

THE HEALTH BENEFITS AND RISKS OF GROWING-YOUR-OWN PRODUCE IN AN URBAN ENVIRONMENT.

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PREFACE

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ACRONYMS

<u>Acronym</u>	<u>Definition</u>	<u>First use (page #)</u>
ADE	Average Daily Exposure	26
ALARP	As Low As Is Reasonably Practicable	9
ANCOVA	Analysis of covariance	60
ANOVA	Analysis of variance	59
BCF	Bio-concentration Factor	86
BDN	Bayesian Decision Network	223
BMD	Benchmark Dose	14
BMI	Body mass index	175
BN	Bayesian Network	VI
C4SL	Category 4 Screening Level	13
CEC	Cation exchange capacity	55
CF	Concentration Factor (used in CLEA model)	25
CHD	Coronary heart disease	35
CHD	Chronic Heart Disease	235
CLEA	Contaminated Land Exposure Assessment Model	3
CPT	Conditional probability table	218
CR	Consumption rate	28
CRM	Certified reference material	?
CVD	Cardiovascular disease	215
DAG	Direct acyclic graph	217
DALY	Disability adjusted life years	35
DBN	Dynamic Bayesian Network	223
DW	Disability weights (<i>used in DALY calculation</i>)	36
EA	Environment Agency (UK)	3
GPM	Grouped produce model	105
GYO	Grow Your Own	VI
HCV	Health Criteria Value	22
HF	Home-grown fraction	28
HI	Hazard Index	26
HIA	Health Impact Assessment	2
HM	Heavy metal	2
HQ	Hazard Quotient	26
HR	Hazard Ratio	33
HSCIC	Health and Social Care Information Centre	182
IC	Inorganic carbon	52
ICP-MS	Inductively coupled plasma mass spectrometry	54
IPM	Individual produce model	105
IR	Intake rate	26
Kd	Soil-to-solution coefficient	88
LF	Lifestyle Factor	236

LLTC	Low Level of Toxicological Concern	22
LOD	Limit of detection	141
LOQ	Limit of quantification	141
MA	Microsoft Access ©	45
MCS	Mental component summary (<i>metric of the SF 36v2</i>)	183
NCC	Nottingham City Council	42
NMF	New Model Framework (Ligand and Trapp, 2010)	87
NOAEL	No Observed Adverse Effect Level	14
OOBN	Object Orientated Bayesian Network	223
OR	Odds Ratio	33
PAH	Polyaromatic Hydrocarbon	2
PBPK	Physiologically based pharmacokinetic model	262
PCB	Polychlorinated Biphenyl	2
PCS	Physical component summary (<i>metric of the SF 36v2</i>)	183
POD	Point of Departure	14
PP	Precautionary Principle	9
PT	Probability table	218
PTS	Potentially Toxic Substance	2
RBCA	Risk-Based Corrective Action Model	3
Rfc	Reference Concentration	12
Rfd	Reference Dose	12
RMSLE	Root mean squared log error (Equation 3.8)	105
RR	Relative Risk	33
SES	Social Economic Status	182
SF 36v2		
©	Short-form 36 health questionnaire	179
SOM	Soil organic matter	40
SPOSH	Significant Possibility Of Significant Harm	10
TBLM	Terrestrial Biotic Ligand Model (Thakali <i>et al.</i> 2006)	87
TC	Total carbon	52
TOC	Total organic carbon (<i>also referred to as Org C</i>)	52
UF	Uncertainty Factor	22
USEPA	US Environment Protection Agency	3
YLD	Years of life lost attributed to disability (<i>used in DALY calculation</i>)	36
YLL	Years of life lost (<i>used in DALY calculation</i>)	36

ABSTRACT

The practice of gardening and growing-your-own (GYO) produce in urban areas, has been associated with many potential benefits to health from increased fruit and vegetable consumption and exercise, but also health risks arising from exposure to potentially toxic elements (such as cadmium: Cd, and lead: Pb) in urban soils. However, the potential health benefits of gardening are currently overlooked by authorities during assessments of contaminated land, which may result in access to urban gardens and allotments being incorrectly restricted or removed because of concerns over the impact to human health. The trade-off between health benefits and risks is investigated in this thesis through: the sampling and analysis of the properties of allotment soils (chapter 2); a comparison of plant uptake models (chapter 3) verified using a pot experiment (chapter 4), and a questionnaire survey investigating the effect of gardeners' routines on benefits and risks (chapter 5). The different areas of study are combined in the creation of a model framework developed to estimate health benefits and risks attributable to urban gardening (chapter 6).

A total of 149 allotment plots were sampled across the city of Nottingham and analysed in the laboratory for trace element concentrations by ICP-MS. Concentrations of lead (Pb) in top-soils exceeded the category 4 screening level of 84 mg Pb kg⁻¹ dw in most plots (n=129), with a median concentration across all plots of 214 mg Pb kg⁻¹ dw. However, other elemental concentrations (As, Cd, Cr) were below the respective C4SLs for

an allotment land-use and unlikely to pose health risks. In comparison, analysis of the results of a questionnaire survey of 120 gardeners' routines, suggested that gardening more regularly in autumn and winter was positively correlated with categories of increased fruit and vegetable consumption and exercise (one-way ANOVA; $p < 0.05$). Furthermore, gardeners over the age of 45 scored significantly better in the physical health (but not the mental health) categories of the SF 36 v 2 questionnaire compared to standardised scores when results were adjusted for age and gender (Table 5.9). Health benefits and risks were combined using a prototype Bayesian Network (BN) model developed according to the principles of CLEA (Contaminated Land Exposure Assessment model). Health outcomes from benefits (e.g. fruit and vegetable consumption) and risks (e.g. lead toxicity) were derived using a sub-model; requiring odds and hazard ratio's for the outcomes of disease. The model predictions suggested that for the study population, whilst there were some benefits attributable to urban gardening, these appeared to be outweighed by health risks from potential exposure to Pb. Further work is required to confirm or refute this finding and to validate and improve the BN model. The study also highlights the importance of considering health benefits of gardening in risk assessment in future for urban gardeners. Future work should also consider weaknesses highlighted in the use of the CLEA model to predict health risks for gardeners, especially in the prediction of plant uptake of toxic elements, and the limited data availability describing the toxic effects of chemicals exceeding a threshold value which may or may not result in SPOSH.

1 INTRODUCTION

1.1 BACKGROUND

There are two contrasting perspectives in the scientific literature when it comes to the health impacts of the use of urban land for growing-your-own (GYO) produce. The first is that growing-your-own produce is likely to be broadly beneficial to health. Gardeners who practice GYO, grow, and therefore consume, a larger range and quantity of fruit and vegetables than their non-gardening counterparts (Alaimo *et al.* 2008; Zezza and Tasciotti 2010). Spending time in green spaces is likely to benefit an individual's emotional health and well-being (Groenewegen *et al.* 2006) and taking part in gardening activities can reduce stress levels (Van den Berg and Custers 2011; Hawkins *et al.* 2011). Gardening is also a moderate form of exercise and requires individuals to engage in a range of activities that can be beneficial for muscle toning and development as well as cardiovascular fitness (Park *et al.* 2011).

There are also a number of indirect socioeconomic and environmental benefits of urban gardening. Urban gardens have the potential to provide corridors through which wildlife can travel (Goddard *et al.* 2010), mitigate against the urban heat-island effect (Kleerekoper *et al.* 2012) and improve flood control and air quality (Wilby and Perry 2006). Urban gardens also help to provide food security: urban agriculture is practised more prominently by the poor, who cannot afford rising food prices in the marketplace (Zezza and Tasciotti 2010).

The second perspective is that an increase in urban gardening could have a tangible adverse effect on the health of the population. Gardeners are likely to be more exposed (see section 1.2.3) than the general population to potentially toxic substance(s) (PTSs) in the soil, which can enter soils as a result of industrial production, waste disposal, burning of fossil fuels, vehicular emissions and waste products, mining and current/historical military actions among other sources (Swartjes 2011). Soils can become contaminated with a long list of PTSs, including heavy metals (HMs) such as cadmium and lead, as well as organic pollutants including polyaromatic hydrocarbons (PAHs) and polychlorinated biphenyls (PCBs) (Alloway and Ayres 1997). Prolonged human exposure to these substances may result in a range of adverse health effects from skin lesions to cancers (Swartjes and Cornelis 2011).

In the present day, many authorities are aware of the issue of soil contamination with PTSs and carry out monitoring programmes to prevent adverse effects to human health (Swartjes 2011). Health impact assessments (HIAs), are designed to safeguard the public against disease and injury, and actively promote health (WHO 2017a). Assessments of contaminated land; commonly referred to as 'risk assessments', are one example of a HIA used by authorities to reduce the risk to the public from exposure to PTSs. Risk assessments are often informed by mathematical models used to estimate exposure to PTSs. Well-known examples of exposure models include the Contaminated Land Exposure Assessment

model (CLEA)- developed by the UK Environment Agency (EA), and the Risk-Based Corrective Action model (RBCA) - developed by the US Environmental Protection Agency (USEPA). Risk assessment models are required to differentiate and quantify exposures to PTSs from soil arising in different scenarios; including activities at home, on allotment gardens, and in industrial or commercial settings (EA 2009d).

1.1.1 Thesis rationale, aims and objectives

Despite a general awareness among policy makers of the health benefits of gardening, health benefits are not currently considered in risk assessment models for land uses involving gardening. This could lead to an unbalanced view of the effect of gardening in urban soils on health, and the potential for unwarranted closure of gardens due to perceived health risks (Leake *et al.* 2009). Furthermore, the process of assessing health risks arising from exposure to PTSs in soil is complex, leading to considerable uncertainty in the outputs of risk assessment models (Swartjes *et al.* 2013) which are normally interpreted cautiously to prevent harm to human health. Because of time and logistical constraints, the thesis was limited to investigating exposures to HMs contained in the soils of urban gardens, with a particular focus on urban allotments and exposure through the consumption of home-grown produce.

The aims of the work in this thesis were:

- To improve the process of estimating health risks to gardeners from exposure to HMs in soils, through a review of current approaches and the development of plant uptake models.
- To develop an approach for modelling the trade-off between exposures to health benefits and risks from gardening on urban soils.

These aims are achieved through five experimental chapters investigating:

The presence and mobility of contaminants in urban garden soils (chapter 2), the transfer of metals into edible produce from soils (chapters 3 and 4), the influence of gardening routines on the degree of exposure to benefits and risks (chapter 5), and the creation of a method to assess the trade-off between health benefits and risks (chapter 6) (Table 1.1). Data from chapters 2-5 are used to create the exposure model presented in chapter 6.

The remainder of this introduction introduces the reader to the concept of urban gardening and the estimation of health risks and benefits arising from taking part in gardening activities.

Table 1.1: An explanation of chapters 2 to 5 in this thesis

<u>Chapter</u>	<u>Explanation</u>
Chapter 2 (Pages 39-84)	A survey of soils taken from 9 allotments across the urban city of Nottingham to investigate the influence of soil management and soil characteristics on heavy metal concentrations and mobility.
Chapter 3 (Pages 85 – 130)	The development of plant uptake models predicting uptake of cationic metals into a number of fruit and vegetables.
Chapter 4 (Pages 131 – 172)	An experiment to investigate the potential uncertainty that can be attributed to incorrectly grouping produce in exposure models.
Chapter 5 (Pages 173 – 207)	A questionnaire survey of gardener's routines and an investigation of the health benefits of gardening for a group of allotment gardeners.
Chapter 6 (Pages 208 – 259)	The development of a model to predict the trade-off between health benefits and risks attributed to exposures from gardening on urban soils.

1.2 WHAT IS URBAN GARDENING?

The term 'urban gardening' used throughout this thesis is defined as recreational gardening activities taking place in areas located in large towns and cities, where the density of the population and current and historical industrial activity is more likely to result in contamination of soils with PTSs (Davies 1978). Urban gardening may involve the growing of edible produce for personal consumption or for sharing within the gardening community – referred to as GYO ('grow-your-own') (Wakefield *et al.* 2007). There are many forms of urban gardening. These include organised gardening projects such as community gardens and allotment gardens, and more spontaneous gardening projects such as rooftop gardening and 'guerrilla gardening' – the sporadic development of neglected plots of land (Hardman and Larkham 2014).

1.2.1 Allotment gardens

An allotment is a "plot of land rented by an individual for the purpose of growing [produce] or flowers" (OED 2017a). They were first introduced to the UK in the early 19th century by wealthy land-owners as a form of payment to their labourers, ensuring that their workers were not undernourished (Burchardt 2000). Allotments became more popular throughout the 19th century and, after recognition of their importance during WWI, they were protected by law in the allotments act of 1925 (Burchardt 2000). Today, allotment plots are normally ~250 m² and cost between £25 and £125 to rent from local councils or other land owners

(Vis *et al.* 2014). Individual councils and allotment associations set their own rules governing how the plots are used. These requirements normally state that the land is used primarily for growing produce for personal consumption, although growing flowers and keeping chickens is also commonly permitted (EA 2009d). Ownership of an allotment plot is common in the UK with more than 300,000 plots across the country (Leake *et al.* 2009) and many allotments in urban towns and cities have long waiting lists (3-4 years is not uncommon) of people wanting their own plot (Campbell and Campbell 2009).

1.2.2 Characteristics of urban garden soils

The soils of urban allotments and gardens are often diverse as a result of human interaction with the soil (Alloway 2004). Inputs into soils may come from the surrounding urban area and the historical use of the site, as well as through the management of sites to maintain fertility. In addition to sources of contamination from industrial emissions and burning of fossil fuels; allotments may become contaminated with metals and other contaminants as a result of burning of materials in bonfires, imports of unclean soil, burial of harmful materials, and applications of fertilizers and pesticides (Alloway 2004). The soils of allotment gardens are normally fertile, containing high amounts of organic matter and maintained at a neutral pH (Alloway 2004). Maintenance of allotment gardens to include high levels of organic matter may reduce the risks of exposure to PTSs in the soil, as some contaminants bind to organic matter reducing their mobility into produce (Brown *et al.* 2016).

1.3 HEALTH RISKS

1.3.1 Defining risk and uncertainty

'Health' can be defined as the "absence of disease" (Gerstman 2003) whereas risk can be defined as the chance of a negative impact occurring to something of value (Swartjes 2011). A health risk is therefore the increased chance, or likelihood, of disease. The health of urban gardeners may be at risk from exposures to contaminants from contact with soils or contaminated produce. Risks to human health can be both voluntary (e.g. smoking cigarettes) and involuntary (e.g. genetic health conditions). Involuntary risks, such as the risks presented to the health of gardeners by contaminants in soil, are usually perceived to be worse than voluntary risks, because the choice to engage with voluntary risks may be exciting and enjoyable whilst involuntary risks are by nature out of the individuals' control (Barnett and Breakwell 2001). Knowledge, and 'fear or dread', also play a role in the perception of risk such that if someone is more knowledgeable about a potential hazard, they are less likely to be as concerned by it (Barnett and Breakwell 2001).

Calculating health risks from exposure to contaminants carries a degree of uncertainty. In Environmental Science, many issues are complex as the environment is complex, so attempting to represent the environment systematically, inevitably results in reduced confidence in the result (Kriebel *et al.* 2001). Uncertainty in environmental models may be

represented numerically of the magnitude is known, but there is also considerable uncertainty in the model development process that cannot be accounted for (Kriebel *et al.* 2001).

Due to the uncertainty of predicting health risks from exposure to PTSs, the precautionary principle is often followed. This states that if an activity poses a threat to human health or the environment, preventative measures should be taken, even if cause and effect relationships are unknown or uncertain (Kriebel *et al.* 2001). The precautionary principle (PP) is used throughout risk assessment, such as in the estimation a 'safe' dose of a potentially toxic chemical (e.g. the 'no observable adverse effect level' : Section 1.3.3) and the use of 'realistic worst case' scenarios used to estimate exposure (Section 1.3.4). Along with PP, the principle of ALARP - 'as low as is reasonably practicable', is often used in risk management decisions to protect the health of the public, but also to reduce the administration of measures to remediate land where this is likely to be ineffective.

1.3.2 Risk assessment

The process of formally estimating health risks arising from exposure to PTSs was derived in the 1980s. Carrying out a risk assessment reduces expenditure on unnecessary remediation whilst simultaneously protecting public health (Cushman *et al.* 2001). In the UK, risk assessments for contaminated land may be initiated either through the planning process - as part of the development or re-development of a site,

or through local authority inspection to identify potentially contaminated areas (DEFRA 2012). Risk assessments are used to identify situations that may result in 'Significant Possibility of Significant Harm' (SPOSH) where action may be required to reduce risks to public health under the Environmental Health Act Part IIA, 1990 (DEFRA 2012).

Risk assessment structure

Risk assessments are comprised of four parts: Data collection and evaluation, toxicity assessment, exposure assessment and risk characterisation (Cushman *et al.* 2001; Troldborg 2010) (Figure 1.1). For SPOSH to occur, there must be a source of a harmful substance to which the receptor (in this thesis: a person or group of people) is exposed, via contact with the substance in the environment (pathway). The combination of source → pathway → receptor is referred to as the contaminant linkage (DEFRA 2012).

1) Data collection and evaluation

The first stage of a risk assessment involves characterising the source of the contamination. This stage will consist of a desktop study to ascertain the source, transport and fate of the chemical of concern within transport media (soils, groundwater, air, rivers etc.). Fieldwork may also be undertaken to determine the concentrations of the chemical in the environmental media which may be compared to guidance values to determine if further assessment of the site is necessary.

2) Toxicity Assessment

Human exposure to a chemical via dermal contact, ingestion or inhalation may result in different mechanisms of toxic action. Toxicity assessments aim to quantify the relationship between the dose of a chemical and toxic response(s), so that a safe level of exposure to a chemical can be determined (See Section 1.3.3). This is normally termed the reference dose (RfD) or reference concentration (RfC). The dose – response relationship may be difficult to estimate for some chemicals for which human data are not available. Uncertainty factors (usually a factor between 1 and 10) may be used to associate toxic effects seen in other organisms to humans (See Section 1.3.3).

3) Exposure Assessment

Contact with a hazardous chemical in environmental media is dependent upon human behaviour. Exposure assessment involves the estimation of the frequency, magnitude and duration of contact between humans and the hazardous chemical, in the present and the future (Troldborg 2010). Exposure assessment models combine models of the fate and transport of a chemical, with population demographics and survey data to estimate the daily intake of a chemical.

4) Risk Characterisation

The final step of the risk assessment process involves combining information on the chemical, pathway and exposure to determine if the risk from the chemical is acceptable or not. The decision as to whether the risk is acceptable is normally made by expert judgement taking into account the views of a number of stakeholders. Following risk characterisation, steps may be taken to manage or reduce the risks from the source of contamination to the public.

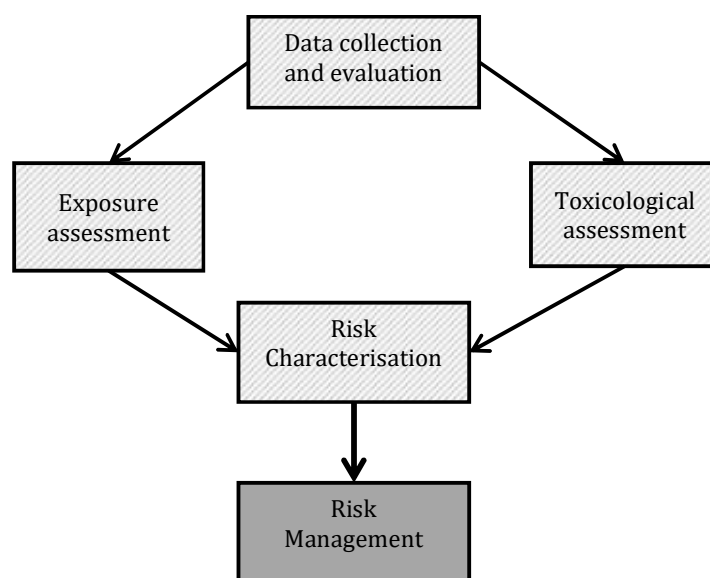


Figure 1.1: A schematic of the risk assessment process adapted from Swartjes, (2011). Boxes which are formally part of the risk assessment process are patterned not shaded.

Generic or site specific assessment

Generic or site specific risk assessment can be used by local authorities to determine if land should be considered to contaminated – requiring action to reduce, or remove, health risks from human exposure. The decision making process regarding what does or does not constitute contaminated land, often follows a tiered structure. In the UK, what does or does not constitute contaminated land can be determined using Table 1.2.

Table 1.2: UK categories for the assessment of contaminated land (DEFRA, 2012). Categories 1 or 2 concern contaminated land whilst categories 3 and 4 do not.

<u>Category</u>	<u>Explanation</u>
<u>1</u>	There is robust scientific evidence that significant harm will or has occurred to the receptor if the site continues to be used in the present form and no action is taken.
<u>2</u>	Following a detailed risk assessment, the local authority decides there is a strong case that significant harm may occur from continued use of the site in its present form.
<u>3</u>	Following a detailed risk assessment, the local authority decides there is not a strong case that significant harm may occur from continued use of the site in its present form.
<u>4</u>	There is no or minimal risk of significant harm occurring to the receptor if the site continues to be used in the present form and no action is taken.

Following an initial screening phase, if it is considered that contamination may cause significant harm but this cannot be proven unequivocally from historical events or epidemiological data (Category 1), then the first step is to compare contaminant concentrations with guidance values - termed category 4 screening levels (C4SLs), through a generic assessment. Generic assessment criteria such as C4SLs are derived from known reference doses that are unlikely to result in significant harm occurring to the receptor. If contaminant concentration(s) fall below the C4SL then the

site is considered category 4 and no further action is warranted. Sites may also be considered category 4 if the concentration of the contaminant(s) within the soil fall below background or there is no evidence of a full contaminant linkage (DEFRA 2012). Following generic assessment, site specific assessments may be used to determine if the site should be considered as category 2 or category 3. Site specific assessments require in-depth analysis of exposure pathways and modelling of the contaminant transport and fate for a specific scenario.

1.3.3 Metal toxicity and evidence for health risks

Toxicity assessment

Where available, toxicity assessments rely on data from epidemiological studies connecting exposure to PTSs (usually expressed as a dose) with human health outcomes (response). For many chemicals, data from epidemiological studies (see section 1.4.2) are not available so data from animal studies are relied upon as an approximation (See: Olson *et al.* 2000). Mechanisms of toxicity vary between substances and the health effects arising from exposure can exhibit both a threshold and non-threshold response (Figure 1.2). Traditionally in risk assessment concerning threshold chemicals, a dose which is considered safe termed the 'point of departure' (POD), was determined by identifying the dose for which no corresponding adverse health outcome was observed (defined as the 'no-observed-adverse-effect-level': NOAEL). More recently, authorities have moved away from this principle and instead adopted the

benchmark dose (BMD), which involves modelling the relationship between dose and response in comparison to a background dose (EFSA Scientific Committee 2017). The BMD is normally the dose which results in a 5 or 10% change in the toxic response relative to the background response in the unexposed population (EFSA Scientific Committee 2017). The BMD can be applied to both threshold and non-threshold chemicals, and the confidence intervals around the BMD reflect the reliability and quantity of the data from which the BMD is derived (Figure 1.2).

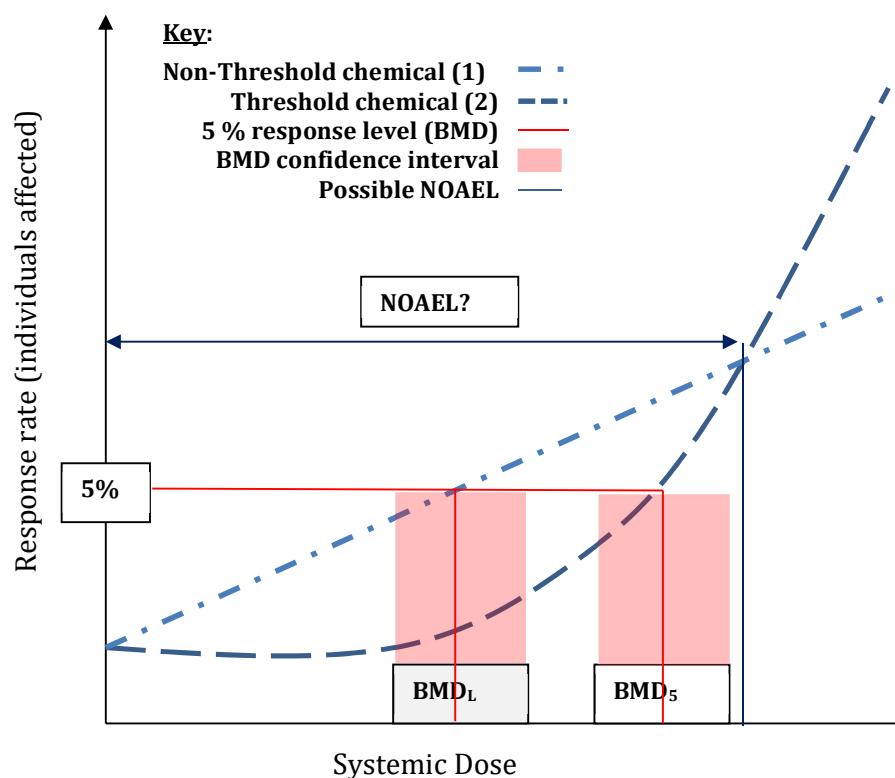


Figure 1.2: An illustration of the relationship between dose and response exhibited by threshold and non-threshold chemicals and the calculation of the benchmark dose (BMD) (here a 5% observed response rate) and no observed adverse effect level (NOAEL). The lower 95% confidence interval of the BMD; the BMDL₅, is often used in preference to the BMD₅ where there is uncertainty over the 5% observed response rate in laboratory studies.

Heavy metals (HMs)

There is a reasonably large body of evidence concerning the toxicity of the HMs covered in this thesis. Heavy metals are referred to as such because of their atomic mass and density; however, this is not true for all elements sometimes associated with this definition including Cu, Ni and Zn (Nieboer and Richardson 1980). Often HMs are present in the environment in small concentrations and some elements in this definition may also be referred to as 'trace elements'. The elements included in this thesis are arsenic (As), cadmium (Cd), copper (Cu), chromium (Cr), nickel (Ni), lead (Pb) and zinc (Zn). These elements are all added to the urban environment through anthropogenic activity, which may lead to higher concentrations of these elements in urban areas than elsewhere.

Arsenic:

Arsenic (As) has been associated with the word 'poison' for centuries but has been consumed voluntarily by several communities in Austria, where consumption of small amounts of inorganic arsenic was linked with fertility and courage (Przygoda *et al.* 2001). Acute exposures can lead to vomiting, diarrhoea, depression and death in extreme cases (National Research Council 2006) whilst long term exposures are associated with cancer Chronic ingestion has been observed to result in cancer of the liver and bladder, leading to the classification of inorganic arsenic as a 'known carcinogen' (ATSDR 2005b). Sources of As in soil include from weathering

of As-rich rock and as a by-product of copper and lead smelting. Arsenic compounds are also used in the manufacture of wood preservatives, glass and enamels and they are a component of phosphate fertilizers, herbicides and insecticides (EA 2009a). The most likely source of intake of inorganic As in food is from soil derivatives (ATSDR 2005b).

Cadmium:

Cadmium (Cd) is a silvery-white solid at room temperature and has been described as “one of the most dangerous elements in the food and environment of man” (Vos *et al.* 1987; Cited in Hough, 2002). Exposure to Cd resulted in many fatalities and injuries in a mass-poisoning event in the Jinsu river basin, Japan, which left many with ‘Itai-Itai’ (‘ouch-ouch’) disease (Inaba *et al.* 2005). Cadmium is very bioavailable in soils and can accumulate in body tissues, especially the kidney where it may reside for up to 14 years (EA 2009f). In the Kidney, Cd substitutes for Zn resulting in the impairment of renal function and renal metabolism of vitamin D, leading to osteoporosis (bone fragility) and osteomalacia (bone softening) (EA 2009f). Whilst cadmium has been linked with lung cancer in animals, it has not been conclusively linked with cancer in humans, leading to the suggestion that it should be deemed ‘probably carcinogenic to humans’ (Sato *et al.* 2002). Cadmium may enter the environment as a by-product of mining and smelting activities and is used in Cd-Ni batteries, and the coatings of heat resistant plastics, ceramics and enamels (National Research Council 2006). Cadmium is normally present in soils as the Cd²⁺ ion (EA 2009f).

Chromium:

Chromium can take any valence between Cr (II) and Cr (VI) but most commonly occurs in the environment as chromite - Cr (III), or chromate, Cr (VI) (Alloway and Ayres 1997). Chromite is a micronutrient; however chromate is the more toxic of the two forms of chromium, and is a known carcinogen (ATSDR 2008). Chronic inhalation of chromate may also lead to irritation to the respiratory tract and ingestion may result in the development of stomach ulcers and anaemia (ATSDR 2008). Contamination of soils with Cr compounds, mainly occurs through waste disposal, such as wastes from textile industries, electroplating and sewage sludge (Alloway and Ayres, 1997). Chromium is also released into the environment as a result of fossil fuel combustion (ATSDR 2008).

Copper:

Copper (Cu) can be found naturally in the environment and may be released into the environment through natural activities including volcanic eruptions as well as through anthropogenic use (ATSDR 2004). It is a reddish brown metal and is normally present in an elemental form as the monovalent (Cu^+) or divalent (Cu^{2+}) cation (ATSDR 2004). Chronic exposure to Cu may result in toxicity to the liver, brain and other organs; leading to symptoms including dizziness, headaches and nausea (ATSDR 2004). However, human toxicity is rare as the human body is able to regulate uptake and excretion except when exposed to very high

concentrations (Gaetke and Chow 2003). Copper is essential to enzymatic activities in humans and plants, and deficiency of copper has also been documented (Alloway and Ayres 1997; Gaetke and Chow 2003). Copper is widely used in industrial processes and can also enter the environment through waste disposal, combustion of fossil fuels, fertilizer production and mining and smelting activities (ATSDR 2004). The use of Cu in pipes may increase concentrations in drinking water, but this is not normally considered hazardous as long as a neutral pH (6.5-8) is maintained (Alloway and Ayres 1997).

Lead:

Lead (Pb) is a blue-gray solid at room temperatures. Lead toxicity can arise as a result of competition between Pb and calcium (Ca) for uptake in the body; especially in binding to proteins (Needleman 2004). It is mainly associated with neurological defects in children who are especially vulnerable, as the neurological effects of lead have been shown to be greater on the developing brain (Canfield *et al.* 2003). The effects of Pb on the brain exhibit a non-threshold response among children which means even low doses (equating to $< 10\mu\text{g Pb dL}^{-1}$ blood) may result in a reduction of a child's Intelligence Quotient (IQ) score (Canfield *et al.* 2003; ATSDR 2007). Among adults, chronic exposures to Pb can cause cardiovascular disease and kidney diseases, as well as neurobehavioral effects such as headaches, fatigue and irritability (ATSDR 2007). The main sources of lead pollution in the environment are derived from petrol and fossil fuel combustion (including the historic use of Pb in petrols in

developed countries), paint flakes (from older paints), solders and varnishes, lead pipes, pesticides, lead shot and pollution from mining and smelting activities (Alloway and Ayres 1997).

Nickel:

Nickel (Ni) is the 24th most abundant element in the earth's crust with an atomic mass of 58.7 units. Nickel toxicity can occur through contact with the skin as well as through ingestion and inhalation. It is estimated that 1-4% of men and 8-20% of women are susceptible to skin rashes and dermatitis as a result of dermal contact with nickel (EA 2009e). Oral ingestion can also result in the formation of skin defects in sensitized individuals (EA 2009e). Nickel is classed as a carcinogen (ATSDR 2005a). Almost all forms of intake have been shown to result in tumours developing in experimental animals after high levels of exposure (Denkhaus and Salnikow 2002) and carcinogenic responses have been observed in humans following inhalation of nickel sub-sulphide and nickel oxide (ATSDR 2005a). However, pure elemental nickel is not thought to be carcinogenic, only Ni-compounds (ATSDR 2005a). Nickel is used as a component of batteries and in metal alloys, primarily in the production of stainless steel (ATSDR 2005a).

Zinc:

Zinc is one of the most common elements in the earth's crust and is a bluish-white metal at room temperature (ATSDR 2003). It is an essential

element to humans and plants, but may become toxic in high concentrations. Intakes of Zn above the recommended daily allowance (RDA) of 100-300 mg have been observed to result in toxic effects including nausea, vomiting and fatigue (Fosmire 1990). Deficiency may also lead to ill-health effects such as decreased immune function and loss of taste and smell (ATSDR 2003). Zinc deficiency may increase the toxic effects of both Cd and Pb which can replace Zn in the body (Goyer 1997). Pollution of the environment with Zn may occur as a result of mining, purification of Zn, Pb and Cd ores, steel production and combustion of fossil fuels and waste materials (ATSDR 2003).

Other metal(loid)s:

Mercury (Hg) is also often considered as a heavy metal (Nieboer and Richardson 1980) while organic, methyl-mercury (CH_3Hg^+) is associated with food poisoning from seafood. The bioaccumulation of CH_3Hg^+ in fish after it was released in industrial waste, led to the mass poisoning of citizens in the Minimata prefecture, Japan (Kurland *et al.* 1961). Inorganic Hg is less toxic; however, all forms have the potential to cause stomach upset, changes in heart rate/blood pressure and kidney damage (ATSDR 1999).

Tolerable intakes

Establishing a relationship between the dose of a chemical and a toxic response is complex. Exposure to chemicals normally exhibits more than

one toxic response in humans and responses may vary between different population groups. To establish a Health Criteria Value (HCV) that consists of a daily or weekly dose likely to cause minimal risk to humans, the POD (see: toxicity assessment, section 1.3.3) for the different toxic effects of a chemical are reviewed (EA 2009d). The HCV is commonly the POD/UF where the UF is the Uncertainty Factor or Factors (EA 2009d). Uncertainty Factors are used to account for intra- and inter- species variation between human-human and animal-human studies respectively. For potentially toxic substances (PTSs) for which the relationship between dose and response exhibits a threshold, UFs are typically $10 \times 10 = 100$ (10 each to account for intra- and inter- species variation: DEFRA 2013a). For PTSs for which there is no threshold between dose and response - including carcinogens or genotoxins - an UF of 10,000 from animal to human data is used to account for the extra human variability in susceptibility to DNA alterations (DEFRA 2013a). In the derivation of C4SLs, a further term, LLTC (low level of toxicological concern), was introduced. LLTCs represent a daily dose which would give rise to a low level of concern of health risks (DEFRA 2013a). They are less protective of health than a HCV, but a daily dose equivalent to an LLTC ($\mu\text{g kg BW}^{-1} \text{ day}^{-1}$) is still unlikely to result in adverse health effects.

To establish a POD using human data, biomarkers of exposure measured in blood or urine are often used. For example, the LLTC for Cd, is derived by examining the relationship between beta-2 microglobulin – a biomarker for the incidence of renal failure, with creatinine-adjusted

concentrations of Cd in urine (EFSA 2011; DEFRA 2013b). Once an LLTC has been derived, backwards exposure modelling can be used to determine a corresponding concentration of the contaminant in soil (C4SLs) for a specific scenario (see section 1.3.4). Since their introduction in 2013, C4SLs have been defined for exposures to As, Cd, Cr (VI) and Pb (Table 1.3).

Table 1.3: LLTCs and C4SLs derived for the daily dose as a result of oral ingestion, and the corresponding concentrations in soils of allotments. LLTCs and C4SLs are given for As, Cd, Cr(VI) and Pb (data collated from DEFRA, 2013a-e).

Metal(loid)	LLTC_(oral) ($\mu\text{g kg BW}^{-1} \text{ day}^{-1}$)	C4SL_(allot.) (mg kg soil^{-1})	Derivation of LLTC
As	0.30	49	Epidemiological data relating As dose from contaminated drinking water to the occurrence of lung and bladder cancers.
Cd	0.54	4.9	The presence of beta 2-microglobulin in urine used as a biomarker for renal failure.
Cr (VI)	0.44	170	Small intestinal tumours in male mice.
Pb	2.1	84	A review of neurobehavioral effects in children; cardiovascular effects and renal toxicity among adults.

1.3.4 Calculating exposure and risk

Routes of Exposure

Human exposure to a chemical in soil may occur over a combination of different pathways. The CLEA model – the most widely used exposure model in the UK, lists several pathways that are relevant to an allotment use. These include: direct ingestion of soil, inhalation of soil dusts and

vapours, skin contact with soil, ingestion of contaminated produce and indirect ingestion of soils adhered to produce (EA 2009d). Where contact to a chemical through these exposure pathways exhibits a common mode of toxicity, the standard approach is to sum the dose attributable to each pathway together (Equation 1.1 - redrawn from Equation 2.1, EA 2009d) and compare the average daily exposure with a HCV or LLTC.

$$ADE = \frac{(IR_{ing} \cdot ED_{ing} \cdot EF_{ing})}{BW \cdot AT} + \frac{(IR_{inh} \cdot ED_{inh} \cdot EF_{inh})}{BW \cdot AT} + \frac{(IR_{der} \cdot ED_{der} \cdot EF_{der})}{BW \cdot AT} \quad 1.1$$

Where: ADE is the average daily exposure in mg kg BW⁻¹ day⁻¹, IR is the intake rate attributable to ingestion (_{ing}), inhalation (_{inh}), or skin contact (_{der}) in mg day⁻¹, ED is the exposure duration in years, EF the exposure frequency in days year⁻¹, BW is the bodyweight in kg, and AT is the 'averaging time' which is normally equivalent to the exposure duration expressed in days.

For chemicals for which a common mode of toxicity is not expressed, the average daily exposure through ingestion, inhalation or dermal exposures may be compared to separate HCVs or LLTCs. Calculating individual exposure pathways can be complex and requires knowledge of the transport and fate of a chemical in the soil and other media as well as knowledge of human activities and routines (e.g. Fryer *et al.* 2006). The basic calculations involved in estimating the intake rate via two pathways relevant to allotment gardeners are explained overleaf.

1) Consumption of contaminated produce

Consumption of contaminated produce is the primary route of exposure for allotment gardeners in the UK to Cd, As and Pb (DEFRA 2013a - results from sensitivity analysis using CLEA model). Exposure via this route depends on the volume of produce grown and consumed by the individual, and the transfer of the chemical from the soil into produce. Heavy metals are often sorbed onto soil constituents which can prevent transfer into plant roots and translocation around the plant. Exposure models covering this pathway, aim to predict this transfer and relate the concentration of chemicals in soils to the concentration in produce, often using a transfer or concentration factor (CF) (Chapter 3). In the CLEA model, the intake rate from consumption of contaminated produce is calculated using Equation 1.2 (adapted from equation 7.16: EA 2009d).

$$IR = \sum_{x1 \dots xn} \{ C_s \cdot CF_x (DC_x \cdot CR_x) HF_x \} \quad 1.2$$

Where: IR is the intake rate from produce consumption (mg day^{-1}), C_s is the concentration of the chemical in soil ($\text{mg g}^{-1} \text{ dw}$), and for each produce group $x1$ to xn ; CF is the concentration factor ($\text{mg g}^{-1} \text{ dw}$), DC is the dry to fresh weight conversion factor (gdw gfw^{-1}), CR is the consumption rate (g fw) and HF is the homegrown fraction (dimensionless).

2) Direct and indirect soil ingestion

Direct ingestion of non-foodstuff ('pica') is common among children who are often considered to be at most risk from chemicals in the soil, due to their comparatively low body weight (EA 2009d). This is considered to be

negligible in the CLEA model for adults (EA 2009d). However, children and adults may ingest small, but significant, quantities of soil adhered to home-grown produce. The amount of soil present on home-grown produce can depend on the type of produce (e.g. is it grown above or below ground?), and the way the produce is prepared (is it washed or peeled?) prior to consumption. Intake rates from direct, and indirect, soil consumption are calculated by the CLEA model according to Equations 1.3 and 1.4 respectively (adapted from equations 6.1 and 6.2: EA 2009d) (adapted from Equations 6.1 and 6.2 in EA, 2009).

$$IR = C_s \cdot SIR \quad 1.3$$

$$IR = \sum_{x1 \dots xn} \{ C_s \cdot CF_x \cdot SL_x \cdot PF_x (DC_x \cdot CR_x) \cdot HF_x \} \quad 1.4$$

Where: IR is the intake rate from produce consumption (mg day⁻¹), SIR is the direct soil ingestion rate (g day⁻¹), C_s is the concentration of the chemical in soil (mg g⁻¹ dw), and for each produce group x1 to xn; CF is the concentration factor (mg g⁻¹ dw), DC is the dry to fresh weight conversion factor (gdw gfw⁻¹), CR is the consumption rate (g fw), SL is the soil loading factor and HF is the homegrown fraction (dimensionless).

Calculating risk and CASLs

To determine whether an individual (or group of individuals) is at risk from exposure to a chemical, their estimated average daily exposure (ADE) can be compared to a HCV or LLTC. The ratio between the ADE and a HCV or LLTC for a specific chemical is called the Hazard Quotient (HQ) (EA 2009d). For all chemicals with the same mode of action, the Hazard

Index (HI) – the summation of HQs for each chemical, can be calculated (Equation 1.5 adapted from Equations 2.6 and 2.7 in EA 2009d).

$$HI = \sum_{i=1}^n HQ_i \quad \text{Where: } HQ = ADE / (HCV \text{ or } LLTC) \quad 1.5$$

If the HI is at or below 1, this suggests that harm is unlikely to occur to the receptor as the HCV/LLTC is indicative of minimal risk. A HI >1 indicates the receptor may be at harm from exposure to this chemical; however the likelihood of harm occurring to the receptor will depend on the dose-response curve (Figure 1.2). To determine the HI or HQ, the CLEA model is run in forwards mode (EA 2009d).

Category 4 screening levels replaced soil guideline values (SGVs) in 2013 after a review of the CLEA model and derivation and use of SGVs (DEFRA 2013a). C4SLs represent concentrations of PTSs in soils that are highly unlikely to result in harm to human health and are used to eliminate the possibility that land could be considered to be ‘contaminated’. The main difference between C4SLs and SGVs is the use of the LLTC in the derivation of a C4SL, rather than the HCV used to determine the replaced SGVs. To determine a C4SL or SGV, the CLEA model is run in backwards mode with a HI/HQ of 1 (EA 2009d). The output is a soil concentration from which exposure would be unlikely to result in harm. C4SLs are determined for different land use scenarios and different chemicals. A

‘realistic worst case scenario’ is used to define exposure to each chemical so that the majority of the population are protected (EA 2009d).

Limitations of the CLEA model addressed by this thesis

In (DEFRA 2013a), a number of limitations of the CLEA model were described. In particular, it was acknowledged that the estimation of exposure for allotments and residential gardens is sensitive to the parameter ‘CF’ (Concentration Factor) (Equation 1.2) - linking the concentration of chemicals in soil to plants. Methods of modelling plant uptake and estimating ‘CFs’ are reviewed in chapters 3 and 4 of this thesis and improvements are suggested. Further uncertainty was acknowledged in the estimation of the home-grown fraction (HF) and consumption rate (CR) (Equation 1.2 and 1.4) (DEFRA 2013a). These parameters are reviewed in a survey of gardener’s routines in chapter 5. Finally, the CLEA model has adopted a deterministic rather than a probabilistic approach which means uncertainty in the model is not represented. A probabilistic approach using a Bayesian network model, is presented and discussed in chapter 6 of this thesis.

1.4 HEALTH BENEFITS

1.4.1 Defining health benefits

To compare health benefits and health risks from urban gardening, they must be presented on the same scale. A health benefit is therefore defined as something that is protective against the risk of disease and ill-health and the opposite of a health risk (section 1.3). This definition is appropriate because in many cases good health, including good dietary health, may actively reduce the adverse health effects from exposure to HMs (Peraza *et al.* 1998; Lall, S.B. *et al.* 1999). Micronutrients, including Zn, Fe and Se which may be enriched alongside other metals in urban soils, as well as vitamins A, D and E which can be provided by fruit and vegetables, help to reduce the toxic effect of HMs, through competition at binding sites and enhancing mechanisms that remove toxic HMs from the body (Figure 1.3) (Peraza *et al.* 1998; Lall, S.B. *et al.* 1999).

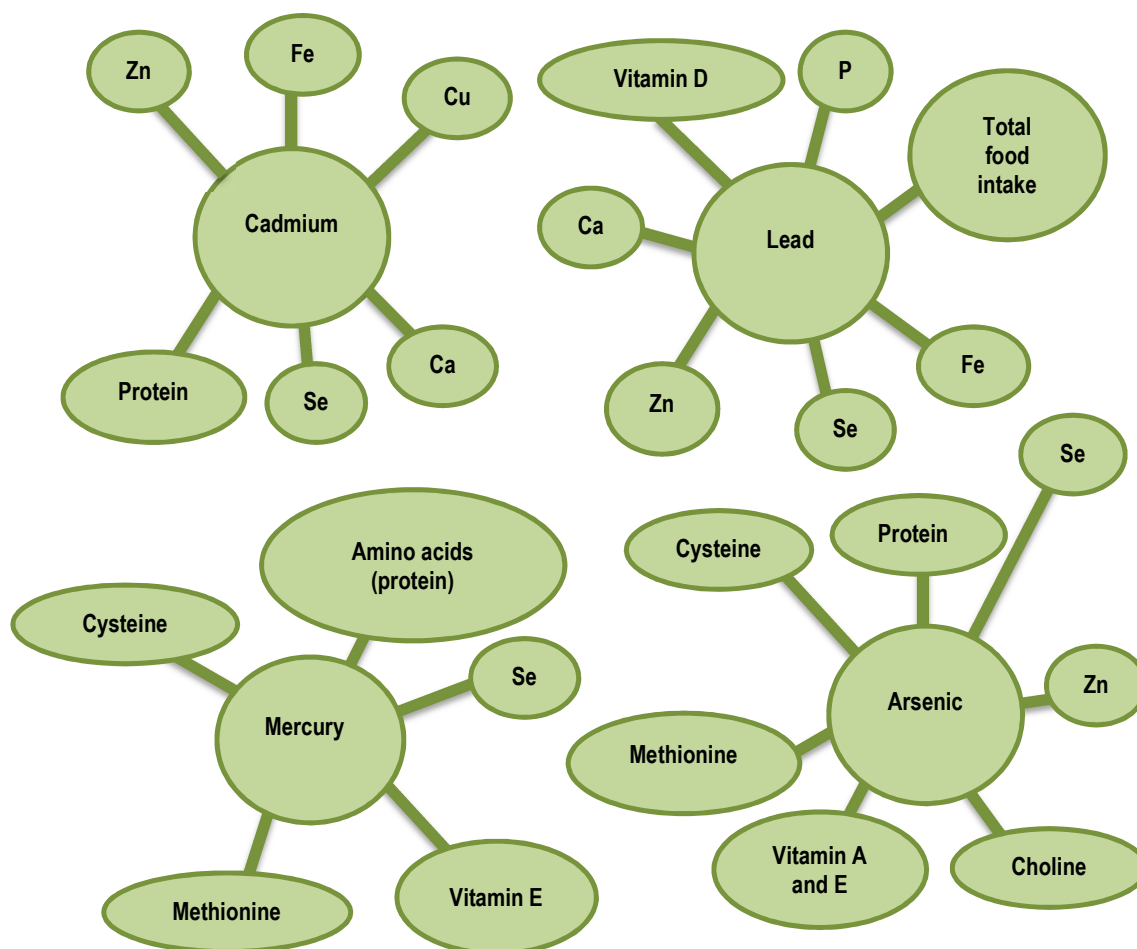


Figure 1.3: The protective interaction of micronutrients and vitamins (outer bubbles) with the toxicity of As, Cd, Hg and Pb (inner bubbles). Adapted from Peraza et al (1998).

1.4.2 Evidence for health benefits

Taking part in gardening and GYO activities appears to be related to a healthy lifestyle that also promotes overall physiological, psychological and dietary health (Leake *et al.* 2009). The effects of having a healthy lifestyle including the consumption of fruit and vegetables, helps protect against diseases, including cardiovascular diseases and cancers which have a large overall burden on human health and well-being (Steinmetz

and Potter 1996; Kruk 2007). There are few published comparisons between gardening and disease (Buck 2016); so to quantify the benefit of gardening for health both the influence of gardening and GYO activities on the lifestyle of gardeners, and the relationship between lifestyle (e.g. 'the amount of exercise achieved in one day) and disease, has to be examined. The relationship between gardening and lifestyle is studied in chapter 5 using a questionnaire survey of allotment gardeners, backed up by literature studies; whilst the relationship between lifestyle and disease is quantified through the use of epidemiological studies and associated metrics.

Epidemiological studies

Epidemiology is the study of "the causes, transmission, incidence and prevalence of health and disease in human populations" (Gerstman 2003). Epidemiological studies can be sub-divided into experimental studies – the researcher controls exposure in some way, or observational studies – the researcher does not intervene (Gerstman 2003). The studies in this thesis used to determine health risks from metal toxicity, and health benefits from lifestyle choices, are observational. Most of the studies are also longitudinal – involving prospective or retrospective observation of a group of people through a period of time.

There are two types of longitudinal study; case-control studies (retrospective) and cohort studies (usually prospective):

- In a cohort study, sub-groups of the study population are identified that are/ are not exposed to a suspected effector of disease (e.g. smokers and the risk of lung cancer: Freedman *et al.* 2005). The number of individuals that in the exposed and unexposed populations that contract the disease are then compared.
- In a case-control study, 'cases' – individuals who have the disease within the study population, are selected by the investigator and compared with individuals - 'controls', that do not have the disease (e.g. tobacco use and cardiovascular disease: Teo *et al.* 2006). The frequency of exposure to the suspected effector of the disease is compared between cases and controls. Case – control studies tend to be preferable to cohort studies when the disease is uncommon in the study population (Gerstman 2003).

Quantifying the risk of disease

It is important that there is a clear definition of a 'case' in epidemiological studies (Coggon *et al.* 2003) as evidence of disease may range from biomarkers in blood or urine to chronic illnesses and death. The simplest metric to quantify disease is to count the number of new cases over a time period, or the number of cases within a population at a given time

(Gerstman 2003). From this, the disease incidence (count of new cases/time) or disease prevalence (proportion of the population who are cases) can be calculated (Coggon *et al.* 2003). To make comparisons between exposed or unexposed groups, more complex metrics are required. Relative risk (RR), hazard (HR) and odds ratios (ORs) allow researchers to predict the likelihood of disease in exposed populations compared to unexposed populations (Figure 1.4). For each metric, a value of 1 can be interpreted as there being no effect of exposure on the disease, whilst a value of <1 indicates that exposure has a protective effect, and a value of >1 indicates that exposure has a harmful effect. A high risk, hazard, or odds ratio accompanied by reasonable statistical certainty, may be used as evidence that the exposed population are being harmed through their exposure.

