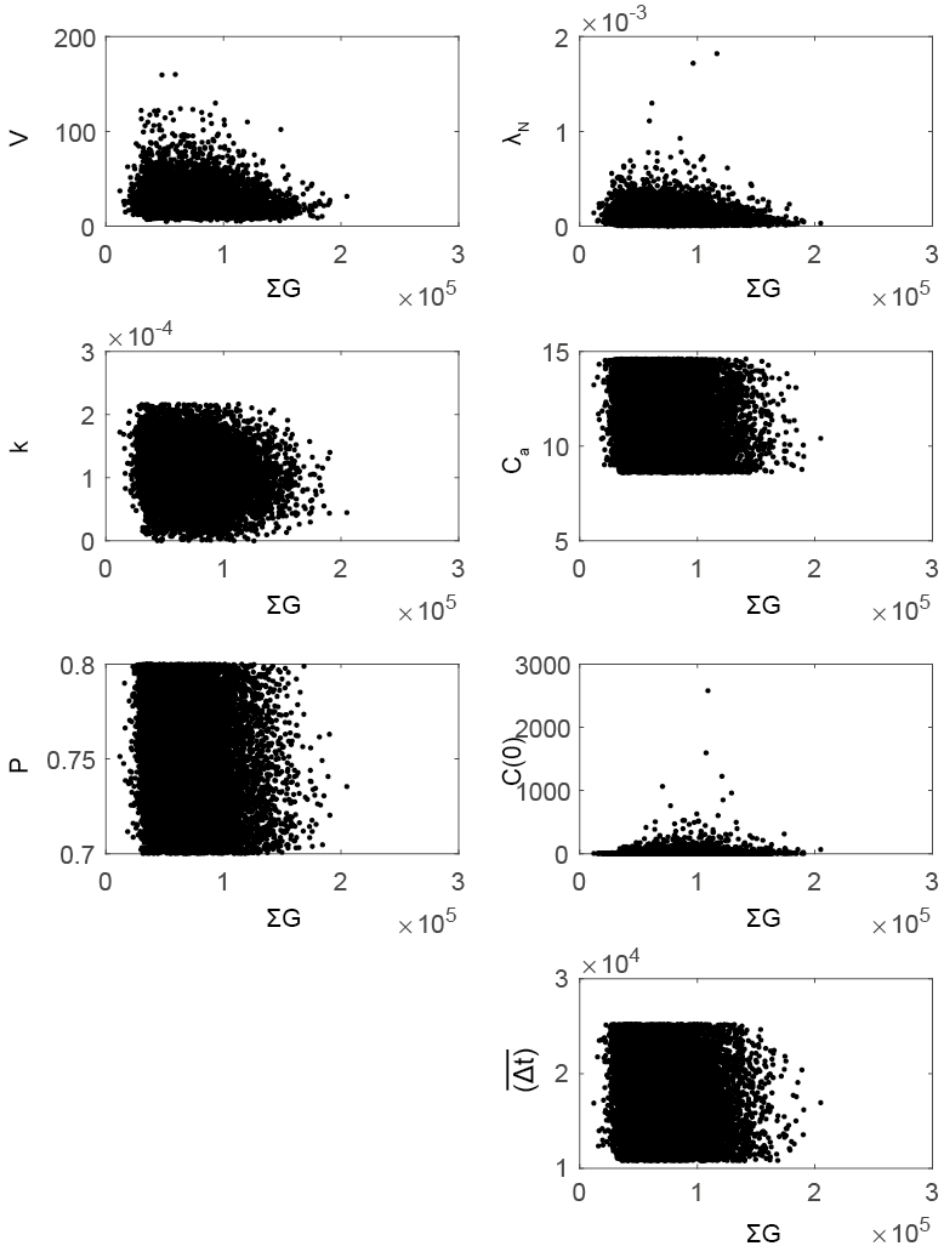
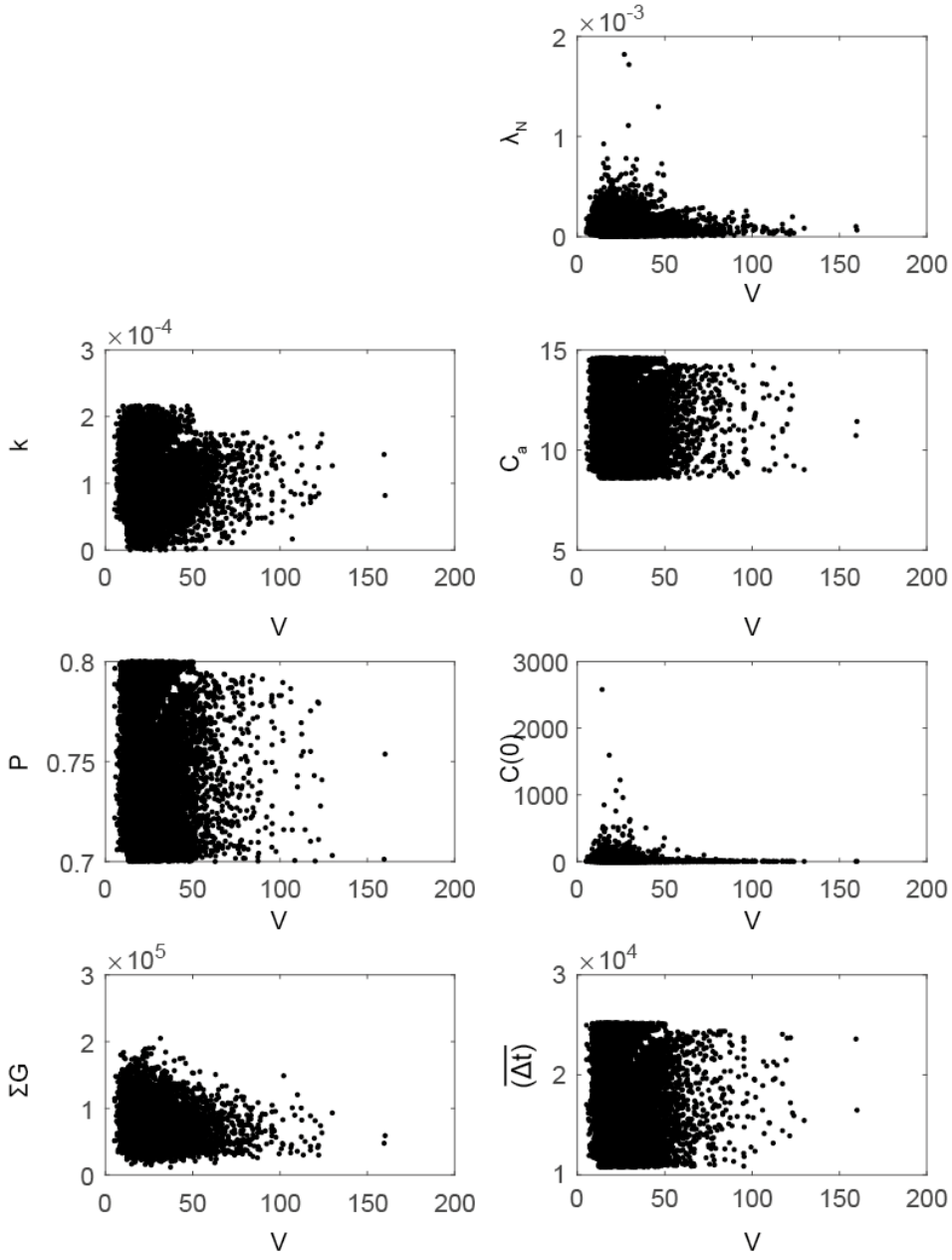