Confounding and bias

Most epidemiological studies have to control for confounding factors and bias in the analysis (Coggon *et al.* 2003). Confounding can arise due to differences between genders, race, age groups, or any other factor, that is not the subject of the epidemiological study but has an influence on both exposure and outcome (Kamangar 2012). The effect of confounding factors in any analysis can be restricted through randomization, matching like-for-like groups of cases and controls, selection of sub-groups of the population unaffected by the confounding factor (e.g. non-smokers), and by accounting for confounding factors in the statistical analysis (Kamangar 2012).

Calculating Odds, Risk and Hazard Ratio's

Disease?	Yes	No
Exposed counts:	A	B
Unexposed counts:	C	D

Odds ratio (OR)

- Calculated for case-control studies.
- The odds of an event (e.g. cancer) occurring in the exposed (e.g. to cigarette smoke) group compared to the odds in the non-exposed group over the period of study
- $OR = (a+b)/(c+d)$

Relative Risk (RR)

- Calculated for cohort studies.
- The ratio between the **cumulative** prevalence of disease in the exposed vs unexposed group over the study period.
- $RR = [a/(a+b)]/[c/(c+d)]$

Hazard Ratio (HR)

- Calculated for cohort studies.
- The **average** ratio between the prevalence of disease (e.g. cancer) in the exposed vs unexposed group throughout the study period.
- $HR = [a/(a+b)]/[c/(c+d)]$ between t_0 and t_1 .

Comparing OR, HR and RR

- The OR and RR is similar when the likelihood of disease is small but ORs are more extreme when the likelihood of disease is high. ORs can be converted to RRs if the baseline risk is known (Grant, 2014).
- The $RR \approx HR$ when the time period over which RRs and HRs are estimated is similar. However, as the RR is cumulative, the RR will approach 1 over a lifetime, so for longer time periods it is not appropriate to equate RR and HR (Stare and Maucourt-Boulch, 2017).

Figure 1.4: Relating the odds ratio (OR), the relative risk (RR) and the hazard ratio (HR) from epidemiological studies.

1.4.3 Combining benefits and risks

The primary aim of this thesis was to derive a way to compare the health benefits and health risks of urban gardening. Three methods were considered to combine benefits and risks: the combination of risk ratios (RRs, HRs or ORs) describing risks and benefits, the use of the disability-adjusted life years metric (DALYs), and the use of expert elicitation.

Combining risk ratios

It is common for risk ratios to be combined in the literature through the use of meta-analyses. Meta-analyses, when conducted properly allow scientists to combine epidemiological evidence concerning a common theme (e.g. the relationship between fruit and vegetable consumption and Coronary Heart Disease (CHD) - Dauchet *et al.* 2006). The combination of a number of different studies helps to reduce bias in the risk ratios and gives the combined result a reduced level of uncertainty (Haidich 2010). The difficulty with using a meta-analysis for combining health benefits and risks from gardening is that often the cause and outcome of exposure are different. Martin *et al.* (2008) introduced a method for the construction of an odds model of CHD using odds ratios which is used as a framework for the combination of risk ratios in chapter 6. There are a number of challenges associated with using this approach. Firstly, it is unclear how risk ratios should be combined: summation may under predict the risk, whilst multiplication may over predict risks, from multiple exposures. Secondly, risk ratios may be presented for different

levels of exposure but do not give any indication of the severity of a disease to the human population.

Disability-adjusted life years (DALYs)

Leake *et al.* (2009) suggested that disability-adjusted life years (DALYs) could be used to combine the health benefits and risks attributable to urban gardening. DALYs can be used to estimate the burden of disease within a population and estimate the difference between ideal health and reality (WHO 2017b). One DALY is equivalent to the number of years of life lost (YLL) due to disease, plus the number of years estimated to be affected through disability (WHO 2017b) (Equation 1.6).

$$\text{DALY} = \text{YLL} + \text{YLD} \quad 1.6$$

The number of years of life lost (YLL) are calculated using the number of deaths in the population (N) and the standard life expectancy (L) (Equation 1.7). The estimated years of life lost through disability (YLD) is calculated using the number of incident cases (I) or the prevalence of the disease within the population (P), the disability weight (DW) and the duration of exposure (L) (not required if P used) (Equation 1.8).

$$\text{YLL} = N \times L \quad 1.7$$

$$\text{YLD} = I \times \text{DW} \times L \quad \text{or} \quad \text{YLD} = P \times \text{DW} \quad 1.8$$

Disability Weights (DWs) were initially derived through expert judgement (Murray 1994) but have since been thoroughly reviewed using global surveys of the general population (Salomon *et al.* 2013).

The use of DALYs would allow for comparison between the severity of different health outcomes, which risk ratios and HIs do not, and quantifies the relationship between exposure and the severity of disease. However, the main disadvantage with the use of DALYs for estimating the health benefits and risks of gardening, is that DWs have not generally been established for exposure to HMs. Whilst exposures to HMs may lead to health outcomes which can be assigned DWs, some health outcomes of exposure to HMs included in risk assessment are very specific (e.g. The presence of beta 2-microglobulin in urine – Cd) or very broad (lung and bladder cancers – As) making this process difficult (Lokuge *et al.* 2004). DALYs have been calculated to estimate the global burden of disease attributable to Pb (Fewtrell *et al.* 2004) and the effect of interventions on the burden of disease attributable to As in drinking water in Bangladesh (Lokuge *et al.* 2004) (the latter was only able to calculate YLL and not YLD because of difficulties obtaining disability weights for arsenic toxicity).

Expert knowledge elicitation

In risk assessment, models are often fraught with uncertainty and assessments often have to cope with an absence of measured data (Kandlikar *et al.* 2007). Consulting experts is one approach to reducing uncertainty when there are significant knowledge gaps in either the

design, or parameters, of a model (Knol *et al.* 2010). Experts do not have to be professionals in a particular field: the term can also refer to the general public and other stakeholders involved in a particular problem (Knol *et al.* 2010).

There are several opportunities in the assessment of the health benefits and risks of gardening for experts to be involved. Responses from experts regarding the perception of risk from exposures to HMs or healthy lifestyles, could help to make a judgement on how each should be regarded in risk assessment models. The knowledge of health professionals could be used to make a judgement over the severity of benefits and risks, and the conceptualisation of a benefits-risks model could also be informed by experts. As an emerging field, expert knowledge elicitation has been criticised (Bolger and Wright 2017). However, in many circumstances the use of expert elicitation may help to prevent over-reliance on empirical evidence which may be inappropriate for risk assessment purposes (Kandlikar *et al.* 2007; Knol *et al.* 2010).

2 THE SOIL PROPERTIES OF URBAN ALLOTMENTS

2.1 INTRODUCTION

Human interaction with soils, including through the addition of a wide range of materials, can change the soil's biological, physical and chemical properties (Linde *et al.* 2001; Markiewicz-Patkowska *et al.* 2005; Bretzel and Calderisi 2006). Heavy metals (or metalloids) (HMs) such as Cd and Pb, are released into the local environment from industries, the combustion of fossil fuels, and from waste disposal (Alloway and Ayres 1997). In urban gardens and allotments, HMs are also added to the soil through the application of fertilizers and pesticides, the burning and burial of metal-containing materials, imported soils, the weathering of metal ornaments, and recreational activities (Alloway 2004). Heavy metals are persistent in soil, and can accumulate in allotment gardens to concentrations which could pose a threat to human health through exposure to contaminated soils and produce (Mulloli *et al.* 2004). Because of this, HM concentrations have been previously studied in the soils of allotments and gardens across the UK (Davies 1978; Moir and Thornton 1988; Hursthouse *et al.* 2004; Nathanail *et al.* 2004; Giusti 2011) and elsewhere (Anglin-Brown *et al.* 1995; Hursthouse *et al.* 2004; Szolnoki *et al.* 2013).

Soil properties (such as clay content and soil organic matter: SOM) affect the mobility, and subsequent human exposure, of HMs. In common with other urban soils, the properties of urban gardens and allotments tend to

be very spatially diverse (Alloway 2004). However, the management of urban garden and allotment soils may result in soils containing higher concentrations of soil organic matter (SOM), oxides and clay minerals than the surrounding area (Alloway 2004). Soils containing higher proportions of soil colloids such as humus, metal oxides and clays, are likely to contain lower proportions of metals in an exchangeable form, as metals typically diffuse further into the colloidal structures contained in soils over time (Degryse *et al.* 2009a). Urban garden and allotment soils also normally have a pH value between 5 and 8, in contrast to other urban soils which can be strongly acidic or strongly alkaline (Alloway 2004). Alkalisiation of urban allotment and garden soils resulting from surface run-off coming into contact with concrete buildings and roads (Messenger, 1986), may increase binding of HM cations to soil colloids. Heavy metals bound to soil colloids are less likely to accumulate in edible plant tissues (Reichenberg and Mayer 2006; Degryse *et al.* 2009a; McLaughlin, M.J. *et al.* 2011), reducing exposures from the consumption of contaminated produce.

The methods that individual plot holders use to cultivate their soils also have an effect on the mobility of HMs in the soil. Gardens and allotment soils usually contain high concentrations of plant nutrients (Alloway 2004). The addition of metal cations such as magnesium (Mg) and potassium (K) in fertilizers, can also result in competition for binding sites and release HM cations into the soil solution (Noraho and Gaur 1995). Phytoavailability may be reduced through the binding of metals to

phosphorus (P) compounds (Bolan and Duraisamy 2003). The addition of organic matter in composts and liming to raise the soil pH, also reduces metal mobility in the soil (McLaughlin *et al.* 1997).

In this chapter, the results from a survey of allotment soils collected across the urban city of Nottingham are presented and discussed. This was the first extensive survey of HM concentrations in allotment gardens in Nottingham to the authors' knowledge, and one of a small number of studies to investigate the influence of soil management practices on HM concentrations in allotment gardens. The aims of the survey were as follows:

1. To determine HM concentrations and soil properties of urban allotments that could be used in the development of a health benefits vs health risks model (Chapter 3).
2. To investigate differences in metal concentrations and soil properties within and between allotment sites.
3. To investigate the influence of soil properties and agronomic management on HM concentrations.

2.2 MATERIALS AND METHODS

2.2.1 Sample location

The city of Nottingham has a rich industrial history and was known for textile manufacturing and coal production industries, although other industries including brick and iron works, also existed across the city. The city has had a large number of growing spaces ever since the 1830s (Burchardt 2002) including the St Ann's allotments; a site covering 30 hectares on which plots were reportedly first rented in 1604 (BBC 2009). Today, the city of Nottingham contains over 40 allotment sites serving a population of around 300,000 (Nottingham City Council 2017; Nottinghamshire County Council 2017).

Heavy metal concentrations and soil properties of allotment soils were determined from samples collected from the individual plots of 9 allotment sites (Blenhiem -BH, Canal -CN, Mapperly -MP, Moorfields -MR, Old Park Farm -PF, Sneinton Dale -SN, St Ann's -ST, Windmill -WM and Woodthorpe -WT) (Figure 2.1). Plots are rented from Nottingham City Council (NCC) or allotment associations (on behalf of the NCC), by individual tenants. St Ann's (ST) was the largest site containing ~550 individual plots. This compares to some of the smaller sites, WT and MP, both of which contained fewer than 30 plots (Table 2.1). At several of the allotment sites (SN, ST, WM), plots were separated with large hedgerows. At others, plots were separated by smaller hedgerows or fences (BH, CN, MR, PF) or by boundaries marked by stakes in the ground (MP and WT).

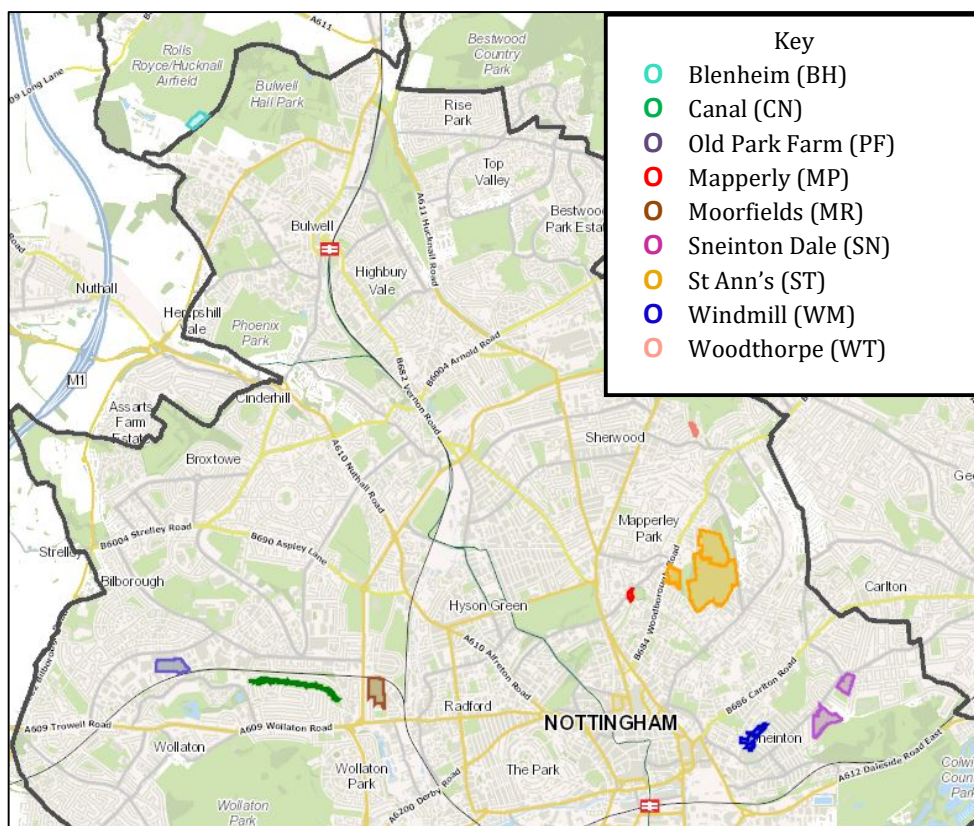


Figure 2.1: Location of sampled allotments across Nottingham city.

Table 2.1: Description of the size and number of plots and sampling regime at each allotment site. For some plots (number indicated in parenthesis) only topsoils were sampled (see section 2.2.4)

Allotment	Total number of plots	Number of plots sampled	Date of sampling
Blenheim (BH)	~120	20	Sept – Nov 2015
Canal (CN)	~200	21	Sept – Nov 2015
Mapperly (MP)	~20	9	Oct 2015
Moorfields (MR)	~250	12	Sept – Nov 2015
Old Park Farm (PF)	~90	14	March– June 2015
Sneinton Dale (SN)	~140	7(5)	Sept 2014
St Ann's (ST)	~550	50(3)	Sept 2014 and June – Sept 2015
Windmill (WM)	~100	6	Sept – Nov 2015
Woodthorpe (WT)	~25	7(7)	Sept 2014
Total	~1500	146	

2.2.2 Permissions and administration

Initial contact was made with the city council through telephone conversations and a meeting between the researcher and the 'parks and spaces' team which oversaw the running of the allotment sites. The study was subject to ethical approval which was granted on 5th September 2014 by the School of Biosciences, University Ethics Committee (Reference: SB1415/03 PGR- Stubberfield (Young)). The process of contacting the NCC, allotment associations and ethical approval is documented in Appendix I.

Permission to carry out the study and to take soil samples was granted by Nottingham City Council 'Parks and Open spaces team' in mid - September 2014. Allotment associations are responsible for day-to-day maintenance and annual rent collections, at 4 of the 9 allotment sites (PF, SN, ST and WT), and were contacted by the researcher using contact details provided by the NCC. Contact with plot holders was made through:

- (a) Written communication with plot holders
- (b) Advertisement at allotment events such as open days

A) Written communication

Postal invitations were sent to plot holders explaining the aims and objectives of the study. Participants indicated a preferred day/time for sampling to take place and provided their contact details. Participants were asked to return consent forms on the day.

B) Allotment events

In addition to written communication, Allotment events such as open days or annual general meetings were targeted when lots of plot holders were on site. Plot holders were personally invited to take part in the study by the research team. Participants were asked to indicate a preferred day/time for sampling on consent forms which were returned to the research team.

All allotment plot holders were asked for permission to take a small, < 1 kg sample from their plot(s) and given a research pack. The research packs contained:

- A short questionnaire (A4 sheet) asking plot holders to describe any amendments they had made to the soils on their plots.
- A longer questionnaire asking participants to provide answers to a range of questions concerning their gardening activities, lifestyle and health and well-being (described in Section 5.2.1).
- A short summary of the aims and objectives, and consent forms.

Plot holders were encouraged to return the short questionnaire and consent forms on the day of sampling. Two separate Microsoft Access (MA) 2010 © databases were created to store personal data and questionnaire responses. The databases are described in more detail in Section 5.2.4. The databases were linked by a unique identification

number present on all samples and paperwork. Hard copies of signed consent forms with personal data removed were retained in a locked cabinet. Copies of all paperwork in the research packs can be found in Appendix II.

In return for giving permission for samples to be taken, plot holders received a copy of the agronomic data for their plot which included soil pH, texture, organic matter and available nutrients; Mg, P and K. The rest of the data was passed on to the NCC to be disseminated to plot holders on request. Entry into a prize draw for a £100 cash prize or membership to the Royal Horticultural Society / National Trust + spending money, was offered for the return of completed questionnaires (Chapter 5), and one winner was chosen at random.

2.2.3 Soil amendment questions

Participants were asked to respond “YES” or “NO” to whether they had amended the soil by: adding mineral fertilizer (MF), liquid feed (LF), manure or slurry (MS), poultry litter (PO), mushroom compost (MC), shop-bought compost (SB), organic compost (OC), bark chippings (BC), lime (LM), ploughing or turning over the soil (PL) or any other amendment (OT). The inclusion of soil amendments was based on prior knowledge of soil amendments commonly added to an allotment plot. There was space provided to write other amendments (e.g. fish, blood and bone). Participants were also asked to indicate how regularly plots were amended in a calendar year using a Likert scale with the responses

“never”, “0-1”, “2-5”, “6-10”, “more than 10”. Responses were coded 0 (for “never”) and 1-4 (for the other responses) and entered into the database (section 5.2).

2.2.4 Soil sampling strategy

Sampled plots were distributed across the allotments at random according to the participation of plot holders in the study. Soil samples were collected from 146 individual plots, distributed across the 9 allotment sites, between September 2014 and November 2015 with most of the allotment plots sampled in the autumn to avoid disrupting the growing season (Table 2.1. The exception to this rule was site OPF from which samples were taken between March-June when the researchers were given access to the site). Initially, topsoil samples alone were taken from 15 plots (7 from WT, 5 from SN and 3 from ST). It was subsequently decided to sample sub-soils as well. Top- and sub-soil samples were taken from plots using a Dutch auger in a ‘W’ transect, with samples taken at each corner in the ‘W’, giving 5 replicate samples which were then aggregated together. Topsoil samples were taken using a 10 cm diameter auger to a depth of 15 cm. Subsoil samples were taken with a narrower, 4 cm diameter auger at the start of the second soil horizon (located by a change in the soil colour and constituency with increased sample depth). Aggregated samples were stored in polyethylene (plastic) bags labelled with a unique ID to the respondent, the allotment code (BH, CN etc.), the plot number and ‘TOP’ for top-soil or ‘SUB’ for sub-soil.

The sampling strategy was complicated, on occasion, by the preference of plot holders for some areas not to be sampled. Many allotments also contained paths and small out-buildings which altered the shape of the plot. Designated bonfire areas, and other areas which were not intended for growing produce, were avoided as they may have contained concentrations of metals and/or soil properties that did not reflect the rest of the plot. The amount each plot was cultivated varied within allotment sites (Figure 2.2). On some occasions where a plot holder had inherited two smaller plots side-by-side, it was deemed appropriate to sample them as one.



Figure 2.2: A selection of sampled allotment plots. Image A: Plots were occasionally separated with large hedges (site: ST). Images B and C: Paths and sheds complicated the sampling strategy (site: CN). Image D: Some plots were barely cultivated at all (site: CN).

2.2.5 Sample storage and processing

Subsoil and topsoil samples were transported back to the laboratory and either dried immediately or refrigerated for a short period (2 to 3 days). Soils were transferred to labelled plastic tubs and air dried on a heated rack for between 2 and 3 days. Once they had been dried sufficiently, soils were mixed by hand with clean gloves to avoid cross contamination and sieved to remove particles > 2 mm in diameter. They were then returned to the drying rack until fully dry (at least 3 days), sealed, and stored in a cool, dry environment. A subsample of approximately 15 g from each individual sub- or top-soil sample was ball milled to < 40 µm for 4 minutes at 2750 rpm using an agate ball mill (Retch: PM 400), and stored in labelled, sealed polyethylene bags.

2.2.6 Laboratory analysis of soils

Soil texture, pH, organic carbon and trace element concentrations were determined for top-soil samples; trace element concentrations were also determined for sub-soil samples. Unless otherwise specified, all chemical reagents used in analyses were of Fisher © trace element or analytical reagent grade.

Soil texture

Particle size was estimated using a laser granulometry method. Due to time constraints over the use of the equipment, particle size was analysed for allotment sites as a whole rather than for individual plots. The number

of samples used to estimate soil texture for each allotment site was based on the total number of plots at the site (Table 2.1).

Samples were prepared by the chemical removal of organic matter with hydrogen peroxide and the addition of calgon to separate soil particles. Firstly, 0.5 g of (sieved and dried) soil was added to 30 ml of H₂O₂ (hydrogen peroxide: >100 volumes) in centrifuge tubes. The tubes were then heated to 80 °C in a water bath for 2 hours and centrifuged to 3,000 rpm (Thermo Scientific: Megafuge 40) for 10 minutes and the remaining H₂O₂ was decanted to leave a plug of soil sediment and soil grains. The sediment was rinsed three times with 30 ml of deionised water to remove the hydrogen peroxide and centrifuged at 3,000 rpm between rinses. Finally, calgon (7 g sodium hexametaphosphate and 35 g sodium bicarbonate diluted in 1 L deionised water) was added to each sample and samples were then placed in an ultrasonic bath (Grant: XUBA 1) for a minimum of 30 minutes. One-in-ten samples was replicated to estimate the accuracy of the sample measurements.

Prepared samples were decanted into a particle size analyser (Coulter: LS200) using a liquids module, and the fraction of the sample occupying different particle size diameters was estimated by light scattering of the laser beam. Results were obtained on a percentage-by-volume basis with increasing particle size. Percent sand was calculated by the addition of percentages for particle sizes with diameters > 50 µm, silt between 8 µm and 50 µm and clay < 8 µm. The clay fraction was estimated using a

diameter of $< 8 \mu\text{m}$, rather than the usual $< 2 \mu\text{m}$, as this gives a better estimation of the fraction of clay particles using a laser granulometry method (clay particles are not spherical so do not conform to the assumptions of the laser granulometry method: (Konert and Vandenberghe 1997). Soil textural class was estimated from particle size distributions with the UK soil textural classification system using (Moeys 2014) soil texture wizard in statistical package R v 3.2.1.

Soil pH

Soil pH was determined on top-soils collected from all of the allotment plots using a pH meter (Hanna Instruments: pH 209) in the laboratory. The pH meter was first calibrated using buffer solutions at pH values of 4.01 and 7.00. Sieved and dried soil samples (5 g) were then combined with 12.5 ml of deionised water in a centrifuge tube. Samples were shaken end-over-end at 35 rpm for 30 minutes and allowed to settle prior to analysis. The pH meter was placed in the resulting solution and the value read after a one minute equilibration period. The meter was thoroughly rinsed between samples, using deionised water, and then blotted dry to reduce cross-contamination. The pH meter was recalibrated after every ~ 10 samples using the buffer solutions.

Total organic carbon

Total organic carbon (TOC) was estimated for each topsoil sample by analysing the sample for inorganic carbon (IC) and total carbon (TC) (Equation 2.1).

$$\text{TOC} = \text{TC} - \text{IC} \quad 2.1$$

Total carbon:

A carbon – nitrogen – sulphur analyser (CNS; Flash EA 1112) was used to measure total carbon. Approximately 20 mg of ball milled soil was added to tin foil capsules; folded shut with a clean pair of tweezers. With every batch of 54 samples (including replicates); one blank, one bypass (5-10 mg aspartic acid), two standards (20 mg of 'Peaty soil', Elemental Microanalysis: B2176) and approximately six samples of certified reference material (20 mg 'Peaty Soil') were included for quality control. One-in-ten soil samples were replicated to check for accuracy in individual CNS measurements. The mean recovery of total C from certified reference materials was 101 % (n = 29). Samples, blanks and standards were loaded into the auto sampler. Settings for the CNS are described in Figure 2.3.

Temperatures	System Timings
Furnace – <u>Right</u> 900°C	Cycle (Run time) – 720 s
Oven – 65 °C	Sampling Delay – 12 s
	Oxygen inject end – 17 s
Gas Flows	Detector
Carrier – 140 ml min ⁻¹	Filament – On 1000 µV
Oxygen – 250 ml min ⁻¹	
Reference – 100 ml min ⁻¹	

Figure 2.3: Instrument settings for the measurement of total carbon in soils using the CNS (Flash EA 1112).

Inorganic carbon:

Inorganic carbon was measured using a total organic carbon analyser (TOC; Shimadzu Vcph) connected to a sample solids module (SSM; Shimadzu SSM-5000a). Approximately 100 mg of soil was weighed into ceramic sample boats which were cleaned (with Mili-Q® water) and heated in a muffle furnace (Carbolite: AAF 1100) for 6 hours at 450 °C, prior to the analysis, to reduce contamination with exterior carbon. During each run (of approximately 35 samples), a calibration curve was first calculated using the Shimadzu software with three blank samples; three standards containing 50 mg, 75 mg and 100 mg of 'chalky soil' (Elemental Microanalysis: B2188 containing 2.1 % inorganic carbon by weight) and three sodium carbonate standards (50 mg, 75 mg and 100 mg containing 11.3 % inorganic carbon by weight). One-in-ten samples were replicated to check the accuracy of the inorganic carbon measurements. The software was set to calculate inorganic carbon as a percent of the total weight and weights were entered manually into the software for each sample.

Ceramic boats were placed into the solids module in turn and 0.7 ml phosphoric acid (>85 %) added to each sample to remove organic carbon. Samples were then placed in the furnace and heated to 200 °C. Inorganic carbon present in the sample is converted to carbon dioxide which is measured by the non-dispersive infrared detector (NDIR) in the TOC analyser. The calibration curve; re-calculated for each run by plotting the known concentrations of blanks and standards against the observed

concentration, had a Pearson correlation coefficient (r) between 0.97 and 1. Organic carbon content was converted to soil organic matter by multiplying by 1.58 (Rowell 1994).

Available nutrients and cations

Available nutrient concentrations for Mg, P and K were measured for each individual top-soil sample. Ten percent of the samples were replicated to estimate the accuracy of available nutrient concentrations.

Extraction with ammonium nitrate:

Available nutrient concentrations for cations Ca, K, Mg and Na were measured by extraction with 1 M ammonium nitrate (NH_4NO_3) followed by elemental analysis using inductively coupled plasma mass spectrometry (ICP-MS: Thermo Fisher X series II). Two grams (2 g) of air dried, sieved soil was combined with 20 ml of 1 M NH_4NO_3 and shaken end-over-end for 1 hour (35rpm) and centrifuged at 3,750 rpm for 15 minutes. Supernatants were syringe-filtered to 2 μm , and 9 ml of the filtered supernatant added to 1 ml 0.1M nitric acid (HNO_3 : >68 %) to acidify the sample prior to ICP-MS. Samples were run on the ICP-MS using an autosampler (Cetac ASX-520) and inserted through a concentric glass venturi nebuliser (Thermo-Fisher Scientific, Bremen, Germany; 1 ml min⁻¹). Samples were run in 'collision cell mode' with 7% hydrogen in helium gas. This removes polyatomic ions which collide more regularly with the He ions in the ICP-MS quadrupole.

Available nutrient concentrations were used to estimate cation exchange capacity (CEC) by converting concentrations in mg kg^{-1} to cmol C kg^{-1} using the valence of each nutrient (cmol C kg^{-1} : centi-mols of positive charge per kg of soil). Concentrations of cations (cmol C kg^{-1}) were summed to give an estimate of the CEC.

Olsen P method:

Available P ($\mu\text{g P g}^{-1}$ dw soil $\equiv$ mg P kg^{-1} dw soil) was measured using the Olsen P method with 2 g of sieved and dried soil for each topsoil sample weighed into centrifuge tubes. This was combined with half a spoon of phosphate charcoal and 30 ml of sodium bicarbonate before being shaken end-over-end (35 rpm) for a minimum of 30 minutes. Samples were then centrifuged at 3,750 rpm for 15 minutes. One-in-ten samples were replicated to check for inconsistency in the analysis. A 3 ml volume of supernatant per sample was used and combined with 4 ml of acid molybdate reagent, 4 ml ascorbic acid in a 50 ml volumetric flask and made up to 50 ml with milli-Q® water. Samples were left for 30 minutes for the colour to develop. A calibration curve was set up using increasing aliquots of 10 mg l^{-1} P potassium di-hydrogen from 0 mg l^{-1} to 60 mg l^{-1} . A new calibration curve and three blanks were used for each new batch of samples (roughly 30 samples). Light absorbance was measured using a spectrophotometer (Cecil: 1011) calibrated with the 0 mg l^{-1} P standard. Available phosphate was calculated for each sample using Equation 2.2.

$$P_{Olsen} = Slope \cdot \left\{ \frac{\left(P_{soil} \cdot \frac{V_{bicarb}}{V_{soil}} \right) - \left(P_{blank} \cdot \frac{V_{bicarb}}{V_{blank}} \right)}{W} \right\} \quad 2.2$$

Where: P_{soil} is the light absorbance of the sample (nm), V_{bicarb} the volume of sodium bicarbonate used (30 ml), V_{soil} the volume of supernatant used in the analysis (3 ml), P_{blank} the average light absorbance of the blank samples (nm), V_{blank} the volume of supernatant in blank samples (3 ml), W the exact mass of soil used in the analysis (g), and *slope* is the regression line relating the light absorbance (nm) of the standards with the known concentration of P (μg). The mean recovery rate (see above) for samples was 104% ($n = 13$).

Trace elements

Total trace element concentrations for B, Na, Mg, P, S, K, Ca, Ti, Li, Be, Al, V, Cr, Mn, Fe, Co, Ni, Cu, Zn, As, Se, Rb, Sr, Mo, Ag, Cd, Cs, Ba, Tl, Pb and U were determined by ICP-MS (Thermo Fisher X series II) following acid digestion. Acid digestion and trace element analysis was undertaken for all individual top-soils and sub-soils in duplicate with an average and standard deviation obtained. Firstly, c. 200 mg ball milled soil for each duplicate sample was weighed into Perfluoroalkoxy alkane (PFA) vessels. The vessels were placed onto a programmable hot plate (Analysco 2416) and 2 ml of HNO_3 and 1 ml perchloric acid (HClO_4 : 70 %) was added to each sample. The hot plate was set to programme 2 (Figure 2.4) overnight. The following day 2.5 ml of HF was added and the hot plate set

to programme 3 (Figure 2.4) overnight. Lastly, 2.5 ml HNO₃ and 2.5 ml milli-Q® water was added and heated to 50 °C for 45 minutes on the hot block. The remaining liquid was decanted into a 50 ml volumetric flask with the digestion pots rinsed three times with milli-Q® water and made up to 50 ml (with milli-Q® water). Volumetric flasks were shaken end-over-end by hand and after a settling period, 6 ml of liquid transferred to

Programme 2:	Programme 3:
STEP: 80°C	STEP: 120°C
DWELL: 8 hrs	DWELL: 1 hr
STEP: 100°C	STEP: 140°C
DWELL: 2 hrs	DWELL: 3 hrs
END	STEP: 160°C
	DWELL: 4 hrs
	STEP: 50°C

Figure 2.4. Programme options for Analysco 2416 hot block

a small sealed storage tube. Prepared samples were further diluted by a factor of ten initially, prior to measurement with ICP-MS. Nine samples (in duplicate) were repeated with a dilution factor of one hundred, due to high

Pb concentrations which initially caused the ICP-MS to switch to analogue mode when they were first run. In each run of the ICP-MS (there were two runs initially followed by a subsequent run in which the nine samples above were included) three blank, acid digested samples were included to account for background element concentrations, and three samples of certified reference material (Montana II soil: NIST 2711a) were included as quality control. Samples were run on the ICP-MS using an autosampler (Cetac ASX-520) in 'collision cell mode' (see above) and inserted through a concentric glass venturi nebuliser (Thermo-Fisher Scientific, Bremen, Germany; 1 ml min⁻¹). The mean recovery of elements from the certified reference material (Montana II soil: NIST 2711a) is reported in Table 2.2.

Table 2.2. Mean recoveries of certified reference material after acid digestion and determination by ICP-MS.

Metal(loid)	Potentially toxic elements							Nutrient cations			
	As	Cd	Cr	Cu	Ni	Pb	Zn	Ca	K	Mg	Na
Recovery rate (%)	94	91	79	82	106	89	99	99	99	99	98

The deviation between duplicates was high ($> 100 \text{ mg kg}^{-1}$) for a small number of samples containing high concentrations (above 800 mg kg^{-1}) of Pb. It was suspected that this could be a result of particulate lead (such as particles of leaded paint) in the samples which may not have been equally distributed across the milled sample from which the duplicate samples for digestion and element analysis were taken.

2.2.7 Data analysis and reporting

Comparison to guidance values

Data from the soil analyses were compared with guidance values used to determine concentrations of metals which will not result in SPOSH (significant possibility of significant harm) (DEFRA, 2013). Concentrations of As, Cd, Cr and Pb were determined and compared to Category 4 Screening Levels (C4SLs) for an allotment use (DEFRA 2013c-e) and the withdrawn soil guideline values they replaced in 2013 (EA 2009b-e) (Ni concentrations were compared with the SGV only as a C4SL for Ni has not been developed by DEFRA). Elemental concentrations (Cd, Cu, Pb, Ni) were also compared with Normal Background Concentrations (NBCs) for an urban or (As) principle domain (Ander *et al.* 2013). An enrichment factor (EF) was calculated for all HMs by dividing top-soil

concentrations by sub-soil concentrations. Available nutrients; P, K and Mg were compared with recommended concentrations (DEFRA 2010) which represent levels at which further application will not improve yield and further application results in unmerited expense for the grower.

Variation between allotment sites

Differences in HM concentrations (As, Cd, Cr, Cu, Ni, Pb and Zn), soil properties (TOC, pH and CEC) and plant nutrients (Mg, P and K), between allotment sites, were assessed separately by one-way analysis of variance (ANOVA) in Minitab © v 17.2.1. Normality was assessed in the dependent variables (HM concentrations, soil properties and plant nutrients) using the Anderson-Darling test. Concentrations of Cd, Cu, Pb and Zn in top-soils were found to be positively skewed and were transformed using a log-linear (box-cox) transformation during the analysis. Tukey analysis was used to make comparisons between soil properties, nutrients and HM concentrations at different allotment sites.

Influence of soil properties and agronomic management

Analysis of co-variance (ANCOVA) (Minitab v 17.2.1) was used to determine correlations between soil properties, nutrients or agronomic management with HM concentrations in top-soils. Heavy metal concentrations were included as the dependent variable. Allotment site and responses to soil amendment questions (recorded in a binary 'YES' or 'NO' format) were included in the model as cofounding factors, and soil

pH, TOC and available nutrient concentrations (Mg, K and P) were included as covariates. Top-soil concentrations of Cd, Cu, Pb and Zn were transformed in the analysis (log-linear box-cox). Factors and covariates were removed from the statistical model on a top-down basis until the simplest model was obtained. Factors and covariates were removed in order of adjusted sums of squares (low to high) and only replaced if they were found to be significant to the model. Between two and six factors and covariates were retained in each ANCOVA model.

Limitations of data analysis and assumptions of ANOVA and ANCOVA

The main assumptions of a generic linear model (GLM) are that samples within separate groups are random, the data is normally distributed, scores in different groups are independent, and the variance accounted for by each group is homogenous (Rutherford, 2001). A further assumption in ANCOVA is that the relationship between the dependent and independent variable across each group is linear (Rutherford, 2001). The data here inevitably are limited by differences in sample size between the different allotments because of the differences between the size of site and access (Table 2.1). This could result in differences in the variation accounted for between the different allotment sites, although Levene's test for equal variances (performed using Minitab v 17.2.1) carried out retrospectively, showed that differences in variances between allotment sites were not significant for the different responses tested. Differences in the size of groups in an ANCOVA model can also influence the scores of one group being independent from another in the analysis as

the group with the larger sample size may have more weighting on the relationship between one factor and another. As a result, the output from the ANCOVA model should and was treated with discretion. Relationships between discrete and continuous variables highlighted in the ANCOVA model were explored further at the allotment site with the largest sample size - St Ann's (ST); to observe if the relationship remained when the influence of allotment site was removed from the analysis.

2.3 RESULTS

2.3.1 Heavy metals

Metal/metalloid concentrations in top-soils are displayed in boxplots in Figure 2.5 and Figure 2.6. Top-soil concentrations of all metals and metalloids studied (As, Cd, Cr, Cu, Ni, Pb and Zn) were found to be significantly different between samples taken from different allotment sites (two-way ANOVA; $p < 0.001$). Soils from allotment sites; WM and MR, contained higher median concentrations of As, Cd, Cu, Pb and Zn than the other allotment sites (Table 2.3). Median concentrations of Cu, Ni, Pb and Zn were also high in soils from ST. In contrast, soils from BH contained lower concentrations of Cd, Cr and Pb than any of the other allotment sites (tukey; $p < 0.05$). Site CN contained high concentrations of Cd, but did not contain high concentrations of any of the other metals.

Metal/metalloid concentrations in top-soils were compared with guidance values used for the assessment of contaminated land (Table 2.3). Lead concentrations were found to exceed the C4SL for an allotment of 84 mg kg^{-1} (DEFRA 2013e) in all samples from 7 of the 9 allotment sites. Ten samples from ST, WM and MR also exceeded the NBC for Pb of 830 mg kg^{-1} (Ander *et al.* 2013). Samples from site BH were all below the C4SL for Pb, with one exception containing 138 mg kg^{-1} Pb. The maximum concentrations of As, Cd and Cr in sampled top-soils were below the C4SLs for an allotment land-use of 49, 4.9 and 170 mg kg^{-1} respectively

(DEFRA 2013c, b; d). Four of the soil samples from site CN exceeded the withdrawn SGV for Cd of 1.8 mg kg^{-1} (EA 2009b).

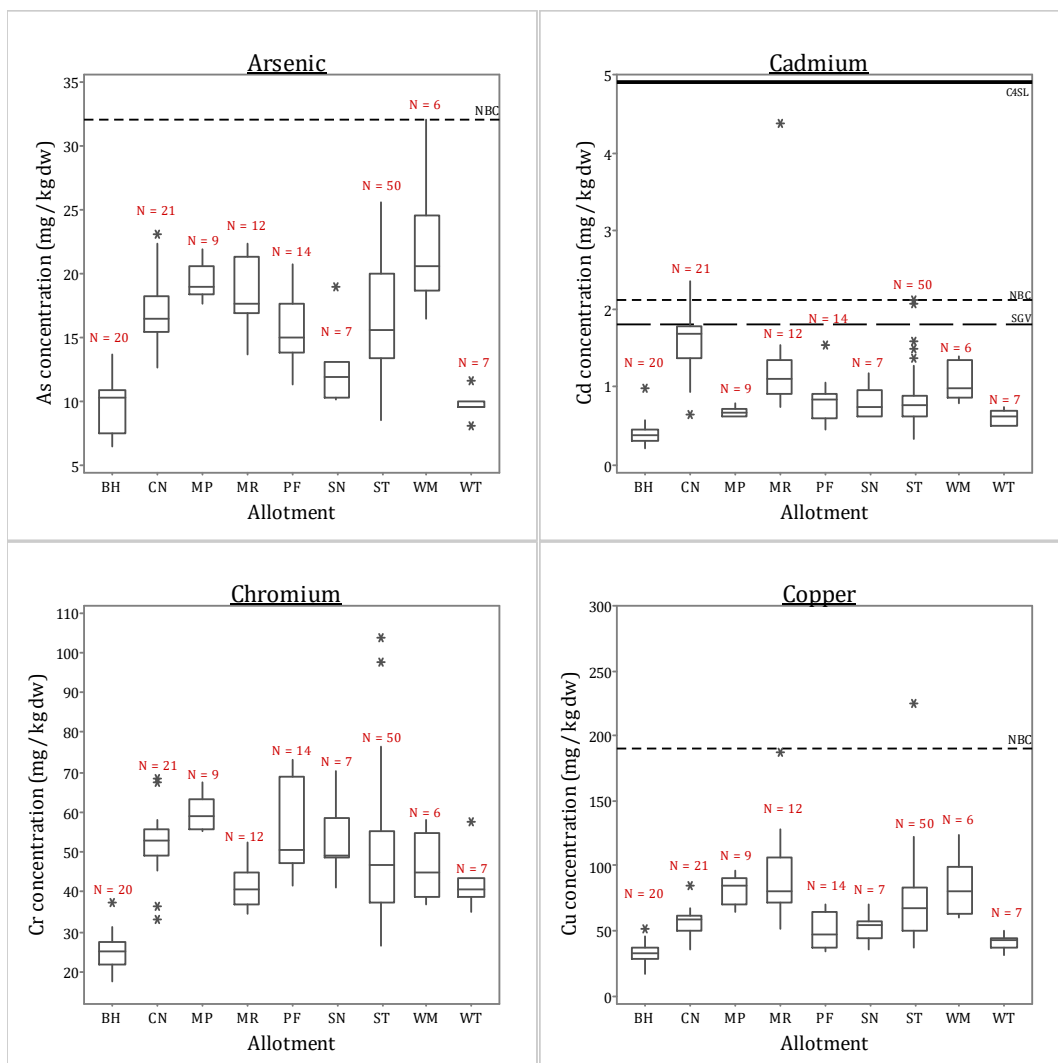


Figure 2.5: Boxplots showing the distribution of top-soil concentrations of As, Cd, Cr and Cu in soil samples from the 9 allotment sites: Blenheim (BH), Canal (CN), Mapperly (MP), Moorfield (MR), Old Park Farm (OPF), Sneinton (SN), St Ann's (ST), Windmill (WM) and Woodthorpe (WT). Boxes indicate median and inter-quartile range (IQR). Asterisks show outlying values exceeding the bounds of upper/lower quartile $\pm 1.5 \times \text{IQR}$. One sample from ST contained a concentration of $545.7 \text{ mg Cu kg}^{-1} \text{ dw}$ which is not shown in the graph. Reference lines represent soil guideline concentrations: Category 4 Screening Levels (C4SLs: DEFRA, 2013 a-d); Soil Guideline Values (SGVs: EA, 2009a); and Normal Background Concentrations (NBCs) (Ander *et al.*, 2013).

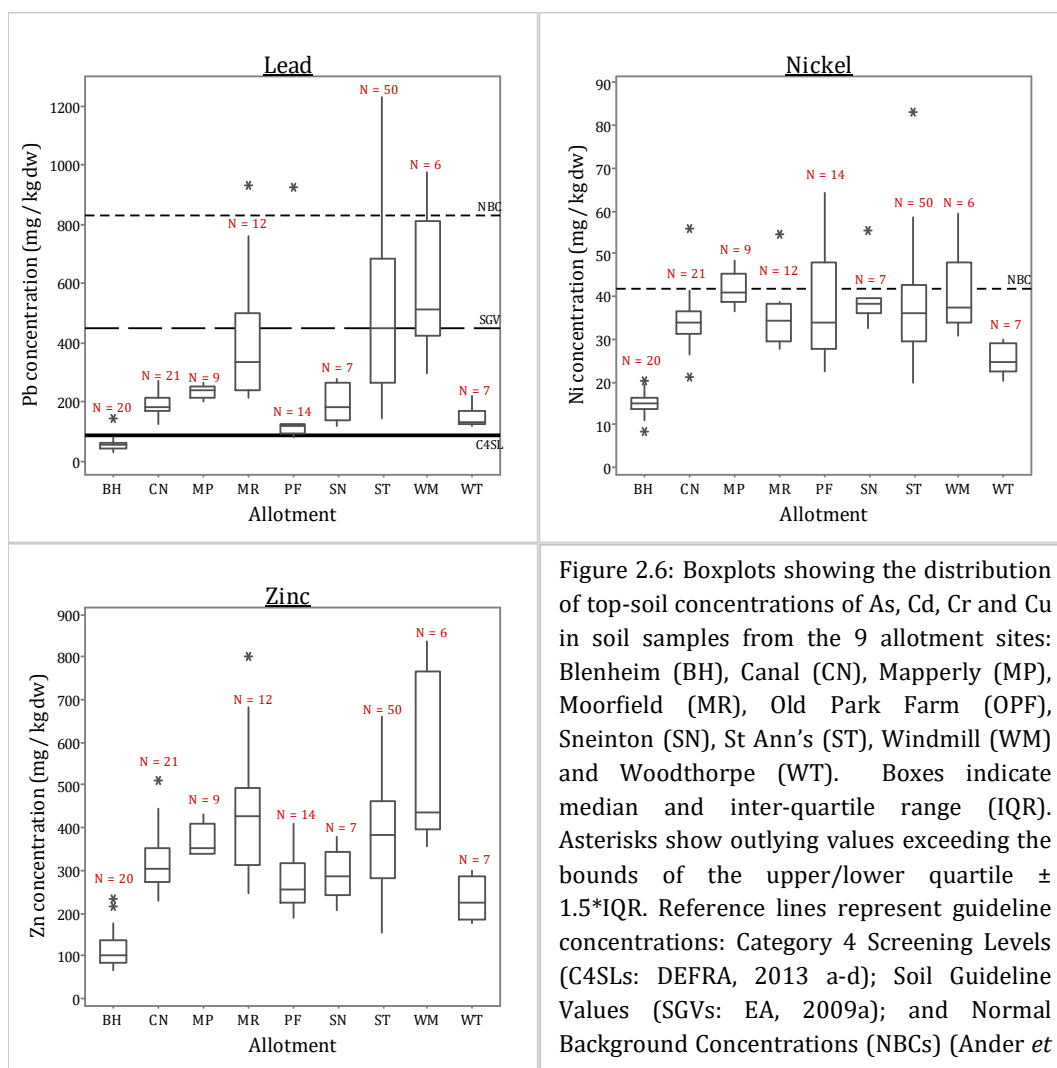


Figure 2.6: Boxplots showing the distribution of top-soil concentrations of As, Cd, Cr and Cu in soil samples from the 9 allotment sites: Blenheim (BH), Canal (CN), Mapperly (MP), Moorfield (MR), Old Park Farm (OPF), Sneinton (SN), St Ann's (ST), Windmill (WM) and Woodthorpe (WT). Boxes indicate median and inter-quartile range (IQR). Asterisks show outlying values exceeding the bounds of the upper/lower quartile $\pm 1.5 \times \text{IQR}$. Reference lines represent guideline concentrations: Category 4 Screening Levels (C4SLs: DEFRA, 2013 a-d); Soil Guideline Values (SGVs: EA, 2009a); and Normal Background Concentrations (NBCs) (Ander *et*

Category 4 Screening levels have not yet been developed for Cu, Ni and Zn. Concentrations of Ni were found to be below the discontinued SGV of 230 mg Ni kg⁻¹ dw (EA 2009c) across all of the allotment sites, but samples from several sites did exceed the NBC of 42 mg Ni kg⁻¹ dw for an urban soil (Ander *et al.* 2013). Two soil samples from site ST exceeded the NBC derived for copper in urban soils of 190 mg Cu kg⁻¹ dw (Ander *et al.*, 2013). No assessment criteria exists from the EA for Zn. Soil concentrations of Zn in all samples ranged from 63.3 mg Zn kg⁻¹ dw (BH) to 837.1 mg Zn kg⁻¹ dw (ST).

Top-soils of the allotment plots were enriched relative to sub-soils in Cd, Cu, Pb and Zn. Concentrations of Cd and Pb were, on average, over two-fold higher in top-soils compared to sub-soils. Top-soils were not enriched with As, Cr or Ni. The average EFs for these metals across all sites were between 0.98 (Cr) and 1.12 (Cu) (Table 2.4). Enrichment factors for all metals were statistically different between allotment sites (two-way ANOVA; $p < 0.001$ for As, Cd, Cu, Ni, Pb and Zn, $p = 0.017$ for Cr). Top-soil concentrations of all metals (except Cr: EF = 1.14) from site WM were higher than sub-soils (EF: 1.56 – 6.74). Concentrations of metals in top-soils were approximately equal to concentrations of metals in sub-soils from site BH (EF: 0.88 – 1.05).

Table 2.3: Descriptive statistics and guideline values for metal concentrations in **top-soils** for each allotment site. Values are given in mg metal kg⁻¹ dw soil except the coefficient of variation (CV) – the ratio of the standard deviation to the mean - expressed as a %. ¹⁾ Category 4 screening levels (DEFRA, 2013 a-d); ²⁾ Soil Guideline values (EA, 2009a-d); ³⁾ NBCs (Ander *et al.*, 2013).

Metal(loid)/allotment site		BH	CN	MP	MR	PF	SN	ST	WM	WT	All sites	C4SL ₁	SGV ₂	NBC ₃
N (count)		20	21	9	12	14	7	50	6	7	146			
Arsenic (As)	Median	10.3	16.3	18.9	17.5	14.9	11.9	15.6	20.5	9.5	15.5	49	43	32
	Mean	9.7	16.9	19.3	18.4	15.5	12.4	16.2	21.8	9.7	15.4			
	CV (%)	22	15	7	15	18	25	26	24	11	29			
	Min.	6.4	12.5	17.6	13.6	11.2	10.1	8.4	16.4	8.0	6.4			
	Max.	13.7	23.1	21.8	22.3	20.7	18.9	25.5	31.9	11.5	31.9			
Cadmium (Cd)	Median	0.351	1.658	0.649	1.092	0.822	0.728	0.742	0.975	0.605	0.762	4.9	1.8	2.1
	Mean	0.383	1.582	0.652	1.374	0.805	0.776	0.811	1.047	0.591	0.897			
	CV (%)	43	25	10	71	33	27	46	23	16	60			
	Min.	0.206	0.635	0.576	0.725	0.444	0.578	0.317	0.780	0.473	0.206			
	Max.	0.964	2.349	0.780	4.365	1.523	1.155	2.095	1.374	0.734	4.365			
Chromium (Cr)	Median	24.6	52.9	58.9	40.5	50.4	49.0	46.7	44.7	40.6	47.2	170	-	-
	Mean	24.8	52.4	59.6	41.2	55.7	52.8	48.2	46.1	42.0	46.3			
	CV (%)	18	16	7	13	21	18	32	19	17	31			
	Min.	17.3	32.7	55.1	34.5	41.4	41.1	26.3	36.6	34.8	17.3			
	Max.	37.3	68.2	67.1	52.0	73.2	70.0	103.8	58.0	57.4	103.8			
Copper (Cu)	Median	32.5	57.9	84.7	80.2	46.0	53.1	67.2	80.0	42.7	57.8	-	-	190
	Mean	31.4	56.4	81.4	92.3	48.7	51.3	79.8	82.5	41.4	64.9			
	CV (%)	28	18	14	39	27	22	92	28	15	76			
	Min.	16.3	35.4	63.7	50.2	33.5	34.4	35.8	59.0	30.4	16.3			
	Max.	50.9	83.6	96.0	186.3	69.3	70.3	545.7	122.9	50.0	545.7			

Table 2.3 continued														
Metal/allotment		BH	CN	MP	MR	PF	SN	ST	WM	WT	All sites	C4SL ₁	SGV ₂	NBC ₃
Lead (Pb)	Median	49	180	235	330	113	179	444	510	127	214	84	450	830
	Mean	52	187	231	407	163	190	489	583	148	305			
	CV (%)	47	19	9	56	135	32	54	41	25	84			
	Min.	27	122	198	209	78	113	141	293	114	27			
	Max.	138	270	260	932	921	273	1227	976	218	1227			
Nickel (Ni)	Median	14.6	33.7	40.8	34.0	33.7	38.0	36.1	37.4	24.4	34.0	-	230	42
	Mean	14.8	34.3	41.5	35.0	37.3	39.7	37.1	40.7	25.0	33.4			
	CV (%)	19	19	9	21	34	19	31	25	14	36			
	Min.	8.4	21.0	36.6	27.7	22.4	32.5	19.6	30.9	20.3	8.4			
	Max.	20.0	55.6	48.2	54.6	64.0	55.6	83.1	59.5	29.7	83.1			
Zinc (Zn)	Median	101	302	349	427	256	285	382	435	224	308	-	-	-
	Mean	116	317	368	443	268	287	375	536	229	322			
	CV (%)	41	22	10	36	23	20	32	38	20	45			
	Min.	63	226	335	244	190	205	151	357	175	63			
	Max.	232	509	429	800	410	377	658	837	296	837			

Table 2.4: Mean (minimum and maximum) enrichment factors (EFs) for each metal(loid) for each allotment site.

Metal(loid)/site	BH	CN	MP	MR	PF	SN	ST	WM
As	0.88 (0.47-1.26)	1.04 (0.73-1.34)	1.25 (0.81-1.47)	1.22 (0.87-1.53)	0.99 (0.64-1.35)	1.32 (0.97-1.67)	1.15 (0.45-1.75)	1.56 (1.09-2.65)
Cd	0.96 (0.58-1.56)	1.42 (0.82-3.10)	3.14 (1.21-8.55)	2.26 (1.39-4.02)	1.89 (1.19-3.99)	3.83 (1.78-5.88)	2.73 (0.47-6.07)	3.80 (1.77-6.69)
Cr	0.92 (0.56-1.27)	0.96 (0.60-1.10)	0.89 (0.80-1.01)	1.16 (0.79-1.39)	0.86 (0.59-1.11)	0.95 (0.91-0.99)	1.02 (0.42-2.00)	1.14 (0.79-1.51)
Cu	0.92 (0.45-1.50)	1.29 (0.75-2.55)	2.25 (1.59-3.08)	2.24 (1.46-3.31)	1.71 (0.64-2.62)	2.23 (1.35-3.11)	1.99 (0.61-9.30)	2.62 (1.58-4.73)
Pb	0.90 (0.49-2.02)	1.34 (0.85-2.58)	2.81 (1.23-5.06)	3.08 (1.27-5.70)	2.88 (1.35-11.76)	3.53 (1.81-5.26)	3.07 (0.79-7.39)	6.74 (3.18-10.14)
Ni	0.86 (0.57-1.53)	0.96 (0.27-1.72)	1.10 (1.02-1.32)	1.43 (0.91-1.96)	1.12 (0.57-1.69)	1.19 (1.03-1.34)	1.18 (0.51-2.04)	1.57 (0.96-2.59)
Zn	1.05 (0.57-2.02)	1.42 (0.82-2.65)	1.77 (1.28-2.19)	2.53 (1.63-3.85)	1.63 (0.97-2.42)	1.76 (1.22-2.31)	2.30 (0.69-4.46)	3.16 (2.00-6.36)
n	20	21	9	12	14	2	46	6

2.3.2 Soil properties and plant nutrients

Soil Properties

The soil texture of the allotment plots appeared to be influenced to some extent by the local geology. Soil samples collected from site BH to the north and west of the city contained a higher percentage of sand compared to samples collected from the other allotment sites (Table 2.5). The site was located on an underlying geology of dolomized limestone and dolomite but also had a recent addition of top soil which may have contained high percentages of sand. The rest of the sites were located on a mixed geology comprised of mudstone, siltstone and sandstone (Figure 2.7).

Organic carbon content, pH and estimated CEC varied significantly between top-soil samples taken from different allotment sites (two-way ANOVA; $p < 0.001$). Sites BH and WT contained lower organic carbon compared with the rest of the sites (Table 2.6). Sites MR and PF contained slightly higher organic carbon. The pH range across plots at all sites was between 5.75 and 7.88. Estimated CEC was higher in SN than any of the other allotment sites (Tukey comparisons; $p < 0.05$).

Table 2.5: Soil physical properties of the allotment sites. Textural class was estimated with Moeys, (2014) soil texture wizard in R v 3.2.1 using the UK soil textural classification system (rows may not add up to 100% due to rounding errors). The mean sand, silt or clay $\pm$ standard deviation were calculated using representative numbers of plots to each allotment (n) out of estimated total number of plots (N).

Allotment	n:N (Count)	Sand (%)	Silt (%)	Clay (%)	Textural class (UK)
BH	3:~120	83 $\pm$ 1	9 $\pm$ 1	8 $\pm$ 1	Loamy sand
CN	5: ~200	51 $\pm$ 11	26 $\pm$ 7	22 $\pm$ 4	Sandy clay loam
MP	2: ~20	23 $\pm$ 20	43 $\pm$ 11	34 $\pm$ 9	Clay loam
MR	5: ~250	67 $\pm$ 4	19 $\pm$ 2	14 $\pm$ 2	Sandy loam
PF	2: ~90	34 $\pm$ 22	30 $\pm$ 5	36 $\pm$ 17	Clay
SN	4: ~140	41 $\pm$	36 $\pm$	23 $\pm$	Clay loam
ST	12: ~550	55 $\pm$ 7	25 $\pm$ 3	20 $\pm$ 4	Sandy clay loam
WM	4: ~100	56 $\pm$ 11	24 $\pm$ 7	20 $\pm$ 4	Sandy clay loam
WT	2: ~25	51 $\pm$	29 $\pm$	20 $\pm$	Sandy clay loam

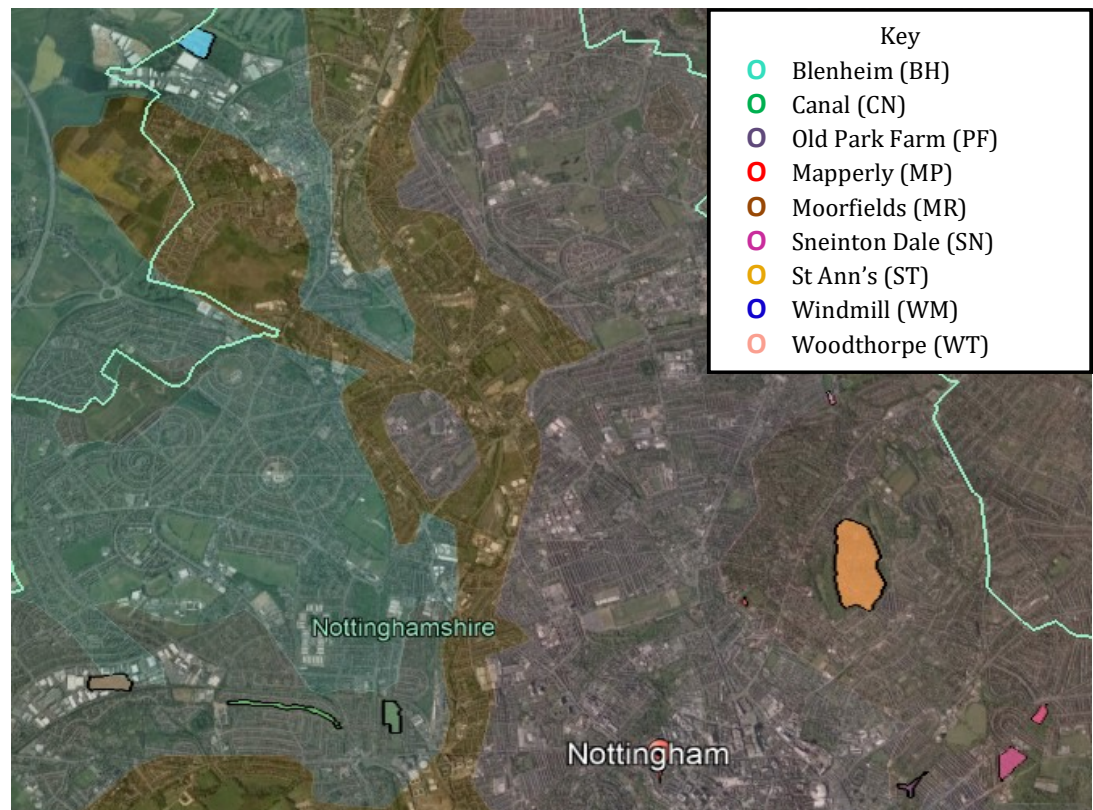


Figure 2.7: The geology around the allotment sites sampled across the city of Nottingham. Changes in the background colour show distinctions in the local geology. Allotment sites are indicated by filled polygons. Site BH located in the north west of Nottingham City is located in the Zechstein Group comprised of dolomized limestone and dolomite. Sites CN, PF and MR in the south east are located in the penine middle coal measures group comprised of a mixture of mudstone, siltstone and sandstone. Similarly, the other 5 sites located to the east of the city are located on unclassified Triassic rocks comprised of a mix of mudstone, siltstone and sandstone. This map was created using Google Earth Pro version 7.3.0 and a layer describing onshore geology of the UK (BGS, 2008).

Table 2.6: Summary statistics describing soil chemical properties for each allotment site. The coefficient of variation (CV) is calculated as a percentage of the mean value.

Chemical property/site		BH	CN	MP	MR	PF	SN	ST	WM	WT	All sites
pH	Mean	6.99	7.32	6.69	6.98	6.66	7.57	6.89	6.57	7.22	6.97
	CV	3	3	3	4	6	4	6	6	5	6
	Min.	6.61	6.79	6.42	6.51	5.88	7.08	5.75	5.98	6.53	5.75
	Max.	7.42	7.63	6.96	7.62	7.18	7.88	7.42	7.12	7.56	7.88
TOC (%)	Mean	4.12	5.46	5.72	7.46	7.43	5.64	6.27	5.97	3.87	5.88
	CV	48	26	11	17	29	25	26	28	18	32
	Min.	1.73	3.25	4.95	5.48	2.86	3.34	3.86	4.18	3.16	1.73
	Max.	9.86	8.80	7.02	9.58	10.66	8.12	10.41	7.82	5.22	10.66
CEC (cmol C / kg)	Mean	17.9	29.7	22.8	22.5	22.9	39.7	23.1	21.1	28.7	24.2
	CV	32	14	7	8	21	22	27	18	22	30
	Min.	10.1	19.7	20.7	19.0	13.4	26.4	10.4	16.4	21.5	10.1
	Max.	31.8	35.9	26.0	25.7	29.3	52.7	42.1	27.5	37.7	52.7
	n	20	21	9	12	14	7	50	6	7	146

Plant nutrients

Concentrations of available plant nutrients; Mg, P and K, varied significantly between allotment sites (two-way ANOVA; $p < 0.01$). Site SN contained higher-than-average concentrations of available Mg and K, but lower-than-average concentrations of P (two-way ANOVA; $p < 0.05$) (Figure 2.8). Mean concentrations of available plant nutrients; Mg, P and K, in all allotment sites, met or exceeded recommended concentrations (RCs) required for growing arable crops, suggested by (DEFRA 2010) of 101-175, 50-85 and 181-240 mg kg⁻¹ respectively (Table 2.7). Minimum concentrations of Mg in site SN, and K in sites PF and BH, were slightly below the RCs. However, maximum concentrations of available nutrients exceeded RCs by a factor of 4-8 fold (Table 2.7).

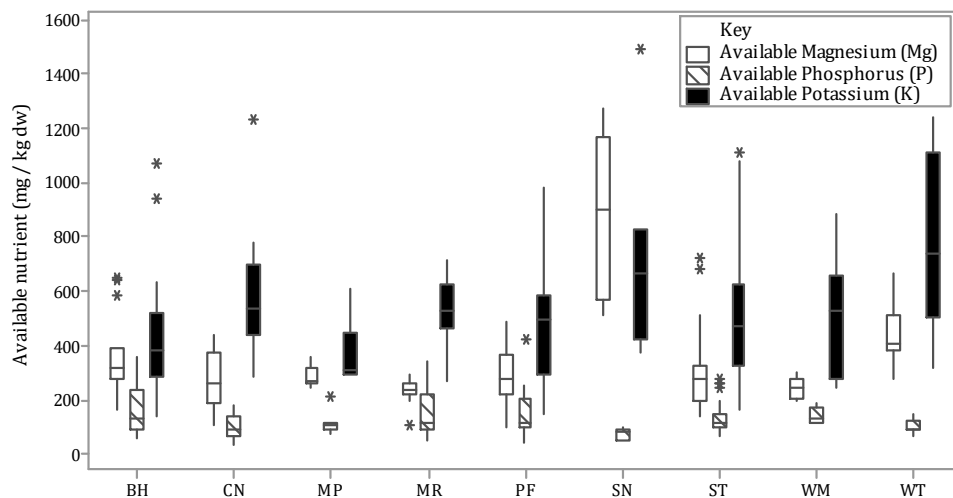


Figure 2.8: Available nutrient (Mg, P and K) concentrations (mg / kg dw) displayed by allotment site: BH = Blenheim (n=20), CN = Canal (n=21), MP = Mapperly (n = 9), MR = Moorfields (n = 12), PF = Old Park Farm (n=14), SN = Sneinton (n=7), ST = St Ann's (n=50), WM = Windmill (n=6) and WT = Woodthorpe (n=7).

Table 2.7. Summary statistics describing available nutrient concentrations (Mg, P and K) for each of the allotment sites. A description of how available P, Mg and K concentrations were derived is provided in Section 2.2.6). 1) Recommended concentrations (RCs) represent concentrations at which further additions of these nutrients would not be expected to result in an increased yield of arable vegetables (DEFRA 2010). The coefficient of variation (CV) is given as a percentage of the mean value.

Chemical property/site		BH	CN	MP	MR	PF	SN	ST	WM	WT	All sites	RC ₁
Available Mg (mg kg ⁻¹)	Mean	342	268	284	227	291	880	288	243	435	321	101-175
	CV	41	38	14	21	35	34	43	17	28	56	
	Min.	162	105	243	107	95	514	135	192	278	95	
	Max.	649	436	359	288	486	1275	720	298	666	1275	
Available P (mg kg ⁻¹)	Mean	167	98	109	153	148	72	128	138	102	128	50-85
	CV	56	46	36	61	67	35	39	22	26	53	
	Min.	56	31	70	52	43	44	61	109	69	31	
	Max.	356	178	207	342	424	101	278	185	148	424	
Available K (mg kg ⁻¹)	Mean	428	578	377	528	491	728	514	508	782	525	181-240
	CV	54	36	29	23	46	51	46	45	43	46	
	Min.	138	282	285	266	143	376	163	244	318	138	
	Max.	1067	1230	605	712.3	983	1493	1111	885	1238	1493	
N		20	21	9	12	14	7	50	6	7	146	

2.3.3 Soil amendments

The results of the short questionnaire describing soil amendments are displayed in Figure 2.9. The soil amendment questionnaire was answered and returned by 126 (86 %) participants. The vast majority of respondents (90 %) reported that they would normally plough or turn over their soil at least once in a calendar year. Other common soil amendments included adding organic compost (75 %), manure or slurry (66 %) and shop-bought composts (54 %). Liquid and mineral fertilizers and poultry litter were commonly added several times a year. In contrast, plot holders rarely added lime to their soil more than once in a calendar year (Figure 2.9).

There were differences in soil amendments between allotment sites (Figure 2.10). Mushroom compost, for example, was widely available at site WT, so 5 of the 7 participants from that site reported that they would normally add it to their plot. For comparison, only 4 out of the remaining 119 respondents, reported adding mushroom compost to their plots. A greater proportion (60 and 75 % respectively) of respondents from site BH reported adding mineral or liquid fertilizers to their plots compared to (40 % and 39 % respectively) respondents across all sites. Plot holders were asked to record any other amendments they added to their allotment plots. Five participants reporting adding blood, fish and bone meal to their plots. Another four participants reported adding a composted nettle and comfrey mixture to their plots.

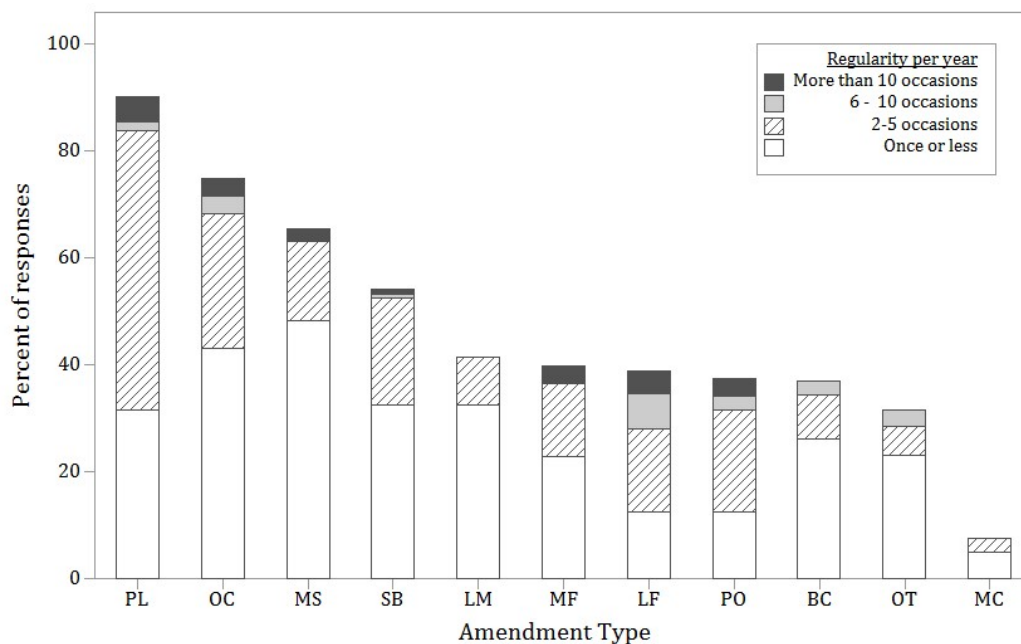


Figure 2.9: The percent of respondents (n = 126) who reported amending the soil by: Ploughing or turning over the soil (PL), adding organic compost (OC), adding manures or slurries (MS), adding shop-bought compost (SB), adding lime (LM), adding mineral fertilizers (MF), adding liquid fertilizers (LF), adding poultry litter (PO), adding bark chippings (BC), other amendments (OT) and adding mushroom compost (MC). Bars are split into categories representing the frequency with which soil amendments occurred.

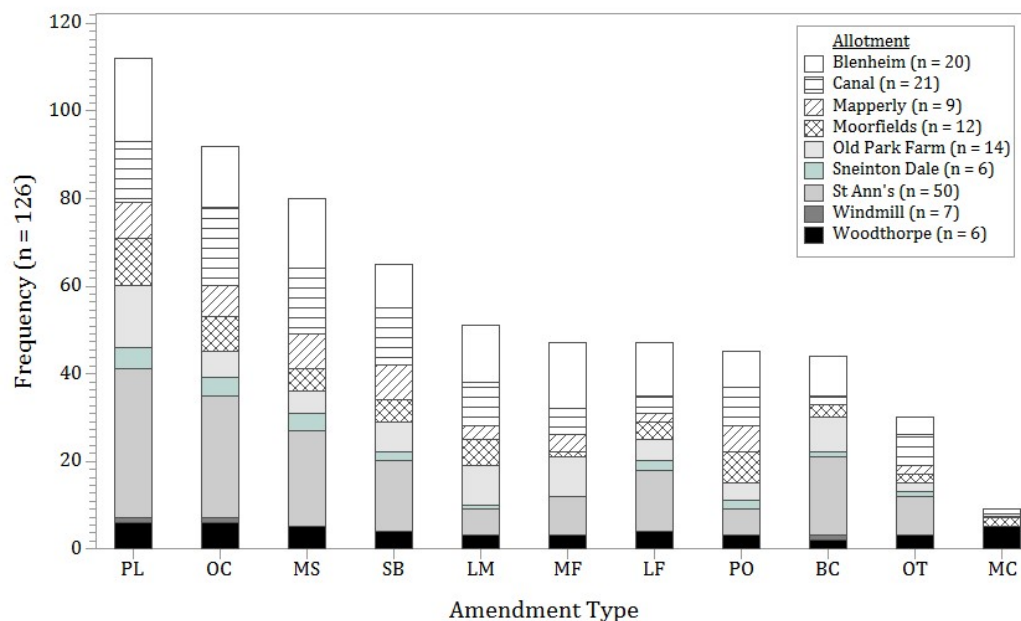


Figure 2.10: The number of respondents (n = 126) who reported amending the soil by: Ploughing or turning over the soil (PL), adding organic compost (OC), adding manures or slurries (MS), adding shop-bought compost (SB), adding lime (LM), adding mineral fertilizers (MF), adding liquid fertilizers (LF), adding poultry litter (PO), adding bark chippings (BC), other amendments (OT) and adding mushroom compost (MC). Bars are split according to the allotment site from which corresponding soil samples were taken.

2.3.4 Influences of soil properties and soil amendments on HMs

The influence of soil properties and soil amendments on top-soil concentrations of HMs, was determined using a top-down ANCOVA model (Section 2.2.7). Much of the variation (adjusted sums of squares in the ANCOVA model) in top-soil concentrations of HMs was accounted for by differences between allotment sites, which were significant in all statistical models ($p < 0.001$). Total organic carbon was positively correlated with top-soil concentrations of Cu, Ni, Pb and Zn ($p = 0.031$, 0.006 , 0.001 and 0.005 respectively) (Figure 2.9). Soil pH values were also positively correlated with top-soil concentrations of As, Cr, Cu, Ni and Pb ($p = 0.014$, 0.014 , 0.013 , 0.002 and 0.008 respectively). However, the correlation between soil pH and HMs was weak and explained a low proportion of the variation in HM concentrations. Top-soil concentrations of Cd were negatively correlated with available Mg and available P ($p = 0.005$ and 0.004 respectively). The addition of bark chippings (BC) to allotment plots was found to be negatively correlated with chromium and lead ($p = 0.011$ and 0.035 respectively). Furthermore, the addition of liquid fertilizer (LF) was negatively correlated with top-soil concentrations of Cr and Pb ($p = 0.001$ and 0.011). The addition of shop-bought compost (SB) was positively correlated with top-soil Pb concentrations ($p = 0.038$) and mineral fertilizer (MF) was positively correlated with top-soil Cr ($p = 0.007$).

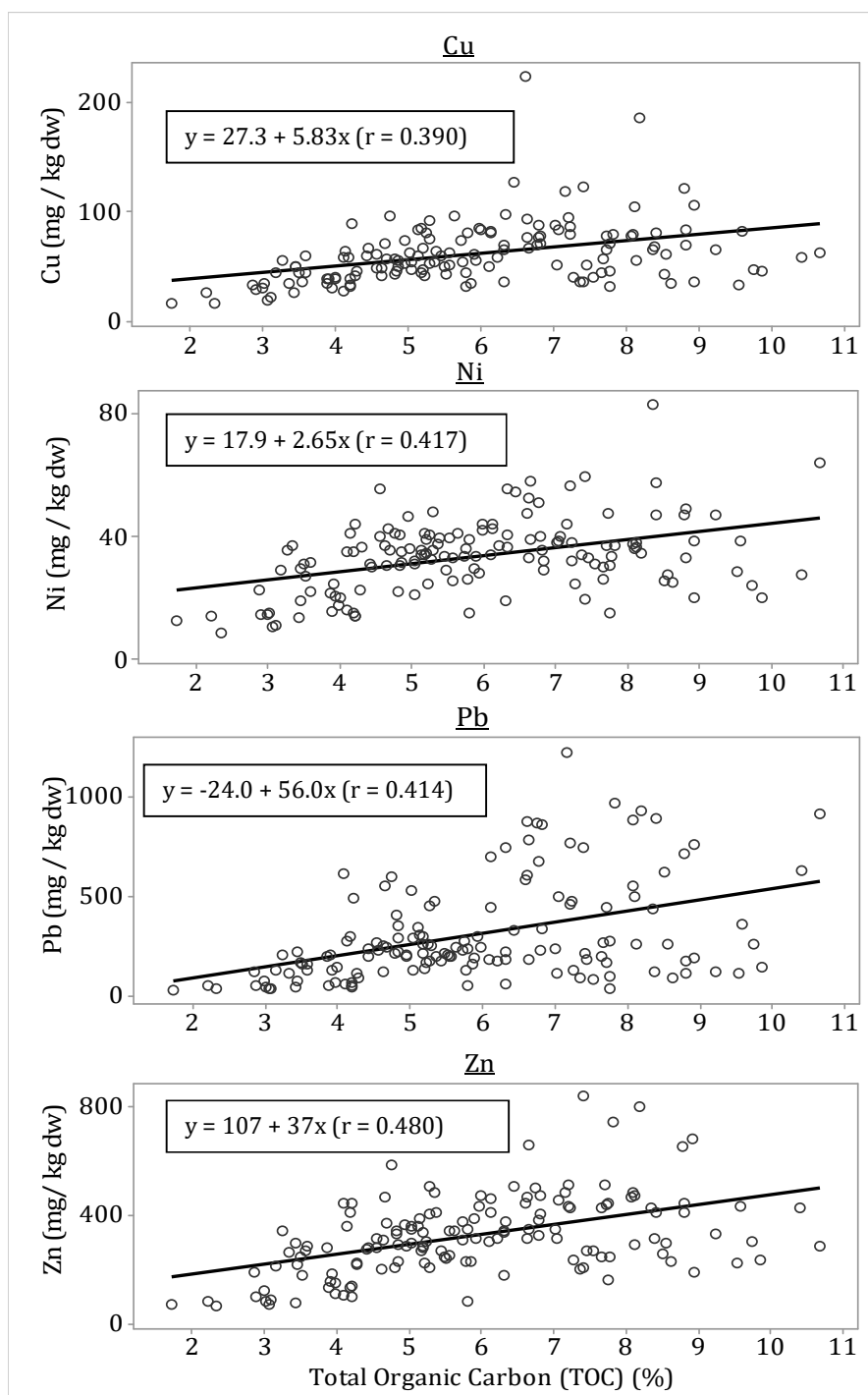


Figure 2.9: Scatterplots showing the relationship between HM concentrations and total organic carbon in top-soils from all of the allotment sites.

A lot of the variation in the ANCOVA model was determined by differences in HM concentrations between allotment sites. This may have resulted in type 1 errors occurring in the statistical model. The interactions between significant variables and allotment site on top-soil concentrations of HMs were further investigated in separate ANCOVA models. It was expected that if an association between one of the independent variables and the allotment site was genuine, the correlation between the independent variable and HM concentrations would exist at several allotment sites, or at least the majority, and therefore be significant ($p < 0.05$) in the ANCOVA model. The interaction between TOC and allotment site did not have a significant influence on top-soil concentrations of HMs ($p > 0.05$). Soil pH was positively correlated, however, with top-soil concentrations of Pb at site ST which contained a relatively large number of samples ($n = 50$, $p = 0.004$) (Figure 2.113). The interaction between soil pH and allotment site explained some of the variation in top-soil Pb, but not in any of the other HMs studied. Available magnesium was positively correlated with top-soil concentrations of Cd at site ST ($p = 0.006$), although two outlying values were identified that exaggerated this relationship (Figure 2.124). It was not possible to estimate the effect of an interaction between soil management options and allotment site on HM concentrations. Bark chippings and liquid fertilizer were not added to a large proportion of plots at sites CN and MP; both of which contained high concentrations of Cr. This may have resulted in a significant effect of the amendment of soils with bark chippings and liquid fertilizer on concentrations of Cr (Figure 2.102).

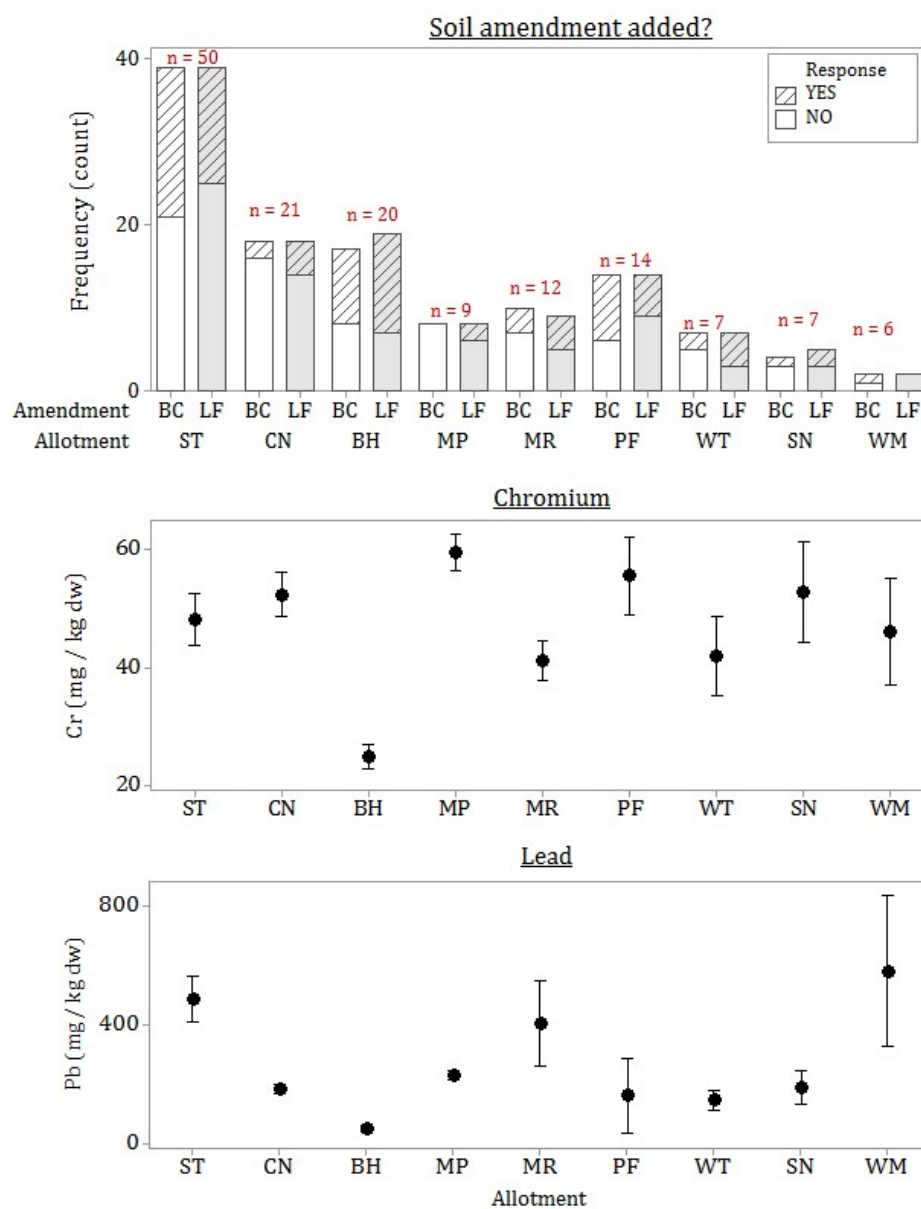


Figure 2.10: Bar chart and interval plots showing the association between the addition of soil amendments; bark chippings (BC) and liquid fertilizer (LF) and concentrations of Pb and Cr by allotment site (see Table 2.1 for site descriptions). Allotment sites are ordered according to the number of participants that replied negatively to adding bark chippings. Concentrations of Cr were high for site CN and MP which also had a large proportion of negative responses for the addition of bark chippings or liquid fertilizer. Concentrations of Pb were also high in site ST which would have counted for a large proportion of the negative responses to adding bark chippings or liquid fertilizer.

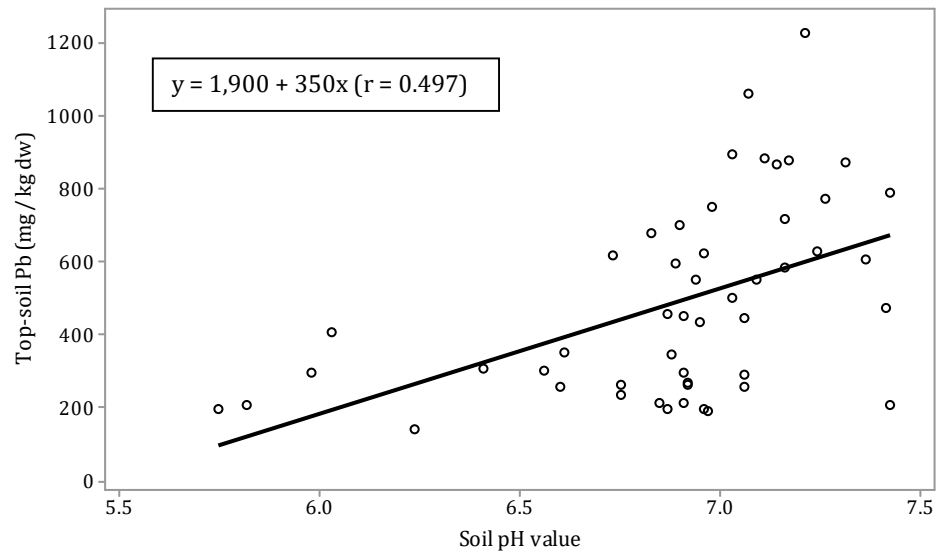


Figure 2.11: Scatterplot showing the association between soil Pb concentrations and pH across samples taken from the allotment site at St Ann's (ST).

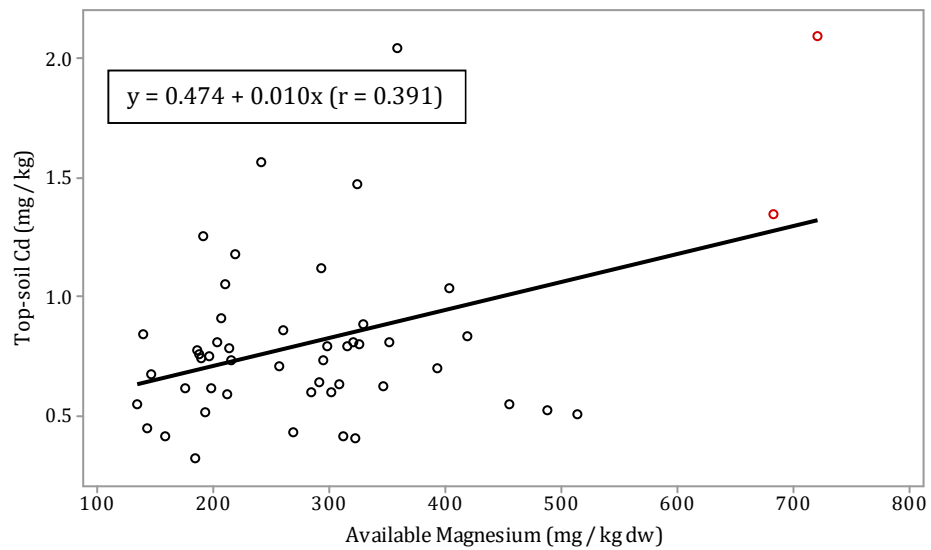


Figure 2.12: Scatterplot showing the association between available magnesium concentrations and concentrations of soil Cd across samples taken from the allotment site at St Ann's (ST). Points highlighted in red were considered to be outliers and later removed.

2.4 DISCUSSION

Comparison of metal concentrations with guidance values

There is a growing concern from allotment plot holders over the quality of their soil and whether it is safe to use to grow produce (Wakefield *et al.* 2007; Giusti 2011). Concentrations of As, Cd, Cr were found to be below the respective C4SLs in all topsoil samples collected for this study. However, concentrations of Pb were found to exceed the C4SL in 125 of the 146 top-soil samples, suggesting that plot holders in the present study could be exposed to Pb. Other metals, including Cd, Ni and Cu, were present in some of the allotment plots at concentrations above the NBC for an urban soil, but did not exceed relevant guidance values, suggesting that gardeners are not at risk from these metals.

Exposure to lead

The median concentration of Pb in top-soils across all allotment sites from the present study was 214 mg kg^{-1} ($49 - 510 \text{ mg kg}^{-1}$) - nearly three-fold higher than the C4SL of 84 mg kg^{-1} for an allotment. This is a concern as Pb exposures resulting in relatively low ($< 10 \mu\text{g dL}^{-1}$) Pb concentrations in blood can lead to intellectual impairment for children (Canfield *et al.* 2003), and are associated with renal failure and hypertension in adults (DEFRA 2013d). The enrichment of allotment soils with Pb has also been reported in a number of other studies in the UK (Davies 1978; Hursthouse *et al.* 2004; Mulloli *et al.* 2004; Giusti 2011) and

elsewhere (Hursthouse *et al.*, 2004; Kabala *et al.*, 2009) (Hursthouse *et al.* 2004; Mulloli *et al.* 2004; Giusti 2011). In particular, Mulloli *et al.* (2004) found median values of 869 mg kg⁻¹ Pb in top-soil samples from an allotment site in Newcastle-Upon-Tyne prior to its' remediation. However, median concentrations of Pb in the present study are not uncommon for an urban soil in the UK (NBC = 830 mg kg⁻¹) (Ander *et al.* 2013). To remediate the allotment sites in this study (and other allotments in the UK) to below the C4SL for Pb is therefore likely to be impractical due to the possibility of recontamination of the soil by dusts from surrounding urban areas (Brown *et al.* 2016).

Variation in metal concentrations between allotment sites

The main source of variation in HM concentrations was found to be due to differences between allotment sites. Differences in HM concentrations are likely to have arisen as a result of differences in the location and history of the allotment sites. The top-soils of allotment sites which were located closest to the city centre (MP, MR, ST, SN, WM) contained higher concentrations of HMs (Table 2.3), and were enriched relative to sub-soils (Table 2.4). Samples taken from further away from the city centre (BH, CN, PF and WT) were not enriched with these metals. There was also a contrast between the HM concentrations at older sites, ST and WM, compared to the relatively new site at BH, which had received a new top-soil covering within the last decade (Table 2.3). Several of the allotments lie close to major roads (MR, SN, ST), which could have acted as a source of Pb (Maher *et al.* 2008) or a railway line (CN, PF, MR) (Figure 2.1) which

used to run alongside a local canal that was later filled and turned into an allotment (site CN). Both the railway and canal historically supplied coal to the city. Railways are associated with higher metal emissions (Malawska and Wiołkomirski 2000) and coal can contain high concentrations of Cd and other HMs (Fernandez-Turiel *et al.* 1994). It is possible that the close proximity to the railway or canal caused the higher median concentrations of Cd at the CN site relative to the other allotment sites (Table 2.3). Metals accumulate over time in top-soils and both the proximity of an allotment site to anthropogenic activity as well as the age of exposure, are likely to be important factors in determining HM concentrations in soils (Davies 1978; Imperato *et al.* 2003).

Soil properties and soil management

The effect of differences in soil properties and soil management on HM concentrations in top-soil, was overshadowed by differences in HM concentrations and soil properties between plots on different allotment sites. There was also a large amount of intra-allotment variability in HM concentrations that could not be accounted for by differences in soil properties or soil management. The allotment top-soils were generally characterised by high amounts of organic matter (on average > 10 % SOM), a fine texture (sandy-clay loam) and a neutral pH (~7) (Table 2.5). It is expected that these soils would retain metal cations (Cd, Cu etc.), reducing their availability to be taken up by plants (McLaughlin *et al.* 2011). The soils also contained high concentrations of plant nutrients, Mg, K and P (Table 2.7). Magnesium and potassium are both likely to bind

to organic matter and clay particles, releasing HMs into solution and increasing plant uptake of HMs (Noraho and Gaur 1995). HMs may bind to phosphorus compounds thus reducing bioavailability (Bolan and Duraisamy 2003); however, there was limited evidence in the present study for a relationship between HM concentrations in top-soils and available plant nutrients. Available Mg and available P were both negatively correlated with Cd concentrations but it was unclear if these relationships were significant, or artefacts of differences in Cd or plant nutrients between the different allotment sites.

The management of allotment gardens has the potential to increase or decrease exposure to HMs. For example, some gardening practices including additions of compost and regularly turning over the soil, have the effect of diluting the concentrations of HMs in top-soils (Brown et al 2016). It was not possible to identify clear relationships between soil management practices and HM concentrations in this study, as the relatively low sample numbers from each allotment site meant that it was difficult to untangle these relationships from other sources of variation such as plot location. The majority of the plot holders reported turning over their soil or adding organic compost (90 % and 75 % of participants respectively) at least once per calendar year. Historical misuse of one allotment plot to dispose waste in one case (reported by the present plot holder), may have led to concentrations of Cd that were four fold higher than the median for the respective allotment site, and the enrichment of the soil with Cr, Cu and Ni. Punishments for the misuse of allotment plots

should be considered that reflect the severity of impact the misuse could have on subsequent plot holders to reduce exposures to HMs and other contaminants.

Considerations for risk assessment

There are a few implications of the result of this study for risk assessment practices. Firstly, the large variation in metal concentrations between allotment plots on the same site may lead to sampling errors if the whole site is not sampled prior to comparison of HM concentrations with guidance values. Sampling strategies for the assessment of contaminated land that quantify the extent of the variation in HM concentrations, should be adhered to (e.g. Taylor and Ramsey 2005). Furthermore, the bioavailability of the sites studied should be considered to derive appropriate guidance values. In the present study, the allotment soils contained higher concentrations of organic matter and had a finer soil texture than the CLEA model defaults used to derive C4SLs (6% SOM and a sandy-loam: EA 2009c). The allotment soils in the present study are therefore likely to retain cationic metals more strongly, resulting in reduced exposure from consumption of contaminated produce, compared to the CLEA model defaults.

2.5 CONCLUSIONS

This study was the first to extensively measure HM concentrations and the influence of soil properties at allotment sites across Nottingham: a city which has a long history of allotment and industrial use. Furthermore, few studies have attempted to investigate the influence of soil amendments on HM concentrations of allotment soils. The main source of variation in heavy metal concentrations was found to be between allotment sites; probably as a result of differences in location and exposure to adjacent anthropogenic activities. This appeared to outweigh the variation in HM concentrations accounted for by soil properties or management practices of allotment plots included in this study. The allotments were found to contain median concentrations of Pb above the C4SL but below the NBC. This suggests that plot holders could be at low-level risk from Pb exposure. However, remediating the soils to below the C4SL is likely to be impractical due to recontamination with Pb from the surrounding area. The heterogeneity of HM concentrations and soil properties in top-soils of allotments suggests that care should be taken when sampling for the purposes of a risk assessment. If variation in top-soil concentrations of HMs and soil properties between plots and allotment sites is not taken into account through correct sampling practices, this could lead to soils being misclassified as “contaminated” or “uncontaminated” land. Plot holders should be educated on methods such as wearing gloves and washing/peeling produce which can reduce exposure to HMs (Kim *et al.* 2014).

3 MODELLING PLANT UPTAKE OF HEAVY METALS

3.1 INTRODUCTION

Consumption of edible produce grown on soils contaminated with metals has led to concerns over public health (Murray *et al.* 2011) with exposures from dietary sources often found to be of greater concern than exposures from soil ingestion/inhalation (Zhuang *et al.* 2014). Plant uptake models are an important part of contaminated land assessments which are used by regulatory bodies to help identify and manage public health risks. However, modelling plant uptake of metals is challenging due to the variability and uncertainty of soil and plant processes (Antunes *et al.* 2006). Soil properties such as pH, organic carbon content, redox potential, cation exchange capacity, volume and type of soil colloids, all influence the bioavailability of metals to plants (Reichenberg and Mayer 2006; Degryse *et al.* 2009b). Plant processes that control uptake and translocation of metals to edible tissues vary between genotypes (Clemens 2006; Alexander *et al.* 2006) and metal concentrations tend to be higher in roots > leaves > fruit/seeds/tubers (Clemens 2006). Metal uptake is also influenced by the growing conditions for the plant such as light intensity and water/nutrient availability (Greger 1999).

Different approaches for modelling plant uptake of metals have been developed in the literature. Models have been developed based on a) the simple ratio between soil and plant concentrations referred to here as a bioconcentration factor (BCF: e.g. Baes III *et al.* 1984), b) using the

fugacity approach (e.g. New model framework (NMF), Legind and Trapp 2010), c) Freundlich – style regression models (e.g. Hough *et al.* 2004) and d) the availability of the free metal ion (Morel 1983) and modelling of interactions between elements at biological ligands (Terrestrial biotic ligand model (TBLM) – Thakali *et al.* 2006). Plant uptake models for different produce are sometimes grouped together by regulatory bodies, usually according to differences in concentration in edible plant tissues (e.g. leaves, fruits, roots), for ease-of-use in contaminated land assessment. This practice of grouping produce ignores genotypic variation in plant uptake of metals between produce and could exacerbate uncertainty in predictions made by contaminated land assessment models.

A) Bioconcentration factors

Bioconcentration factors (BCFs) (Equation 3.1.) are calculated using a simple ratio between metal concentrations in soils and plant tissues:

$$BCF = M_{plant} / M_{soil} \quad 3.1$$

Where 'M_{plant}' is the total concentration of the metal in a particular plant tissue (mg kg⁻¹ dw) and 'M_{soil}' is the total concentration of the metal in soil (mg kg⁻¹ dw). BCFs have been published by a number of authors, including for uptake of cadmium into spinach (Liang *et al.* 2013), transfer of Cd and Pb into lettuce and potato (Augustsson *et al.* 2015), plant uptake of several heavy metals into a number of leafy vegetables (Chang *et al.* 2014), for other produce such as cassava and plantain (Bortey-Sam *et al.*

2015) and from glasshouse experiments, sewage sludge amendments and field trials (Ramirez-Andreotta *et al.* 2013). Baes III et al (1984) contains a large number of BCFs for plant uptake of radionuclides and has been widely used by regulatory bodies, including the United States Environmental Protection Agency (USEPA) and The Dutch National Institute for Public Health and the Environment (RIVM), in contaminated land assessments. The RBCA model (USEPA) uses BCFs from Baes III et al (1984) and from a technical document evaluating the risks of sewage sludge application (USEPA 1992) to estimate plant uptake of As, Cd, Hg, Ni, Se into edible plant tissues.

B) Fugacity models

Models based on a fugacity approach have been developed by Legind and Trapp (2010) and the Environment Agency (EA 2009d). The model, 'new model framework' (NMF) developed by Legind and Trapp (2010) consists of a BCF from soils to edible plant tissues, adjusted for foliar uptake of metals through leaf surfaces. Plant uptake estimates in the CLEA model developed by the Environment Agency, are based on Thorne *et al.*, (2004) and make use of soil - to - solution partition coefficients (K_d) to estimate a concentration factor from soils to roots (CR). This is combined with a parameter (F_{int}) to estimate a concentration factor (BCF) for uptake of metals into different plant tissues (EA 2009d) (Equation 3.2 and 3.3).

$$CR = \frac{\delta}{(\theta_w + \rho_s \cdot K_d)} \quad 3.2$$

$$BCF = CR \cdot F_{int} \quad 3.3$$

Where δ is the soil-plant availability correction, θ_w is the water-filled soil porosity and ρ_s is the dry soil bulk density (g cm^{-3}) (EA 2009d).

C) Regression models

An advancement on the simple BCF, is to derive a regression model to describe the relationship between metal concentrations in soils and plants. Regression models have been developed by a number of authors including (Swartjes *et al.* 2013) for the CSOIL RIVM model; Jackson and Alloway (1991) for the prediction of the uptake of Cd into potato; Bacigalupo and Hale (2011), to predict uptake of arsenic and lead into a number of leaf, fruit and root vegetables, Hough *et al.* (2004) for 5 metals and 9 vegetables, and Bešter *et al.* (2013), who created models for uptake of cadmium into 9 different vegetables. Regression models can be adapted to include soil properties, such as pH and organic carbon content, known to influence plant uptake of metals. Freundlich - style regression models (e.g. Hough *et al.* 2004; Swartjes *et al.* 2013) consider plant uptake as a non-linear process: roots become saturated with metals over time reducing the diffusion gradient between root surfaces and the soil solution (Krauss *et al.* 2002) (Equation 3.4).

$$\text{Log}_{10}M_{plant} = A + B \cdot \text{Log}_{10}M_{soil} + R \quad 3.4$$

Where 'M_{plant}' is the total concentration of the metal in the plant tissue (mg kg⁻¹ dw), 'M_{soil}' is the total concentration of the metal in the soil (mg kg⁻¹ dw) and A and B are constants estimated by fitting the model. The model used by the RIVM is a Freundlich regression which includes soil pH, clay content (clay) and organic carbon content (OrgC) (Equation 3.5).

$$\text{Log}_{10} M_{\text{plant}} = A + B \cdot \text{Log}_{10} M_{\text{soil}} + C \cdot \text{pH} + D \cdot \text{Log}_{10} \text{Org C} + E \cdot \text{Log}_{10} \text{Clay} \quad 3.5$$

Models have been developed in this way by the RIVM for uptake of Cd and Pb by a number of different produce (Swartjes *et al.* 2013).