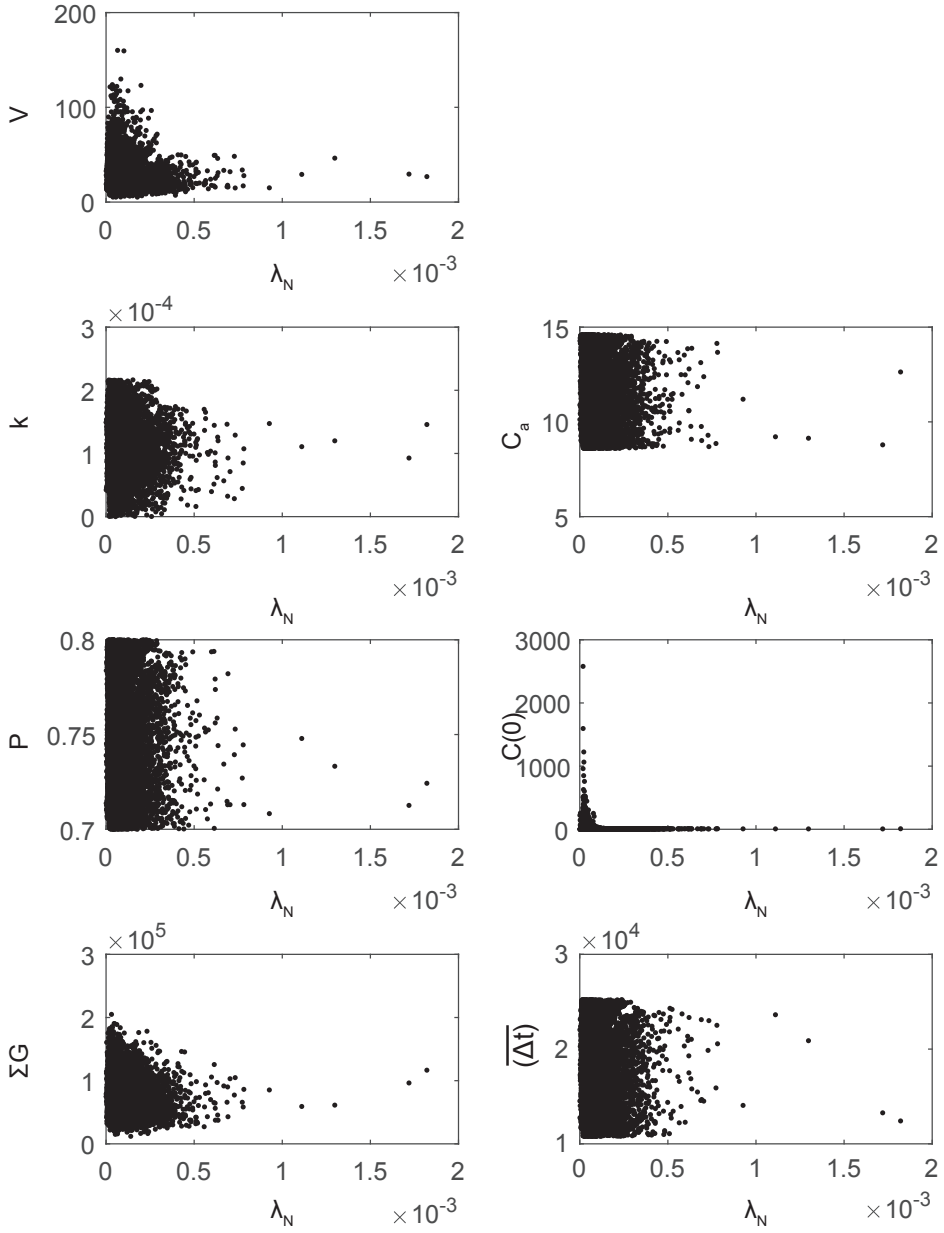
FIGURE F.3: Plotted correlations between all inputs and $\overline{C(T)}$



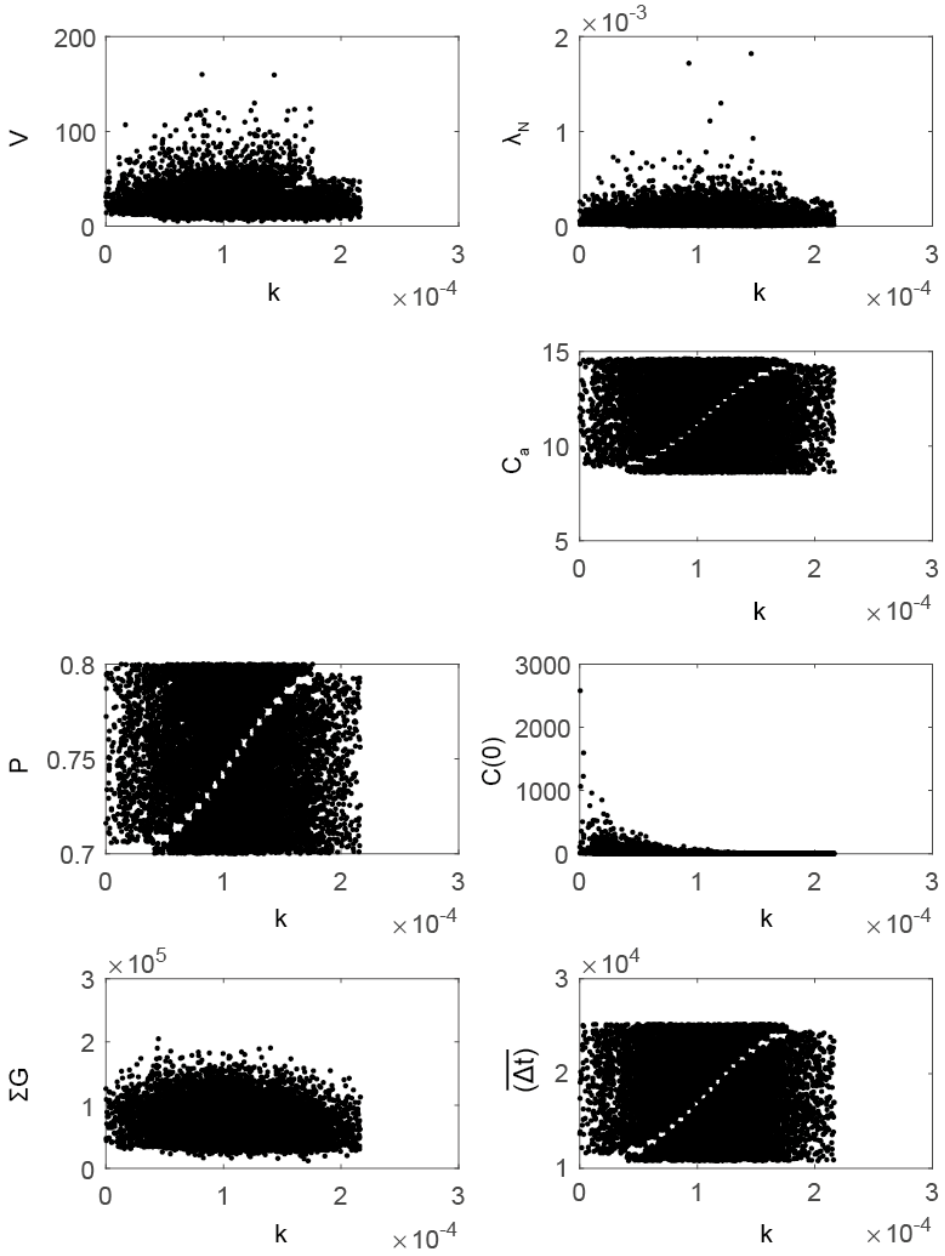
(A) Source strength