D) Free ion activity models

Lastly, plant uptake models have been developed around the principle of the activity of the free metal ion (Morel 1983) which suggests that plant uptake primarily occurs through uptake of the metal ion in solution. The terrestrial biotic ligand model (Thakali *et al.* 2006), an adaption of the biotic ligand model (BLM: Di Toro *et al.* 2001) was developed from similar principles to consider interactions of metal ions with biological ligands, and has been successfully employed to predict toxicity of metals to reduce elongation of barley roots and tomato yields (Thakali *et al.* 2006). A similar approach was used to successfully predict uptake of Cd, Cu and Zn into spinach grown on sewage sludge (Datta and Young 2004). Biotic ligand models offer an improvement to simple free ion activity models as plant uptake of metal complexes and competition between metal ions is considered (McLaughlin *et al.* 2011).

Data that can be used to create useful models to predict plant uptake of metals for the assessment of contaminated land, is scarce in the literature, as studies of metal uptake are often confined to a small number of produce and a limited range of soil characteristics (pH, organic carbon content etc.) (Hough 2002). Furthermore, the models need to be understood and easily applied by a range of stakeholders using measurable soil or plant characteristics (Hough 2002). In this chapter, plant uptake models imbedded in the exposure assessment models: Contaminated Land Exposure Assessment (CLEA: EA), CSOIL (RIVM) and Risk-Based Corrective Action (RBCA: USEPA) are compared with the Hough v2 models, updated from an existing publication (Hough *et al.* 2004), to be applicable to a greater range of produce. Comparison of these models has only been achieved once before in the literature (Legind and Trapp 2010) and only for uptake of two metals (Cd and Pb) into carrots and potatoes. The process of grouping produce for modelling plant uptake is also reviewed.

The aims of this chapter were to:

- i) Develop and evaluate Freundlich-style regression models and BCFs for uptake of 5 metals; Cd, Cu, Pb, Ni and Zn into a number of different produce from a large database containing paired soil – plant concentrations.

- ii) Develop and evaluate regression models and BCFs for uptake of metals into produce, grouped according to edible plant tissue (flower, fruit, leaf, root, tuber). Compare models created for individual produce and produce groups.

- iii) Compare and contrast the models produced in i) and ii) with those used by regulatory bodies, EA, USEPA and RIVM.

3.2 METHODS

Freundlich regression models and bioconcentration factors (BCFs) describing plant uptake of 5 metals (cadmium - Cd, copper - Cu, lead - Pb, nickel - Ni and zinc - Zn) into a number of different fruit and vegetables, were developed using the Hough database (Hough 2002). The database is comprised of paired soil-plant metal concentrations and geochemical data from peer-reviewed literature. This work follows on from a previous study (Hough *et al.* 2004) in which regression models were created to predict uptake of 5 different metals into 9 different types of fruit and vegetables. The previous models (Hough *et al.* 2004) were extended to include a wider range of produce and new models were also developed by grouping produce together according to edible plant tissues (leaf, root, fruit etc.). Models were validated using an independent database collated from peer-reviewed data and grey literature at INRA, Bordeaux (the Nguyen database: Nguyen 2014.).

3.2.1 Database management

The Hough database contains ~4,000 records of paired metal concentrations covering 5 cationic metals (Cd, Cu, Ni, Pb and Zn) and 34 different plants. Criterion for the selection of data into the database required that records contained total metal concentrations in soils which could be paired directly with total metal concentrations in plant tissues ($\text{mg kg}^{-1} \text{ dw}$). Data for soil pH and organic carbon content (in many cases this was estimated using soil organic matter (SOM) or loss on ignition

(LOI) data by multiplying by 0.58 - (Rowell 1994) were also required. Field studies were preferred to pot trials and experiments where soils were spiked with metals: a small number of pot trails and experiments involving spiked soils were included to increase the range of parameters such as soil pH and organic matter. Other conditions for acceptance of literature studies within the database can be found in (Hough 2002).

The Nguyen database (Nyguen, 2014) contains ~1500 records that are independent of the Hough database. The Nguyen database contains paired plant and soil data for the same 5 metals as the Hough database and 31 different plants - the Nguyen and Hough databases share a total of 15 different plants between them. The databases were stored together in a relational database created as part of this work using Microsoft Access 2010® (MA) (Table 3.1) (Appendix III).

Table 3.1 (a) - (h): The organisation of data in data tables within the Microsoft Access ® database.

a) Main data table

Field	Field type	Description
ID	Number	Unique indexed number for each new data entry
Metal	Text	Elemental symbols (Cd, Cu etc.)
M Soil mg/kg	Number	The concentration of the metal in the soil in mg/kg dry weight (dw)
M plant mg/kg	Number	The concentration of the metal in the plant in mg/kg dw
% Org C	Number	The Organic carbon content (%) of the soil
pH	Number	The soil pH
RefID	Number	An index relating the record to the references table
PTID	Number	An index relating the record to the plant type table
PGID	Number	An index relating the record to the plant group table
MetalID	Number	An index relating the record to the metals table

b) Additional soil data * table contains soil information not present for all records

<u>Field</u>	<u>Field Type</u>	<u>Description</u>
ID	Number	Unique indexed number related to main data table
Texture ID	Number	An index relating record to soil texture table
% sand	Number	The percentage of sand in the inorganic soil fraction
% silt	Number	The percentage of silt in the inorganic soil fraction
% clay	Number	The percentage of clay in the inorganic soil fraction
CEC (meq/100g)	Number	Cation exchange capacity of soil (milli equivalents of charge / 100g soil)
% SOM or %LOI	Number	Soil organic matter content as percentage of soil mass
Na (mg/kg)	Number	Sodium concentration in mg/kg dw soil
P (mg/kg)	Number	Phosphorus concentration in mg/kg dw soil
K (mg/kg)	Number	Potassium concentration in mg/kg dw soil
Labile M (mg/kg)	Number	The concentration of metal estimated to be available for plant uptake in mg/kg dw soil

c) Extraction Methods

<u>Field</u>	<u>Field type</u>	<u>Description</u>
ID	Number	Unique indexed number related to main data table
Total metal extraction	Text	The method used to estimate total metal concentration in soil
Labile M extraction	Text	The method used to estimate labile metal concentration in soil

d) Metals

<u>Field</u>	<u>Field Type</u>	<u>Description</u>
MetalID	Number	Indexed number relating to main data table
Metal	Text	The elemental symbol
Metal Full	Text	The full elemental name of the metal

e) Plant Group

<u>Field</u>	<u>Field Type</u>	<u>Description</u>
PGID	Number	Indexed number relating to main data table
Plant group	Text	The produce group to which the plant belongs (fruit, flower, leaf, root or tuber)

f) Plant Type

<u>Field</u>	<u>Field Type</u>	<u>Description</u>
PTID	Number	Indexed number relating to main data table
Plant type	Text	The plant being studied (lettuce, carrot, potato etc.)

g) Soil texture

<u>Field</u>	<u>Field Type</u>	<u>Description</u>
Texture ID	Number	Indexed number relating to the additional soil data table
Soil texture	Text	E.g. Sand, loamy sand, sandy loam (UK classification system)

h) Reference list

<u>Field</u>	<u>Field Type</u>	<u>Description</u>
Ref ID	Number	Indexed number relating to the main data table
Short Reference	Text	Short reference for the author of the record
Long Reference	Text	Long reference for the author of the record
Database	Number	1: Corresponds to the original Hough database 2: Corresponds to the added Nguyen database
Location	Text	Where the research was carried out
Experiment	Text	The purpose of the experiment
Field/pot/amended	Text	The type of study: field trial, pot experiment or an unoriginal spiked soil

A number of logical checks such as the removal of blanks and duplicates were performed on the MA database before it was used. An error with the derivation of some values of soil organic carbon from proxy measures (SOM or LOI) was also corrected. The produce category for broccoli was changed from 'leaf' to 'flower' after a review of the source literature to determine which plant tissues had been harvested. An additional category 'tuber' was added for tuber vegetables including potatoes and sweet potatoes (the database originally contained just 4 produce groups: leaf, fruit, flower and root).

3.2.2 Model Development

Regression models and BCFs were developed using data from the Hough database. Regression models were developed according to Hough *et al.* (2004). The regression models require paired data for the concentration

of each metal in the soil and plant (both mg/kg dw), soil pH and soil organic carbon content (% of soil mass). The calculation of BCFs required only paired data for the concentration of metal in the soil and plant (both mg/kg dw). Models were only developed for produce-metal combinations present in the Hough database for which there were a minimum of 10 data points. This allowed inclusion of some other produce such as beans that were not included in Hough *et al.* (2004) and is consistent with the number of data points often used for model development by regulatory bodies (e.g. Swartjes *et al.* 2013).

Regression models

The regression models follow a Freundlich equation from Hough, (2002) written as:

$$\text{Log}_{10}M_{\text{plant}} = A + B_1 * \text{Log}_{10}M_c + B_2 * pH \quad 3.6$$

Where M_{plant} is the total concentration of the metal in edible plant tissues (mg kg⁻¹ dw) and A, B₁ and B₂ are constants are estimated by fitting the models. The model is designed to assume that all metals are bound to humic surfaces in the soil, hence the concentration of metal available to the plant (M_c) is a function of the amount of organic carbon present in the soil (Equation 3.7).

$$M_c = \frac{(M_{\text{soil}})}{(\text{Org C}/100)} \quad 3.7$$

Where M_{soil} is the concentration of the metal in soil in mg kg⁻¹ dw and Org C is the organic carbon content (%).

Regression models were developed for uptake of each of the metals into each plant type (broccoli, cabbage, carrot etc.). A query was set up in the database to calculate/extract data for the parameters 'LogM_{plant}', 'LogM_c' and 'pH', according to plant type and metal, from the Hough database. These data were then exported to Minitab® v 17.2.1 and general linear regression models were fitted with 'LogM_{plant}' as the response variable and 'LogM_c', 'pH' as covariates.

Bioconcentration factors

BCFs were developed to describe metal uptake of each of the metals into each plant type (Equation 3.1). A query was set up in the MA database to extract paired metal concentrations in plants and soils, according to plant type and metal, from the Hough database. BCFs were calculated for each paired metal and plant data point in Microsoft Excel 2010®. The geometric mean of the BCFs was calculated and this was then used in model predictions.

Grouped Produce Models (GPMs)

Produce was grouped according to edible plant tissue (flower, fruit, leaf, root or tuber) on the basis that transport mechanisms to each plant tissue are different (Clemens 2006). Regression models and BCFs were calculated for each produce group. The data used to calculate the models for each produce type (cabbage, lettuce etc.) was combined across the

whole produce group (e.g. leafy veg) and used to derive BCFs and regression models. Summary statistics covering the Hough and Nguyen databases are presented in Table 3.2 - Table 3.6.

Table 3.2: Summary statistics (mean, minimum and maximum) for data in both the Hough and Nguyen databases used in the development and validation of produce models for **cadmium**. Models highlighted in bold font were added to the 9 models developed in Hough *et al.* (2004). Grouped produce models were developed using the combined data from individual produce. * Not used for individual produce model validations.

Produce Group	Model	Hough Database					Nguyen database				
		N	Mplant _{av.} (min-max)	Msoil _{av.} (min-max)	OrgC _{av.} (min-max)	pH _{av.} (min-max)	N	Mplant _{av.} (min-max)	Msoil _{av.} (min-max)	OrgC _{av.} (min-max)	pH _{av.} (min-max)
Flower	Broccoli	84	0.157 (0.01-0.7)	0.832 (0.01-5.8)	5.48 (2.6-13)	7.21 (5.7-8.1)	0	-	-	-	-
Fruit	Corn Cob	12	0.113 (0.03-0.2)	0.548 (0.1-1.6)	8.51 (1-45)	6.03 (4.4-7.1)	0	-	-	-	-
	Peanut	13	0.822 (0.01-2)	0.147 (0.03-0.8)	1.01 (0.4-1.9)	5.99 (4.9-6.7)	0	-	-	-	-
	Tomato	30	0.531 (0.01-1.4)	0.536 (0.1-3.2)	2.62 (1-13)	6.18 (4.4-7.1)	19	1.58 (0.3-4.2)	11.2 (0.2-34)	2.72 (0.1-5.3)	7.28 (6.4-8.0)
Leaf	Cabbage	126	0.495 (0.01-13)	1.63 (0.01-19)	5.34 (0.5-34)	6.80 (4.3-8.1)	21	2.18 (0.01-13)	3.53 (0.4-19)	2.68 (1-10)	6.56 (4.3-7.9)
	Celery	20	5.05 (1-12)	0.407 (0.01-1.2)	1.42 (1.42)	6.15 (4.8-7.5)	0	-	-	-	-
	Chard	38	2.77 (0.6-6.5)	89.3 (0.1-210)	0.784 (0.7-1.9)	6.44 (4.9-8.0)	11	1.14 (0.04-3.9)	0.833 (0.06-1.5)	2.46 (1.1-3.5)	6.92 (6.1-7.9)
	Lettuce	208	2.27 (0.05-29)	1.58 (0.01-19)	4.54 (0.9-43)	6.54 (4.4-8.1)	110	8.28 (0.05-53)	4.45 (0.09-34)	1.67 (0.6-9.8)	6.69 (4.8-9.1)
	Spinach	116	3.33 (0.2-28)	0.725 (0.01-6)	4.20 (1-10)	6.93 (4.8-8.1)	37	10.2 (0.1-110)	2.57 (0.1-20)	2.03 (0.6-5.9)	6.29 (4.2-8.0)
Root	Beet	27	0.932 (0.05-7)	4.91 (0.01-82)	2.32 (1-17)	6.35 (4.4-7.6)	3*	0.343 (0.2-0.4)	1.25 (0.7-1.5)	3.34 (3.0-3.5)	6.37 (6.1-6.5)
	Carrot	159	0.630 (0.04-7)	0.890 (0.01-6)	4.23 (1-40)	6.73 (4.8-8.1)	22	2.13 (0.08-9.2)	13.6 (0.2-34)	3.26 (0.1-5.9)	7.02 (6.1-8.1)
	Parsnip	84	0.099 (0-0.6)	0.784 (0.01-6)	5.26 (3-10)	7.24 (5.7-8.1)	0	-	-	-	-
	Radish	28	1.01 (0.01-5)	2.30 (0.1-17)	2.28 (0.9-18)	6.59 (5.7-7.8)	19	2.63 (0.01-10)	14.5 (0.7-34)	3.22 (0.7-5.3)	7.05 (6.1-8.1)
Tuber	Potato	75	0.232 (0.02-1.7)	0.651 (0.01-5.8)	4.36 (0.98-43)	6.44 (4.1-8.0)	56	0.682 (0.01-3.9)	15.9 (0.2-107)	2.09 (0.4-5.9)	7.12 (5.4-8.4)

Table 3.3: Summary statistics (mean, minimum and maximum) for data in both the Hough and Nguyen databases used in the development and validation of produce models for **copper**. Models highlighted in bold font were added to the 9 models developed in Hough *et al.* (2004). Grouped produce models were developed using the combined data from individual produce. * Not used for individual produce model validations.

Produce Group	Model	Hough Database					Nguyen database				
		N	Mplant _{av.} (min-max)	Msoil _{av.} (min-max)	OrgC _{av.} (min-max)	pH _{av.} (min-max)	N	Mplant _{av.} (min-max)	Msoil _{av.} (min-max)	OrgC _{av.} (min-max)	pH _{av.} (min-max)
Flower	Broccoli	82	5.80 (3.4-10.2)	0.845 (0.01-5.8)	5.32 (2.64-10)	7.24 (5.7-8.1)	0	-	-	-	-
Fruit	Bean	16	11.4 (9.5-16)	38.4 (2.9-180)	1.04 (1.04)	6.48 (5.9-7.1)	10	7.86 (5.5-11)	110 (9-430)	2.75 (1.1-5.8)	6.48 (5.6-7.4)
	Tomato	24	11.9 (9.7-15)	38.4 (2.9-180)	1.04 (1.04)	6.48 (5.9-7.1)	2*	11.8 (12)	10.6 (6.5-15)	0.165 (0.1-0.2)	7.73 (6.5-9.0)
Leaf	Cabbage	92	4.92 (0.7-9.6)	4.52 (0.01-180)	4.92 (0.5-10)	7.13 (5.7-8.1)	17	2.33 (0.01-6.8)	76.2 (12-430)	1.97 (1.0-5.9)	6.70 (5.0-7.9)
	Chard	12	25.0 (15-34)	497 (4.2-1400)	3.82 (1.3-5.0)	6.21 (4.9-8.0)	5*	13.6 (7.9-23)	174 (18-430)	1.75 (1.1-2.4)	6.32 (6.1-7.0)
	Lettuce	90	13.1 (3.8-29)	12.8 (0.01-290)	5.64 (1.3-24)	7.10 (4.9-8.1)	53	9.37 (0.02-40)	85.8 (6.5-640)	1.72 (0.2-15)	7.52 (5.4-9.1)
	Spinach	80	12.3 (5.9-19)	0.895 (0.01-5.8)	5.33 (2.6-10)	7.23 (5.7-8.1)	30	12.4 (0.07-19)	39.7 (9.0-210)	1.72 (0.6-5.9)	6.62 (4.7-8.0)
Root	Carrot	94	5.20 (2.8-13)	7.21 (0.01-180)	4.55 (1.0-10)	7.11 (5.7-8.1)	10	7.06 (4.4-13)	121 (9.0-430)	2.62 (0.1-5.9)	6.68 (5.6-7.4)
	Parsnip	84	5.30 (3.1-11)	0.78 (0.01-5.5)	5.26 (2.6-10)	7.24 (5.7-8.1)	0	-	-	-	-
	Radish	26	15.0 (2.4-40)	135 (2.9-640)	3.85 (1.0-24)	6.82 (5.9-7.6)	3*	11.9 (0.01-36)	34.8 (30-38)	1.59 (1.5-1.7)	6.33 (5.4-7.3)
Tuber	Potato	31	5.99 (2.3-12)	1.11 (0.03-5.8)	5.49 (3.2-9.5)	7.20 (5.7-8.0)	29	5.64 (0.3-13)	56.6 (3-210)	2.36 (0.5-10)	6.84 (4.3-8.4)

Table 3.4: Summary statistics (mean, minimum and maximum) for data in both the Hough and Nguyen databases used in the development and validation of produce models for **lead**. Models highlighted in bold font were added to the 9 models developed in Hough *et al.* (2004). Grouped produce models were developed using the combined data from individual produce.

Produce Group	Model	Hough Database					Nguyen database				
		N	Mplant _{av.} (min-max)	Msoil _{av.} (min-max)	OrgC _{av.} (min-max)	pH _{av.} (min-max)	N	Mplant _{av.} (min-max)	Msoil _{av.} (min-max)	OrgC _{av.} (min-max)	pH _{av.} (min-max)
Flower	Broccoli	82	1.68 (0.04-7.3)	1.07 (0.01-6.4)	5.13 (1.1-10)	7.20 (5.5-8.1)	0	-	-	-	-
Fruit	Bean	16	8.91 (0.5-21)	58.8 (6.5-110)	0.899 (0.8-1.0)	6.42 (5.8-7.1)	19	0.03 (0.02-0.04)	39.2 (34-44)	1.91 (1.3-2.5)	7.70 (7.6-7.8)
	Tomato	16	0.911 (0.4-1.6)	14.1 (6.5-29)	1.04 (1.04)	6.48 (5.9-7.1)	19	2.91 (0.01-23)	939 (39-5300)	2.72 (0.1-5.3)	7.28 (6.4-8.0)
Leaf	Cabbage	100	1.48 (0.03-10)	2.96 (0.01-29)	5.41 (1.0-34)	6.98 (4.7-8.1)	24	4.42 (0.01-29)	234 (20-2700)	2.20 (1.0-4.8)	6.60 (4.3-7.9)
	Lettuce	109	15.4 (0.09-120)	130 (0.01-550)	6.18 (0.6-43)	6.77 (4.4-8.1)	57	9.05 (0.01-87)	497 (16-5300)	2.16 (0.6-5.8)	7.03 (4.8-8.4)
	Spinach	82	2.29 (0.1-17)	1.36 (0.01-20)	5.23 (1.0-10)	7.22 (5.7-8.1)	32	3.70 (0.01-29)	185 (16-670)	1.79 (0.6-5.9)	6.39 (4.7-8.0)
Root	Carrot	109	1.12 (0.07-4.9)	4.04 (0.01-29)	5.51 (1.0-40)	6.96 (5.1-8.1)	32	3.37 (0.01-48)	818 (16-5300)	2.65 (0.1-5.3)	6.81 (4.8-8.1)
	Parsnip	84	0.876 (0.2-3.2)	0.784 (0.01-5.5)	5.26 (2.6-10)	7.24 (5.7-8.1)	0	-	-	-	-
	Radish	16	2.70 (0.6-7.9)	14.1 (6.5-29)	1.04 (1.04)	6.48 (5.9-7.1)	23	7.32 (0.01-33)	889 (31-5300)	2.93 (0.7-5.3)	6.98 (5.4-8.1)
Tuber	Potato	37	0.472 (0.03-2.1)	3.15 (0.03-26.4)	7.31 (1.2-43)	6.85 (4.3-8.0)	80	0.482 (0.02-15)	577 (7-5500)	2.33 (0.4-5.3)	6.62 (4.8-8.4)

Table 3.5: Summary statistics (mean, minimum and maximum) for data in both the Hough and Nguyen databases used in the development and validation of produce models for **nickel**. Grouped produce models were developed using the combined data from individual produce. *Not used for model validations of individual produce models.

Produce Group	Model	Hough Database					Nguyen database				
		N	Mplant _{av.} (min-max)	Msoil _{av.} (min-max)	OrgC _{av.} (min-max)	pH _{av.} (min-max)	N	Mplant _{av.} (min-max)	Msoil _{av.} (min-max)	OrgC _{av.} (min-max)	pH _{av.} (min-max)
Flower	Broccoli	82	1.01 (0.3-6.8)	0.845 (0.01-5.8)	5.32 (2.6-10)	7.24 (5.7-8.1)	0	-	-	-	-
Fruit	Tomato	24	2.54 (0.9-8.7)	127 (0.5-660)	1.04 (1.04)	6.48 (5.9-7.1)	0	-	-	-	-
Leaf	Cabbage	90	1.41 (0.3-22)	12.1 (0.01-660)	5.02 (1.0-10)	7.16 (5.7-8.1)	2*	0.855 (0.8-0.9)	94.6 (74-120)	2.18 (2.0-2.4)	6.20 (6.1-6.3)
	Lettuce	78	0.957 (0.3-3.5)	0.808 (0.01-5.8)	5.24 (2.6-10)	7.23 (5.7-8.1)	20	2.66 (0.1-12)	29 (13-120)	0.962 (0.6-2.4)	7.94 (6.1-9.1)
	Spinach	80	0.732 (0.09-2.1)	0.895 (0.01-5.8)	5.33 (2.6-10)	7.23 (5.7-8.1)	12	8.83 (2.6-17)	22.6 (8.4-63)	1.94 (0.6-5.9)	6.62 (4.7-7.8)
Root	Carrot	94	0.976 (0.08-13)	22.3 (0.01-660)	4.55 (1.0-10)	7.11 (5.7-8.1)	0	-	-	-	-
	Parsnip	84	0.610 (0-2.7)	0.784 (0.01-5.5)	5.26 (2.6-10)	7.24 (5.7-8.1)	0	-	-	-	-
	Radish	16	6.77 (1.3-20)	127 (0.5-660)	1.04 (1.04)	6.48 (5.9-7.1)	1*	8.25	19.6	1.69	7.3
Tuber	Potato	31	0.360 (0.08-1.5)	1.11 (0.03-5.8)	5.49 (3.2-9.5)	7.20 (5.7-8.0)	21	1.12 (0.04-7.0)	22.1 (3-180)	2.12 (0.7-4.6)	6.57 (5.4-8.4)

Table 3.6: Summary statistics (mean, minimum and maximum) for data in both the Hough and Nguyen databases used in the development and validation of produce models for **zinc**. Models highlighted in bold font were added to the 9 models developed in Hough *et al.* (2004). Grouped produce models were developed using the combined data from individual produce.

Produce Group	Model	Hough Database					Nguyen database				
		N	Mplant _{av.} (min-max)	Msoil _{av.} (min-max)	OrgC _{av.} (min-max)	pH _{av.} (min-max)	N	Mplant _{av.} (min-max)	Msoil _{av.} (min-max)	OrgC _{av.} (min-max)	pH _{av.} (min-max)
Flower	Broccoli	82	70.6 (37-170)	0.85 (0.01-5.8)	5.32 (2.6-10)	7.24 (5.7-8.1)	0	-	-	-	-
Fruit	Tomato	16	51.0 (44-65)	54.4 (16-120)	1.04 (1.04)	6.48 (5.9-7.1)	6	45.8 (0.01-79)	1410 (9.0-7600)	2.81 (0.1-5.3)	7.23 (6.4-9.0)
Leaf	Cabbage	102	75.5 (12-1030)	41.3 (0.01-830)	4.75 (0.5-10)	7.01 (4.3-8.1)	24	65.9 (0.02-450)	535 (19-3000)	2.09 (1.0-4.8)	6.47 (4.3-7.9)
	Chard	12	240 (41-460)	1360 (8.9-4100)	1.31 (0.7-2.8)	6.21 (4.9-8)	7	136 (0.01-630)	1320 (18-3000)	1.77 (1.1-2.4)	6.69 (6.1-7.6)
	Lettuce	83	85.5 (42.1-180)	1.82 (0.01-38)	5.06 (1.35-10)	7.15 (4.9-8.1)	57	105 (0.01-500)	778 (8.3-7600)	2.10 (0.2-15)	7.45 (5.4-9.1)
	Spinach	80	218 (73-590)	0.895 (0.01-5.8)	5.33 (2.6-10)	7.23 (5.7-8.1)	24	119 (0.9-210)	325 (45-1400)	1.48 (0.6-5.9)	6.80 (4.7-8.0)
Root	Carrot	94	33.2 (10-230)	9.93 (0.01-120)	4.56 (1.0-10)	7.11 (5.7-8.1)	23	48.5 (0.01-120)	1630 (8.8-7600)	2.85 (0.1-5.9)	7.13 (6.1-8.1)
	Parsnip	84	24.0 (12-65)	0.784 (0.01-5.5)	5.26 (2.6-10)	7.24 (5.7-8.1)	0	-	-	-	-
	Radish	16	65.9 (42-190)	54.4 (16-120)	1.04 (1.04)	6.48 (5.9-7.1)	30	152 (0.01-860)	1370 (18-7600)	2.56 (0.7-5.3)	7.00 (5.4-8.1)
Tuber	Potato	31	18.2 (9.2-45)	1.11 (0.03-5.8)	5.49 (3.2-9.5)	7.20 (5.7-8.0)	69	28.6 (0.5-170)	1150 (1.5-11000)	2.12 (0.4-10.3)	6.93 (4.3-8.4)

3.2.3 Model validation

Regression models and BCFs were evaluated using the Hough database, used to derive the models, and validated with independent data from the Nguyen database. Predictions of M_{plant} were made using the regression equations and by multiplying BCFs by M_{soil} . Root Mean Squared Logarithmic Errors (RMSLEs) were calculated between the measured and modelled data using the logarithmic scale so that the errors were not skewed by large differences in the measured data between plant groups (Equation 3.8) (Hosking and Wallis, 2005).

$$RMSLE = \sqrt{\left[\frac{\sum_{i=1}^n (\log_{10} X_i - \log_{10} Y_i)^2}{n} \right]} \quad 3.8$$

Where RMSE is an expansion on the more commonly used Root Mean Square Error (RMSLE) (Hosking and Wallis, 2005), X_i represents the i^{th} measured value, Y_i the i^{th} modelled value and n the total number of values of X and Y .

A comparison of regressions method using analysis of variance (ANOVA), was employed to test the null hypothesis that there is no difference in model fit between using individual produce models (IPMs) and grouped produce models (GPMs). The regression sum of squares from the GPMs (e.g. leaf) was compared to the total regression sum of squares from IPMs (e.g. lettuce, spinach, cabbage) of the produce that formed the group (estimated from residual sums of squares:

Table 3.7: ANOVA table example for comparison of regression models between individual and grouped produce regressions where: n = the number of individual produce in the produce in the group, SSR = sum of squares of the regression; SSE = the sum of squares of the error (residuals) and SST = the total sum of squares.

Model	DF	SS	MS	F	P
Group	A:2	a: Group SSR	a/A	$(a/A)/(c/C)$	(Calculated from statistical tables)
Individual	B: 2n	b: Group SSE - C	b/B	$(b/B)/(c/C)$	
Error	C: Total - (2n+2)	c: Σ Ind SSE	c/C		
Totals	D: Total	d: Group SST			

3.2.4 Comparisons to other models

The regression models (subsequently referred to as the Hough v2 models) and BCFs were compared to models developed by EA, USEPA and RIVM using the Nguyen database. Errors (RMSLEs) were calculated between measured and predicted metal concentrations in plant tissues. Descriptions of each of the models were provided in Section 3.1.

EA, CLEA model

Models are published as BCFs in DEFRA (2013b) and DEFRA (DEFRA 2013e) for Cd and Pb respectively. Sub-models for 'green vegetables', 'herbaceous fruits' and 'potatoes' from DEFRA (2013b) and DEFRA (DEFRA 2013e) were used to predict metal accumulations in leaves, fruits and tubers respectively.

USEPA, RBCA toolkit

USEPA models (BCFs) for plant uptake from soils published in (USEPA 1996). The 'garden fruits' and 'potato' sub-models were used to estimate uptake into edible fruits and tubers.

RIVM, CSOIL

RIVM regression models for Cd and Pb are published in (Swartjes *et al.* 2013) for the CSOIL model for individual produce. Produce are subsequently grouped in the CSOIL model according to edible tissues and weighted by produce consumption. A set of consumption-weighted BCFs for produce groups from Swartjes *et al.*, (2013) based on a standard set of soil parameters (pH = 7, OrgC = 2%, Clay = 5%, Cd = 3 mg kg⁻¹ dw, Pb = 100 mg kg⁻¹ dw) was used for comparisons with models from other regulatory bodies. Regression models for individual produce from CSOIL were compared with the Hough v2 models.

Table 3.8: BCFs produced for the purpose of modelling plant uptake by a) the Environment agency for cadmium (DEFRA 2013b), lead (DEFRA 2013d) and nickel (EA, 2009d); b) USEPA (USEPA 1996); c) RIVM (Swartjes et al 2013) and adapted for comparisons with the Hough v2 models. Dashes indicate occasions where models had not been developed for metal – produce combinations.

<u>Metal</u>	<u>Model</u>	<u>EA (CLEA)_a</u>	<u>USEPA (RBCA)_b</u>	<u>RIVM (CSOIL)_c</u>
<u>Cd</u>	Leaf	0.542	0.364	0.362
	Fruits	0.276	0.090 ₂	-
	Tuber	0.148	0.008 ₃	0.156 ₃
	Root	0.282	0.064	0.260
<u>Pb</u>	Leaf	0.044	-	0.026
	Fruits	0.013	-	0.010
	Tuber	0.003	-	0.006
	Root	0.039	-	0.015
<u>Ni</u>	Leaf	0.040	0.032	-
	Fruits	0.043	0.006	-
	Tuber	0.009	0.010	-
	Root	0.042	0.008	-
<u>Zn</u>	Leaf	-	0.250	-
	Fruits	-	0.046	-
	Tuber	-	0.024	-
	Root	-	0.044	-

Table 3.9: Model parameters used to estimate plant uptake using RIVM regression models (Swartjes *et al*, 2007). Models listed here were found to be statistically significant (Versluijs and Otte, 2001). Individual CSOIL regression models were used to estimate M_{plant} in the Hough database as the models required clay content; not included in the Nguyen database.

<u>Metal</u>	<u>Model</u>	<u>A (const.)</u>	<u>B (Msoil)</u>	<u>C (pH)</u>	<u>D (% OC)</u>	<u>E (% clay)</u>
<u>Cd</u>	Beetroot	1.90	0.37	-0.18	-0.30	-0.80
	Carrot	0.74	0.45	-0.16	0.20	0.09
	Lettuce	1.00	0.28	-0.18	-0.19	0.16
	Radish	0.00	0.12	-0.32	0.00	0.00
	Spinach	1.30	0.28	-0.22	-0.64	0.37
<u>Pb</u>	Beans	3.20	-0.10	-0.20	-5.30	-
	Beetroot	0.50	0.75	-0.08	-0.64	-1.20
	Cabbage	7.00	0.60	-0.30	-3.10	-5.00
	Carrot	-0.64	0.56	-0.04	-0.16	-0.03
	Lettuce	-0.60	0.90	-0.07	-0.34	-0.19
	Spinach	-0.12	0.36	-0.03	0.25	-0.23

3.3 RESULTS AND DISCUSSION

3.3.1 Modelling metal uptake into individual produce

Freundlich regression models (Hough v2) and BCFs, were derived from the Hough database to predict concentrations of Cd, Cu, Ni, Pb and Zn in edible plant tissues (Table 3.11 - Table 3.15) from different types of produce (lettuce, parsnip, potato etc.). The Hough v2 models for the uptake of Cd, Ni and Zn by individual produce, yielded R^2_{adj} values in the range ($<0.001:0.886$; $n = 12:208$), consistent with results from a similar model in the literature (Swartjes *et al.* 2013). Parameter values for β_1 (Mc) and β_2 (pH) were significantly different from zero ($P < 0.05$) for most of the plant uptake models for Cd, Ni and Zn (exceptions include models for Zn – tomato for which both parameters were insignificant; Ni – lettuce, Ni – spinach and Ni – potato for which β_1 (Mc) was insignificant and Cd – radish for which β_2 (pH) was insignificant). The correlations between modelled and measured data using the Hough v2 models are displayed in Figure 3.1- Figure 3.5 starting on page 110.

The Hough v2 models for plant uptake of Cu and Pb, were not a good fit to data from the Hough database from which they were derived. Many of the models for both Cu and Pb had low R^2_{adj} values (R^2_{adj} values were <0.001 for Cu – tomato ($n=24$), Cu– spinach ($n=80$), Pb – spinach ($n=82$) and Pb – radish ($n=16$)). On several occasions, the model parameters β_1 and β_2 were also not significant in the models ($P > 0.05$). The Hough v2 models for Cu seemed to predict measured concentrations (of Cu) in edible plant

tissues from the Hough database (RMSLE values in the range 0.142 – 0.418), and the Nguyen database (RMSLE values in the range 0.138 – 0.660), reasonably well. However, the variation around mean values of Cu in edible plant tissues in both databases is small, which may have resulted in similar RMSLE values compared to predictions of Cd, Ni and Zn (Figure 3.2).

The Hough v2 models performed better at predicting metal concentrations in edible plant tissues than BCFs, yielding lower RMSLE values (Table 3.11 - Table 3.15). The prediction of Cd concentrations in plants from the Nguyen database, yielded similar RMSLE values (in the range 0.3 – 1.2) regardless of whether the Hough v2 models or BCFs were used (Table 3.11). However, errors for the prediction of concentrations of Cu, Ni, Pb and Zn in produce from the Nguyen database using BCFs were an order of magnitude higher for some produce-metal combinations (e.g. Pb – spinach, Pb – carrot: Table 3.13) than errors using the Hough v2 models. The calculation of BCFs is highly dependent on metal concentrations in the soil in the data from which they are derived, with lower metal concentrations yielding higher BCFs and visa-versa (Wang *et al.* 2006; Hao *et al.* 2011). For some produce-metal combinations in the Hough database (e.g. Ni – lettuce: Table 3.5), the range of metal concentrations in the database was restricted due to the availability of data in the literature.

Parameter values for the Hough v2 models of Cu and Pb, were sometimes negative for B_1 (Mc) and/or positive for B_2 (pH). This is at odds with the consensus in the literature which suggests that concentrations of cationic metals in edible plant tissues should increase with higher concentrations of metals in the soil, lower organic carbon content and lower pH (Greger 1999; McLaughlin *et al.* 2011). Some authors have suggested more complex responses to pH for metals (Podar and Ramsey 2005) and accumulations of organic matter from compost (Murray *et al.* 2011). It is more likely, though, that the poor model fits for both the Hough v2 models and BCFs for Cu and Pb, relate to differences in plant uptake of these metals compared to the other cationic metals. Predicting metal uptake into edible plant tissues for Cu and Pb may be difficult as Cu and Pb are often strongly bound to soil particles (Alloway and Ayres 1997). Copper has been observed to accumulate primarily in the root zone of lettuces (Intawongse and Dean 2006) and foliar uptake of Pb from atmospheric pollution may contribute to Pb accumulations in aerial plant tissues, especially in gardens and allotments near roads or industrial polluters (Harrison and Chirgawi 1989; Alegría *et al.* 1991; Säumel *et al.* 2012; Schreck, Foucault, *et al.* 2012).

Table 3.10. Derivation and evaluation of regression models and BCFs for plant uptake of Cd. P-values for the significance of regression parameters are given in brackets. In all cases, models were derived using data from the Hough database and evaluated using both Hough and Nguyen databases. H = Hough database, N = Nguyen database. Dashes indicate where data from Nguyen database were insufficient to make comparisons.

<u>Metal: cadmium</u>			<u>Regression Models</u>						<u>Bioconcentration Factors</u>			
Group	Produce	N	<i>Development</i>			<i>Performance</i>			<i>Development</i>		<i>Performance</i>	
			α : Const.	B_1 : Mc	B_2 : pH	R^2_{adj}	RMSLE-H	RMSLE-N	BCF _(Geomean.)	Min – Max	RMSLE-H	RMSLE-N
Flower	Broccoli	84	-0.632	0.342 (<0.001)	-0.078 (0.089)	0.330	0.207	-	0.298	0.04 – 8.0	0.442	-
Fruit	Corn Cob	12	-2.471	0.246 (0.003)	0.192 (0.001)	0.886	0.466	-	0.248	0.03 – 1.1	0.525	-
	Peanut	13	0.910	2.084 (0.019)	-0.604 (0.067)	0.496	0.581	-	2.41	0.14 – 20	0.724	-
	Tomato	30	-5.308	0.790 (<0.001)	0.598 (<0.001)	0.822	0.295	0.954	0.802	0.003 – 7.9	0.838	0.825
Leaf	Cabbage	126	-0.401	0.427 (<0.001)	-0.135 (0.001)	0.416	0.357	0.695	0.250	0.02 – 13	0.498	0.573
	Celery	20	1.180	0.560 (0.138)	-0.210 (0.002)	0.912	0.095	-	14.5	3.9 – 170	0.430	-
	Chard	38	0.585	0.130 (0.001)	-0.131 (0.035)	0.347	0.299	0.821	0.215	0.02 – 26	1.06	0.720
	Lettuce	208	-0.543	0.605 (<0.001)	-0.045 (0.186)	0.492	0.398	0.511	1.24	0.08 – 38	0.441	0.449
	Spinach	116	1.240	0.619 (<0.001)	-0.237 (<0.001)	0.611	0.272	0.727	4.44	0.35 – 260	0.541	0.819
Root	Beetroot	27	0.472	0.539 (<0.001)	-0.253 (<0.001)	0.610	0.282	-	0.839	0.018 – 40	0.741	-
	Carrot	159	0.685	0.253 (<0.001)	-0.220 (<0.001)	0.303	0.366	0.514	0.662	0.04 – 160	0.641	0.691
	Parsnip	84	0.377	0.194 (0.011)	-0.232 (0.001)	0.144	0.232	-	0.173	0.009 – 1.7	0.674	-
	Radish	28	-0.720	0.322 (0.045)	-0.003 (0.985)	0.091	0.437	0.781	0.875	0.02 – 11	0.802	1.20
Tuber	Potato	75	-1.212	0.421 (<0.001)	-0.009 (0.836)	0.165	0.391	0.347	0.496	0.013 – 19	0.543	0.923

Table 3.12. Derivation and evaluation of regression models and BCFs for plant uptake of Cu. P-values for the significance of regression parameters are given in brackets. In all cases, models were derived using data from the Hough database and evaluated using both Hough and Nguyen databases. H = Hough database, N = Nguyen database. Dashes indicate where data from Nguyen database were insufficient to make comparisons.

Metal: copper			Regression Models						Bioconcentration Factors			
Group	Produce	N	Development			Performance			Development		Performance	
			α : Const.	B ₁ : Mc	B ₂ : pH	R ² _{adj}	RMSLE-H	RMSLE-N	BCF _(Geomean.)	Min – Max	RMSLE-H	RMSLE-N
Flower	Broccoli	82	0.921	0.067 (0.002)	-0.032 (0.102)	0.098	0.084	-	12.6	1.3 – 560	0.492	-
Fruit	Bean	16	0.389	0.076 (0.100)	0.0658 (0.287)	0.100	0.050	0.227	0.894	0.06 – 4.1	0.583	0.933
	Tomato	24	0.892	0.009 (0.791)	0.0236 (0.598)	<0.001	0.047	-	0.911	0.06 – 4.7	0.607	-
Leaf	Cabbage	92	0.550	-0.074 (<0.001)	0.030 (0.259)	0.192	0.137	0.660	6.99	0.03 – 420	0.754	2.26
	Chard	12	2.392	-0.117 (0.015)	-0.101 (0.022)	0.482	0.095	-	0.323	0.01 – 8.24	1.10	-
	Lettuce	90	1.200	-0.037 (0.040)	-0.008 (0.719)	0.027	0.121	0.531	15.6	0.03 – 890	0.880	2.00
	Spinach	80	1.096	-0.0213 (0.289)	0.001 (0.942)	<0.001	0.080	0.415	25.8	2.0 – 1200	0.527	1.89
Root	Carrot	94	0.848	0.094 (<0.001)	-0.040 (0.079)	0.453	0.108	0.138	6.23	0.05 – 300	0.680	0.302
	Parsnip	84	0.791	0.0875 (<0.001)	-0.022 (0.286)	0.131	0.091	-	11.8	1.3 – 340	0.467	-
	Radish	26	-2.827	0.310 (<0.001)	0.416 (<0.001)	0.655	0.206	-	0.296	0.04 – 3.0	0.557	-
Tuber	Potato	31	0.134	0.079 (0.200)	0.097 (0.097)	0.053	0.148	0.631	10.7	0.6 – 240	0.548	2.02

Table 3.13. Derivation and evaluation of regression models and BCFs for plant uptake of Pb. P-values for the significance of regression parameters are given in brackets. In all cases, models were derived using data from the Hough database and evaluated using both Hough and Nguyen databases. H = Hough database, N = Nguyen database. Dashes indicate where data from Nguyen database were insufficient to make comparisons.

<u>Metal: lead</u>			<u>Regression Models</u>						<u>Bioconcentration Factors</u>			
Group	Produce	N	<i>Development</i>			<i>Performance</i>			<i>Development</i>		<i>Performance</i>	
			α : Const.	B ₁ : Mc	B ₂ : pH	R ² _{adj}	RMSLE-H	RMSLE-N	BCF _(Geomean.)	Min – Max	RMSLE-H	RMSLE-N
Flower	Broccoli	82	-0.321	-0.028 (0.682)	0.073 (0.144)	0.002	0.217	-	3.42	0.2 – 320	0.570	-
Fruit	Bean	16	-2.465	1.211 (<0.001)	-0.211 (0.063)	0.950	0.142	1.866	0.0982	0.03 – 0.21	0.259	2.13
	Tomato	16	-0.568	0.404 (0.131)	-0.117 (0.315)	0.052	0.164	1.265	0.0655	0.02 – 0.18	0.218	1.56
Leaf	Cabbage	100	-2.077	-0.169 (<0.001)	0.330 (<0.001)	0.546	0.177	1.238	1.26	0.002 – 220	1.03	1.91
	Lettuce	109	0.304	0.433 (<0.001)	-0.091 (0.094)	0.742	0.246	1.277	0.967	0.007 – 110	0.843	1.99
	Spinach	82	-0.391	-0.021 (0.742)	0.089 (0.229)	<0.001	0.174	0.847	3.26	0.006 – 110	0.635	2.57
Root	Carrot	109	-1.430	-0.005 (0.884)	0.199 (<0.001)	0.168	0.311	0.889	0.882	0.003 – 58	0.917	1.88
	Parsnip	84	0.702	-0.049 (0.492)	-0.112 (0.085)	0.027	0.283	-	1.61	0.04 – 36	0.581	-
	Radish	16	-1.080	0.325 (0.599)	0.053 (0.846)	<0.001	0.418	0.741	0.143	0.03 – 0.58	0.414	1.45
Tuber	Potato	37	-1.849	-0.236 (0.014)	0.237 (<0.001)	0.493	0.312	0.738	0.354	0.001 – 29	1.03	2.65

Table 3.14. Derivation and evaluation of regression models and BCFs for plant uptake of Ni. P values for the significance of regression parameters are given in brackets. Models were derived using data from the Hough database and evaluated using both Hough and Nguyen databases: H = Hough database, N = Nguyen database. Dashes indicate where data from Nguyen database were insufficient to make comparisons.

<u>Metal: nickel</u>			<u>Regression Models</u>						<u>Bioconcentration Factors</u>			
Group	Produce	N	<i>Development</i>			<i>Performance</i>			<i>Development</i>		<i>Performance</i>	
			α : Const.	B ₁ : Mc	B ₂ : pH	R ² _{adj}	RMSLE-H	RMSLE-N	BCF _(Geomean.)	Min – Max	RMSLE-H	RMSLE-N
Flower	Broccoli	82	1.594	0.200 (<0.001)	-0.260 (<0.001)	0.293	0.210	-	1.77	0.1 – 45	0.524	-
Fruit	Tomato	24	-2.297	0.141 (0.002)	0.341 (0.002)	0.392	0.172	-	0.222	0.003 – 3.5	0.999	-
Leaf	Cabbage	90	1.229	0.254 (<0.001)	-0.227 (<0.001)	0.497	0.255	-	1.27	0.009 – 41	0.605	-
	Lettuce	78	1.310	0.067 (0.194)	-0.201 (<0.001)	0.186	0.198	0.521	1.86	0.07 – 55	0.544	1.55
	Spinach	80	0.808	0.066 (0.221)	-0.147 (0.004)	0.087	0.214	0.875	1.36	0.03 – 28	1.70	0.553
Root	Carrot	94	1.218	0.160 (<0.001)	-0.234 (0.001)	0.336	0.328	-	0.768	0.001 – 20	0.739	-
	Parsnip	84	2.691	0.463 (<0.001)	-0.489 (<0.001)	0.322	0.404	-	0.897	0.02 – 30	0.563	-
	Radish	16	-3.620	0.328 (<0.001)	0.514 (0.005)	0.638	0.211	-	0.490	0.01 – 7.0	0.864	-
Tuber	Potato	31	0.888	0.147 (0.133)	-0.219 (0.017)	0.148	0.235	0.801	0.550	0.03 – 9.3	0.575	1.43

Table 3.15. Derivation and evaluation of regression models and BCFs for plant uptake of Zn. P-values for the significance of regression parameters are given in brackets. In all cases, models were derived using data from the Hough database and evaluated using both Hough and Nguyen databases. H = Hough database, N = Nguyen database. Dashes indicate where data from Nguyen database were insufficient to make comparisons.

<u>Metal: zinc</u>			<u>Regression Models</u>						<u>Bioconcentration Factors</u>			
Group	Produce	N	<i>Development</i>			<i>Performance</i>			<i>Development</i>		<i>Performance</i>	
			α : Const.	B ₁ : Mc	B ₂ : pH	R ² _{adj}	RMSLE-H	RMSLE-N	BCF _(Geomean.)	Min – Max	RMSLE-H	RMSLE-N
Flower	Broccoli	82	2.779	0.127 (<0.001)	-0.149 (<0.001)	0.315	0.118	-	148	13 – 5900	0.493	-
Fruit	Tomato	16	2.107	-0.049 (0.554)	-0.035 (0.515)	<0.001	0.048	0.285	1.14	0.4 – 3.2	0.286	1.50
Leaf	Cabbage	102	2.881	0.010 (0.689)	-0.169 (<0.001)	0.190	0.252	1.373	37.5	0.08 – 4200	1.02	2.91
	Chard	12	4.178	-0.247 (0.001)	-0.150 (0.066)	0.692	0.200	-	0.954	0.02 – 52	1.39	1.95
	Lettuce	83	2.901	0.068 (0.001)	-0.150 (<0.001)	0.495	0.111	0.872	145	4.8 – 7100	0.574	2.85
	Spinach	80	3.269	0.137 (0.003)	-0.154 (<0.001)	0.183	0.180	0.938	411	30 – 1000	0.503	2.84
Root	Carrot	94	2.246	0.103 (<0.001)	-0.133 (<0.001)	0.436	0.178	1.063	28.4	0.2 – 1900	0.780	3.01
	Parsnip	84	2.408	0.107 (0.007)	-0.161 (<0.001)	0.213	0.154	-	50.0	4.8 – 1800	0.501	-
	Radish	16	-1.940	0.306 (0.166)	0.402 (0.010)	0.392	0.125	1.045	1.35	0.3 – 6.8	0.373	1.39
Tuber	Potato	31	1.805	0.115 (0.046)	-0.096 (0.063)	0.126	0.135	0.573	32.5	2.9 - 830	0.533	2.77

Cadmium

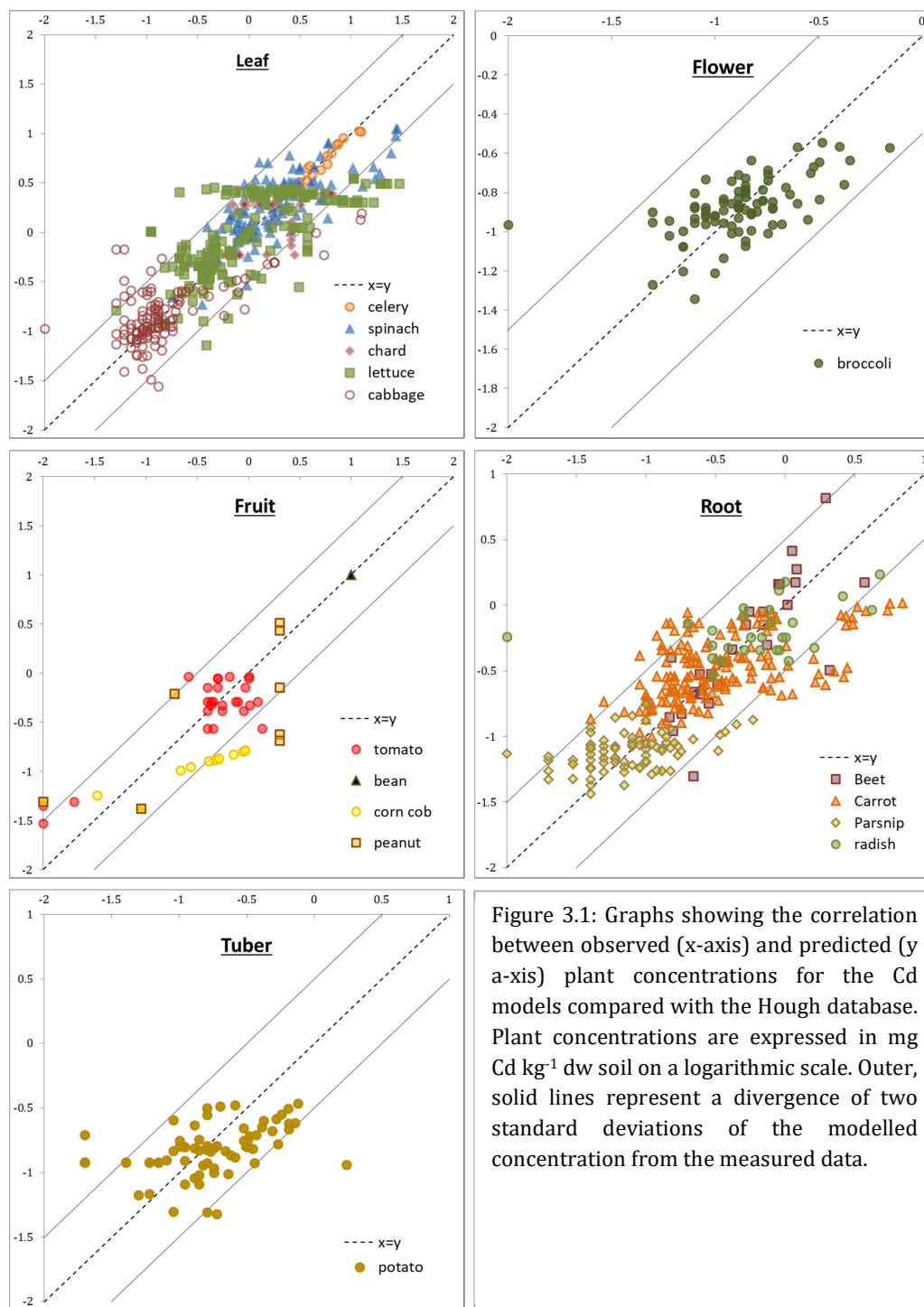


Figure 3.1: Graphs showing the correlation between observed (x-axis) and predicted (y-axis) plant concentrations for the Cd models compared with the Hough database. Plant concentrations are expressed in $\text{mg Cd kg}^{-1} \text{ dw soil}$ on a logarithmic scale. Outer, solid lines represent a divergence of two standard deviations of the modelled concentration from the measured data.

Copper

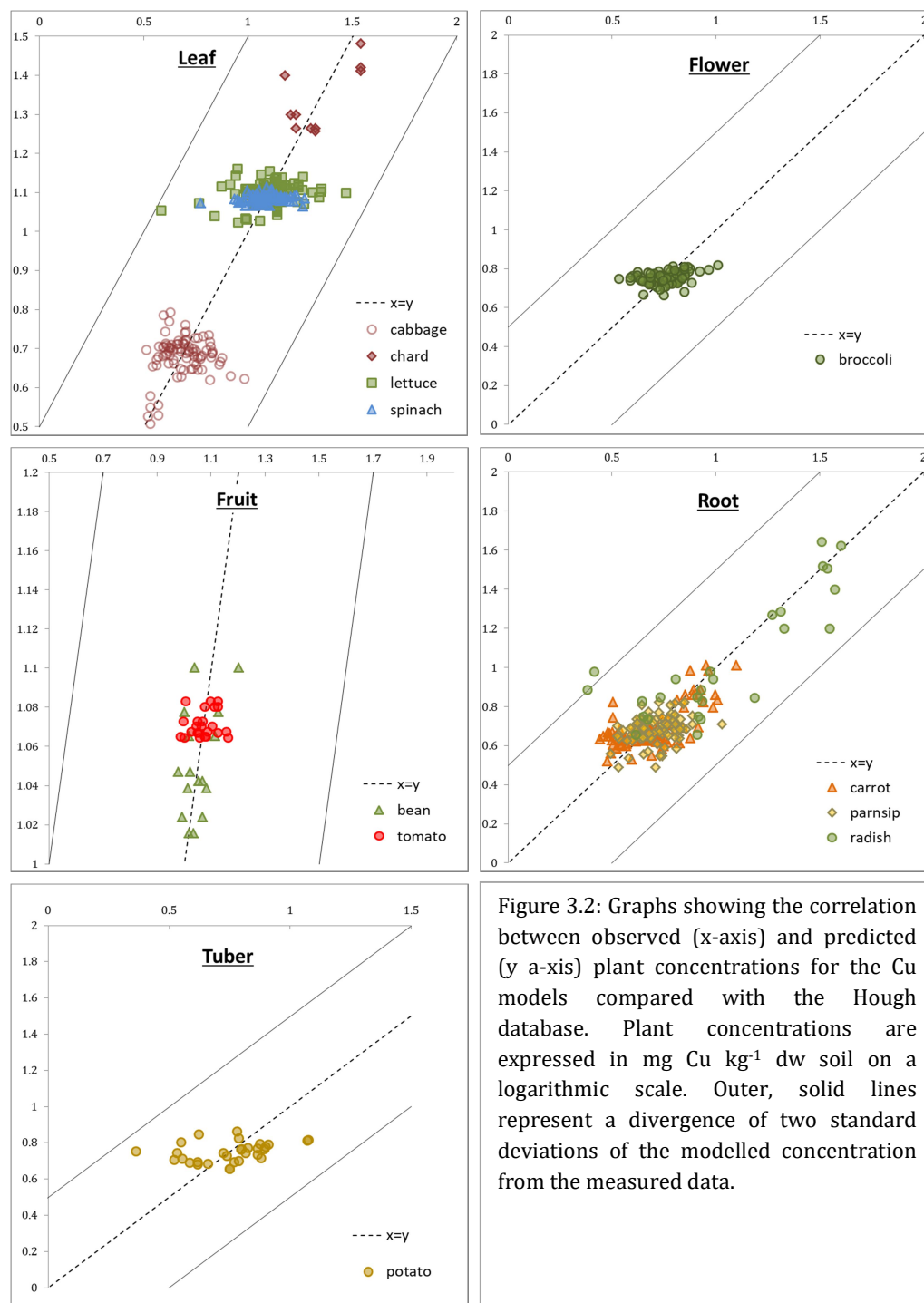


Figure 3.2: Graphs showing the correlation between observed (x-axis) and predicted (y a-xis) plant concentrations for the Cu models compared with the Hough database. Plant concentrations are expressed in $\text{mg Cu kg}^{-1} \text{ dw soil}$ on a logarithmic scale. Outer, solid lines represent a divergence of two standard deviations of the modelled concentration from the measured data.

Lead

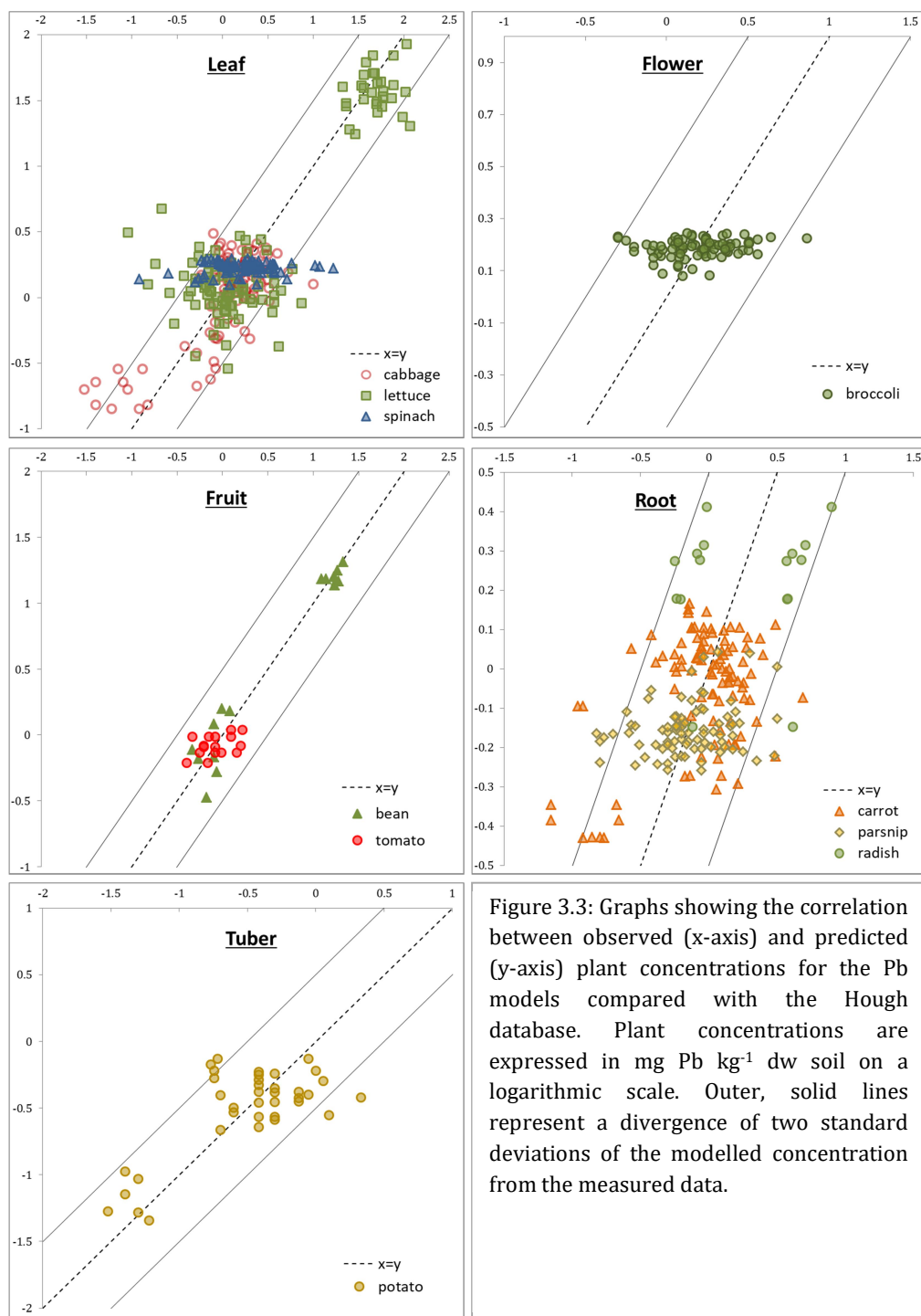


Figure 3.3: Graphs showing the correlation between observed (x-axis) and predicted (y-axis) plant concentrations for the Pb models compared with the Hough database. Plant concentrations are expressed in $\text{mg Pb kg}^{-1} \text{ dw soil}$ on a logarithmic scale. Outer, solid lines represent a divergence of two standard deviations of the modelled concentration from the measured data.

Nickel

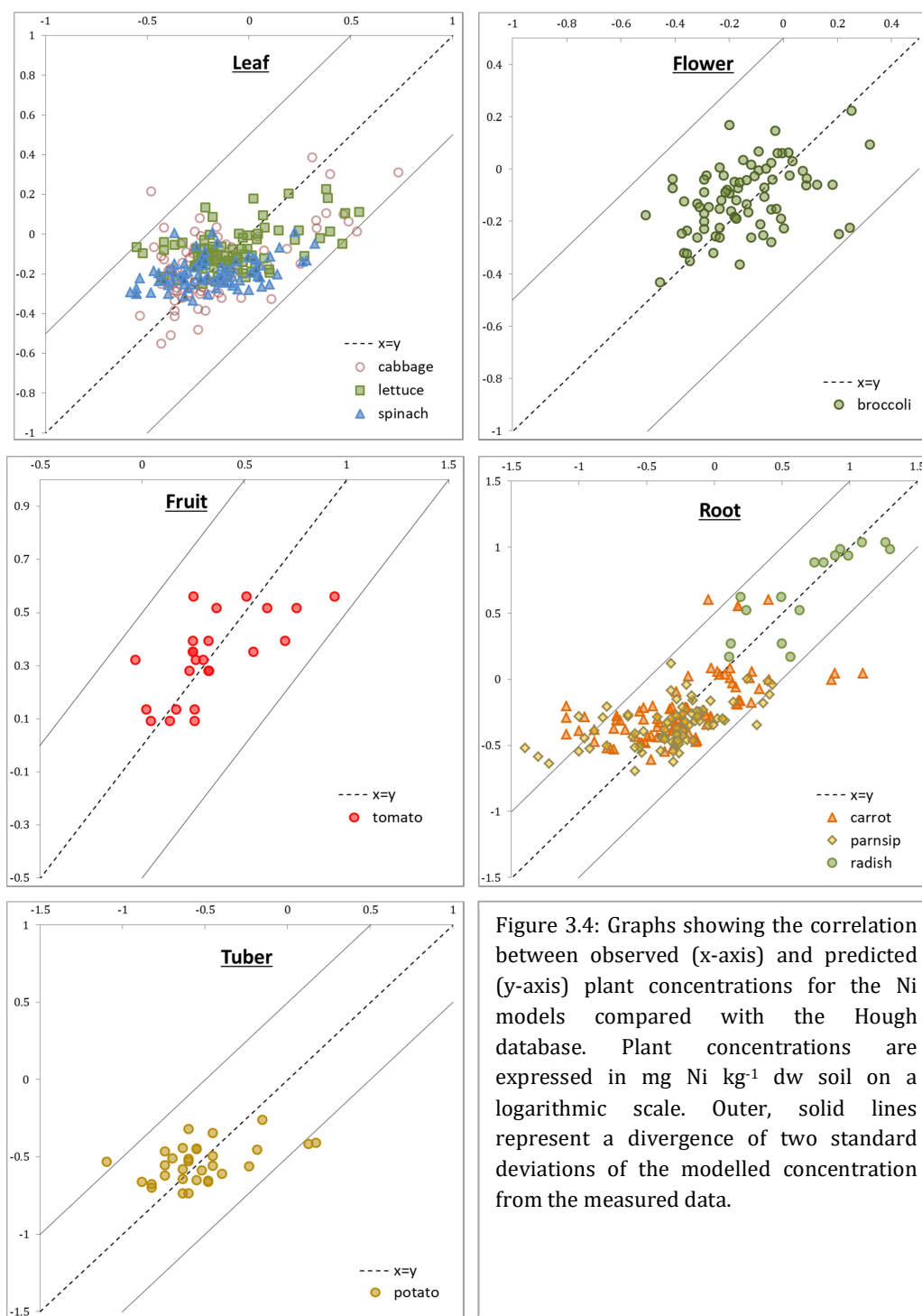


Figure 3.4: Graphs showing the correlation between observed (x-axis) and predicted (y-axis) plant concentrations for the Ni models compared with the Hough database. Plant concentrations are expressed in $\text{mg Ni kg}^{-1} \text{ dw soil}$ on a logarithmic scale. Outer, solid lines represent a divergence of two standard deviations of the modelled concentration from the measured data.

Zinc

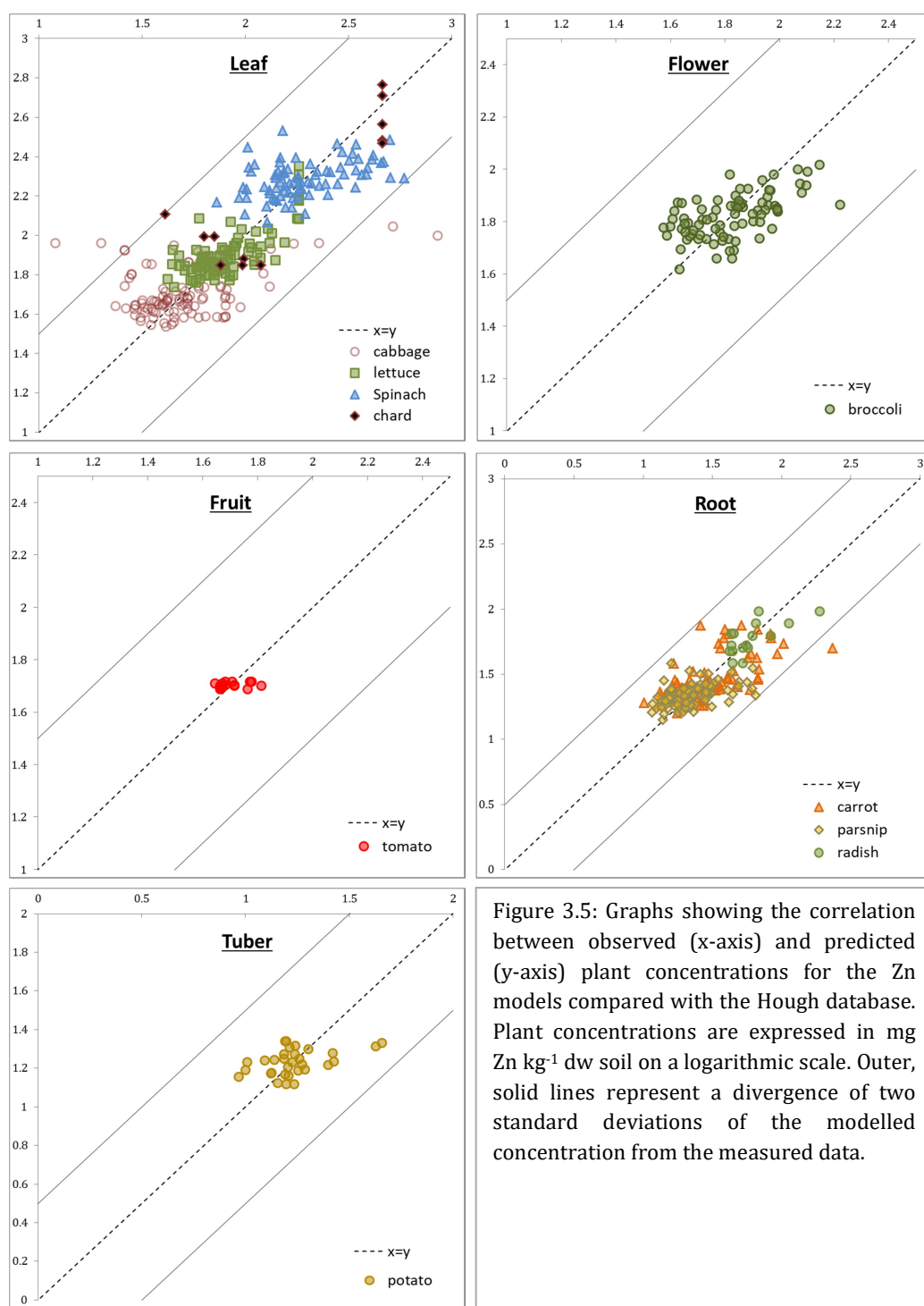


Figure 3.5: Graphs showing the correlation between observed (x-axis) and predicted (y-axis) plant concentrations for the Zn models compared with the Hough database. Plant concentrations are expressed in $\text{mg Zn kg}^{-1} \text{ dw soil}$ on a logarithmic scale. Outer, solid lines represent a divergence of two standard deviations of the modelled concentration from the measured data.

3.3.2 Modelling metal uptake into produce grouped by edible tissue

Freundlich regression models (Hough v2) and BCFs were derived across whole produce groups (GPMs), grouped according to edible plant tissue (fruit, flower, leaf, root, tuber) (Table 3.17 and Table 3.16). There were similarities with the models derived for individual produce (IPMs). Predictions of metal concentrations in edible produce tissues using the GPMs were closer to measured values (lower RMSLE values) when the Hough v2 models were used to make predictions as opposed to BCFs. Model fits (R^2_{adj} values) for Hough v2 models of Cu and Pb uptake were generally worse than for Cd, Ni and Zn. The Hough v2 models for predicting uptake of Cu into leaf and fruit tissues, did not correlate well with data from the Hough database, and model parameters β_1 (Mc) and β_2 (pH) were not significant ($R^2_{adj} < 0.001$; $n=40$ and 274 respectively; $p > 0.05$). Similarly, the Hough v2 models for Pb – Roots and Zn – Leaves did not fit well to data from the Hough database ($R^2_{adj} = 0.014$ and 0.063 ; $n=207$ and 277 respectively). For the rest of the models (Except Pb – leaf), both parameters β_1 (Mc) and β_2 (pH) were significantly different from zero ($P < 0.05$) (Table 3.16).

The Hough v2 IPM and GPM models, were compared using a comparison of regressions method (Section 3.2.3). Combining IPMs (e.g. lettuce, spinach) to predict metal concentrations in edible tissues across the produce group (e.g. leaves) was found to offer a highly significant improvement in model fit ($P < 0.01$) compared to GPMs, for most produce groups (Table 3.17). Predictions of plant concentrations in the Hough

database were also improved using IPMs compared to GPMs, resulting in lower RMSLE values. However, predictions of plant concentrations in an independent database (Nguyen), were closer to measured values when GPMs were used, compared to IPMs, to model uptake of metals. Model predictions of the Nguyen database may have been improved simply due to the greater number of data points used to derive GPMs compared to IPMs. Therefore the improved RMSLE values may reflect the number of data points used to parameterise the model, rather than a genuine improvement (Figure 3.6). The finding that plant uptake models are improved by treating plant species separately, is supported by other authors including Alexander *et al.* (2006) and Yang *et al.* (2010), who have suggested that there are significant differences in plant uptake of metals at all taxonomic levels.

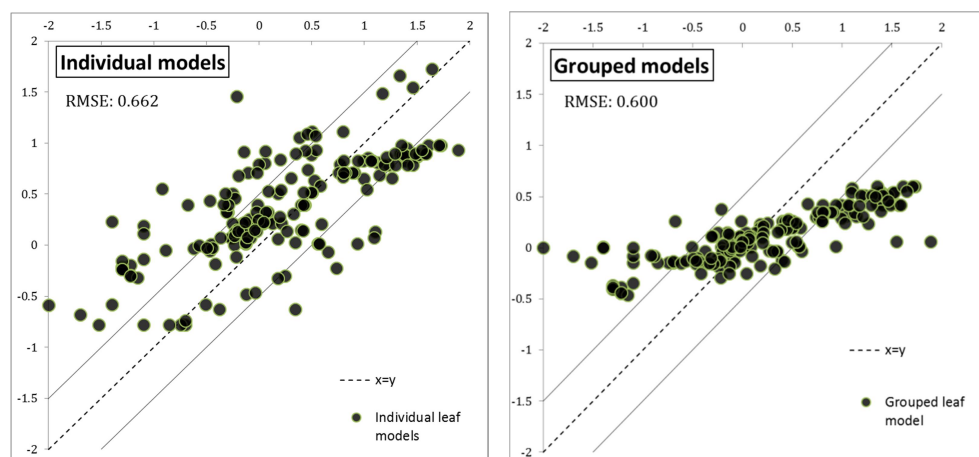


Figure 3.6: Observed (x-axis) and modelled (y-axis) cadmium concentrations (mg / kg dw displayed on log scale) in edible leaves using data from the Nguyen database. The graph on the left shows the combined predictions of individual models within the leaf – cadmium model group. The graph on the right shows the predictions of the grouped produce model for leaf and cadmium. Whilst the grouped produce model better predicts the datapoints (RMSLE of 0.600 compared to 0.662), the individual produce models are doing a better job of explaining the variation in the data: errors for this model are more stochastic than systematic.

Table 3.16. Derivation and evaluation of BCFs for produce groups. RMSLE values (Equation 3.8) were obtained for the performance of the models to the Hough database (H) and Nguyen database (N).

Model			BCFs			
Metal	Group _(a)	N	<i>Development</i>		<i>Performance</i>	
			BCF _(geomean)	Min – Max	RMSLE-H	RMSLE-N
Cd	Fruit	55	0.805	(0.003 – 20)	0.823	0.826
	Leaf	508	1.10	(0.02 – 260)	0.757	0.619
	Root	297	0.671	(0.02 – 160)	0.671	0.797
Cu	Fruit	40	0.894	(0.06 – 4.7)	0.598	0.865
	Leaf	274	11.6	(0.01 – 1200)	0.862	1.91
	Root	202	5.49	(0.04 – 340)	0.771	2.25
Pb	Fruit	32	0.0802	(0.02 – 0.21)	0.255	1.68
	Leaf	291	1.49	(0.002 – 220)	0.888	2.15
	Root	207	0.981	(0.003 – 58)	0.810	2.51
Ni _(b)	Leaf	248	1.46	(0.009 – 55)	0.571	1.29
	Root	192	0.791	(0.001 – 30)	0.686	-
Zn _(b)	Leaf	277	95.7	(0.02 – 1000)	1.00	2.85
	Root	192	28.3	(0.2 – 1900)	0.765	2.79

Table 3.17. Derivation and evaluation of regression models for produce groups. A) Produce groups for tuber and flower only consisted of one type of produce where n<10 in the Hough database, so models for these groups are essentially the same as the models for 'potato' and 'broccoli' in tables x-y. B) The fruit produce group also only consisted of tomatoes in the case of Ni and Zn. C) The large difference in RMSLE-N value between IPMs and PGMS can be explained by the absence of Cd data for corn cob and peanut in the Nguyen database. Surprisingly, the presence of these produce in the GPM improved the predictions of Cd in the Nguyen database. D) This column indicates the significance of the improvement in the model for the IPMs compared to GPMs using a comparison of regressions method: *** indicates a significance of $p < 0.001$, ** $p < 0.01$, * $p < 0.05$ and NS $p \geq 0.05$. RMSLE values are given for the performance of the group models and combined individual produce models to the Hough (H) database and the Nguyen (N) database.

Model			Hough v2 models derived for produce groups (GPMs)						Hough v2models derived for individual produce (IPMs)		
Metal	Group _(A)	N	<i>Development</i>			<i>Validation</i>			<i>Validation</i>		
			α : Const.	B_1 : Mc	B_2 : pH	R^2_{adj}	RMSLE-H	RMSLE-N	Ind RMSLE-H	Ind RMSLE-N	Significance of improvement
Cd	Fruit	55	-2.865	0.570 (<0.001)	0.230 (0.027)	0.343	0.579	0.287	0.416	0.954 ^(C)	***
	Leaf	508	0.098	0.357 (<0.001)	-0.104 (<0.001)	0.246	0.561	0.662	0.347	0.600	***
	Root	297	0.764	0.407 (<0.001)	-0.275 (<0.001)	0.416	0.409	0.609	0.361	0.631	***
Cu	Fruit	40	0.691	0.036 (0.180)	0.041 (0.267)	<0.001	0.051	0.207	0.048	0.208	NS.
	Leaf	274	1.138	-0.007 (0.714)	-0.023 (0.364)	<0.001	0.249	0.577	0.116	0.517	***
	Root	202	0.242	0.138 (<0.001)	0.0431 (0.040)	0.431	0.156	0.998	0.119	1.05	***
Pb	Fruit	32	-2.104	1.143 (<0.001)	-0.231 (0.009)	0.898	0.183	1.558	0.154	1.594	NS.
	Leaf	291	-0.708	0.315 (<0.001)	0.073 (0.106)	0.272	0.523	1.080	0.350	1.158	***
	Root	207	-0.659	0.051 (0.080)	0.074 (0.063)	0.014	0.342	0.883	0.310	0.837	***
Ni _(B)	Leaf	248	1.237	0.184 (<0.001)	-0.216 (<0.001)	0.313	0.237	0.505	0.225	0.685	**
	Root	192	1.590	0.258 (<0.001)	-0.303 (<0.001)	0.440	0.393	-	0.356	-	***
Zn _(B)	Leaf	277	2.989	-0.020 (0.298)	-0.142 (<0.001)	0.063	0.324	1.009	0.196	1.048	***
	Root	192	2.156	0.111 (<0.001)	-0.124 (<0.001)	0.462	0.174	1.047	0.164	1.053	*

3.3.3 Comparing the Hough v2 models with existing alternatives used for contaminated land assessment

The Hough v2 models for predicting uptake of metals into individual produce (IPMs) and produce grouped by edible tissue (GPMs), were compared to existing models of plant uptake contained in the exposure assessment models CLEA (EA), CSOIL (RIVM) and RBCA (USEPA). The CLEA and RBCA models do not differentiate between different produce types, whereas the RIVM model does. In the CLEA and RBCA models, produce are not grouped according to edible tissues (leaf, fruit, root etc.) so produce groupings may not match with the groupings developed for the Hough v2 GPM models. Models were compared in their ability to predict concentrations of Cd, Ni, Pb and Zn in edible plant tissues from easily-measured soil criteria using the Hough database, from which the Hough v2 models were derived, and the Nguyen database. Both databases contained paired metal concentrations in soils and edible plant tissues.

The CLEA, CSOIL and RIVM models predicted measured metal concentrations better in the Nguyen database than the Hough database. This may be explained by the differences in the range of explanatory variables between the databases, especially the total concentrations of metals in soils, which were higher in the Nguyen database, and may be more similar to those from which the CLEA, CSOIL and RBCA models were derived (Table 3.2 - Table 3.6). The CSOIL models made better predictions of data in both the Hough and Nguyen database than the CLEA and RBCA models for Cd (Table 3.19). The CLEA model made better

predictions of data than the USEPA models for uptake of Cd and Ni (Table 3.19). Freundlich regression models derived by RIVM for individual produce could only be compared using data from the Hough database as they required clay content as a parameter which was not available for data in the Nguyen database. Predictions using the RIVM regression models yielded low RMSLE values, similar to the Hough v2 models for the uptake of cadmium by lettuce, carrot and spinach. Predictions made using the RIVM regression models were much worse than the Hough v2 models for other metal-produce combinations (Table 3.18).

The Hough v2 models made better predictions of Cd and Zn concentrations (mean RMSLE: 0.643 and 0.743 respectively) in edible plant tissues from the Nguyen database, than the CLEA, CSOIL and RBCA models (Table 3.19). The performance of the Hough v2 models in predicting uptake of Pb and Ni into the different produce groups (mean RMSLE: 1.08 and 0.740 respectively), was similar to those made by the CLEA and CSOIL models. Two other studies have previously investigated the performance of the Hough models (Hough *et al.*, 2004) compared to other models produced in the literature. Legind and Trapp (2010) also compared model predictions of the CLEA, CSOIL and RBCA models, as well as the original Hough models, to measured data for Cd and Pb uptake by carrots and potatoes. The best model predictions, estimated by mean normalised average error (MNAE), were observed for the CSOIL and Hough models, followed by CLEA and RBCA. In the Legind and Trapp study, K_d values used by the CLEA models to calculate BCFs were

significantly improved with soil organic matter and pH data, using an approach published in Thorne *et al.*, (2004). This was not achieved for the present study as predicted metal concentrations in plants were not found to vary in the CLEA model v 6 (EA, 2016) with changes to pH and organic matter. A study by Novotna *et al.*, (2015), also found that the original Hough models for uptake of Cd, Cu and Zn by potato, outperformed several other models from the literature, when used to predict data from a Czech database. Based on these two studies and comparisons made here, it is reasonable to suggest that the Hough v2 models, which are similar to those produced by Hough *et al.* (2004), would perform similarly well or better than existing alternatives for the assessment of contaminated land.

Table 3.18. RMSLE values for model predictions using the RIVM models and the new Hough models developed here for uptake of cadmium and lead by a variety of produce. RIVM models could only be used to predict metal concentrations in plant tissues from the Hough database as the Nguyen database did not contain clay content which was a required parameter.

<u>Model</u>		<u>Hough v2 individual produce models</u>		<u>RIVM models</u>
<u>Metal</u>	<u>Produce/database used for comparisons:</u>	<u>Hough</u>	<u>Nguyen</u>	<u>Hough</u>
<u>Cd</u>	Carrot	0.366	0.514	0.430
	Lettuce	0.398	0.511	0.283
	Radish	0.437	0.781	2.05
	Spinach	0.272	0.727	0.547
<u>Pb</u>	Beans	0.142	1.87	1.53
	Cabbage	0.177	1.24	3.25
	Carrot	0.311	0.889	1.22
	Lettuce	0.246	1.28	1.74
	Spinach	0.174	0.847	0.819

Table 3.19. RMSLE values for model predictions using EA, USEPA and RIVM models as described in section 3.2.4. The new Hough models developed here are also given for comparison. Dashes indicate where models did not exist to make predictions. For nickel, data in the Nguyen database were sparse and could not be used to make predictions of fruit and root vegetables.

Model		Hough v2 models		EA (CLEA)		USEPA (RBCA)		RIVM (CSOIL)	
Metal	Produce group	Grouped	Individual	Hough	Nguyen	Hough	Nguyen	Hough	Nguyen
		<i>Database:</i> <i>Nguyen</i>	<i>Nguyen</i>						
<u>Cd</u>	Leaf	0.662	0.600	1.35	1.52	0.779	0.830	0.780	0.831
	Fruits	0.287	0.954	0.946	0.497	1.26	0.503	-	-
	Tuber	0.347	0.347	0.756	0.568	1.87	1.15	0.600	0.725
	Root	0.609	0.631	0.736	0.687	1.12	0.761	0.747	0.676
	Average	0.476	0.633	0.947	0.818	1.257	0.811	0.709	0.744
<u>Pb</u>	Leaf	1.080	1.158	1.77	0.971	-	-	1.97	0.893
	Fruits	1.558	1.594	0.738	1.01	-	-	0.845	0.946
	Tuber	0.738	0.738	2.26	0.910	-	-	2.05	1.07
	Root	0.883	0.837	1.62	1.23	-	-	1.99	0.919
	Average	1.06	1.08	1.60	1.03	-	-	1.71	0.957
<u>Ni*</u>	Leaf	0.505	0.685	1.67	0.663	1.76	0.881	-	-
	Fruits	-	-	1.22	-	1.86	-	-	-
	Tuber	0.801	0.801	1.87	0.812	1.83	0.785	-	-
	Root	-	-	1.45	-	2.11	-	-	-
	Average	0.653	0.743	1.55	0.738	1.89	0.833	-	-
<u>Zn</u>	Leaf	1.009	1.048	-	-	2.77	0.872	-	-
	Fruits	0.285	0.285	-	-	1.42	0.729	-	-
	Tuber	0.573	0.573	-	-	3.18	0.804	-	-
	Root	1.047	1.053	-	-	2.91	0.761	-	-
	Average	0.729	0.740	-	-	2.57	0.792	-	-

3.3.4 Study limitations and future direction for plant uptake models for use in the assessment of contaminated land

There are several limitations to the use of the Hough v2 models for the purpose of risk assessment. Firstly, the range of metal concentrations in soils for some produce in the Hough database, did not exceed the soil guidance values meaning the range of data could be inappropriate for predicting health risks from consumption of these produce. The maximum Cd concentrations used to derive Hough v2 models for corn cob, peanut and celery were below the category 4 screening level (C4SL) for cadmium in allotment soils of $4.9 \text{ mg kg}^{-1} \text{ dw}$ as proposed by DEFRA (2013b). Similarly, many of the models for Pb were below the C4SL of $80 \text{ mg kg}^{-1} \text{ dw}$ (DEFRA 2013e). The range of other soil parameters including soil pH and organic carbon for some metal-produce combinations was also small (e.g. Ni – tomato; Table 3.5). There is a significant increase in uncertainty when models are applied to data outside the range with which they were parameterized. Despite this, the models seemed to predict measured data reasonably well in the Nguyen database compared to the existing alternatives (CSOIL, CLEA and RBCA), for which the concentrations of metals in soil were higher (Section 3.3.3).

Secondly, whilst the Hough database is large and encompasses a wide variety of produce, it cannot be claimed that data is available for most produce which are grown by gardeners; all types of contaminants or even all HMs. Further work is required to develop the database further to include other produce and metals to extend the applicability of the

models for risk assessment. Developing the Hough database further to include more paired studies of metal concentrations in soils and plants, and other soil characteristics (e.g. soil texture), may help improve the model fits (R^2_{adj} values) and predictions. Field studies in urban gardens and allotments, present perfect opportunities for further study on the relationship between metal concentrations in soils and edible plant tissues.

Finally, the Hough v2 models could be improved by taking into account other processes that might influence plant uptake such as internal partitioning of metals in plant tissues and foliar uptake. Making sense of these processes will undoubtedly improve plant uptake models for the assessment of contaminated land. Models such as the TBLM (Thakali, Allen, Di Toro, Ponizovsky, Rooney, Zhao, McGrath, *et al.* 2006) do try to comprehend the mechanics of plant uptake and could be a useful part of risk assessment models in the future (Antunes *et al.* 2006). Foliar uptake of metals has been extensively researched (e.g. Harrison and Chirgawi 1989; Schreck, Foucault, *et al.* 2012; Xiong *et al.* 2014) and already incorporated into some models (Legind and Trapp 2010; Schreck, Bonnard, *et al.* 2012). Obvious signs of metal stress and toxicity on plants act as a barrier to toxicity for humans and should be included in risk assessment models (Chaney *et al.* 1984; McLaughlin *et al.* 2011). However, the toxicity barrier varies for different plants with some crop plants able to accumulate these elements in concentrations that could lead to human exposure (Ciura *et al.* 2005).

3.4 CONCLUSIONS

This study is the first to derive and compare models of plant uptake for several metals into different types of edible produce, for the purpose of risk assessment. The Hough v 2 models presented here performed similarly, or better than, alternatives currently used in risk assessment for the prediction of plant uptake of Cd, Cu, Pb, Ni and Zn. The Hough v 2 models do have some limitations but present a viable alternative to existing options for predicting plant uptake of these metals. Future work should focus on extending the Hough database from which the models are derived, to other types of produce and contaminants. The development and use of other models, including the TBLM, for the purpose of risk assessment, should be explored. Other recommendations for risk assessment include:

- Freundlich regression models should be preferred to BCFs to model plant uptake of metals.
- Grouping produce should be avoided if possible as this reduces the performance of the model.
- The margin of error in plant uptake models is large (up to 3 orders of magnitude for models of some produce) and should be incorporated into risk assessment.
- Models used to predict uptake of Cu and Pb should be applied with caution as these elements are less mobile in soils and model predictions are often worse. Alternative methods for predicting uptake of these metals should be considered.

4 THE DIFFERENCE IN METAL UPTAKE BETWEEN CULTIVARS OF LETTUCE AND BEAN

4.1 INTRODUCTION

Predicting plant uptake of metals for the purpose of risk assessment is challenging, as uptake and translocation of metals varies depending on the metal, soil properties and plant type (McLaughlin *et al.* 2011). Grouping produce (e.g. by edible produce tissues such as fruits, leaves and roots) reduces the amount of computation required by risk assessment models and enables estimates of plant uptake to be made for crops when data is unavailable. However, inappropriately grouping produce can increase uncertainty in model predictions. In this chapter, the grouping of 'green vegetables' together in the Contaminated Land Exposure Assessment model (CLEA: EA 2009d), is reviewed using a pot experiment to compare plant uptake of metals into lettuces and French/broad beans; produce considered to be similar in the CLEA model and grouped together under 'green vegetables' (Table 4.1). Differences in the uptake of metals between different cultivars of lettuces and beans were also investigated.

There are a number of internal plant mechanisms that affect plant uptake of metals. For immediate transfer of metals to occur, metals must be present in the soil solution, in close physical proximity to the root and in the form of the free metal ion or a simple chelate (Semple *et al.* 2004). Metals enter the plant through initial binding to the cell wall, followed by

transport across the plasma membrane through secondary transporter proteins (Clemens *et al.* 2002).

Table 4.1: Crops included in each produce group category in the CLEA model. Reproduced from EA, (2009c).

Produce group	Crops included
Green Vegetable	Beans (broad, French, green and runner beans), Brussels sprouts, cabbage (red, white, greens and kale), cauliflower, lettuce, spinach, peas (garden and mangetout), stem vegetables (broccoli, celery, asparagus), okra, globe artichoke, Chinese leaves, endives, chicory, chard, dandelion, watercress and fresh herbs (basil, coriander, tarragon, sage, parsley and mint).
Root Vegetable	Beetroot, carrot, cassava, garlic, ginger, Jerusalem artichoke, leeks, onions, parsnips, radish, rhubarb, salsify, swede, sweet potato, turnips and yam.
Tuber	Potatoes.
Herbaceous Fruit	Aubergine, courgettes, cucumber, marrow, pumpkin, strawberries, tomatoes.
Shrub Fruit	Bilberries, blackberries, cranberries, gooseberries, loganberries, mulberries, physalis, raspberries, blackcurrants, redcurrants and whitecurrants.
Tree Fruit	Apples, apricots, cherries, peaches, pears and plums

Uptake of metals into the xylem for transport around the rest of the plant is tightly controlled by membrane transporter proteins (Clemens *et al.* 2002). Resistance mechanisms including chelation (binding of the metal to high affinity ligands), compartmentalization (storing of metals in cell vacuoles) and biotransformation (reduction of mechanism of toxic action through chemical reduction or binding with organic compounds), help to reduce transport of non-essential and excess essential metals around the plant (Salt *et al.* 1998). Non-essential metals (e.g. Cd, Pb) tend to accumulate mostly in the roots of most plants with small amounts

translocated to shoots and leaves and less still to fruits, seeds and tubers (Clemens 2006).

The expression of genes which control metal resistance mechanisms are not the same for all plants, as plant traits evolve over time (Broadley *et al.* 2010). A number of authors have suggested that there are large differences in plant uptake of metals (especially Cd) between leguminous produce, such as pea and bean, and leafy vegetables including lettuce and spinach (Kuboi, T. *et al.* 1986; Alexander *et al.* 2006; Yang *et al.* 2010). Furthermore, several authors have found evidence of differences in plant uptake between different cultivars of the same produce and at other taxonomic levels (e.g. Alexander *et al.* 2006; Yang *et al.* 2010). Differences in metal accumulations between cultivars of high-energy crops used in agriculture such as barley (Chen *et al.* 2007), wheat (Stolt *et al.* 2006; Liu *et al.* 2014) and rice (Yu *et al.* 2006; Cao *et al.* 2014) have been more extensively researched than home-grown produce. Uptake of metals into lettuce leaves has been observed to be cultivar-dependent (Crews and Davies 1985; Zhang *et al.* 2013), but few authors have previously investigated cultivar differences in French or broad beans.

The produce in this study were grown in soil historically amended with sewage sludge. Sewage sludge is known to contain high concentrations of metals but is often applied to agricultural soils to improve fertility because of its high content of nitrogen, phosphate and organic matter (Gardiner *et al.* 1995; Singh and Agrawal 2008). Soils amended with

sewage sludge may be a better proxy for contaminated urban soils compared to spiking clean soils with metal salts; a process which leads to over-predictions of bioavailability (Efroymson *et al.* 2004).

The aim of this study was to investigate differences in plant uptake of trace metals between lettuces and beans; plants which are known to exhibit different responses to metal concentrations in the soil (Alexander *et al.*, 2006). Investigating these differences will help to improve understanding of plant uptake and identify trends that could be used to simplify plant uptake models. The objectives of the study were to:

- i) investigate differences in metal uptake by lettuces and beans;
- ii) investigate cultivar differences in the accumulation of metals into edible tissues of lettuce and bean;
- iii) evaluate the use of the CLEA 'green vegetable' model and the Hough v2- model for predicting metal uptake by lettuces and beans.

4.2 METHODS

4.2.1 Soil selection and preparation

Bean and lettuce cultivars were grown in a glasshouse in soil historically amended with sewage sludge at contrasting application rates: a soil (L) containing Cd and Pb at concentrations similar to the Category 4 Screening level (C4SL) for an allotment (DEFRA 2013b; e), and a soil containing higher concentrations of trace metals (H). Soils were collected from adjacent fields at the Stoke Bardolph waste management site in Nottingham owned by Severn Trent Ltd. Sample locations were identified from previous surveys of the site.

The soils were air-dried, sieved to <4 mm and homogenised using the cone method with four passes (see Schumacher *et al.* 1990), prior to potting. Four replicate samples were taken from each homogenised soil sample for analysis of trace element concentrations, total and inorganic carbon concentrations and soil pH value. The soils were homogenised again, after lettuces were grown and prior to growing beans, with a further four replicate samples taken from each soil type.

4.2.2 Cultivar selection and experimental design

Broad bean (*Vicia faba*), French bean (*Phaseolus vulgaris*) and lettuce (*Lactuca sativa*) cultivars were selected from Kings Seeds ©; a commonly used supplier to garden and allotment associations across the UK. Cultivars were chosen that presented a range of phenotypes in an attempt

to broaden the range of genetic variation across the plants (Table 4.2). A total of 15 lettuce cultivars and subsequently 15 bean cultivars (8 French and 7 broad beans) were grown in 1 L pots containing both types of soil. Each cultivar-soil combination was replicated 5 times; a total of 150 pots for growing lettuces and beans (5 X 2 X 15). Seeds were sown directly into pots and grown in a glasshouse under a randomised blocking structure to account for natural variations in light intensity (Appendix V).

Table 4.2: List of lettuce and bean cultivars grown in the experiment

Lettuce		Bean	
Cultivar	Description	Cultivar	Description
All The Year Round	Butterhead	Express	Broad
Buttercrunch	Butterhead	Aquadulce Claudia	Broad
Trocadero	Butterhead	Bunyards Exhibition	Broad
Little Gem Cos	Cos	Deadnought	Broad
Lobjoits Cos	Cos	Witkeim Manita	Broad
Valmaine	Cos	Karmazyn	Broad
Great lakes	Crisp and Iceberg	Robin Hood	Broad
Saladin	Crisp and Iceberg	Sungold	French Dwarf
Webbs Wonderful	Crisp and Iceberg	Masterpiece	French Dwarf
Lollo Biondi	Loose leaf	Royalty	French Dwarf
Parella Green	Over-wintering	Tendergreen	French Dwarf
Winter Density	Over-wintering	The Prince	French Dwarf
Little Leprechaun	Red type	Faraday	French Dwarf
Marvel of four seasons	Red type	Cantare	Snap Bean
New Red Fire	Red type	Purple Teepee	Snap Bean

Lettuces were grown from early July to mid-August and were harvested after a period of approximately 60 days. The soils were removed and homogenised (according to their soil types L and H) and re-sown with bean cultivars in mid-September. Bean leaf samples, fruits and stems were all harvested separately. French bean leaves were harvested after

approximately 60 days post-germination, with bean fruits harvested after reaching maturity. The remaining above-ground plant material ('stems') was harvested approximately 80 days after germination of the French beans. Young, and edible, broad bean leaves were harvested approximately 60 days after germination, fruits were harvested after reaching maturity and the rest of the plant was harvested approximately 120 days after germination. Broad beans were pollinated by hand due to a lack of insect pollinators in the winter months by manually transferring pollen from one flower-head to another (Figure 4.1).

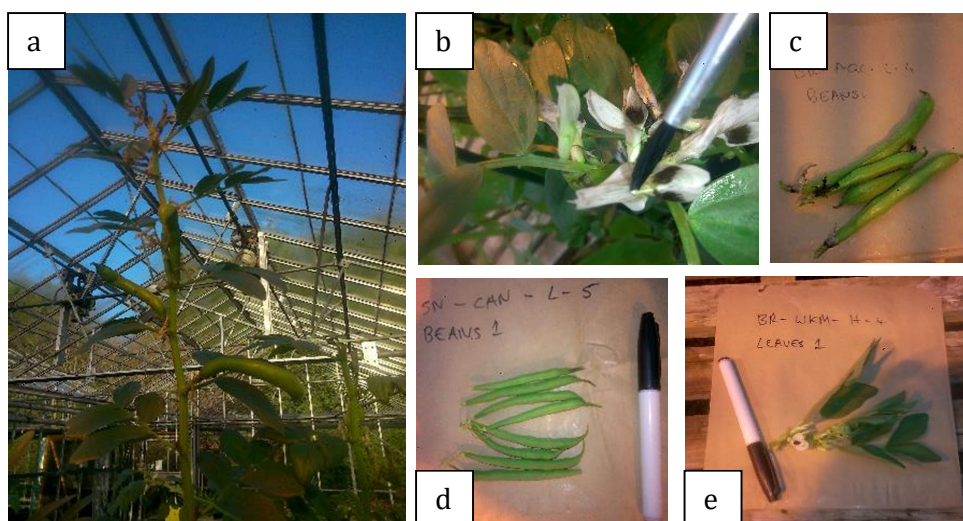


Figure 4.1: Beans during the various stages of growth and harvesting. Images a) and b) show growth and pollination of broad bean plants. Images c), d) and e) show broad bean fruits, French bean fruits and broad bean leaves respectively after harvesting.

4.2.3 Laboratory analysis of soils

The four replicate soil samples from each soil type (L and H), taken before and after lettuces were grown and harvested, were analysed for trace element concentrations, soil pH and organic carbon content (calculated

from separate analyses of total carbon and inorganic carbon). The laboratory methods used for analysis of soils were consistent with Section 2.2.6. A two-way Analysis of Variance (ANOVA) in Minitab v 17.2.1 was used to test for differences between metal concentrations, total organic carbon and soil pH before and after growing lettuces. Metal concentrations, soil pH and organic carbon content (TOC) were not significantly different ($p > 0.05$) between soil samples taken before and after lettuces were grown and harvested. Mean metal concentrations, soil pH and organic carbon content for each homogenised soil (L and H) are reported in Table 4.3.

Table 4.3: Mean and standard deviations of the metal concentrations, soil pH and organic carbon content in both soil types used for growing lettuces and beans ($n = 8$). 1) C4SLs were obtained from DEFRA (2009a) and DEFRA (2009b) for an allotment land use.

Soil type	Cd (mg/kg)	Cu (mg/kg)	Pb (mg/kg)	Ni (mg/kg)	Zn (mg/kg)	pH	OrgC (%)
L	2.71 (0.101)	85.4 (3.33)	96.1 (4.24)	57.9 (3.63)	292 (13.1)	6.86 (0.05)	2.95 (0.150)
H	13.0 (0.741)	298 (11.0)	332 (15.6)	164 (7.41)	789 (34.3)	5.94 (0.09)	6.73 (0.404)
C4SL ₁	4.9	-	84	-	-		

4.2.4 Laboratory analysis of plant material

After harvesting, all plant material was oven dried for a minimum of 72 hours at 50-55 °C and digested prior to trace element analysis by ICP-MS (Thermo Fisher X series II) (see Section 2.2.6 for ICP-MS methods). The preparation process was different for bean stems and fruits/seeds, which required milling, compared to lettuces and bean leaves which did not. Broad bean seeds were separated from their pods with pods removed prior to milling, as the seed is normally consumed rather than the pod. In contrast, French bean fruits were milled whole. Bean fruits from the same plant were grouped together for analysis. Replicate plants were treated separately.