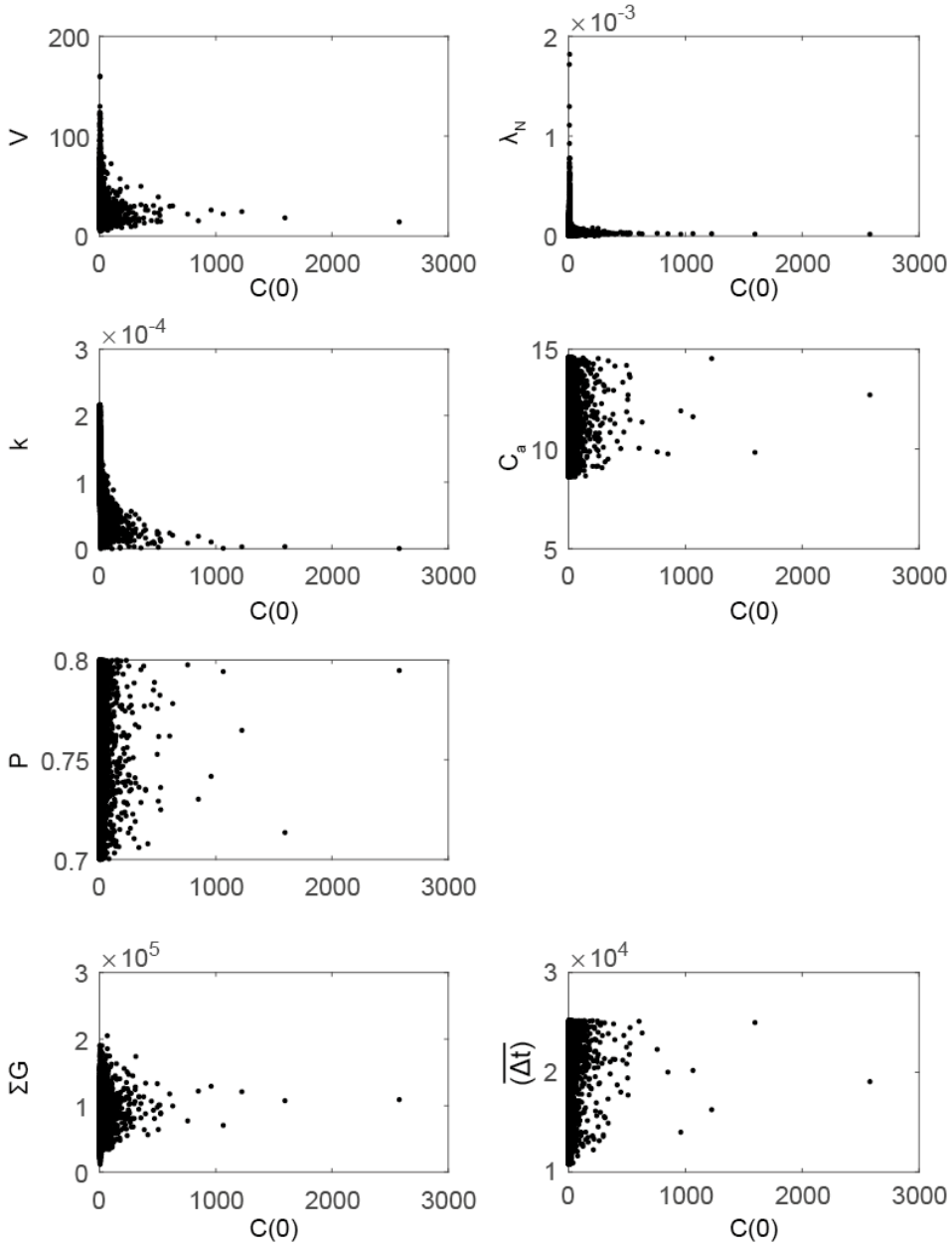
(B) Volume



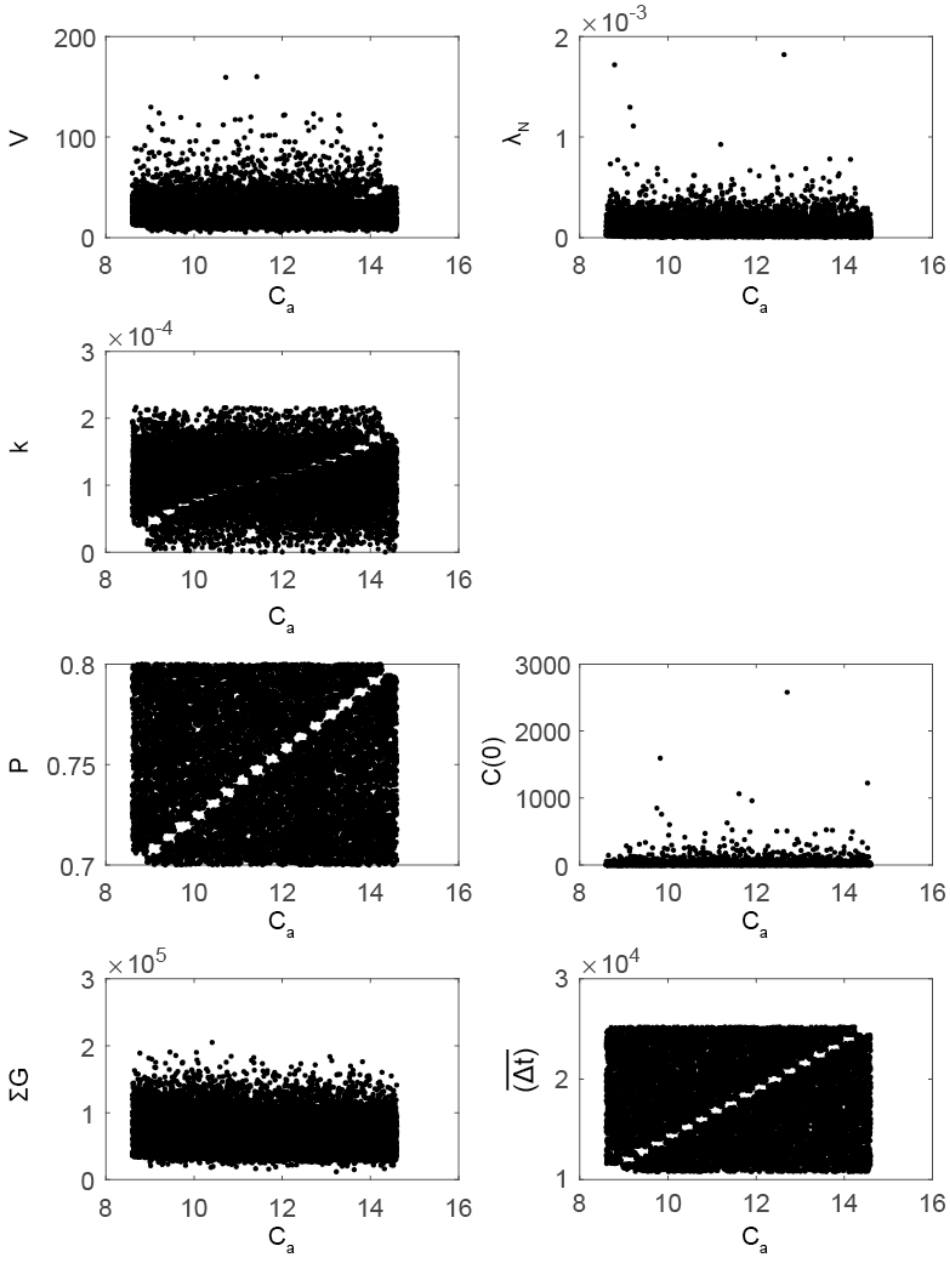