Bean stems and bean fruits

Harvested beans (fruits/seeds) and bean stems were milled with a centrifugal mill (Retsch : ZM200) and stored in sealed, labelled test tubes. Approximately 200 mg of plant material (beans or stems) for each sample was added to 50 ml SMART VENT™ digestion vessels with 8 ml of Primar grade nitric acid (HNO_3 : >68 %) and heated in a microwave (Anton Paar : Multiwave Pro with a 24HVT50 rotor) for 35 minutes at 1400 W. Samples were allowed to cool and made up to 20 ml with milli-Q water, prior to a further factor of 5 dilution with milli-Q water, and trace element analysis by ICPMS. Five Blanks (8 ml HNO_3 in 50 ml water) were included, and five samples of certified reference material (CRM: tomato leaf – *National*

Institute of Standards and Technology, NIST, 1573a) were included for quality control.

Lettuces and bean leaves

Lettuces and bean leaves, were crushed by hand (using gloves) and the entire sample digested in a mixed acid solution (HNO₃: 6 mL/ H₂O₂: 2 mL). The samples were initially left in the acid solution overnight and then heated using a programmable hot plate (Analysco 2416) at 95 °C for 2 hours. The lettuce and bean leaf digests were diluted to 2% HNO₃ using milli-Q water prior to trace element analysis by ICPMS. Five Blanks containing the diluted, mixed acid solution, were included to correct the results from the analysis, and four samples of certified reference material (CRM: tomato leaf - NIST 1573a) were included for quality control. Measured concentrations of Cd, Cu, Ni, Pb and Zn were compared with accredited values for the CRM. Limits of quantification (LOQ) and detection (LOD) were reported by the Plasmalab software used to operate the ICP-MS. If the reported value was below the LOQ then this was considered to be negligible. An explanation of the difference between LOQ and LOD can be found in Saadati *et al.* (2013).

Table 4.4. Percentage recovery (%) of element concentrations of the CRM. Lead (Pb) concentrations were not available for the CRM.

<u>Preparation type/metal</u>	<u>Cd</u>	<u>Cu</u>	<u>Pb</u>	<u>Ni</u>	<u>Zn</u>
Digestion using hot plate (leaves)	92	86	N/A	57	88
Milling and microwave digestion (fruits and seeds)	105	96	N/A	92	97

Table 4.5: Limits of quantification (LOQ) and detection (LOD) reported for the analysis of lettuces and bean leaves, fruits/seeds and stems. All concentrations are reported in mg kg⁻¹ dw.

<u>Analysis type</u>	<u>Limit</u>	<u>Cd</u>	<u>Cu</u>	<u>Pb</u>	<u>Ni</u>	<u>Zn</u>
Digestion using hot plate (leaves)	LOQ	0.024	4.35	0.047	0.45	1.21
	LOD	0.007	1.30	0.014	0.13	0.36
Milling and microwave digestion (fruits and seeds)	LOQ	0.020	0.16	0.042	0.16	4.50
	LOD	0.006	0.05	0.012	0.05	1.35

4.2.5 Statistical analysis of plant samples

Differences between metal accumulations in lettuces and beans

A two-way ANOVA was used to test for differences between metal concentrations in lettuces leaves; French bean fruits, stems and leaves and broad bean seeds, stems and leaves. Soil type and replicate were included as confounding factors in the analysis. Tukey's test was further used to identify differences in metal accumulation between plant tissues.

Differences in metal accumulations between cultivars.

A two-way ANOVA was used to test for differences between metal concentrations in lettuce, French bean and broad bean cultivars. Subsets of the data were taken according to plant tissue (leaf, stem, fruit/seed) and the ANOVA was set up with soil type and replicate included as cofounding factors. Tukey's test was used to identify differences in accumulations of metals between individual cultivars of lettuce and bean.

Predicting uptake of metals into lettuce and bean

Two models; the CLEA 'green vegetable model' (EA, 2009) and the Hough v 2 model, were used to estimate accumulation of metals in lettuces and beans. An explanation of both models can be found in section 3.2.

CLEA model:

To predict uptake into lettuces and beans using the CLEA model, the 'green vegetable' model was used. BCF's were calculated from DEFRA, (2012b) and converted to dry weight using conversion factors from the EA, (2009). Conversion factors can be found in Table 3.8.

Hough v2 models:

Regression models for uptake of Cd, Cu, Ni, Pb and Zn into lettuce, and Cu and Pb into beans, were used (Table 3.11- Table 3.17). The Hough database did not contain data for uptake of Cd, Ni and Zn into beans, so the fruit model was used to estimate uptake of these metals (Table 3.16 – for uptake of Ni and Zn, the fruit model only contains data for tomatoes).

Model predictions were compared with measured data by calculating RMSLE values (Equation 3.8). The following formula was developed to enable a graphical comparison of the modelled data with the measured mean:

$$Fd = (x - \bar{y})/\bar{y} \quad 4.1$$

Equation 4.2 was used to make a graphical comparison between individual measured values and the measured mean to show the spread of the measured data:

$$Fd = (y - \bar{y})/\bar{y} \quad 4.2$$

Where Fd is termed the 'factor difference', x or y refers to an individual measured or modelled value respectively and $\bar{y}$ is the measured mean. A smaller Fd indicates that the value is closer to the measured mean.

4.3 RESULTS AND DISCUSSION

4.3.1 Metal concentrations in beans and lettuces

Metal concentrations in bean and lettuce tissues varied depending upon the soil in which the plant was grown, the plant species (lettuce, French or broad bean) and the plant tissue sampled (leaf, stem or pod/seed). Trends in metal accumulation into different plant tissues also differed between metals (Cd, Cu, Pb, Ni and Zn).

Cadmium

Concentrations of Cd were two orders of magnitude higher in lettuce compared to French bean fruits and broad bean seeds, regardless of soil used (L or H) (Figure 4.2). The mean concentrations of Cd in lettuces, French bean fruits and broad bean seeds from plants grown in soil H were 13.0, 0.18 and 0.21 mg kg⁻¹ dw respectively; equivalent values for soil L were 1.53, 0.04 and 0.05 mg kg⁻¹ dw). Concentrations of Cd were significantly different in stems, leaves and beans (fruits/seeds) of French bean and broad bean plants (both $p < 0.001$) and accumulations of Cd followed the order: stems > leaves > fruits/seeds. Concentrations of Cd in the various plant tissues analysed, were also significantly different ($p < 0.001$) between plants grown on different soil types.

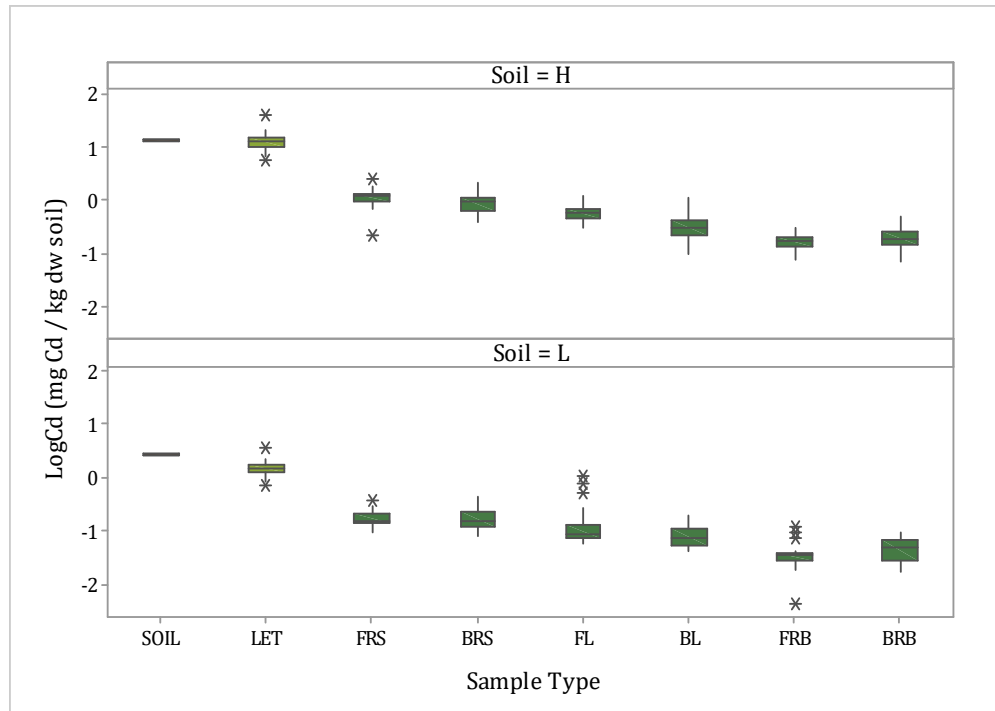


Figure 4.2: Box plots showing cadmium concentrations in soil (SOIL) (brown), lettuce (LET) (light green) and bean (dark green) tissues grown in soil type L, containing lower concentrations of Cd, and soil type H, containing high concentrations of Cd (table 4.3). Bean tissues are subdivided into French bean stems (FRS), leaves (FRL) and fruits (FRB); broad bean stems (BRS), leaves (BRL) and seeds (BRB). Boxes represent the median, interquartile range (IQR) and range of the data. Asterisks indicate outlying values calculated using the upper/lower quartile $\pm 1.5 \times$ IQR.

Copper

Concentrations of Cu were significantly higher in the leaves of broad beans than the other sampled plant tissues (Tukey: $p < 0.05$) (Figure 4.3). Concentrations of Cu in broad beans followed the sequence leaves > seeds > stems. This was different to the order of Cu accumulation in French bean tissues was stems > fruits > leaves. There was no difference between concentrations of Cu in plants grown on the different soil types, L and H ($p = 0.383$).

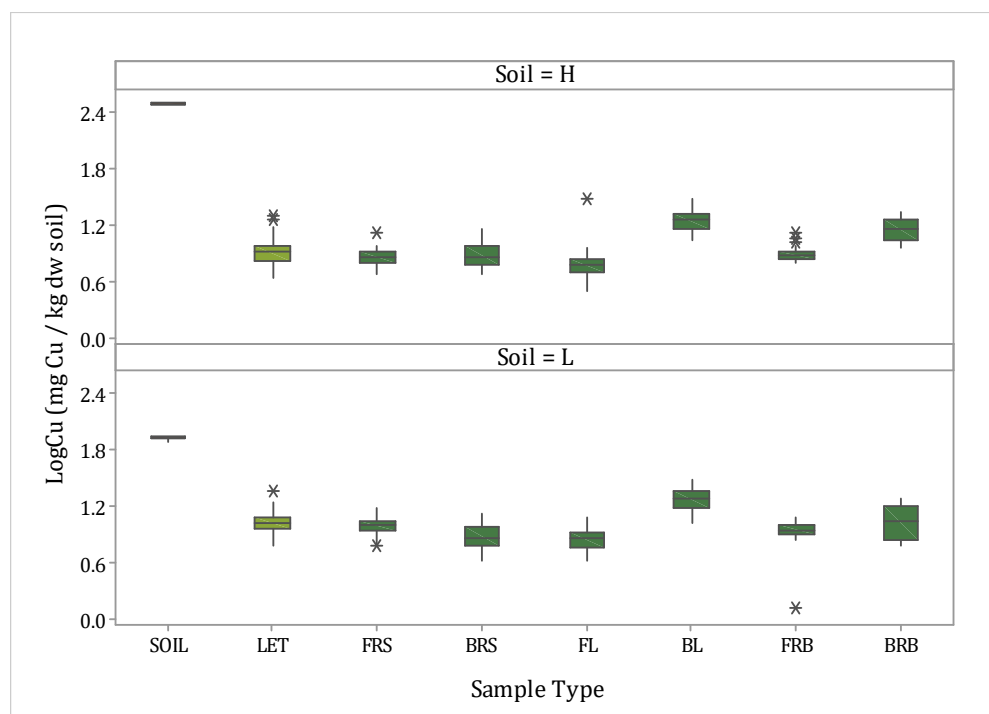


Figure 4.3: Box plots showing copper concentrations in soil (SOIL) (brown), lettuce (LET) (light green) and bean (dark green) tissues grown in soil type L, containing lower concentrations of Cu, and soil type H, containing high concentrations of Cu (table 4.3). Bean tissues are sub-divided into French bean stems (FRS), leaves (FRL) and fruits (FRB); broad bean stems (BRS), leaves (BRL) and seeds (BRB). Boxes represent the median, interquartile range (IQR) and range of the data. Asterisks indicate outlying values calculated using the upper/lower quartile $\pm 1.5 \times \text{IQR}$.

Lead

Concentrations of Pb were significantly higher in lettuces than in bean tissues (Tukey: $p < 0.05$) and below the LOQ (

Table 4.5) in French bean fruits, broad bean seeds and broad bean leaves.

Lead concentrations were also higher in stems and leaves of both varieties of bean. There was no difference in the accumulation of Pb in plants grown on the different soil types ($p = 0.183$).

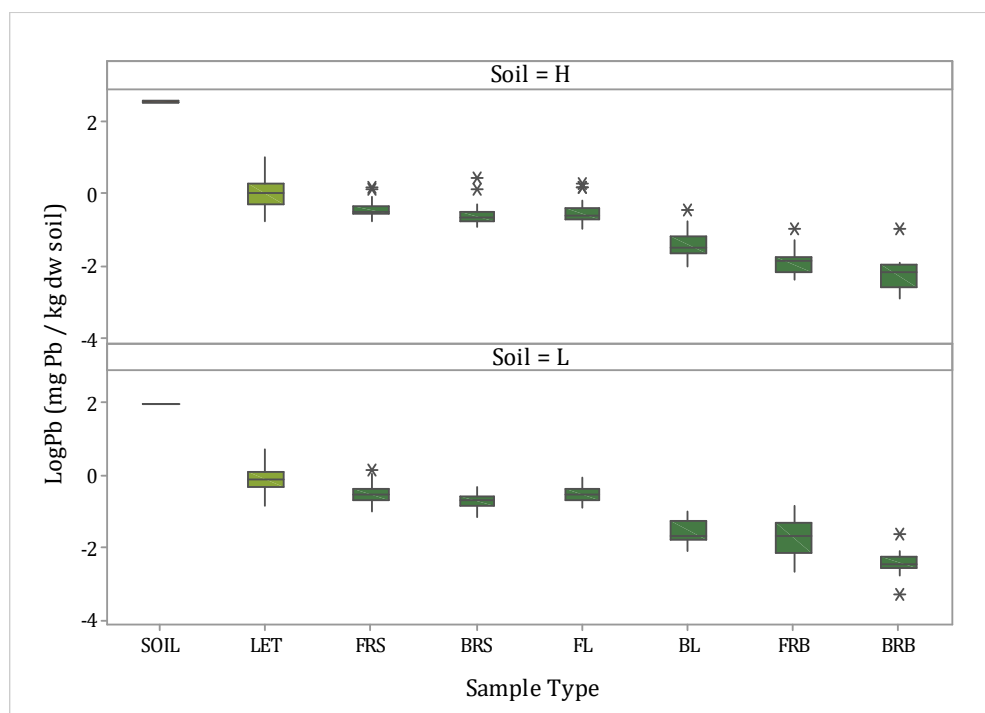


Figure 4.4: Box plots showing lead concentrations in soil (SOIL) (brown), lettuce (LET) (light green) and bean (dark green) tissues grown in soil type L, containing lower concentrations of Pb, and soil type H, containing high concentrations of Pb (table 4.3). Bean tissues are sub-divided into French bean stems (FRS), leaves (FRL) and fruits (FRB); broad bean stems (BRS), leaves (BRL) and seeds (BRB). Boxes represent the median, interquartile range (IQR) and range of the data. Asterisks indicate outlying values calculated using the upper/lower quartile $\pm 1.5 \times \text{IQR}$.

Nickel

Concentrations of Ni were significantly higher in French bean fruits than lettuces and other bean tissues (Tukey: $p < 0.05$) and were lower in lettuce tissues compared to beans. Accumulation of Ni in French bean tissues followed the order fruits > stems > leaves and in broad bean tissues, followed the order leaves > seeds > stems. Accumulations of Ni were significantly higher in plants grown on the more contaminated soil (H) ($p < 0.001$).

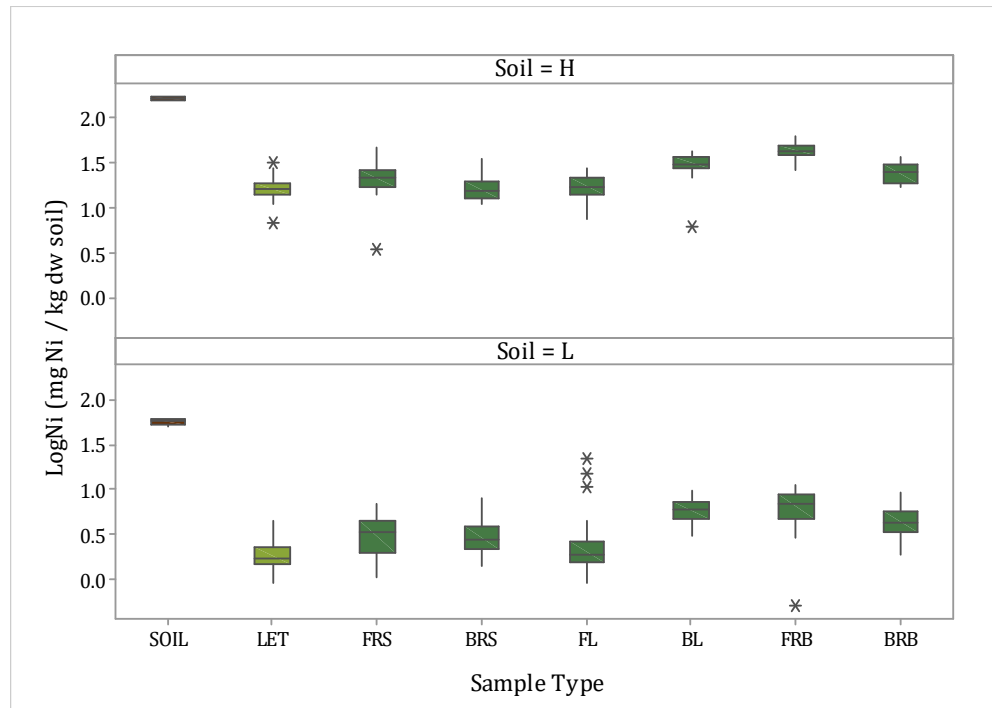


Figure 4.5: Box plots showing nickel concentrations in soil (SOIL) (brown), lettuce (LET) (light green) and bean (dark green) tissues grown in soil type L, containing lower concentrations of Ni, and soil type H, containing high concentrations of Ni (table 4.3). Bean tissues are sub-divided into French bean stems (FRS), leaves (FRL) and fruits (FRB); broad bean stems (BRS), leaves (BRL) and seeds (BRB). Boxes represent the median, interquartile range (IQR) and range of the data. Asterisks indicate outlying values calculated using the upper/lower quartile $\pm 1.5 \times$ IQR.

Zinc

Trends in the accumulation of Zn into bean and lettuce tissues were dependent upon the soil type on which the plants were grown. Concentrations of Zn were significantly higher in lettuces than in bean tissues in plants grown on the more contaminated soil (H) (Tukey; $p < 0.05$). However, concentrations of Zn were higher in French bean and broad bean tissues in plants grown on the less contaminated soil (L). Accumulation of Zn into French bean tissues followed the order stems > leaves > beans. The order of accumulation of Zn in broad bean tissues

depended in which soil the plants were grown. Concentrations of Zn in lettuce and bean tissues were significantly higher ($p < 0.001$) when plants were grown in soil H.

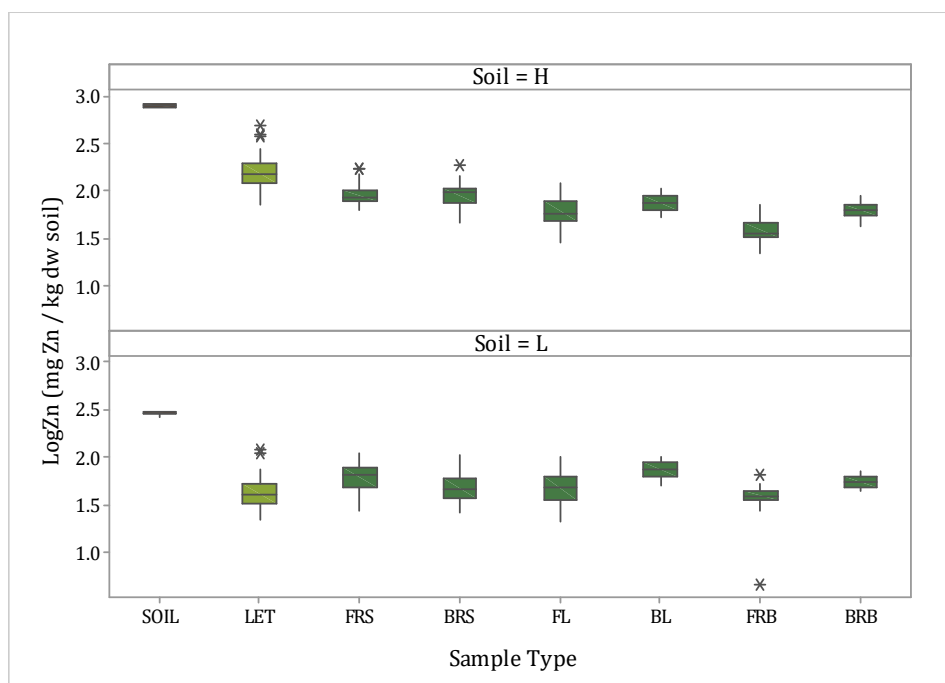


Figure 4.6: Box plots showing zinc concentrations in soil (SOIL) (brown), lettuce (LET) (light green) and bean (dark green) tissues grown in soil type L, containing lower concentrations of Zn, and soil type H, containing high concentrations of Zn (table 4.3). Bean tissues are sub-divided into French bean stems (FRS), leaves (FRL) and fruits (FRB); broad bean stems (BRS), leaves (BRL) and seeds (BRB). Boxes represent the median, interquartile range (IQR) and range of the data. Asterisks indicate outlying values calculated using the upper/lower quartile $\pm 1.5 \times$ IQR.

Discussion

The response of lettuces and beans to metals in the two soils may have reflected the different responses of these plants to essential and non-essential elements. Concentrations of the non-essential metals, Cd and Pb, were higher in lettuce tissues than in bean tissues. The observed difference of two orders of magnitude in accumulation of Cd into lettuce leaves compared to French bean fruits is consistent with the findings of

(Alexander *et al.* 2006). Similar patterns were not observed between lettuces and beans for accumulations of the essential elements, Cu and Ni. Plant resistance mechanisms may have evolved in *V. fabia* (broad beans) and *Ph. Vulgaris* (French beans) to regulate plant uptake of essential and non-essential metals (Liao *et al.* 2003, 2005). Evidence of several responses to toxic metals, including thickening of cell walls, synthesis of phytochelatins and aggregation of metals in calcium – phosphate crystals have been found in *V. fabia* (Probst *et al.* 2009). In contrast, Cd has been found to accumulate in the leaves of lettuce and endive (Baldantoni *et al.* 2016), suggesting that the same regulatory mechanisms may not exist in lettuce. This is further supported by the observed differences in Zn accumulation in lettuces and beans in the present study, which were similar when grown in soil L containing lower concentrations of soil Zn, but much higher in lettuces than beans when the plants were grown in soil H. This suggests that when concentrations of available Zn in soils are high, beans are able to regulate uptake and translocation to prevent excessive Zn assimilation, whereas lettuces are not. For the purposes of growing produce on mildly contaminated land, beans should be preferred to lettuces, especially if the soil is contaminated with Cd.

The concentrations of non-essential metals, Cd and Pb, followed the order: stems > leaves > fruits/seeds. This is also true for Zn concentrations in French beans grown in soil H. However, accumulation of the other essential metals, Cu, Ni (and Zn in plants grown in soil L) did not follow the same trend: In broad bean plants, Cu, Ni and Zn

accumulated mostly in the leaves, followed by seeds and stems. In French bean plants, Cu and Ni accumulated mostly in stems or fruits. Nadgorska-Socha et al (2012) recorded higher concentrations of Cd, Ni, Pb and Zn, but not Cu, in leaves compared to stems of *V. fabia* (Broad bean plants). Kumar and Chopra (2013) observed that for all analysed metals (Cd, Cr, Cu, Fe, Pb and Zn), metal concentrations were lowest in the fruits of French beans compared to shoots, roots and leaves. This was not consistent with the results from the present study.

There were no significant differences between concentrations of Cu and Pb in plants grown in either soil type, suggesting that uptake of these elements is minimal. Copper and Pb are strongly bound to soil colloids - humus and Fe oxides - in arable soils with neutral pH values, so the bioavailability of these metals is low (e.g. McLaughlin *et al.* 2011). Whilst there were some differences in soil properties as well as metal concentrations between the two soils (Table 4.3), there was no clear evidence of an effect of the different soil properties on metal uptake by lettuces or beans.

4.3.2 Differences in metal accumulation in cultivars of French beans, broad beans and lettuces.

Metal accumulation into different cultivars of French beans, broad beans and lettuces, were studied to determine if some cultivars could be grown preferentially to others to minimise metal exposure. Cultivar differences are also not currently taken into account in plant uptake models and could contribute to uncertainty in model predictions.

Lettuces

Differences in the concentrations of Cd, Cu, Ni and Zn in lettuce cultivars, were found to be highly significant ($p \leq 0.001$) with differences in the concentrations of Pb in lettuce leaves, also significantly influenced by cultivar ($P = 0.017$). Differences in concentrations of Cd, Ni and Zn in lettuce cultivars were significantly influenced by soil type ($P = 0.009$, 0.003 and <0.001 respectively) whereas concentrations of Cu and Pb were not ($P = 0.458$ and 0.406 respectively).

Selecting lettuce cultivars:

There were no cultivars of lettuce that accumulated significantly higher, or lower, concentrations of Cd, Cu, Pb, Ni and Zn than the rest, although some trends were observed. The cultivar, '*New Red Fire*' (NRF) consistently accumulated higher concentrations of metals when grown on both soils suggesting that this cultivar should not be grown on soils containing high concentrations of metals (Figure 4.7 and Figure 4.8). The cultivar, '*Winter Density*' (WID), also accumulated lower concentrations of

metals when grown in both soils compared to other cultivars (Figure 4.7 and Figure 4.8). Average (mean) concentrations of metals in lettuce cultivars are reported in Table 4.6.

Differences in metal accumulation between lettuce cultivars have been previously researched by Crews and Davies (1985), Thomas and Harrison (1991), Florijn et al (1991), Alexander et al (2006), Wang et al (2006) and Zhang et al (2013). Of these studies, only Zhang et al (2013) studied a sufficient range ($n = 28$) of lettuce cultivars to truly investigate differences between cultivars in the accumulation of Cd and Pb. The cultivars used in the present study overlapped with Crews and Davies (1985) and Alexander et al (2006); although the latter did not observe intraspecific variation in uptake of Cd, Cu, Pb and Zn. In the study by Crews and Davies (1985), the cultivar '*All year Round*' (AYR) was found to accumulate the highest concentrations of Cd and Zn with cultivars '*Winter Density*' (WID) and '*Webbs Wonderful*' (WW) found to be moderate - low accumulators. This supports the conclusion from the present study that '*Winter Density*' (WID) could be grown as a low accumulating cultivar of these metals.

Table 4.6: Average (mean) metal concentrations in lettuce cultivars reported in mg kg⁻¹ dw, grown in soil L and soil H.

<u>Lettuce: Soil L</u>						
<u>Cultivar</u>	<u>N</u>	<u>Cd</u>	<u>Cu</u>	<u>Ni</u>	<u>Pb</u>	<u>Zn</u>
All Year Round	5	1.55	12.5	1.49	0.76	38.2
Buttercrunch	4	1.24	11.0	1.94	0.76	33.9
Great Lakes	5	1.78	9.5	1.59	0.73	40.5
Little Gem Cos	5	1.15	10.1	1.84	0.70	48.9
Little Leprechaun	5	1.47	10.5	2.20	1.07	42.3
Lollo Biondi	3	1.75	16.8	2.68	2.34	55.9
Lobjoits Cos	1	1.24	10.6	1.28	0.41	38.1
Marvel.	5	1.95	13.6	2.53	2.08	59.3
New Red Fire	5	1.96	13.1	2.67	1.63	61.6
Parella Green	5	1.39	10.1	1.71	0.77	33.4
Saladin	4	1.95	9.4	2.51	0.99	53.4
Trocadero	4	1.31	11.1	1.53	0.92	37.7
Valmaine	5	1.15	10.2	2.55	0.89	37.3
Webbs Wonderful	4	1.68	10.8	1.96	0.65	44.1
Winter Density	5	1.24	8.5	1.58	0.70	41.6
Average	65	1.53	11.1	2.03	1.04	44.6
<u>Lettuce: Soil H</u>						
<u>Cultivar</u>	<u>N</u>	<u>Cd</u>	<u>Cu</u>	<u>Ni</u>	<u>Pb</u>	<u>Zn</u>
All Year Round	5	14.7	9.3	13.6	1.28	139
Buttercrunch	4	8.3	6.4	13.5	0.49	105
Great Lakes	5	18.5	7.8	17.0	0.95	228
Little Gem Cos	5	8.2	7.3	15.9	1.90	124
Little Leprechaun	5	13.1	8.4	14.9	2.75	168
Lollo Biondi	3	16.6	13.8	18.2	2.32	234
Lobjoits Cos	1	11.9	6.3	16.7	0.51	172
Marvel.	5	14.0	10.1	17.6	1.86	159
New Red Fire	5	17.6	13.0	24.5	3.65	310
Parella Green	5	13.6	7.2	13.8	0.60	119
Saladin	4	12.1	8.9	21.0	2.02	152
Trocadero	4	11.4	7.8	14.8	1.50	108
Valmaine	5	12.2	6.4	16.2	0.76	176
Webbs Wonderful	4	13.6	10.2	19.7	2.87	178
Winter Density	5	10.1	6.0	16.1	0.80	103
Average	71	13.0	8.6	16.8	1.62	163

Lettuce cultivars grown in soil L

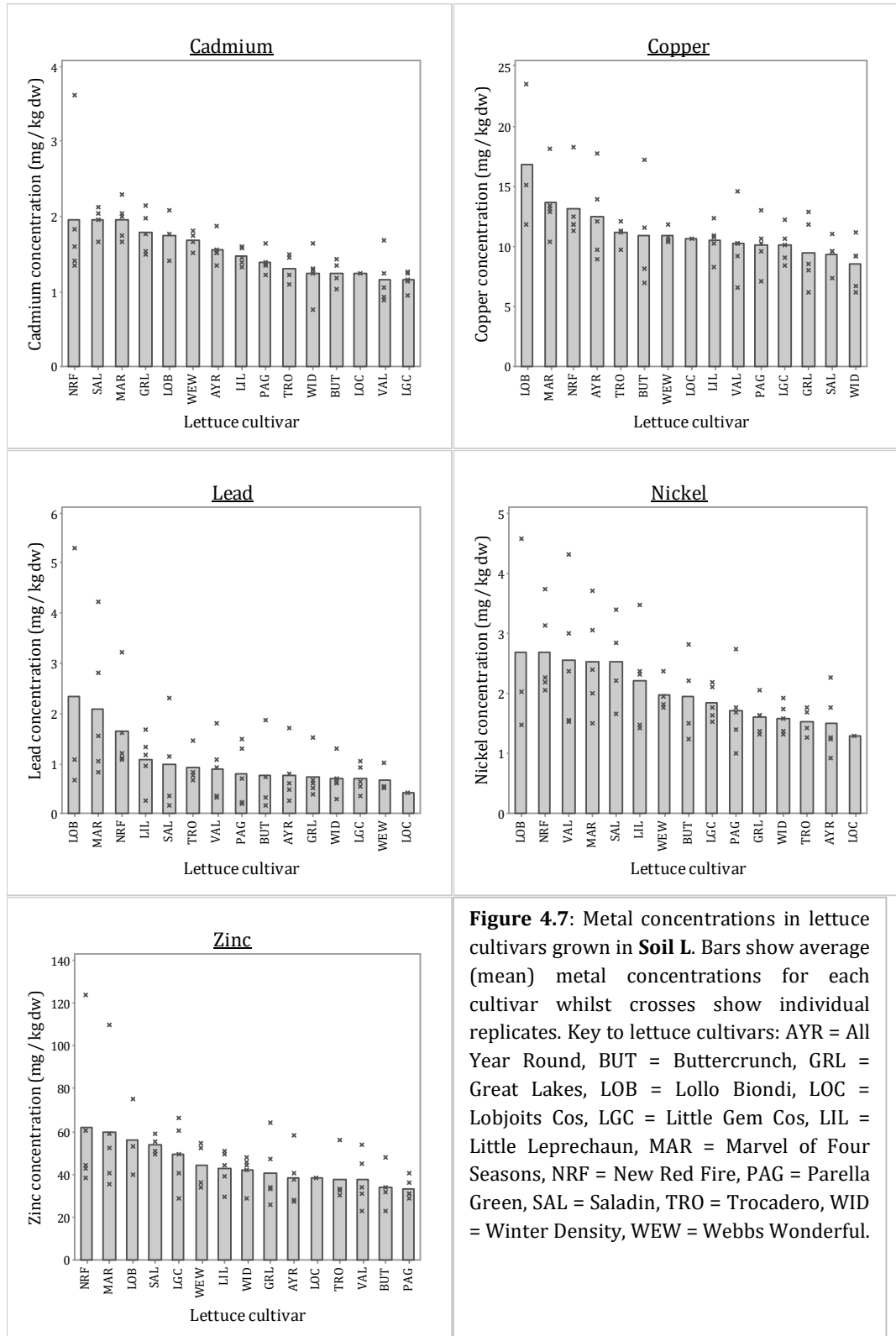


Figure 4.7: Metal concentrations in lettuce cultivars grown in **Soil L**. Bars show average (mean) metal concentrations for each cultivar whilst crosses show individual replicates. Key to lettuce cultivars: AYR = All Year Round, BUT = Buttercrunch, GRL = Great Lakes, LOB = Lollo Biondi, LOC = Lobjoits Cos, LGC = Little Gem Cos, LIL = Little Leprechaun, MAR = Marvel of Four Seasons, NRF = New Red Fire, PAG = Parella Green, SAL = Saladin, TRO = Trocadero, WID = Winter Density, WEW = Webbs Wonderful.

Lettuce cultivars grown in soil H

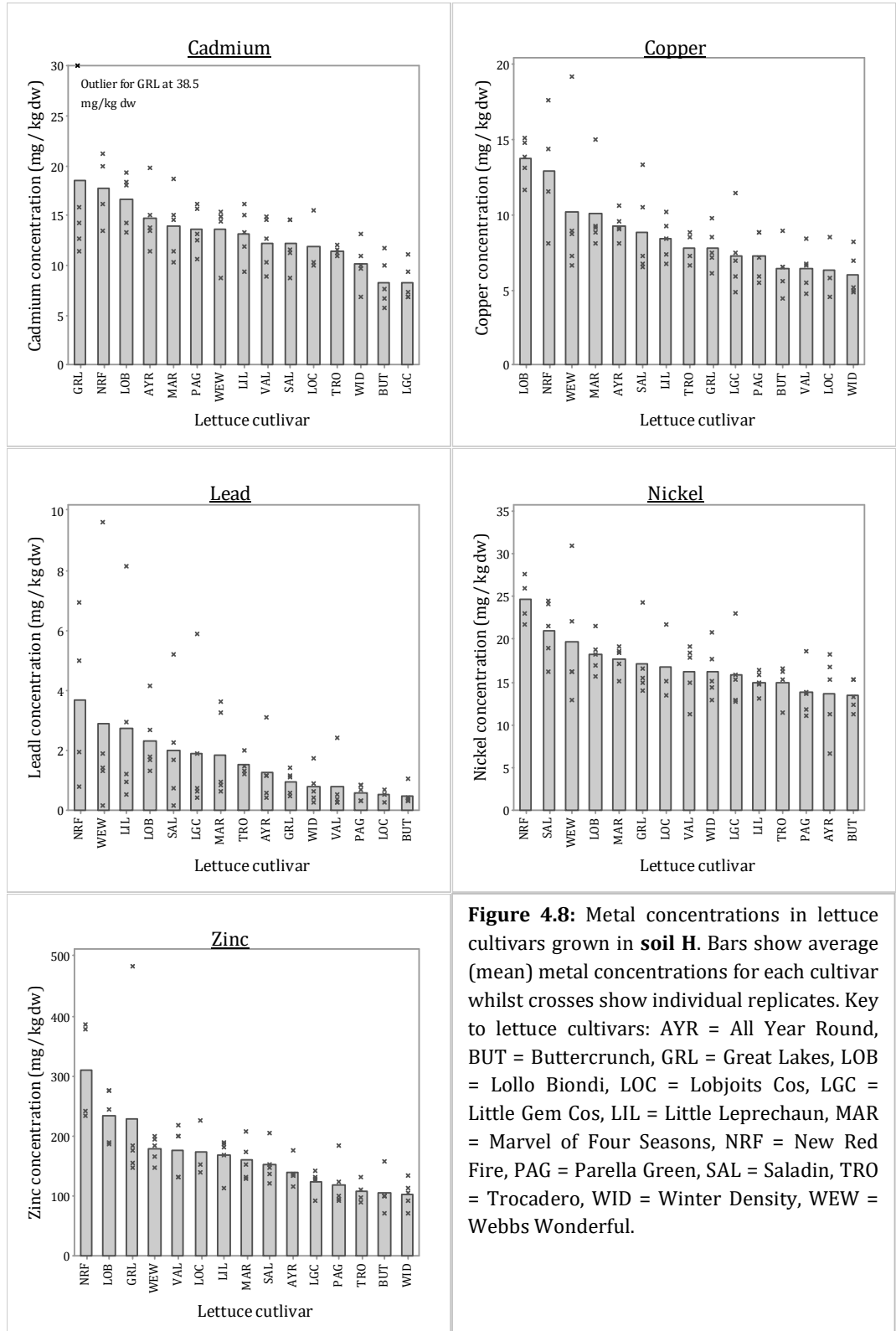


Figure 4.8: Metal concentrations in lettuce cultivars grown in **soil H**. Bars show average (mean) metal concentrations for each cultivar whilst crosses show individual replicates. Key to lettuce cultivars: AYR = All Year Round, BUT = Buttercrunch, GRL = Great Lakes, LOB = Lollo Biondi, LOC = Lobjoits Cos, LGC = Little Gem Cos, LIL = Little Leprechaun, MAR = Marvel of Four Seasons, NRF = New Red Fire, PAG = Parella Green, SAL = Saladin, TRO = Trocadero, WID = Winter Density, WEW = Webbs Wonderful.

Beans

French beans:

There were significant differences in the accumulation of Cd, Ni and Zn in the fruits of French bean cultivars ($p = 0.001$, 0.023 and <0.001 respectively). Differences in the accumulation of Cu in French bean fruits were not significant ($p = 0.064$) between cultivars, and concentrations of Pb in French bean fruits were below the LOQ (Table 4.5), so differences between cultivars in Pb accumulation in fruits could not be reliably assessed. Differences in the accumulation of Cd and Ni between cultivars, were significantly influenced by soil type ($p = < 0.001$ and 0.032 respectively) whereas differences in the accumulation of Cu and Zn were not ($p = 0.734$ and 0.458 respectively). There were significant differences also in the concentrations of some metals in French bean stems and leaves between cultivars; however, differences were not consistent. For example; there were significant differences in the accumulation of Ni in fruits and stems ($p = 0.019$) between cultivars, but not between accumulation of Ni in French bean leaves ($p = 0.138$). Average (mean) concentrations of metals in the tissues of French bean cultivars are reported in Table 4.7 and Table 4.8.

Table 4.7: Average (mean) metal concentrations in French bean fruits, leaves and stems. French beans were grown on soil L. Metal concentrations are reported in mg kg⁻¹ dw.

<u>French beans: Soil L</u>						
<u>Fruits</u>						
<u>Cultivar</u>	<u>N</u>	<u>Cd</u>	<u>Cu</u>	<u>Ni</u>	<u>Pb</u>	<u>Zn</u>
Cantare	4	0.059	9.4	7.93	0.049	47.4
Faraday	5	0.039	8.1	8.31	< LOQ	37.5
Masterpiece	5	0.028	10.1	8.61	0.053	40.7
Purple T.	4	0.029	8.9	8.41	0.055	46.1
Royalty	5	0.029	10.1	9.24	0.100	47.0
Sungold	3	0.043	8.8	5.85	< LOQ	37.9
Tendergreen	5	0.032	9.6	3.51	< LOQ	34.7
The Prince	5	0.043	8.3	3.78	< LOQ	28.7
Average	36	0.038	9.1	6.80	<LOQ	37.9
<u>Leaves</u>						
<u>Cultivar</u>	<u>N</u>	<u>Cd</u>	<u>Cu</u>	<u>Ni</u>	<u>Pb</u>	<u>Zn</u>
Cantare	4	0.161	7.4	2.22	0.33	45.2
Faraday	5	0.146	6.7	2.37	0.24	49.9
Masterpiece	4	0.088	9.0	2.41	0.41	61.6
Purple T.	5	0.191	6.8	3.82	0.55	53.1
Royalty	5	0.288	7.6	5.96	0.35	62.3
Sungold	4	0.275	7.1	5.74	0.29	58.9
Tendergreen	5	0.073	7.7	1.52	0.39	27.4
The Prince	5	0.090	8.5	1.67	0.29	36.1
Average	39	0.159	7.6	3.12	0.36	49.7
<u>Stems</u>						
<u>Cultivar</u>	<u>N</u>	<u>Cd</u>	<u>Cu</u>	<u>Ni</u>	<u>Pb</u>	<u>Zn</u>
Cantare	4	0.251	11.2	5.36	0.267	79.4
Faraday	5	0.198	8.7	3.62	0.735	72.4
Masterpiece	4	0.168	12.3	3.69	0.286	70.0
Purple T.	5	0.248	11.7	4.90	0.500	106.2
Royalty	5	0.164	11.3	4.10	0.230	70.5
Sungold	5	0.141	8.7	3.00	0.278	53.3
Tendergreen	5	0.137	9.8	1.64	0.346	40.1
The Prince	5	0.166	9.8	1.70	0.383	47.5
Average	38	0.183	10.3	3.45	0.383	67.0

Table 4.8: Average (mean) metal concentrations in French bean fruits, leaves and stems. French beans were grown on soil H. Metal concentrations are reported in mg kg⁻¹ dw.

<u>French beans: Soil H</u>						
<u>Fruits</u>						
<u>Cultivar</u>	<u>N</u>	<u>Cd</u>	<u>Cu</u>	<u>Ni</u>	<u>Pb</u>	<u>Zn</u>
Cantare	4	0.192	7.9	43.1	0.044	38.9
Faraday	5	0.168	6.9	44.2	< LOQ	33.8
Masterpiece	5	0.102	7.5	39.2	< LOQ	30.9
Purple T.	4	0.204	7.4	34.4	< LOQ	48.1
Royalty	5	0.235	9.7	49.9	< LOQ	49.5
Sungold	3	0.246	7.7	39.1	< LOQ	39.0
Tendergreen	5	0.180	7.2	40.9	< LOQ	34.4
The Prince	5	0.115	7.9	46.0	< LOQ	31.3
Average	36	0.176	7.8	42.5	<LOQ	37.9
<u>Leaves</u>						
<u>Cultivar</u>	<u>N</u>	<u>Cd</u>	<u>Cu</u>	<u>Ni</u>	<u>Pb</u>	<u>Zn</u>
Cantare	4	0.532	5.4	20.8	0.170	48.4
Faraday	5	0.515	3.9	13.1	0.224	54.3
Masterpiece	5	0.477	6.8	19.9	0.297	80.4
Purple T.	3	1.109	5.3	9.7	0.189	108.4
Royalty	4	0.751	6.5	18.2	0.289	80.0
Sungold	3	0.664	6.4	16.0	0.724	54.8
Tendergreen	5	0.410	11.1	16.1	0.913	39.2
The Prince	4	0.536	6.1	23.9	0.290	58.5
Average	33	0.595	6.6	17.4	0.391	63.9
<u>Stems</u>						
<u>Cultivar</u>	<u>N</u>	<u>Cd</u>	<u>Cu</u>	<u>Ni</u>	<u>Pb</u>	<u>Zn</u>
Cantare	4	0.933	7.6	25.9	0.527	80.6
Faraday	4	0.760	5.3	17.8	0.330	69.5
Masterpiece	5	1.197	7.9	23.6	0.309	92.8
Purple T.	5	1.434	8.2	25.5	0.386	133.1
Royalty	5	1.415	7.8	22.9	0.493	111.1
Sungold	3	1.143	6.4	16.6	0.265	70.8
Tendergreen	4	0.831	7.4	14.8	0.408	72.8
The Prince	5	1.228	7.1	21.0	0.385	82.2
Average	35	1.140	7.3	21.4	0.392	91.4

Broad beans:

There were no significant differences between cultivars in the accumulation of Cd, Cu, Ni and Zn in broad bean seeds ($p > 0.05$). However, differences in the accumulation of Zn between cultivars were significantly influenced by soil type ($p = 0.001$). Concentrations of Pb in broad bean seeds were below the LOQ. Only concentrations of Zn were significantly different between cultivars when measured in broad bean leaves ($p = 0.001$). There were significant differences between cultivars in the accumulation of Cu and Ni in broad bean stems ($p < 0.001$ and $p = 0.004$) but not for the accumulation of Cd, Pb and Zn ($p = 0.178, 0.310$ and 0.066 respectively). Average (mean) concentrations of Cd, Cu, Ni, Pb and Zn in the tissues of broad bean cultivars are reported in Table 4.9 and Table 4.10. Average (mean) concentrations and individual concentrations for each replicate of Cd, Cu, Ni, Pb and Zn, in broad bean seeds and French bean pods, are displayed in Figure 4.9 and Figure 4.10.

Table 4.9: Average (mean) metal concentrations in Broad bean seeds, leaves and stems.
Metal concentrations are reported in mg kg⁻¹ dw.

<u>Broad beans: Soil L</u>						
<u>Fruits</u>						
<u>Cultivar</u>	<u>N</u>	<u>Cd</u>	<u>Cu</u>	<u>Ni</u>	<u>Pb</u>	<u>Zn</u>
Aquadulcia C.	1	0.061	16.0	4.71	<LOQ	63.9
Bunyards	3	0.040	13.3	3.46	<LOQ	62.8
Dreadnought	2	0.056	12.5	4.53	<LOQ	52.1
Express	1	0.061	15.9	5.80	<LOQ	68.6
Karmazyn	1	0.073	19.4	6.40	<LOQ	47.3
Robin Hood	1	0.093	11.0	9.37	<LOQ	71.1
Witkeim M.	4	0.032	6.8	3.58	<LOQ	49.6
Average	36	0.038	9.1	6.80	<LOQ	37.9
<u>Leaves</u>						
<u>Cultivar</u>	<u>N</u>	<u>Cd</u>	<u>Cu</u>	<u>Ni</u>	<u>Pb</u>	<u>Zn</u>
Aquadulcia C.	5	0.086	19.5	6.34	<LOQ	71.4
Bunyards	5	0.072	16.5	5.00	<LOQ	71.2
Dreadnought	4	0.069	22.5	5.44	<LOQ	70.6
Express	3	0.081	22.2	6.98	<LOQ	78.3
Karmazyn	3	0.053	20.7	6.53	<LOQ	74.2
Robin Hood	5	0.161	22.2	8.27	<LOQ	90.9
Witkeim M.	4	0.057	16.0	4.87	<LOQ	63.6
Average	29	0.086	19.8	6.20	<LOQ	74.5
<u>Stems</u>						
<u>Cultivar</u>	<u>N</u>	<u>Cd</u>	<u>Cu</u>	<u>Ni</u>	<u>Pb</u>	<u>Zn</u>
Aquadulcia C.	5	0.197	7.8	3.09	0.178	51.4
Bunyards	4	0.132	6.4	2.02	0.157	41.6
Dreadnought	5	0.152	6.3	2.30	0.213	48.4
Express	5	0.198	8.1	3.22	0.226	49.3
Karmazyn	4	0.186	10.1	4.22	0.249	54.6
Robin Hood	4	0.365	12.3	6.88	0.372	80.7
Witkeim M.	5	0.112	5.5	2.31	0.197	34.4
Average	32	0.188	7.9	3.35	0.224	50.8

Table 4.10: Average (mean) metal concentrations in Broad bean seeds, leaves and stems.
Metal concentrations are reported in mg kg⁻¹ dw.