(c) Infiltration



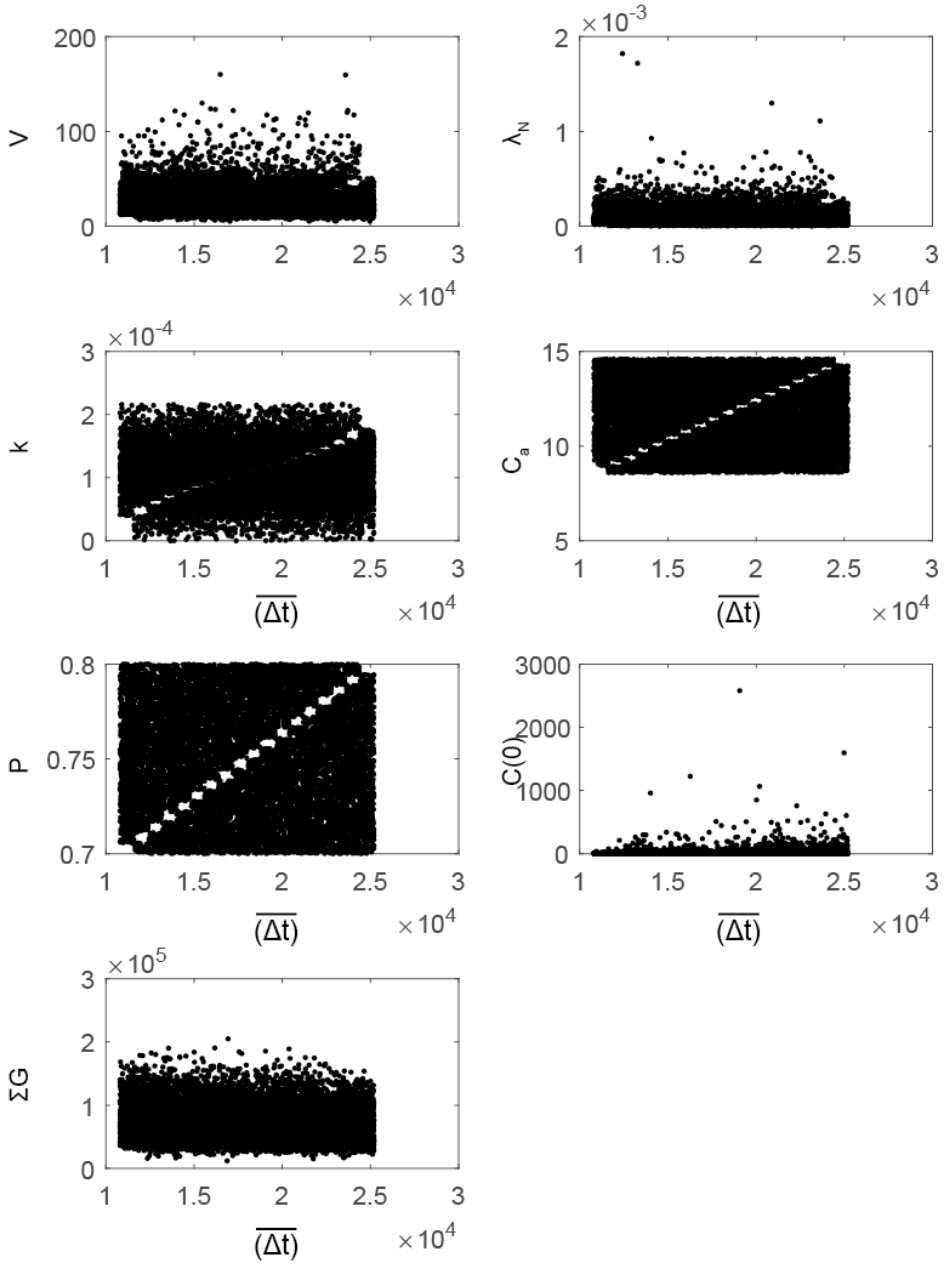
(D) Deposition