<u>Broad beans: Soil H</u>						
<u>Seeds</u>						
<u>Cultivar</u>	<u>N</u>	<u>Cd</u>	<u>Cu</u>	<u>Ni</u>	<u>Pb</u>	<u>Zn</u>
Aquadulcia C.	1	0.154	10.4	18.2	< LOQ	69.1
Bunyards	2	0.209	17.8	28.7	< LOQ	64.8
Dreadnought	3	0.267	14.6	25.5	< LOQ	71.2
Express	2	0.205	12.2	23.1	< LOQ	51.7
Karmazyn	2	0.262	18.0	16.7	< LOQ	75.0
Robin Hood	1	0.149	10.9	22.0	< LOQ	49.6
Witkeim M.	3	0.162	13.7	31.0	< LOQ	56.5
Average	14	0.210	14.4	24.8	<LOQ	63.2
<u>Leaves</u>						
<u>Cultivar</u>	<u>N</u>	<u>Cd</u>	<u>Cu</u>	<u>Ni</u>	<u>Pb</u>	<u>Zn</u>
Aquadulcia C.	4	0.399	17.5	30.5	0.067	69.7
Bunyards	3	0.265	13.7	29.0	0.122	66.1
Dreadnought	5	0.407	17.5	29.8	0.058	80.6
Express	3	0.235	17.8	28.7	0.049	69.0
Karmazyn	3	0.318	18.6	23.2	< LOQ	80.5
Robin Hood	4	0.497	20.8	33.8	0.050	92.6
Witkeim M.	3	0.253	17.8	34.9	< LOQ	60.1
Average	25	0.353	17.8	30.2	0.057	75.2
<u>Stems</u>						
<u>Cultivar</u>	<u>N</u>	<u>Cd</u>	<u>Cu</u>	<u>Ni</u>	<u>Pb</u>	<u>Zn</u>
Aquadulcia C.	5	0.674	6.6	16.6	0.219	82.6
Bunyards	5	0.713	6.3	14.1	0.158	86.3
Dreadnought	5	1.198	7.6	15.5	0.213	126.1
Express	5	1.024	7.8	16.4	0.789	77.2
Karmazyn	4	1.072	8.4	14.4	0.205	97.1
Robin Hood	4	0.893	10.7	26.7	0.344	91.5
Witkeim M.	5	0.839	8.3	18.9	0.411	94.8
Average	33	0.912	7.9	17.3	0.338	93.6

Metal concentrations in bean fruits/seeds from plants grown in soil L

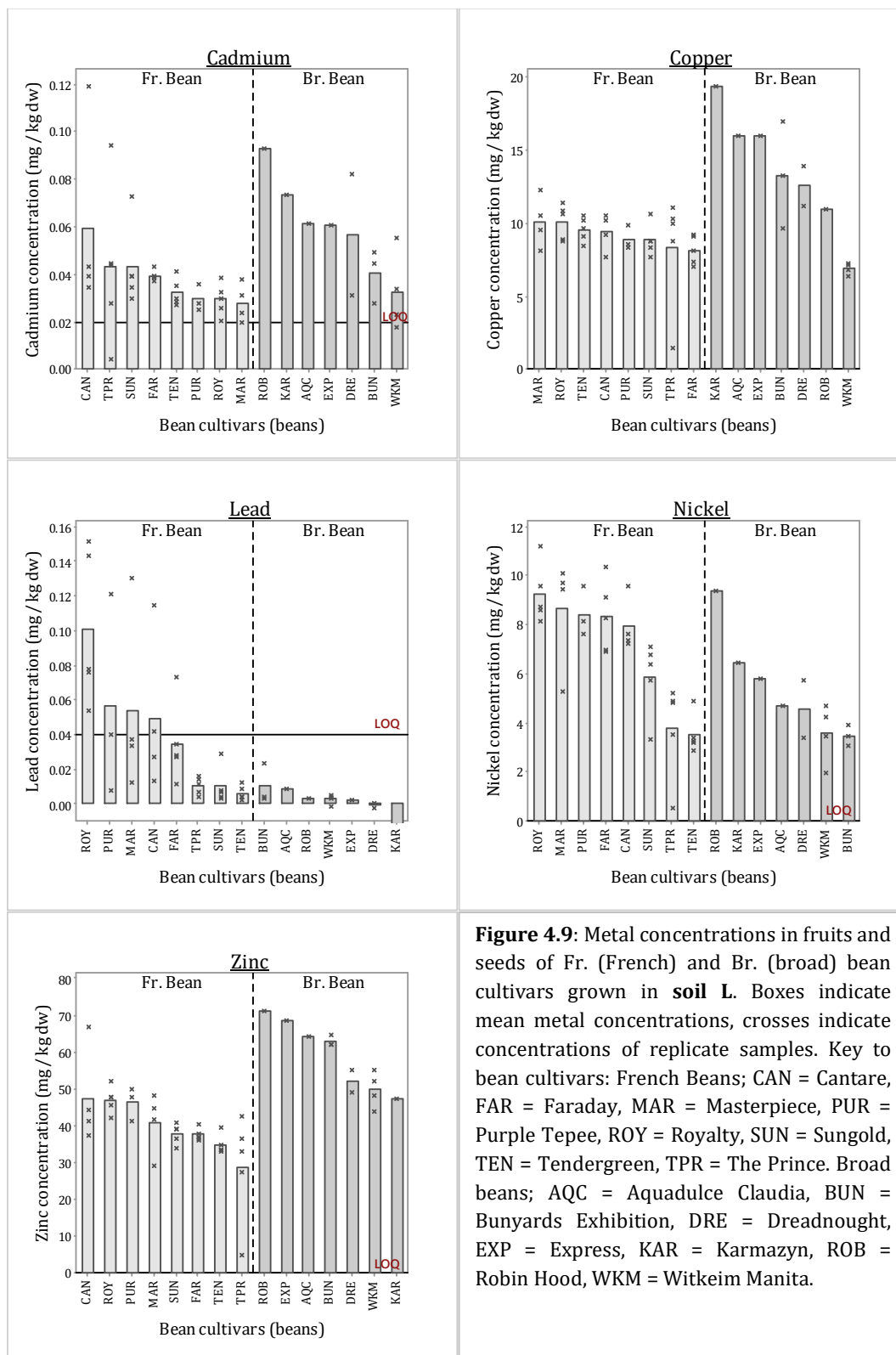


Figure 4.9: Metal concentrations in fruits and seeds of Fr. (French) and Br. (broad) bean cultivars grown in **soil L**. Boxes indicate mean metal concentrations, crosses indicate concentrations of replicate samples. Key to bean cultivars: French Beans; CAN = Cantare, FAR = Faraday, MAR = Masterpiece, PUR = Purple Tepee, ROY = Royalty, SUN = Sungold, TEN = Tendergreen, TPR = The Prince. Broad beans; AQC = Aquadulce Claudia, BUN = Bunyards Exhibition, DRE = Dreadnought, EXP = Express, KAR = Karmazyn, ROB = Robin Hood, WKM = Witkeim Manita.

Metal concentrations in bean fruits/seeds from plants grown in soil H

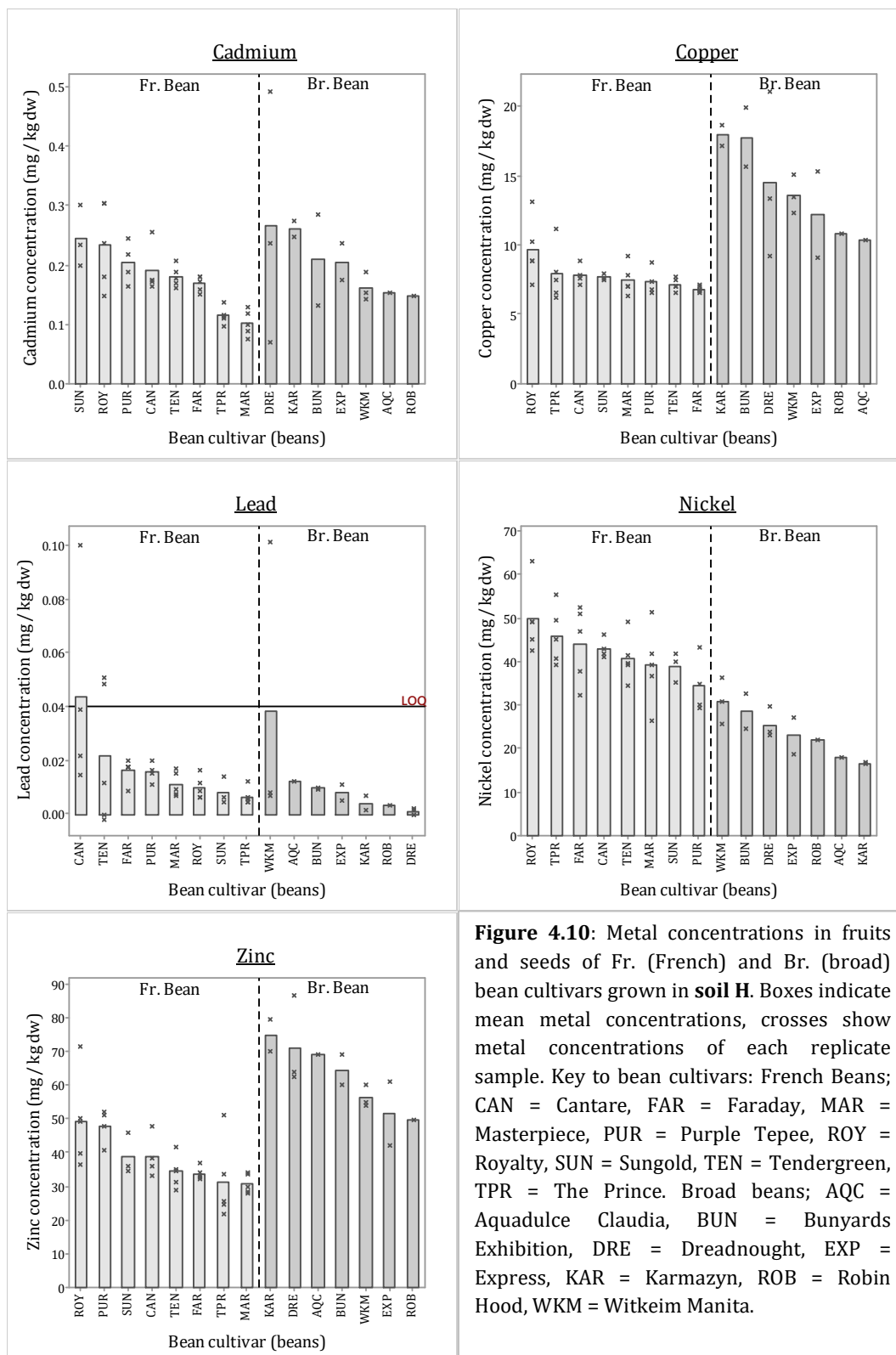


Figure 4.10: Metal concentrations in fruits and seeds of Fr. (French) and Br. (broad) bean cultivars grown in **soil H**. Boxes indicate mean metal concentrations, crosses show metal concentrations of each replicate sample. Key to bean cultivars: French Beans; CAN = Cantare, FAR = Faraday, MAR = Masterpiece, PUR = Purple Tepee, ROY = Royalty, SUN = Sungold, TEN = Tendergreen, TPR = The Prince. Broad beans; AQC = Aquadulce Claudia, BUN = Bunyards Exhibition, DRE = Dreadnought, EXP = Express, KAR = Karmazyn, ROB = Robin Hood, WKM = Witkeim Manita.

Selecting bean cultivars:

From Section 4.3.1 it is apparent that French bean and broad bean plants are able to regulate concentrations of metals reaching edible tissues; French bean fruits, broad bean seeds and young leaves. It is difficult to determine from Figure 4.9 and Figure 4.10 which cultivars, if any, should be preferred for urban horticulture. The French bean cultivar, '*Royalty*' (ROY), was found to accumulate the highest concentrations of Cu, Ni and Zn, and second highest of Cd, in its fruits compared to other varieties of French bean, when grown in soil H. '*Marvel of four seasons*' (MAR) was found to accumulate lower concentrations of Cd than other varieties, in fruits and leaves regardless of soil type. Few studies have investigated differences between cultivars of French beans. Alexander et al (2006) studied differences in a narrow range of cultivars. The suggestion from that paper that the variety '*The Prince*' (TPR) could be used as a low accumulator of Cu cannot be supported or refuted, from the results of the present study alone.

To the authors' knowledge, this is the only study that has investigated differences in metal accumulation between cultivars of broad beans. Differences in metal accumulation between cultivars of broad beans were largely non-significant for the accumulation of metals into broad bean seeds. The present study was limited in the number of French bean ($n = 8$) and broad bean cultivars ($n = 7$) included. The variety of French bean cultivars was further limited by the omission of taller varieties which could not be grown in glasshouse conditions. Future research

investigating differences over a wider range of cultivars would be worthwhile.

4.3.3 Predicting metal uptake into lettuces and beans

The Hough v 2 models (Chapter 3) and the CLEA 'green vegetable model' were assessed for their ability to predict concentrations of Cd, Pb and Ni in lettuces and bean fruits/seeds. The Hough v 2 models were also used to predict accumulations of Cu and Zn in lettuces and bean fruits/seeds. Root mean square logarithmic errors (RMSLEs) (Equation 3.8) were calculated for the comparison between measured and modelled data and are reported in Table 4.11.

Table 4.11: RMSLE values (Equation 3.8) for the predictions of metal concentrations in bean fruits/seeds and lettuces by the CLEA 'green vegetable' model and the Hough v2 models.

Crop	Soil	Model	Cd	Cu	Pb	Ni	Zn
Lettuce	L	Hough v2.	0.200	0.114	1.348	0.197	0.539
		CLEA	0.110	-	0.803	0.172	-
	H	Hough v2.	0.538	0.179	1.415	0.876	0.192
		CLEA	0.281	-	1.227	0.410	-
Fr. Bean (pods)	L	Hough v2.	1.320	0.217	<LOQ	0.370	0.196
		CLEA	1.652	-	<LOQ	0.486	-
	H	Hough v2.	0.810	0.243	<LOQ	1.098	0.157
		CLEA	1.624	-	<LOQ	0.805	-
Br. Bean (seeds)	L	Hough v2.	1.201	0.172	<LOQ	0.208	0.104
		CLEA	1.532	-	<LOQ	0.315	-
	H	Hough v2.	0.788	0.123	<LOQ	0.901	0.154
		CLEA	1.638	-	<LOQ	0.596	-

Cadmium

Both the CLEA model and the Hough v2 models over-predicted Cd concentrations in French and broad bean fruits/seeds. Errors in predictions by the CLEA model were greater, amounting to an over-prediction of 25-40 fold of the mean measured Cd concentration. The Hough v2 models over-predicted Cd concentrations in fruits/seeds by a factor of 4-6 fold when French and broad beans were grown in soil H and approximately 10-20 fold when beans were grown in soil L. Both models predicted Cd concentrations in lettuce reasonably well (Figure 4.11).

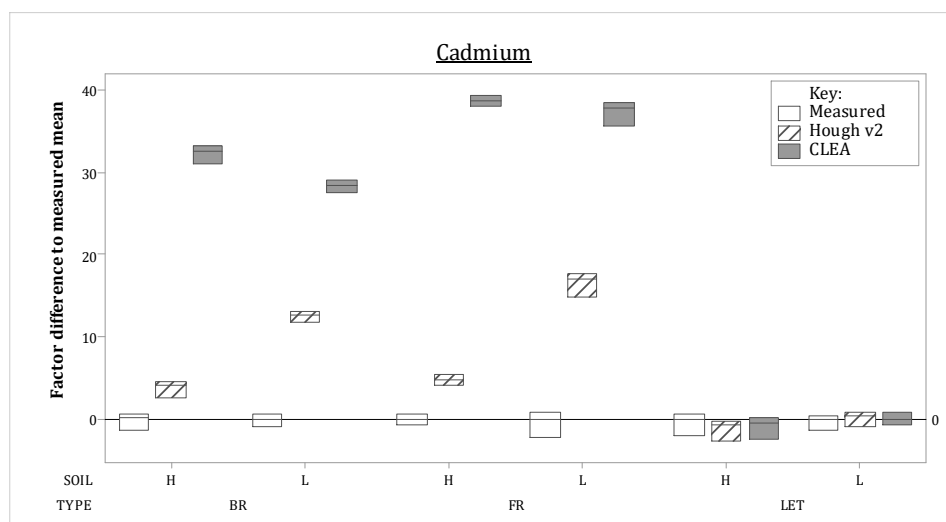


Figure 4.11: Factor differences (Equation 4.1) between measured and predicted concentrations of Cd in broad bean seeds (BR), French bean fruits (FR) and lettuce leaves (LET) from plants grown on soil L and soil H. Boxes show the range and median difference of the data to the measured mean.

Copper

The Hough v2 models predicted Cu concentrations in broad bean seeds, French bean fruits and lettuce leaves, really well. The Hough v 2 models slightly over-predicted Cu concentrations in French bean fruits but otherwise model predictions were within the range of measured data (Figure 4.12).

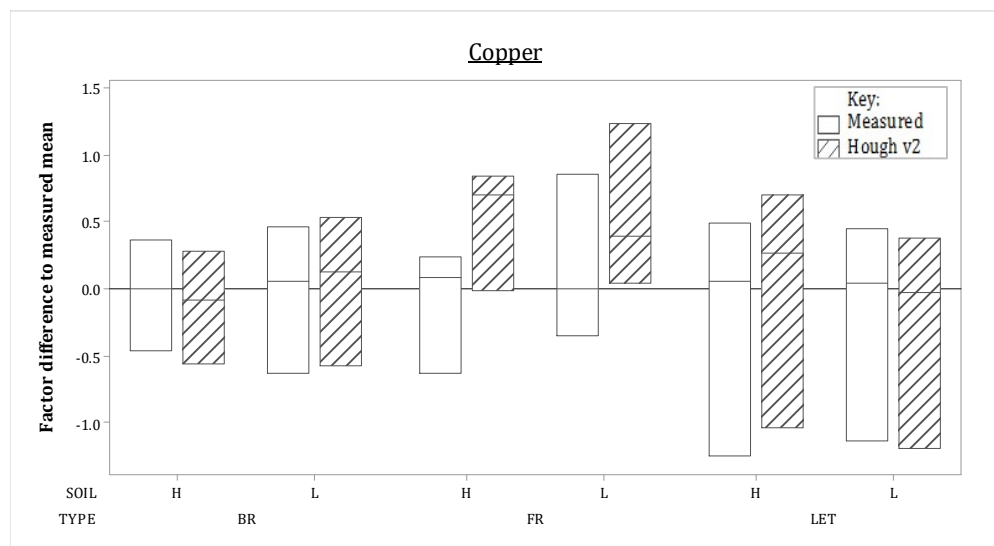


Figure 4.12: Factor differences (Equation 4.1) between measured and predicted concentrations of Cu in broad bean seeds (BR), French bean fruits (FR) and lettuce leaves (LET) from plants grown on soil L and soil H. Boxes show the range and median difference of the data to the measured mean.

Lead

Predictions of Pb concentrations in French and broad bean fruits/seeds were not made as the measured Pb concentrations fell below the LOQ and were therefore unreliable (Table 4.5). The CLEA models made much better predictions of Pb concentrations in lettuces than the Hough v2 models did. The Hough v2 models over-predicted concentrations of Pb in lettuce by a factor of 10-15 fold.

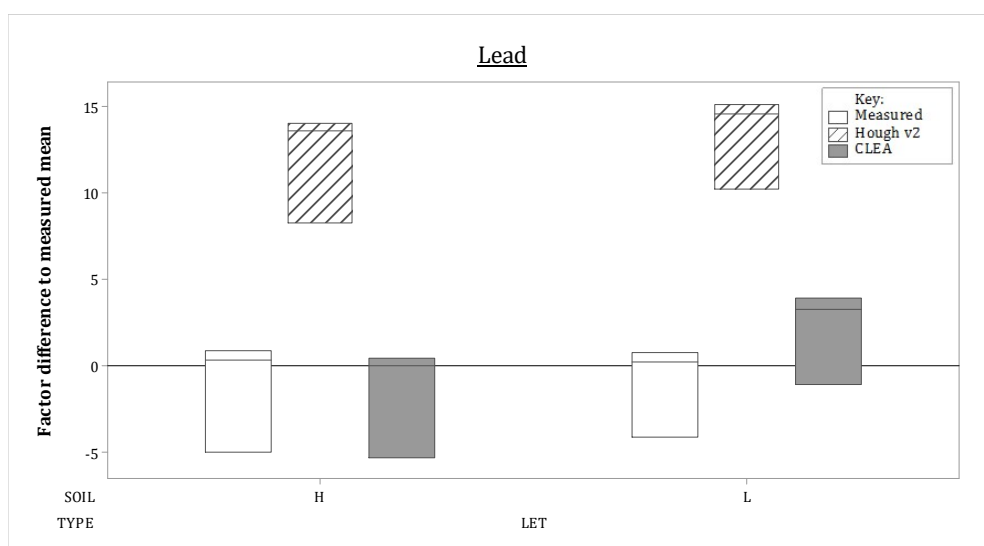


Figure 4.13: Factor differences (Equation 4.1) between measured and predicted concentrations of Pb in broad bean seeds (BR), French bean fruits (FR) and lettuce leaves (LET) from plants grown on soil L and soil H. Boxes show the range and median difference of the data to the measured mean.

Nickel

The CLEA and Hough v2 models both made reasonably good predictions of the concentrations of Ni in French bean fruits, broad bean seeds and lettuce leaves. Both models slightly under-predicted Ni concentrations in French and broad beans when they were grown in the more highly contaminated soil.

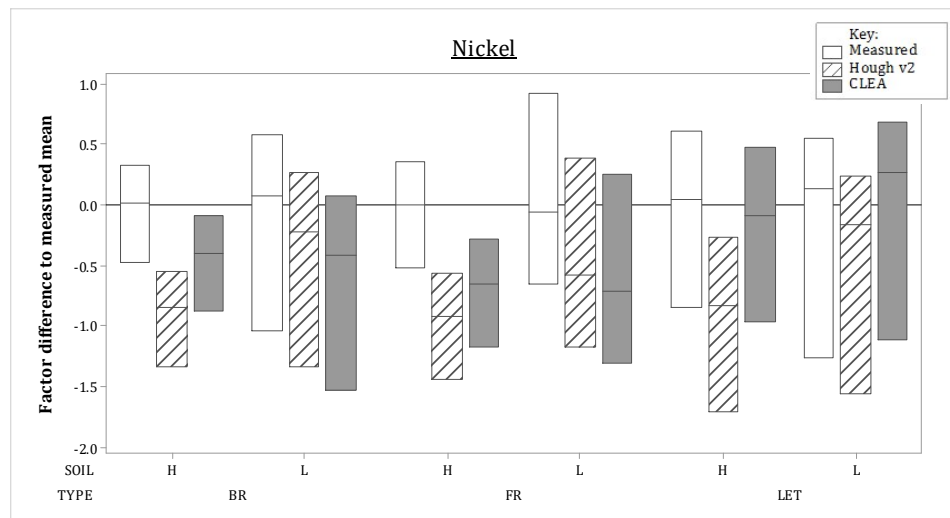


Figure 4.14: Factor differences (Equation 4.1) between measured and predicted concentrations of Ni in broad bean seeds (BR), French bean fruits (FR) and lettuce leaves (LET) from plants grown on soil L and soil H. Boxes show the range and median difference of the data to the measured mean.

Zinc

The Hough v 2 models predicted Zn concentrations in French bean fruits and broad bean seeds, really well. The Hough v2 models predicted Zn concentrations in lettuce well when lettuce was grown in higher soil H but over-predicted Zn concentrations in lettuce when lettuce plants were grown in soil L.

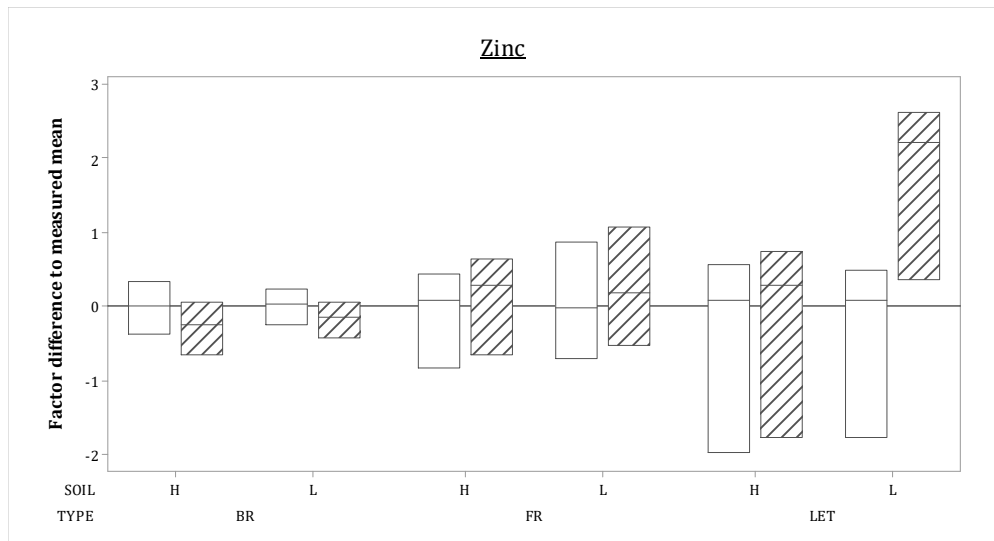


Figure 4.15: Factor differences (Equation 4.1) between measured and predicted concentrations of Zn in broad bean seeds (BR), French bean fruits (FR) and lettuce leaves (LET) from plants grown on soil L and soil H. Boxes show the range and median difference of the data to the measured mean.

Discussion of model predictions

The selection of lettuces and beans in the same group for modelling uptake of metals in the CLEA 'green vegetable' model appears arbitrary and does not satisfactorily consider the mechanisms of plant uptake. This is illustrated in the difference between measured and modelled concentrations of Cd in French bean fruits and broad bean seeds by the CLEA model. The Hough v 2 model used to predict uptake of Cd into French bean fruits and broad bean seeds resulted in a smaller error. However, the Hough v 2 model used to predict Cd uptake into French and broad bean fruits/seeds, was based mainly on the accumulation of Cd into tomato fruits. Whilst tomato fruits may be regarded as more similar to French and broad bean fruits/seeds than lettuce leaves, it is unsurprising that there are differences between French and broad beans and tomatoes in Cd accumulation also. Thorne et al (2004), from which the CLEA 'green vegetable' model is derived, contains separate models for leguminous and non-leguminous plants which may improve predictions of Cd uptake for French and broad beans. A better method still would be to model uptake of metals into produce individually when this option is available.

Differences in the accumulation of Ni, Cu and Zn between French, broad beans and lettuces, were less pronounced than for Cd. This resulted in reasonable predictions of Ni accumulation made by both the Hough v 2 models and the CLEA model. The Hough v 2 models also made reasonable predictions of concentrations of Cu and Zn in lettuces and beans. The Hough v 2 models over-predicted concentrations of Pb in lettuces. The

accumulation of Pb into lettuce from soil is likely to be small and it is possible that foliar deposition of Pb in studies from which the Hough v 2 models were derived, resulted in the over-predictions. The growth of plants in a glasshouse environment with perlite, would have substantially reduced particulate deposition of Pb on lettuces in the present study.

4.4 CONCLUSIONS

Relative accumulation of cationic trace metals (Cd, Cu, Ni, Pb and Zn) in French and broad beans was observed to be significantly different compared to accumulation of the same metals into lettuces. Grouping these produce together in plant uptake models, such as the CLEA 'green vegetable' model, could lead to substantial errors, especially in the prediction of plant uptake of Cd. French bean and broad bean plants may have evolved more effective strategies to prevent uptake and translocation of non-essential metals, Cd and Pb, than lettuces, and would be safer plants to grow on urban soils contaminated with these metals. The lettuce cultivars '*New Red Fire*' and '*Winter Density*' were found to accumulate higher and lower concentrations of metals respectively compared with other lettuce cultivars. It was not possible to identify cultivars of French and Broad bean which accumulate lower concentrations of Cd and Pb than others. Further research should focus on finding low metal-accumulating varieties of French and broad bean which could be grown on mildly contaminated soils.

5 THE HEALTH BENEFITS AND RISKS FOR ALLOTMENT GARDENERS IN AN URBAN CITY

5.1 INTRODUCTION

Several authors have alluded that both the health benefits (from increased fruit and vegetable consumption, exercise etc.), and health risks (exposure to contaminants), should be considered in the regulation of gardening activities on urban land (Brown and Jameton 2000; Frumkin 2001; Leake *et al.* 2009; Brown *et al.* 2016). Taking part in gardening activities is associated with a lifestyle that is beneficial to health. Gardeners have been observed to partake in greater levels of physical activity than non-gardeners, and consume a larger number of portions of fruit and vegetables (Alaimo *et al.* 2008; Park *et al.* 2009; Carney *et al.* 2011). Gardeners have also been found to have a lower average body-mass-index (BMI) compared to non-gardeners, meaning they are less likely to be overweight or obese (Zick *et al.* 2013; Wood *et al.* 2015). Finally, gardening activities have been shown to reduce stress levels (Van den Berg and Custers 2011; Hawkins *et al.* 2011) and are beneficial to psychological health in other ways too; including through community cohesion (Wakefield *et al.* 2007) and through a sense of achievement associated with successfully growing produce (Wakefield *et al.* 2007; Wood *et al.* 2015). Increased fruit and vegetable consumption and physical activity, has been reported to have a protective effect against a number of diseases including heart diseases, stroke and cancers (Steinmetz and Potter 1996; Kruk 2007); diseases which accounted for

the top 5 causes of death in Western Europe between 1990 and 2010 (Lozano *et al.* 2012).

In contrast to the health benefits arising from gardening, there have been many studies which have found an association between neighbourhood soil contamination and ill-health: Neighbourhood soil contamination with Pb, has been associated with high blood-Pb levels in children and linked to a reduction in IQ (Zahran *et al.* 2013). Soil contamination with arsenic has also been found to be correlated with mental retardation (Liu *et al.* 2010). Furthermore, environmental exposure to Cd has been associated with an increase in the likelihood of all-cause mortality, cardiovascular disease and cancer in two prospective studies (Nawrot *et al.* 2008, 2010; Menke *et al.* 2009). Staessen *et al.* (1999) also observed that moderate environmental exposures to Cd were correlated with lower bone density in women, increasing the likelihood of fractures.

Health outcomes associated with gardening, are dependent on the types of activities the gardeners are doing, how often they visit their garden and what produce they grow and consume. Gardening routines, including the regularity and duration of visits to the allotment or home garden, could increase both health benefits and health risks (EA 2009d; Hawkins *et al.* 2011). Different types of gardening activities, such as digging, weeding etc. all have different roles in improving muscle strength, co-ordination or improving cardiovascular fitness (Park *et al.* 2011) and likely affect health risks too, through proximity to the soil and soil dusts. The volume of

consumption of home grown produce has not been widely investigated in the literature (Mulloli *et al.* 2004) but is associated with increased exposure to metals from the soil. Preparation of produce by peeling and washing during cooking or otherwise, can reduce exposure to metals contained in soil particles or in the root/fruit exodermis (outer layer) (Arif *et al.* 2011). The increase in fruit and vegetable consumption and physical activity from gardening and GYO, could help tackle 'sloth and obesity' in the western world (Leake *et al.* 2009).

In previous chapters (2-4), it has been shown that potentially toxic metals are present in different concentrations in the soils of urban allotments and can accumulate in different concentrations in edible plant tissues. In this chapter, the influence of gardening routines on exposures to the health benefits and health risks of gardening, is investigated through a questionnaire survey of gardeners, living in and around Nottinghamshire, UK. This is the first study to explicitly evaluate gardening routines from the perspective of impact to both health benefits and risks. Information from this chapter is used to underpin a novel benefit/risk assessment model for the purpose of assessing outcomes of gardening on contaminated land (Chapter 6). The aims of the questionnaire survey were to: obtain data that could be used to estimate health risks and benefits for a group of gardeners; investigate the relationships between participation in gardening, fruit and vegetable consumption, exercise and health, and to investigate the quantity and types of produce grown and consumed by gardeners.

5.2 METHODS

5.2.1 Questionnaire Development

A self-administered 'Gardening and Health' questionnaire was used to capture data on gardening routines that might influence health benefits and risks. A copy of the questionnaire can be found in Appendix II. The questionnaire contained the following sections designed to obtain different information about the routines of gardeners:

Section A. Participation in gardening. Participants were asked where they had access to a garden (e.g. allotment, home garden) and how often they spent some time gardening there.

Section B. Gardening activities. Participants were asked how often they engaged with different gardening activities from a pre-selected list.

Section C. Produce consumption. Participants were asked which produce they grew on their allotment/home garden from a pre-selected list and to estimate how often they consumed produce from their garden in portions per week/month.

Section D. Lifestyle. Participants were asked about their lifestyle including fruit and vegetable consumption, exercise, smoking status, alcohol consumption and socio-economic questions.

Section E. Health. Participants were asked to fill in the short-form (SF) 36 v2[©] questionnaire and to record their height and weight.

Section F. Further participation. Participants were asked if they would like to take part in future studies and to indicate their preferred choice of incentive.

Participants were asked to indicate their age and gender on the consent form.

Except for section E, which included the SF 36 v 2 © (see Section 5.2.2), the questionnaire was designed by the researcher. Questions were designed using previously published questions or responses from previous surveys where available (see individual questionnaire sections below) to reduce bias and allow for comparisons to be made with existing datasets. Most questions used a likert scale format, recognisable in most surveys, with differing numbers of responses.

Section A - Gardening Participation

Questions 1, 2, 3, 5 and 6 were used to determine how many of the participants had access to an allotment or a home garden and the proportion of participants that used their gardens for growing food. Questions 4, 7 and 8 helped to determine how often participants did some gardening either at home or on their allotment: Questions were divided into categories based on research by a previous author (Saunders 1993 - used by the CLEA model: ; EA 2009d) and divided into Spring/Summer, Autumn/Winter to help account for differences in participant responses as a result of seasonality.

Section B - Gardening Activities

The first 10 questions in this section were developed using a 5 point Likert scale and designed to ask respondents about their engagement with a variety of different gardening activities. Activities were chosen

based on Park *et al.*, (2011) which documents intensities of different activities measured in METs (metabolic equivalents) for a group of Korean gardeners. Respondents were asked in an open question (Q 11) to list any other activities they regularly took part in. The contribution of gardening activities to the exercise regime of participants was assessed in Q12 using a 10-point likert scale.

Section C - Produce consumption

Participants were given a table of suggested produce (with space to record their own) and asked to indicate which they grew, where they grew it and how many times per week/month they consumed the produce. They were also asked to indicate whether they normally peeled the produce, or washed it, prior to consumption. Participants were also asked to indicate, for each type of produce, how much of their general consumption came from the supermarket or their home garden/allotment.

Section D - Lifestyle

This section included questions asking participants to describe their fruit and vegetable consumption and regularity of exercise. Participants were also asked about their alcohol consumption, cigarette use, household income before and after retirement and highest qualification achieved. Respondents were asked to indicate their level of each lifestyle activity (e.g. fruit and vegetable consumption) over a recent time period (e.g.

“How many portions of fruit and vegetables (not including potatoes) did you eat yesterday?”) and more generally. Fruit and vegetable consumption questions and responses and those for exercise were designed for comparison with Health Survey England, 2013 data recorded in Health and Social Care Information Centre (HSCIC) (2013a). Questions and answer categories for alcohol consumption and smoking are based on data from HSCIC (2012a, 2013b). Social economic status (SES) was assessed by a composite score calculated from the highest qualification achieved (education) and household income (household income before retirement was calculated for individuals who had retired). Responses were graded 1-n (with n considered the highest) and then multiplied together to give a score for SES (a similar framework to that used by the two-fold or four-fold index; Hollinshead (unpublished) 1975). Qualification responses including vocational and non-vocational qualifications were categorised according to the National Qualifications Framework v 5. Responses for household income were categorised so that possible answers reflected the distribution of household income for the UK, according to the IFS (2016).

Section E - Health

The SF 36 v 2[©] questionnaire was administered as a measure of physiological and psychological health and well-being (see Section 5.2.2). Participants were also asked to record their own height and weight and given the option to record this in metric or imperial units. Height and

weight were converted to metric units (m and kg) and used to calculate BMI.

5.2.2 Short-form 36 v 2[®]

The short-form 36 v 2[®] is recognised internationally as a validated, self-reported measure of physical and psychological health and well-being. The questionnaire contains 36 questions relating to eight categories: *Physical function* (PF), *Role Physical* (RP), *Bodily pain* (BP), *General health* (GH), *Vitality* (VT), *Social Function* (SF), *Role emotional* (RE) and *Mental health* (MH). Composite scores for the first four categories are combined to give the *Physical Component Score* (PCS) and composite scores from the last four categories, the *Mental Component Score* (MCS). Scores for each category are normalised according to population means taken from a large western population. The SF 36 v 2 has also been previously used by a number of other researchers investigating health outcomes arising from gardening (Van den Berg *et al.* 2010; Park *et al.* 2011; Hawkins *et al.* 2011; Wang and MacMillan 2013). An additional question was used to ascertain whether the respondents' responses to the SF 36 v 2[®] were typical or whether they might have been affected by illness, injury or a significant emotional experience such as marriage or becoming a parent. This was previously implemented for a similar study by (2010) and makes sense for this sort of study when a longer period than the usual recall period of 4 months is important.

5.2.3 Study design and administration

Participants were firstly recruited as part of a study investigating the composition of the soils of allotment gardens. Details of access to participants, ethical approval and permissions for this part of the study are explained in Section 2.2. Participants were also recruited at Grow-Your-Own events run by Nottingham City Council (NCC) on 6th September 2014 and 5th September 2015. The research team were invited by the NCC to set-up a stall at the event and hand out questionnaire packs containing questionnaires and consent forms (see Section 2.2 and Appendix I for more information). The packs were pre-stamped and addressed for return. Entry into a prize draw for participants with the chance to win £100 cash prize or a gardening equivalent prize (including membership to the RHS or National Trust) was offered as an incentive for the return of completed questionnaires. One winner was picked at random from the list of entries. All participants were required to be over 18 years old.

5.2.4 Scoring responses and data entry

Questionnaire responses and personal information (names and contact details) were entered into separate databases linked using an ID number unique to each individual questionnaire. The questionnaire was set up as a tabbed form in Microsoft Access 2010[®]. Entered data was automatically recorded in related data tables that were set up for each section of the questionnaire (Section C was split into two due to its size). Questions which required participants to answer using a likert scale (e.g. 'Never',

'Some of the time', 'most of the time', 'all of the time') were recorded numerically by the database software using numbers 0 – nth response. Non-responses were recorded as '999' and removed prior to data analysis. Responses to open-ended questions were recorded in full. Hard copies of questionnaires and consent forms were kept in a locked cabinet for reference with contact details removed and shredded as per the ethical requirements (Appendix I).

5.2.5 Data analysis and limitations

Participation in gardening in this study was considered to be a continuous variable, ranging from those who do little or no gardening, to individuals who spend a lot of their time engaged in gardening activities. As a result, no control group was recruited for this study; the study population are instead compared to baseline studies that have investigated health and well-being, fruit and vegetable consumption and exercise in larger populations. These include national and regional statistics for the consumption of fruit and vegetables, engagement in physical activity, and normalised scores for the SF 36 (Ware and Kosinski 2001); all adjusted by age and gender. Inevitably, comparing data collected from our study with broader data sets has some limitations. We cannot be sure for example that the reason for the study population having a higher consumption of fruit and vegetables or doing more exercise than regional or national averages is because of their participation in gardening activities, as there are many covariates that we were not able to take into account. However, this study builds on other studies which have found similar findings to

give a broader picture of the influence of gardening on health and well-being.

Statistical analyses and graphs were prepared using Minitab v 17.2.1[®]. Analysis of co-variance (ANCOVA) was used to test the null hypothesis that gardening more regularly in spring/summer or autumn/winter (independent variable) is not correlated with an increase in fruit and vegetable consumption (number consumed yesterday), exercise (number of times in the last week) or the physical component or mental component sub-scales of the SF 36 v 2[®] (Section 5.2.2). ANCOVA was also used to test the null hypothesis that gardening more regularly (independent variable) is not correlated with a decrease in BMI. Age and SES scores were included as covariates in the analysis. Gender, alcohol consumption and current cigarette use were included as confounding factors. Data from the produce consumption table (section C) were exported into a pivot table created using Microsoft Excel 2010[®]. The annual consumption of produce (portions per year) was estimated using the number of portions consumed per week/month of each produce multiplied by the number of months per year this was grown (For example, for an individual who grew tomatoes for 5 months of the year and consumed 2 portions per week, the annual consumption would be $2 \times (52.3/12) \times 5 = 44$ portions per year). Means from the SF-36 v2 reported from the study split by age and gender, were compared with means reported by Ware and Kosinski (2001) for a large US cohort using a t-test (Whitely and Ball, 2002). Whilst Ware and Kosinski (2001) is based on

earlier version of the SF 36, the questions and scoring structure is the same with a slight change to the responses concerning role limitations (Bjorner *et al.* 2013). Comparing the SF 36 v2 data to data for an American cohort may also incur error although data for a UK cohort split according to age and gender was not available to this project.

5.3 RESULTS

5.3.1 Survey demographics

One-hundred-and-twenty participants filled in and returned completed questionnaires. The minimum age of the gardeners studied here was 30 years with a maximum of 85 years and a mean of 59 years (under 18s were not included in the study). The study included 58 males and 56 females (6 respondents did not indicate their gender). Half of the respondents (50 %) were currently employed with 38 % indicating that they had retired prior to receiving the survey. Reported household incomes of participants were similar to data for the East Midlands Region (Figure 5.1). The majority of respondents (66 %) stated that they held a qualification equivalent to a bachelor's degree or above.

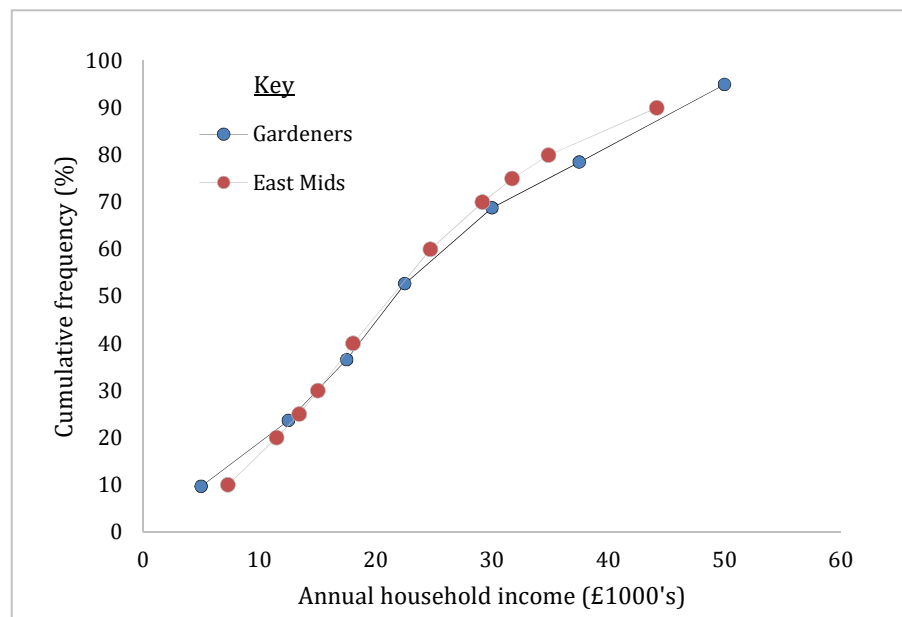


Figure 5.1: Comparison of annual household income (AHI) reported by participants in the gardening and health survey (gardeners), with reported AHI from the East Midlands (IFS, 2015-2016). Annual Household Income for gardeners was calculated using the mid-point of the categories over which AHI was assessed in question: D18 from the questionnaire. Data from above the 90th percentile is not included as it is strongly skewed by high earners in both surveys.

5.3.2 Participation in gardening

Out of 120 participants; 109 had access to a garden at home and 90 had access to an allotment. The participants varied in their engagement with gardening. Some participants reported gardening less than once a month whilst others reported gardening every day. One individual who had access to an allotment, reported not doing any gardening. Individuals that had access to an allotment generally reported gardening more regularly than individuals that did not (Figure 5.2).

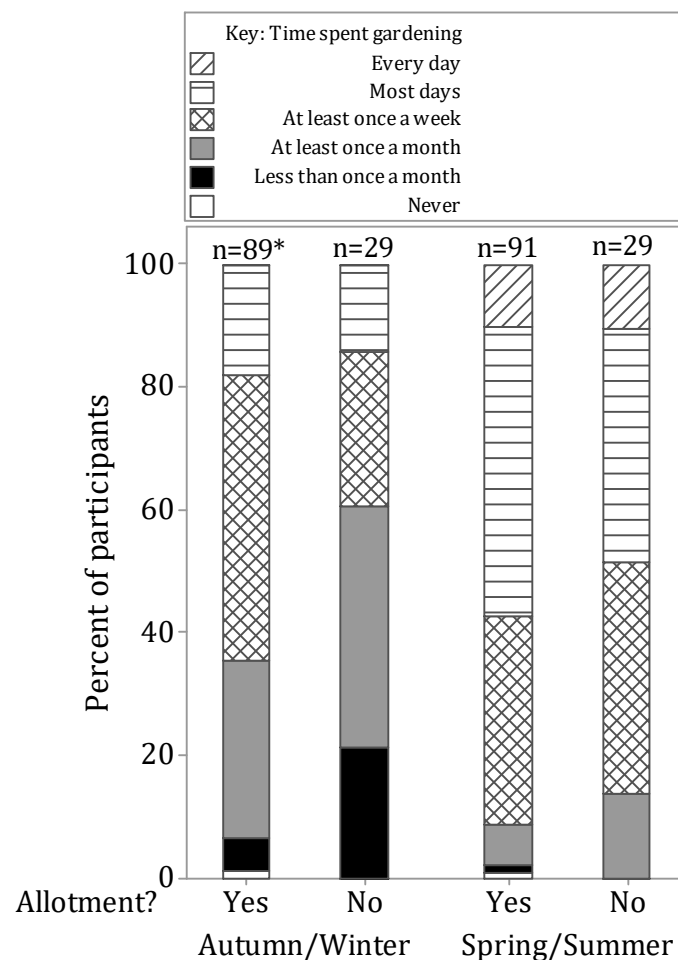


Figure 5.2: Responses to question A6: how often do you spend some time gardening at home or in other locations? *Two participants failed to answer the question and are not included in this graphic.

The regularity with which participants took part in gardening activities was strongly dependent on the season (Figure 5.2). Participants who had an allotment were more likely to spend several hours in their allotment in spring/summer than autumn/winter (Figure 5.3). Most participants that had access to an allotment reported spending at least one hour on their allotment per visit regardless of season (94 % of all respondents in spring/summer and 80 % in autumn/winter).

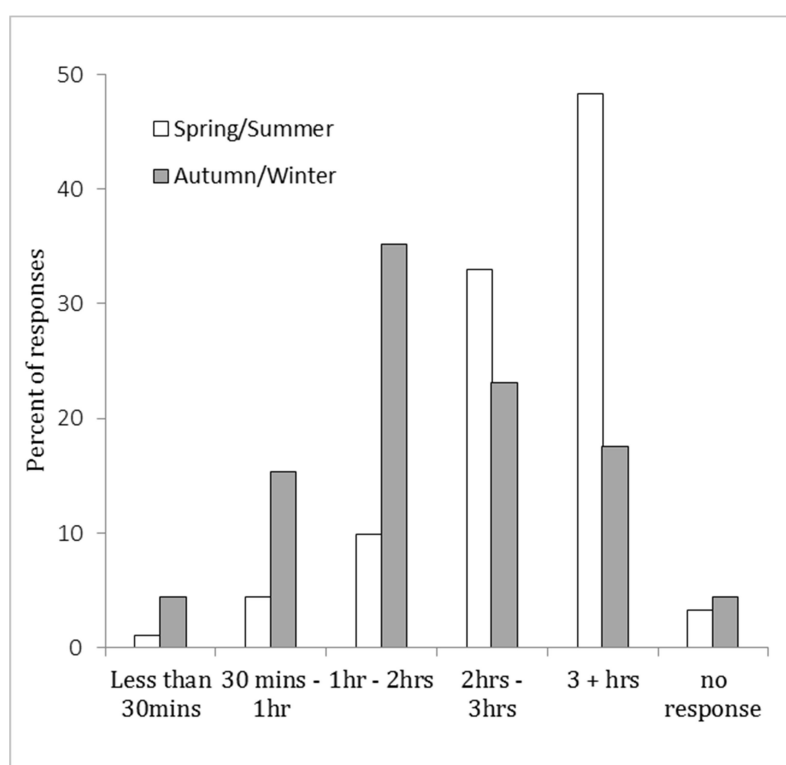


Figure 5.3: Responses to question A9: Length of time of visits to allotment plot (n = 91).

5.3.3 Gardening activities

Participants reported engaging with a wide range of gardening activities. As well as the pre-selected gardening activities in the questionnaire, participants also reported spending a lot of time maintaining hedgerows and outbuildings. Moderate intensity activities (Park *et al.* 2011); including digging, raking, weeding and tying plants to stakes), were performed to some extent by most of the respondents (Figure 5.4). More than two-thirds ($n = 80$) of the participants reporting either spending 'some' or 'a lot' of their time digging or weeding. Tying plants to stakes or raking the soil were less common activities, with the majority (more than 60) of respondents reporting spending 'some' or 'a little' of their time doing them. Low intensity activities, (Park *et al.* 2011; including watering with a watering can, harvesting produce and cutting stems/ flower heads), were also performed to some extent by most participants. Most participants (more than 60), reporting spending 'some' or 'a lot' of their time watering with a watering can and 'some' of their time harvesting produce. Fewer participants reported spending much of their time cutting stems and removing flower heads (Figure 5.4).

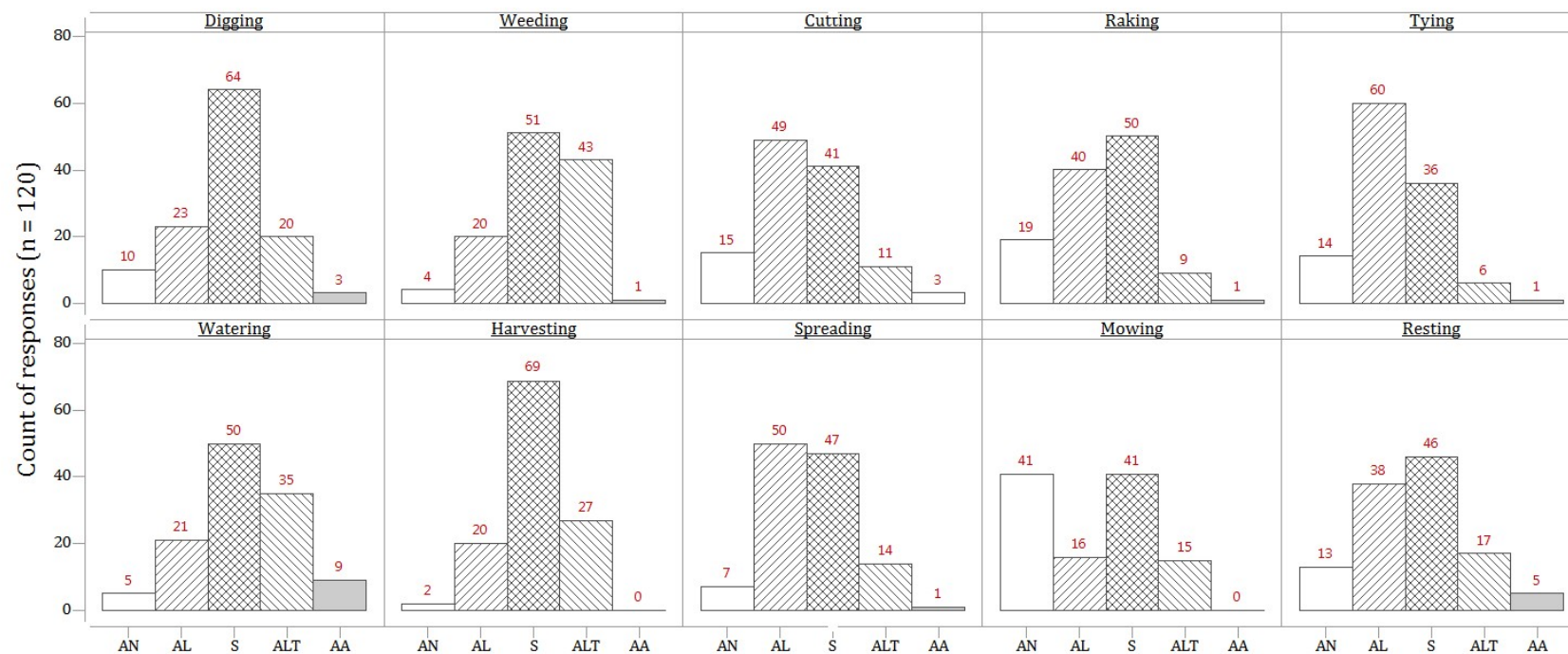


Figure 5.4: Histogram of responses (counts) describing the amount of time (qualitative) gardeners reported spending engaged with different activities. Key: AN = 'almost none', AL = 'a little', S = 'Some', ALT = 'a lot' and AA = 'almost all'. Bars show counts in each category out of 120. Non-responses are not recorded in the figure, but amounted to a maximum of 3 for any one category.