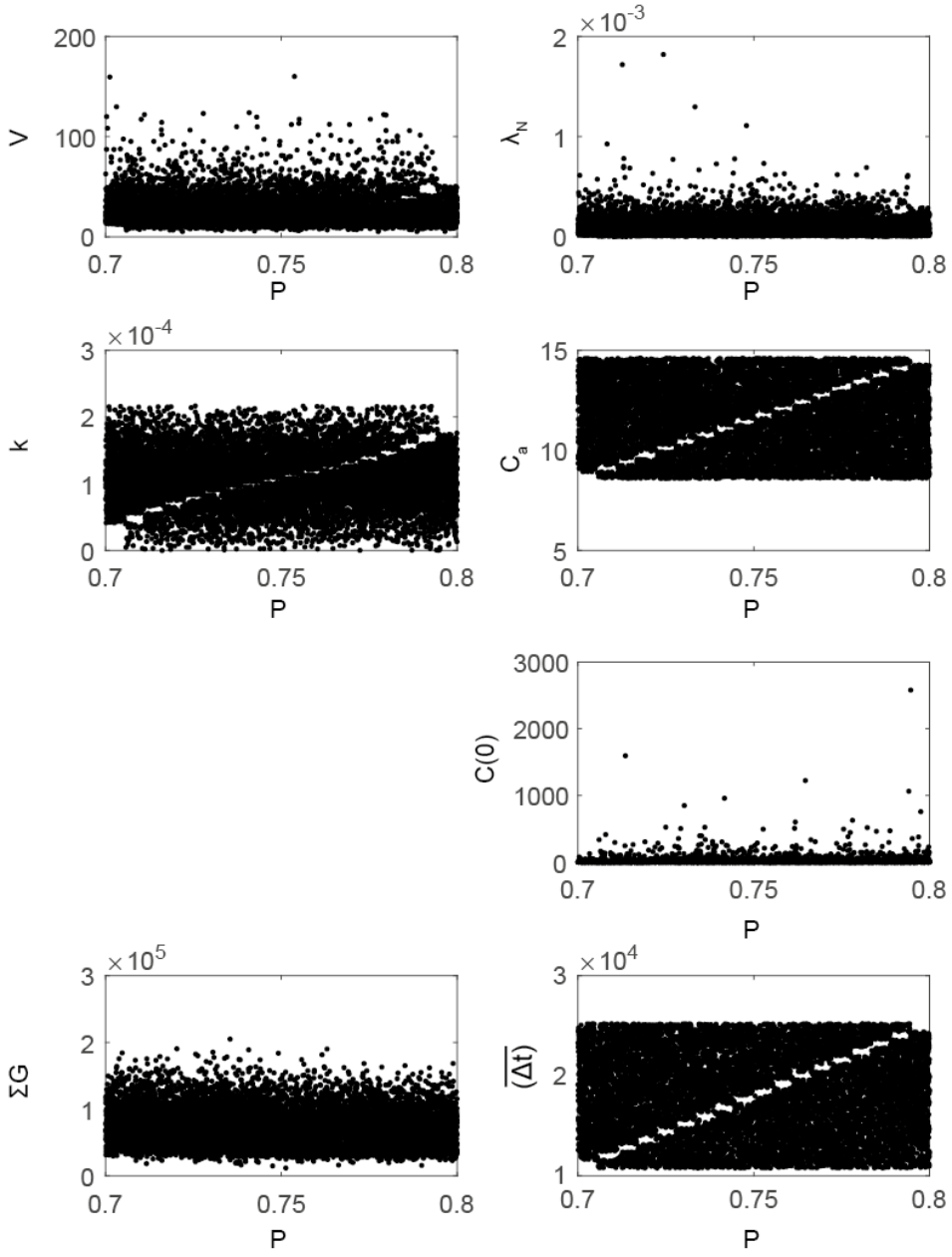
(E) Initial concentration



(F) Ambient concentration



(G) $(\overline{\delta t})$



(H) Penetration coefficient

FIGURE F.4: Plots of $\overline{C(T)}$ inputs to check for correlations

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