5.3.4 Trends in produce growth and consumption

The most commonly grown produce among the survey participants were potatoes and tomatoes, which were grown by 78 % of participants that grew some kind of produce in their home garden, allotment garden or elsewhere (GYO gardeners; n=118). Leafy produce, including lettuces and herbs, known to accumulate Cd (Baldantoni *et al.* 2016) were also grown by a large proportion of GYO gardeners (60 % and 64 % of gardeners grew lettuces and herbs respectively). Consumption of some produce, such as berries, only occurred for a few months of the year when the produce was in season (although some participants did report consuming homemade jams and preserves all year round). Others, such as herbs, were grown and consumed fresh all year. Fruits were found to be grown and consumed more often than leaves, roots or other edible tissues (Table 5.1). Consumption of GYO produce was found to contribute to a mean of 627 portions, per person, per year. Using a crude conversion of 1 portion of fruit and vegetables $\approx$ 80 g fw, this equates to a mean consumption of $\approx$ 960 g fw person⁻¹ wk⁻¹ of all home-grown produce to the diet.

Table 5.1: Produce consumption by produce group. Consumption of produce in g wk⁻¹ was calculated using a conversion factor of 1 portion $\approx$ 80 g fw (NHS, 2015). Values are averages (mean) $\pm$ standard deviation.

Produce Group	Consumption (mean portions person ⁻¹ yr ⁻¹)	Consumption (mean g fw person ⁻¹ wk ⁻¹)
Flower	16 $\pm$ 19	24 $\pm$ 29
Fruit	241 $\pm$ 107	369 $\pm$ 164
Leaf	131 $\pm$ 74	201 $\pm$ 114
Root	137 $\pm$ 71	209 $\pm$ 110
Tuber	66 $\pm$ 33	102 $\pm$ 51
Other	1 $\pm$ 7	2 $\pm$ 10
Totals	592 $\pm$ 230	909 $\pm$ 353

The amount of types of produce grown by participants in the study varied substantially between those who grew and consumed none, to those who grew and consumed 30+ different types of produce (Figure 5.5). Participants who had access to an allotment, generally grew more types of produce than participants that did not; but some home-gardeners still reported growing over 20 different types of produce in their gardens (Figure 5.5).

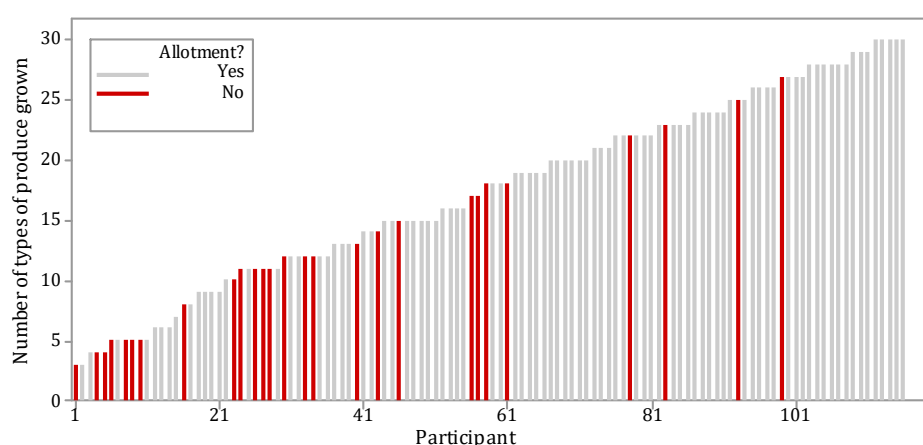


Figure 5.5: Number of types of produce grown per gardener surveyed. Two individuals did not grow produce whilst another two failed to respond to this Section of the survey.

Preparation methods were dependent on the type of produce. Berries such as blueberries were not commonly washed (washed by 33% of growers), whilst root/tuber vegetables such as potatoes and parsnips were more often washed than not (Table 5.2). Root produce were commonly peeled prior to consumption (Table 5.2). The contribution of GYO produce, compared to shop-bought produce, to the diet also depended on the type of produce. Commonly grown and consumed produce such as potatoes and tomatoes, were normally supplemented with shop-bought produce. Less commonly consumed produce or produce that is expensive or a rarity on supermarket shelves, such as

damsons and fennel, tended to be sourced completely from allotments and gardens (Table 5.2).

Table 5.2: Produce consumption patterns. The number of participants who grew some produce in their home garden, allotment or elsewhere was 118. Dashes in rows indicate occasions where data were insufficient or not applicable. The GYO (grow-your-own) contribution column is the average score gardeners reported for column h in the produce consumption table (Section C) (0 = none from GYO, 10 = all from GYO).

<u>Produce</u>	<u>Popularity</u> (count of persons)	<u>Peeled?</u> Yes - %	<u>Wash?</u> Yes - %	<u>Average Consumption</u> Portions person ⁻¹ yr ⁻¹	<u>GYO contribution</u> (0-10)
Potato	92	46	92	83	5
Tomato	92	8	63	58	5
Onion	77	87	52	76	5
Runner bean	77	25	77	34	8
Strawberry	77	-	70	27	6
Herbs	76	-	64	100	7
Raspberry	76	-	53	37	8
Rhubarb	75	36	80	18	8
Beetroot	73	77	89	27	6
Apple	71	37	59	57	6
Lettuce	71	-	85	66	6
Courgette	69	-	81	30	7
Blackberry	68	-	65	24	9
Leek	62	60	90	18	6
Carrot	61	64	87	29	4
Cabbage	60	-	90	30	6
Pea	58	43	48	19	6
Broad bean	53	64	60	23	8
Cucumber	51	-	65	32	5
Garlic	49	98	45	74	6
Parsnip	48	77	96	18	6
Blackcurrant	47	-	68	24	9
Plum	47	-	77	25	7
Broccoli	45	-	87	28	5
Spinach	41	-	93	29	7
Sweetcorn	40	55	65	12	6
Squash	39	72	64	19	8
Brussels sprouts	37	68	89	18	7
Cauliflower	26	-	81	15	6
Gooseberry	17	-	76	14	9
Pepper	14	-	86	26	5
Pear	12	17	75	29	4
Other beans	11	36	64	65	5

Table 5.2 continued					
Produce	N	Peel? (%)	Wash? (%)	Consumption	GYO?
Pumpkin	11	91	55	12	8
Chilli pepper	10	-	90	72	8
French bean	10	-	80	45	8
Blueberry	9	-	33	48	9
Radish	9	22	100	28	8
Artichoke	7	71	71	8	9
Kale	7	-	86	38	9
Redcurrant	7	-	43	12	10
Redcurrants	7	-	86	12	4
Damsons	6	-	67	15	10
Fennel	6	17	83	13	10
Chard	5	-	100	21	10
Other berries	5	-	40	43	10
Asparagus	4	-	75	41	9
Celeriac	4	100	100	17	10
Cherry	4	-	50	13	5
Figs	4	-	25	7	10
Loganberry	4	-	75	16	10
Swede	4	100	100	10	9
Celery	3	-	100	23	9
Chives	3	-	67	59	6
Grapes	3	-	67	2	4
Hazelnuts	3	67	0	22	7
Other currants	3	-	0	7	10
Parsley	3	-	67	59	10
Quince	3	67	67	1	10
Spring onion	3	-	33	33	2
Sweet potato	3	67	100	4	5
Chickpea	2	100	50	30	5
Greengages	2	-	100	3	8
Jostaberry	2	-	0	11	10
Kholrabi	2	100	50	18	10
Mange tout	2	-	100	29	9
Oca	2	-	100	5	10
Tayberry	2	-	0	24	10
Turnip	2	50	100	20	10
Yacon	2	100	100	8	10
Bay	1	-	0	-	-
Camomile	1	-	0	-	-
Cardoon	1	-	0	2	10
Cress	1	-	100	17	10
Cucamelon	1	-	100	87	10
Currants	1	-	100	313	10

Table 5.2 continued					
Produce	N	Peel? (%)	Wash? (%)	Consumption	GYO?
Edible flowers	1	-	0	52	10
Elder	1	-	0	-	-
Endive	1	-	0	-	10
Horseradish	1	100	100	12	3
Lemons	1	0	0	17	5
Marrow	1	0	0	3	10
Mint	1	-	0	-	-
Morjoram	1	-	0	-	-
Mouli	1	100	100	52	10
Mulberry	1	-	100	-	10
Nectarines	1	0	0	13	2
Okra	1	100	100	17	8
Other soft fruit	1	-	100	17	9
Rocket	1	-	0	-	-
Rosemary	1	-	0	-	-
Sage	1	-	0	-	-
Salad leaves	1	-	100	152	6
Sugar snap peas	1	-	0	17	3
Thyme	1	-	0	-	-
Walnuts	1	-	100	-	10

5.3.5 Lifestyle

Fruit and vegetable consumption

The contribution of GYO produce to the diet was hypothesised to correlate with increased fruit and vegetable consumption for participants who more regularly took part in gardening activities than those who did not. The results of a one-way ANOVA, suggested a significant ($p = 0.037$) relationship between regularity of gardening in autumn/winter and an increase in fruit and vegetable consumption. From Figure 5.6, it can be observed that taking part in gardening more regularly in autumn/winter appeared to be correlated with increased fruit and vegetable consumption. The exception to the trend, was for gardeners who took part in gardening activities most days ($n=10$) who appeared to consume fewer portions of fruit and vegetables than gardeners in the other categories.

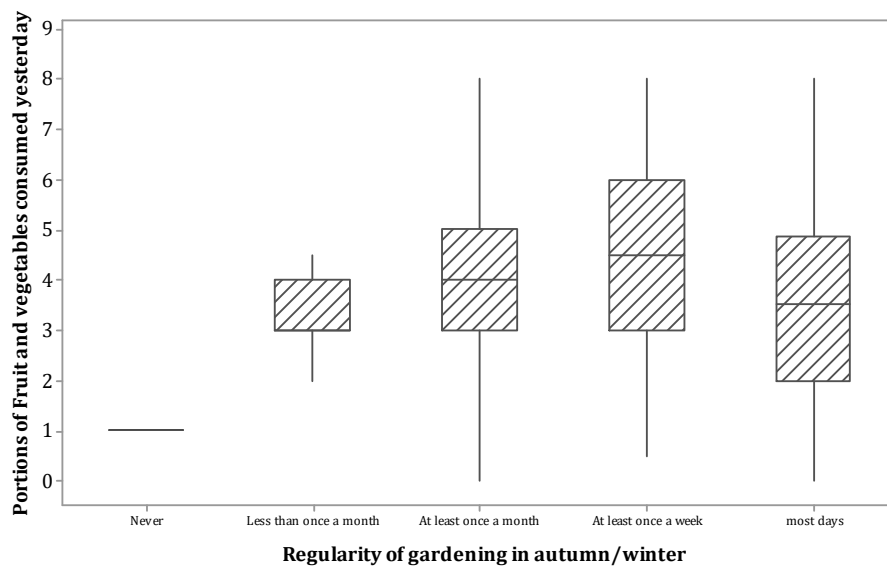


Figure 5.6: Boxplots showing the relationship between self-reported fruit and vegetable consumption (portions) over the previous 24hrs, and regularity of gardening in Autumn/Winter. Boxes show 25th, 50th and 75th percentiles; whisker show 1.5 X IQR.

The mean consumption of fruit and vegetables on the previous day for all participants (n = 119) was 4.1 portions. Men reported consuming 4 portions on average, whilst women reported consuming an average of 4.1 portions. These values are higher than averages for the East Midlands region of 3.4 and 3.6 portions for men and women respectively (HSCIC 2013a)

Physical activity

The UK government recommends that individuals should do some form of moderate (e.g. walking or gardening), or more intensive exercise of 30 minutes or more on five separate occasions per week (HSCIC 2012b). Most of the participants in the present study (71 % of men and 51 % of women) reported meeting the recommendations in the previous week. This compares to just 39 % of men and 29 % of women who met the same government recommendation in 2008 (HSCIC 2012b). Increased regularity of gardening activities in autumn/winter was found to be positively correlated with increased occasions of moderate, or more intensive exercise, by participants (one-way ANOVA: $p = 0.008$).

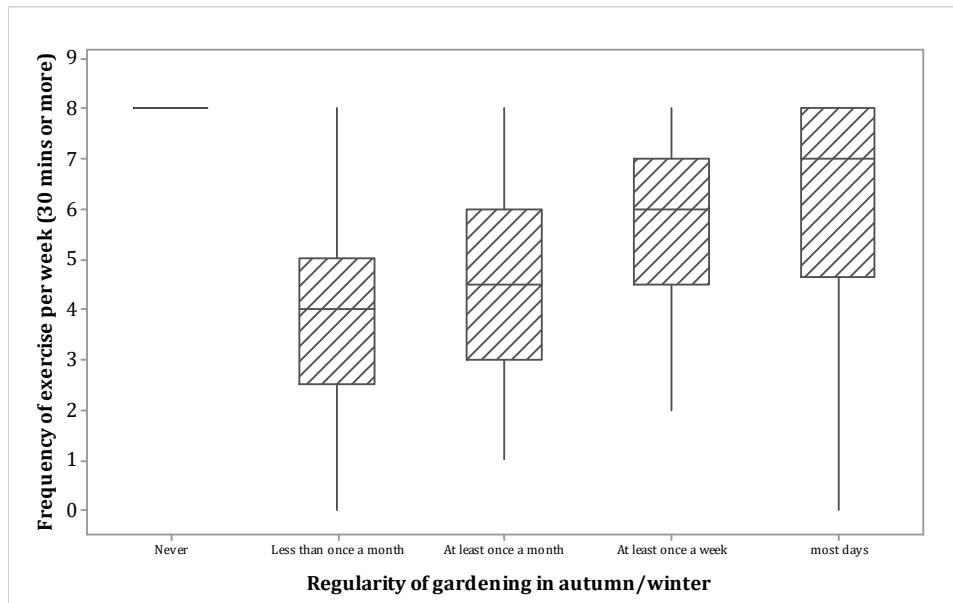


Figure 5.7: Boxplots showing the relationship between self-reported frequency of moderate exercise in the previous week and regularity of gardening in Autumn/Winter. Boxes show 25th, 50th and 75th percentiles; whisker show 1.5 X IQR.

Body-mass index (BMI)

Body-mass index was calculated from self-reported height (m) and weight (kg). The average (mean) BMI was 25.3 kg m⁻² for male participants (n = 56), and 25.5 kg m⁻² for female participants (n = 50) (Six participants did not provide their gender and/or height and weight which were used to calculate BMI). This compares to an average (mean) BMI for the general population (UK) of 27.4 kg m⁻² for men and 27.1 kg m⁻² for women (HSCIC 2012b). There was no significant correlation between reduced BMI and regularity of gardening in spring/summer or autumn/winter (one-way ANOVA: p = 0.675 and 0.341 respectively) (Figure 5.8).

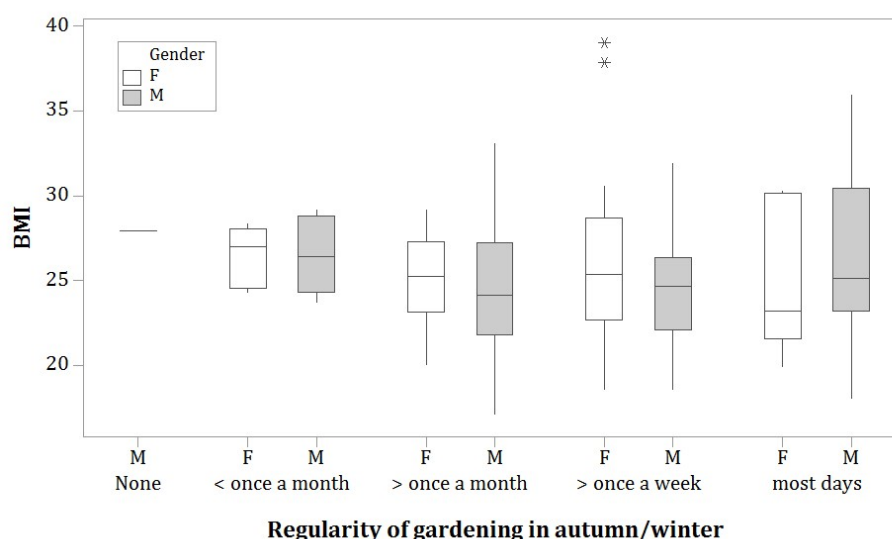


Figure 5.8: Boxplots showing the relationship between the regularity with which participants took part in gardening activities in autumn/winter and BMI (kg / m²). Boxes show 25th, 50th and 75th percentiles; whisker show 1.5 X IQR.

Alcohol consumption and cigarette smoking

Alcohol consumption and cigarette smoking were recorded as confounding factors that could influence health outcomes from gardening lifestyles. In chapter 6, these data are used in the context of a health benefits vs risk model to estimate the net influence on health for the study population. A large majority, 88% of the study population, reported consuming alcohol in the last week to some extent. Over half (53%) of the study population consumed >10 units in the previous 7 days; 5% of the study population consumed more than 30 units in the same time period. The number of participants that reported drinking in the last week was higher than data from a national survey (67% of men and 53% of women respectively: HSCIC 2012a). However, the number of 'binge' drinkers

(those drinking more than double recommended amounts: 14 units for women and 21 units for men respectively) in the national survey was much higher than the data for the current survey; consisting of 31% of men and 24 % of women (HSCIC 2012a).

Nearly half (45 %) of the study participants reported that they had smoked regularly at some point in their lives. Of those who smoked cigarettes, only 22 % reported smoking the previous day. The median number of cigarettes smoked was between 6 and 20; consistent with the national average of 12 cigarettes per day (HSCIC 2013b). No individuals reporting smoking more than 30 cigarettes on the previous day.

5.3.6 Short-form 36 v 2 responses

Physical component scores

Scores from the physical component summary (PCS), suggested that gardeners in the study population had better physical health than the US population from which the survey was constructed. The mean PCS score was 51.8; 51.0 for males and 52.7 for females respectively. The scores were negatively skewed by some individuals in the study population scoring much lower than the 50 norm. When scores were adjusted for age and gender, older participants (age 45+) in the present study, scored significantly higher than normalised scores from the US population (see Figure 5.10 and Table 5.3). Scores from the different sub-components forming the PCS, were fairly similar across age groups and genders.

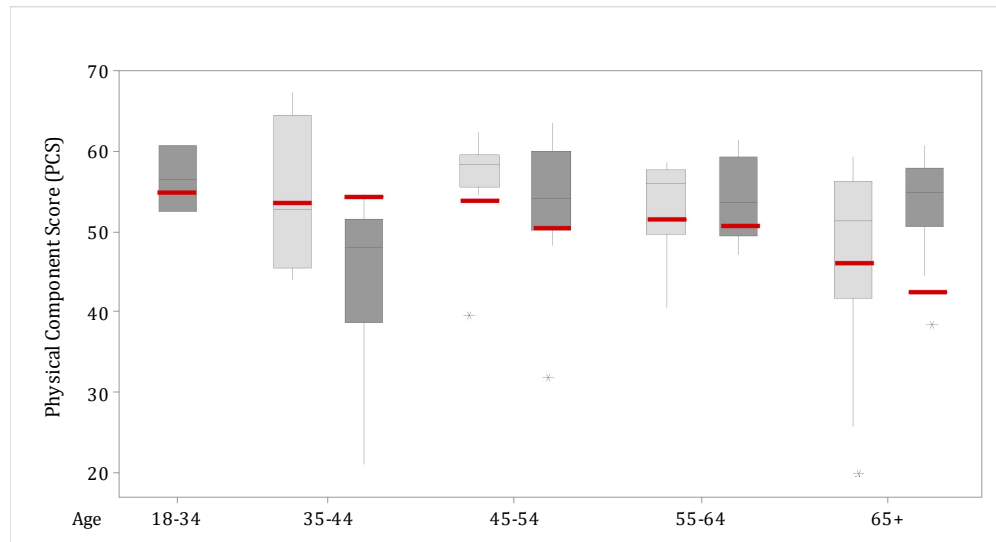


Figure 5.10: Box plots showing physical component scores (PCS) grouped by age category and gender. Boxes show 25th, 50th and 75th percentiles; outliers are outside median $\pm$ 1.5 X IQR. Darker gray boxes = females, and lighter grey = males. Red lines indicate median scores from a US population, according to age and gender (Ware and Kosinski, 2001). There were no males in the 18-34 age group.

There was a significant increase in the PCS scores of gardeners who were gardening more regularly in autumn/winter ($p = 0.011$). PCS scores appeared to increase with gardening regularity, but the increase appeared to reach a threshold for participants gardening once a week or more (Figure 5.9).

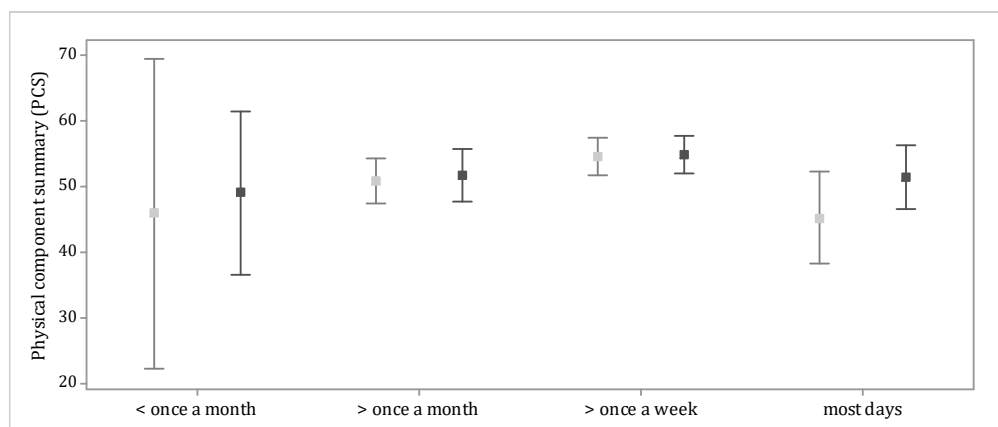


Figure 5.9: Interval plot showing the relationship between regularity of gardening in autumn/winter and PCS scores. Darker grey = females, lighter grey = males. Intervals show means and 95th percent confidence intervals. There were no males in the 18-34 age group.

Mental component scores

Scores from the mental component summary (MCS) were also higher than norms from the US population from which the SF 36 v 2 was constructed. The mean MCS score from across the study population was 52.8; 53.0 for males and 52.5 for females respectively. The scores were negatively skewed by some individuals in the study population scoring much lower than the 50 norm. The scores from the study population, were not significantly different from the mean scores in the US population, when adjusted for age and gender (see Figure 5.11 and Table 5.3). Scores from the different sub-groups forming the MCS, were fairly similar between age and gender categories. There was no increase in MCS scores for participants who took part in gardening activities more regularly in autumn/winter or spring/summer ($p = 0.649$ and 0.165 respectively).

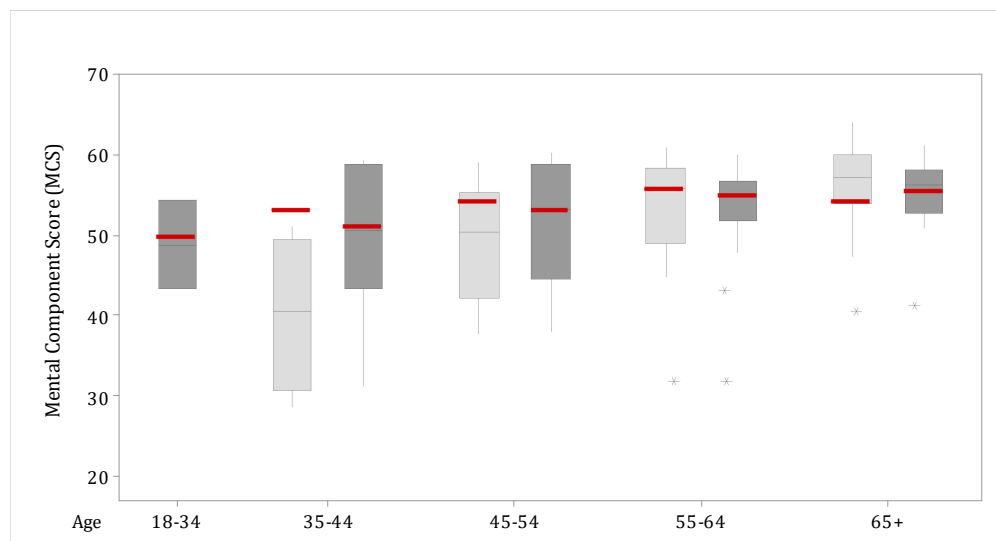


Figure 5.11: Box plots showing mental component scores (MCS) grouped by age category and gender. Boxes show 25th, 50th and 75th percentiles; whiskers show median $\pm$ 1.5 X IQR. Darker gray boxes = females, and lighter grey = males. Red lines indicate median scores from a US population, according to age and gender (Ware and Kosinski, 2001).

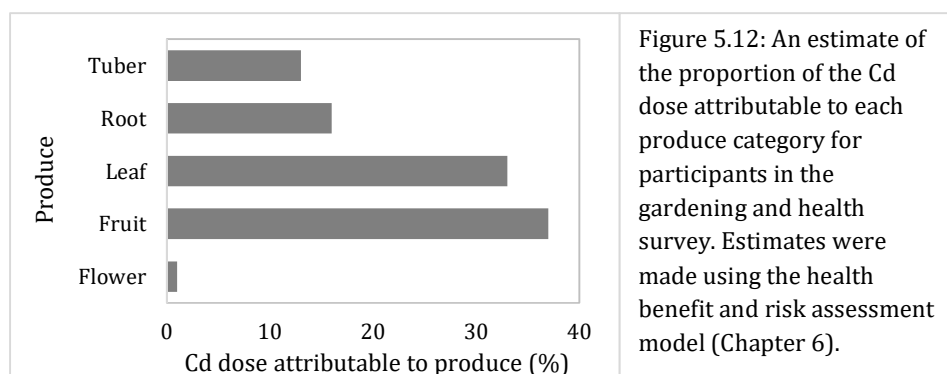
Table 5.3: Physical component summary (PCS) and mental component summary scores from the SF 36 v 2. Means for individual age and gender categories were compared with Ware and Kosinski (2001) by using a t-test to compare means (Whitley and Ball, 2002).

<u>Gender</u>	<u>Age Category</u>	<u>n</u>	PCS				MCS			
			<u>Gardening survey</u>		<u>Ware and Kosinski (2001)</u>	<u>Significance of difference</u>	<u>Gardening survey</u>		<u>Ware and Kosinski (2001)</u>	<u>Significance of difference</u>
			mean	SD	mean	P value	mean	SD	mean	P value
Male	35-44	3	49.86	4.81	51.97	0.527	44.11	5.79	50.80	0.183
	45-54	10	56.32	6.04	50.90	0.019	49.24	7.05	49.93	0.764
	55-64	11	53.23	5.90	48.05	0.015	52.58	8.14	52.48	0.967
	65+	30	47.85	9.84	43.51	0.022	56.18	4.70	51.48	<0.001
Female	18-34	2	56.46	4.07	53.10	0.451	48.76	5.46	46.66	0.683
	35-44	6	44.46	10.82	51.65	0.164	49.64	9.25	48.44	0.764
	45-54	12	53.49	7.99	47.90	0.034	51.59	7.33	50.69	0.679
	55-64	19	54.37	4.76	47.51	<0.001	53.11	6.29	51.79	0.372
	65+	15	53.24	6.05	42.17	<0.001	55.10	4.85	53.02	0.119
	All		52.0	8.20	(50)	-	52.3	7.8	(50)	-

5.4 DISCUSSION

5.4.1 Health risks

Exposure to heavy metals from gardening on urban and allotment soils is increased through increased consumption of GYO produce and increased incidence, and duration, of garden visits (EA 2009d). Plant uptake of metals varies according to the metal and edible plant tissue consumed (See chapter 3 ; Clemens 2006; McLaughlin *et al.* 2011). For exposures to Cd, the main route for gardeners is expected to be through the consumption of produce, as Cd is very bioavailable in soils (Yang *et al.* 2010). Potatoes and tomatoes, the most commonly grown produce by gardeners in this study, do not readily take up Cd (Clemens 2006) whereas leafy vegetables such as lettuces tend to accumulate Cd more readily (Alexander *et al.* 2006). The main contribution of Cd from produce grown on gardens in the study, is estimated to be from fruits and leaf vegetables (Figure 5.12). Fruits were consumed more than the other produce types, and Cd accumulates in leafy vegetables. This suggests that both reducing the amount of produce and reducing the amount of leafy produce consumed from urban gardens and allotments, could be viable methods to reduce exposures to Cd.



The main exposure route to Pb for urban gardeners is likely to be through soil ingestion, as Pb is not readily taken up into produce (e.g. Intawongse and Dean 2006; Clark *et al.* 2008). Inadvertent soil ingestion is linked to the number of hours gardeners spend on their gardens or allotment plots (EA 2009d). The regularity and duration of visits to gardens was found to be strongly dependent on season (Figure 5.2 and Figure 5.3). Lead exposures are particularly pertinent for children who may suffer from developmental toxicity as a result (Canfield *et al.* 2003; Zahran *et al.* 2013). Reducing the amount of time children spend playing in urban soils, may help to reduce their exposure. Furthermore, practices including wearing gloves and removing dust ridden clothing prior to entering the house, could further reduce exposures from soil Pb (Brown *et al.* 2016). At least two-thirds of participants in this study, reported peeling root vegetables prior to consumption. Most participants also washed produce prior to consumption, which also helps to reduce Pb exposure from soil particles adhered to produce (Arif *et al.* 2011). The contribution of produce such as berries, which are commonly consumed without washing, to Pb exposures should be considered.

There was a large amount of variation in gardening practices and routines among participants in the survey. This could result in considerable variability in risk assessment models if it is not taken into account. In a review of risk assessment models, Swartjes (2007) found variations in model outputs of adult exposure to be between one and two orders of magnitude. Swartjes (2007) postulated that variation in model outputs

could arise as a result of differences in model inputs and estimates of exposure. In chapter 3 of this thesis it was shown that risk assessment models CLEA, RBCA and RIVM may all over under-, or over-estimate plant uptake of metals by up to 2 orders of magnitude. The variation in both Cd and Pb exposures between gardeners in this study is likely to vary considerably between gardeners, due to the differences in produce consumption, number, and duration of garden visits, adding further uncertainty to the prediction of health risks. The inclusion of self-reported data on exposure, such as number and duration of visits to the garden, may help improve the accuracy of site-specific assessments. Assessing fruit and vegetable consumption, in Section C of the questionnaire was difficult and a number of participants in a pilot study reported found this section difficult to answer. Pre-selection of produce may have contributed a source of bias but reduced the amount of time participants spent trying to answer this section. A better approach could be to use food diaries to record produce consumption or ask participants to duplicate meals. However, these approaches are also subject to sources of bias and uncertainty, and may not necessarily offer much of an improvement on 24-hr recall designs (Bingham *et al.* 1994).

5.4.2 Health benefits

Similarities between the results of this study and previous studies help to support the argument that gardening could help to promote healthier lifestyles and tackle national sloth and obesity (as suggested by Leake *et al.* 2009). Firstly, fruit and vegetable consumption by gardeners in this study, was found to be correlated with the amount of gardening individuals did in autumn/winter, and was greater than fruit and vegetable consumption by the UK general population. Two other studies have observed an increase in fruit and vegetable consumption among gardeners compared to non-gardeners (Alaimo *et al.* 2008; Carney *et al.* 2011). Levels of physical activity were also found to be higher in our study than regional averages: This is supported by a study which found that physical activity levels in summer, were significantly higher for gardeners compared to non-gardeners (Van den Berg *et al.* 2010). Furthermore, the mean BMI of participants in this study ($25.4 \pm 4.0 \text{ kg / m}^2$), was very similar to the mean BMI of participants in two other UK based studies of allotment gardeners ($25.5 \pm 3.3 \text{ kg / m}^2$ - Hawkins *et al.* 2011; and $25.5 \pm 3.5 \text{ kg / m}^2$ - Wood *et al.* 2015), and lower than national averages.

This is the first study (to the author's knowledge), to find a relationship between gardening more regularly (in autumn/winter), and the physical component of the SF 36 v 2. The median PCS score (53.8) observed in this study, was similar to the median PCS score of 53.5 observed by Hawkins *et al.* (2011) for a group of allotment gardeners. Another study, Park *et al.*

(2009), found a significant improvement in the physical component scores of the SF 36 v2 for gardeners, compared to a non-gardening control group. Park *et al.* (2009) and Hawkins *et al.* (2011), did not observe a difference between gardeners and non-gardeners in the mental component score from the SF 36 v2. The physical component scores from this study, were also significantly higher for older participants, compared to means from a western population (Ware and Kosinski 2001). This supports studies which have suggested that gardening may be more beneficial for a more elderly generation (e.g. Wang and MacMillan 2013).

There was no difference between the MCS scores in the present study and the US population studied by Ware and Kosinski (2001). Van den Berg *et al.* (2010) observed that gardeners had higher life satisfaction compared to their non-gardening counterparts; Wood *et al.* (2015), that gardening improves self-esteem, while Hawkins *et al.* (2011) and Van den Berg and Custers (2011), found a correlation between gardening activities and reduced stress (measured by reduced cortisol production). It is possible that the SF 36 v 2 may not be a sensitive enough tool, due to the number of confounding factors that play a part in psychological health and well-being, to register a potentially small effect of gardening on mental health. The relatively small sample size of 126 participants in this study is also likely to be a limiting factor in identifying correlations between mental health and gardening exposures.

A surprising result of this study was the correlation between increased participation in gardening in autumn/winter, but not spring and summer, with PCS scores, increased fruit and vegetable consumption and increased physical activity. The ANOVA used to determine differences in lifestyle and health as a result of participation in gardening, was unbalanced as the number of individuals in each category (“< once a month”, “> once a month”) etc. was different, so the results should be treated with caution. However, participants who did more gardening in spring/summer may not have been as active in autumn/winter (Figure 5.13), reducing the impact gardening may have had on their overall lifestyle, health and well-being.

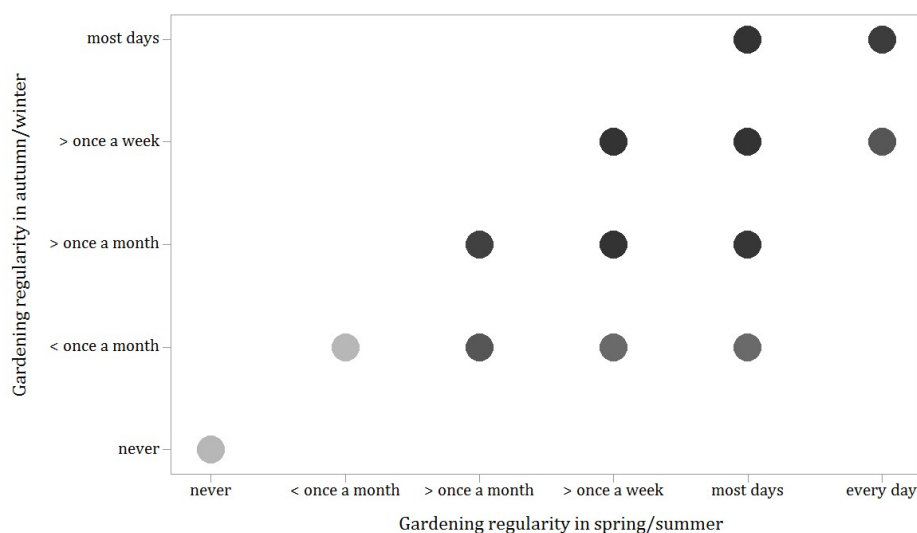


Figure 5.13: Scatter plot showing the relationship between gardening regularity in spring/summer and autumn/winter. Darker points indicate a higher frequency of responses.

5.5 CONCLUSIONS

This study is the first to consider, and estimate, the influence of gardening routines on exposures to both health benefits and health risks. This holistic approach helps to contrast the healthy lifestyle of gardening with health risks from exposures to Cd and Pb in urban environments. The data from this survey have been used to underpin a novel risk assessment model, which balances the trade-off between health benefits and health risks for urban gardeners.

Practical measures may help to reduce environmental Cd and Pb exposure for gardeners, whilst still allowing gardeners to enjoy and maintain a healthier lifestyle. Reduced exposures to Cd could be achieved by reducing the consumption of home-grown produce, especially leafy produce grown on urban gardening spaces, and through soil amendments to reduce the bioavailability of Cd (Brown *et al.* 2016). Reduced exposures to Pb could be achieved through wearing gloves, washing and peeling produce and reducing the amount of time children spend playing in urban garden soils. Participation in gardening, was found to be beneficial for physical health and well-being among older gardeners, and for maintaining a healthy lifestyle through increased fruit and vegetable consumption, physical activity and a reduction in BMI. Participants in this study were generally healthier than national and regional averages. The benefit of being able to maintain a healthy lifestyle is likely to outweigh the health risks of gardening on soils mildly contaminated with Cd and Pb

(Leake *et al.* 2009; Brown *et al.* 2016); a hypothesis which is further considered in the benefits vs risks model in Chapter 6.

6 A MODEL OF THE HEALTH BENEFITS AND RISKS FROM URBAN GARDENING

6.1 INTRODUCTION

Exposure assessment models, such as the CLEA (Contaminated Land Exposure Assessment) and RBCA (Risk-Based Corrective Action) models required in the regulation of contaminated land, do not currently consider the possible health benefits of gardening (Leake *et al.* 2009). This may lead to an incorrect assessment of the impact of urban gardening on health, resulting in risk management decisions which reduce opportunities for urban gardeners through the full or partial closure of existing sites (Leake *et al.* 2009). Subsequent remediation of urban gardens which exceed current guidance values for contaminants in soils, may also be expensive, and possibly ineffective, if surrounding areas are not treated simultaneously (Brown *et al.* 2016). Furthermore, many urban gardens in the UK may contain higher concentrations of Pb compared to current Category 4 screening levels (C4SLs) (Chapter 2), as normal background concentrations of Pb in urban soils exceed C4SLs by a factor of 10 (Chapter 2; Ander *et al.* 2013). There is therefore a requirement for the development of a risk assessment model that considers both the benefits and risks of urban gardening (Leake *et al.* 2009).

6.1.1 Does urban gardening lead to appreciable health risks?

There is no doubt that chronic exposure to high concentrations of HMs such as Cd and Pb, and other contaminants that may be present in urban soils, can lead to adverse health (see Section 1.3.3). However, there is less evidence that exposure to concentrations of HMs at, or slightly above, current standards for allotment soils (Table 1.3), will lead to adverse health effects. Leake *et al.* (2009) reviewed two studies of urban allotments containing concentrations of Pb in excess of 1,000 mg Pb kg⁻¹ dw soil (Prasad and Nazareth 2000; Mulloli *et al.* 2004) and concluded that in neither case was there evidence of adverse health from additional exposure to Pb. A similar study also found that there was no evidence of an increase in urinary Cd for individuals who grew and ingested home-grown vegetables in soils containing > 3 mg Cd kg⁻¹ dw soil (Wood 1996). Whilst other studies have found an association between soil concentrations of Pb and Cd in residential locations and adverse health effects (Nawrot *et al.* 2006; Zahran *et al.* 2013), it is possible that much of the exposure could be attributable to Cd and Pb in household dust (Hogervorst *et al.* 2007) which is not considered to be a relevant pathway for allotment gardeners (EA 2009d).

Further consideration should also be given to the deliberately conservative approach used to develop existing exposure models. Whilst the use of conservative measures, such as using a 'realistic-worst-case-scenario' in exposure assessment and uncertainty factors in toxicity assessment (Chapter 1), may be valid to minimise health risks, the

combination of conservative measures used by exposure assessment models is likely to result in an over-estimation of the health risks from exposure to contaminants in soils (see: Prasad and Nazareth 2000).

6.1.2 Does urban gardening promote health?

There is plenty of evidence to support the role of gardening in promoting healthy lifestyles (Chapter 5) but direct epidemiological evidence linking gardening and GYO to a reduction in disease is sparse (Buck 2016). However, there is a plethora of evidence linking the benefits of gardening to lifestyle, including an increase in fruit and vegetable consumption and exercise, with the reduction of disease.

Regular consumption of fruit and vegetables has a role in protecting against chronic heart disease (CHD), strokes and possibly cataracts and hypertension (Van Duyn and Pivonka 2000). Fruit and vegetable consumption may also have a role in reducing the risk of cancer, as fruit and vegetables contain a number of anti-carcinogens including vitamins C and E (Steinmetz and Potter 1996). The protective effect of fruit and vegetables may only be weakly associated with reduced risk of cancer, compared to a stronger protective effect against cardiovascular diseases (CVDs) (Takachi *et al.* 2007; Willett 2010). However, increased fruit and vegetable consumption may be used as part of a diet to facilitate weight loss (see: Tohill *et al.* 2004) which is an important risk factor for cancers and CVD in the developed world (Danaei *et al.* 2005).

Regular physical activity also reduces the risk of cancer, cardiovascular diseases (CVD) and diabetes mellitus (Warburton 2006). The association of physical activity with a reduction in CVD may be partly explained by the effect of physical activity on other risk factors including blood pressure and homeostasis (Mora *et al.* 2007). Like fruit and vegetable consumption, increased physical activity can also help people who are overweight to lose weight and stay healthy (Fox and Hillsdon 2007). Evidence relating physical activity and cancers is limited to specific cancers including breast cancer and colon cancer (Warburton 2006), with leisure-time activity important in reducing the risk of colon cancer for both males and females (Samad *et al.* 2005). Physical activity also increases bone mineral density, reducing the risk of fractures and osteoporosis (Coupland *et al.* 1999).

6.1.3 Combining health benefits and risks of urban gardening

To the best of the author's knowledge, the evidence for health benefits and risks from urban gardening have not previously been quantified together. A similar study did attempt to quantify the benefits of consumption of fish with the health risks of ingestion of methyl mercury (see section 1.3.3), concluding that the health benefits of fish consumption vastly outweigh the risks for most individuals consuming shop-bought fish (Mozaffarian and Rimm 2006). Other studies have concluded, qualitatively, that the health benefits of gardening are similarly likely to outweigh the health risks from exposure to soil contamination (Leake *et al.* 2009; Brown *et al.* 2016).

In this chapter, the health risks and benefits of gardening are compared in a Bayesian Network (BN) (Section 6.2). Bayesian Networks have been described as “graphical probability calculators”, and help to describe the propagation of uncertainty throughout the model (Tighe *et al.* 2013) which can be useful to stakeholders trying to make meaningful decisions with complex data (Chen and Pollino 2012; Landuyt *et al.* 2013).

6.2 BAYESIAN NETWORKS (BNs)

6.2.1 What are BNs?

Bayesian networks are probabilistic, graphical models of a real-life system represented by boxes (nodes) denoting model variables; and arrows between the boxes (directed edges), symbolising the flow of information between them. This visual network of nodes and directed edges is referred to as a Directed Acyclic Graph (DAG) (Figure 6.1). BNs can be created using existing software or developed independently (Nyberg *et al.* 2006). The software used to create the BNs presented here was NETICA® v 5.18 (Norsys 2014) so some of the applications and methods described here are directly applicable to this software alone.

BNs work on the basis of (conditional) probability - defined as the “extent to which an event is likely to occur” given knowledge of the preceding conditions (OED 2017b). The ‘events’ in the BN are denoted by states within each node (see Figure 6.1 for an example). These can be either

qualitative, quantitative or both (Chen and Pollino 2012). The likelihood of a state occurring in any node depends on the set of built-in probabilities that govern the node, and the influence of any nodes with which it is connected. In BNs this is often represented by probability tables attached to each node. A parent node (input) which is not dependent on the results of other nodes is informed by its own probability table (PT). Child nodes which depend on the conditions of other nodes are informed by conditional probability tables (CPTs). Input nodes which have no parents can be referred to as 'root' nodes, whilst output nodes which have no children may be referred to as 'leaf' nodes (Frank 2015).

6.2.2 Strengths and limitations

Bayesian networks have a number of strengths that are suited to exposure and risk assessment models (Pollino *et al.* 2007). These include the ability to simultaneously model causes and effects (Aguilera *et al.* 2011), to model different types of data including 'expert knowledge' when relationships between variables are uncertain/unknown (Chen and Pollino 2012; Troldborg *et al.* 2013), and the ability to update the model with new data when this becomes available (Pollino *et al.* 2007; Tighe *et al.* 2013).

There are some limitations to using existing BN software such as NETICA, including the requirement for discretisation of continuous data, which can reduce model precision (Aguilera *et al.* 2011; Chen and Pollino 2012;

Landuyt *et al.* 2013). The use of a large number of states to describe continuous variables increases the size of CPTs and results in a loss of performance (Landuyt *et al.* 2013). BN software is also unsuited to modelling systems that require feedback loops as the use of the DAG does not allow this (Nyberg *et al.* 2006; Troldborg *et al.* 2013). Whilst probabilistic modelling is suitable for representing uncertainty, model outcomes must still be interpreted by stakeholders which can be complex if presented as probabilities (Landuyt *et al.* 2013).

6.2.3 Example BN: Smoking and lung cancer

Consider the following example: the UK government wants to investigate a policy to reduce lung cancer incidence by controlling smoking. The problem is presented using a BN with a parent node, 'smoking' linked to a child node, 'lung cancer'. Both nodes are assigned two states; 'yes' and 'no' (Figure 6.1).

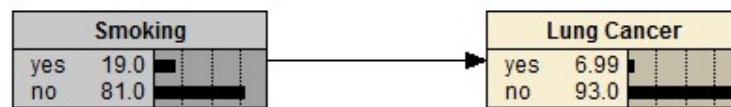


Figure 6.1: The direct acyclic graph (DAG) for the relationship between 'smoking' and 'lung cancer' (NB: the probabilities for the lung cancer states do not total 100% due to rounding errors).

To create the conditional probability table (CPT) for the lung cancer node, we need to know the probability of contracting lung cancer given that someone has smoked previously, $P(LC=y|S=y)$, and the probability of contracting lung cancer for someone who has never smoked $P(LC=y|S=n)$.

It is known that the likelihood of an adult having previously smoked, $P(S=y)$, in the UK is 19% (0.19), and the likelihood of someone contracting lung cancer in their lifetime, $P(LC=y)$, is ~7% (0.07) (CRUK 2015). It is also known that 86% of those diagnosed with lung cancer during their lifetime have smoked: $P(S=y|LC=y) = 0.86$ (CRUK 2015) (the probability of someone being diagnosed with cancer who has not smoked during their lifetime is: $P(S=n|LC=y)$). To calculate the likelihood of lung cancer given smoking, Bayes Theorem can be used (Equation 6.1):

$$P(A|B) = \frac{P(B|A) * P(A)}{P(B)} \quad 6.1$$

Where $P(A)$ and $P(B)$ refer to the probabilities of event A and B occurring and $P(A|B)$ is the probability of event A occurring given that event B has already occurred. The probability of someone contracting lung cancer over their lifetime given that they have smoked is 31.7% (Equation 6.2), and the probability of someone contracting lung cancer in their lifetime if they have never smoked is 1.2% (Equation 6.3).

$$P(LC=y|S=y) = \frac{P(S=y|LC=y)*P(LC=y)}{P(S=y)} = \frac{0.86 * 0.07}{0.19} = 0.317 \quad 6.2$$

$$P(LC=y|S=n) = \frac{P(S=n|LC=y)*P(LC=y)}{P(S=n)} = \frac{0.14 * 0.07}{0.81} = 0.012 \quad 6.3$$

This information can be used to create the CPT for lung cancer (Figure 6.2)

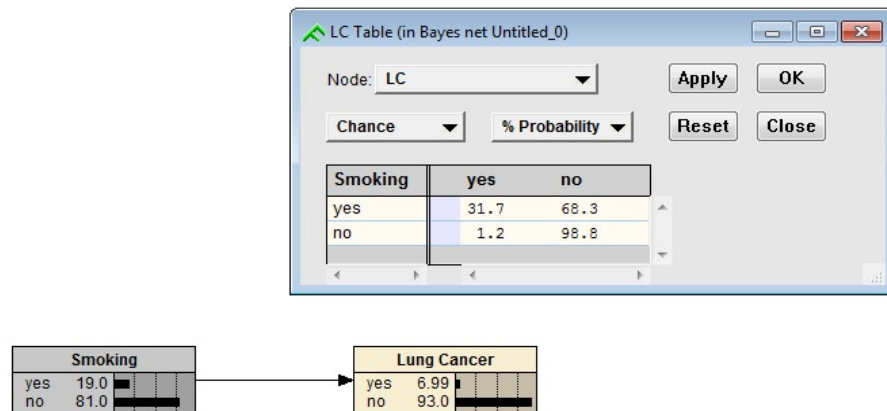


Figure 6.2: The DAG for the smoking and lung cancer example with the CPT displayed for the Lung Cancer (LC) node. Given that the probabilities for LC=y are known (left column), the probabilities for LC=n (right column) can be determined through the rule of subtraction ($A' = 100\% - A$).

Top-down modelling

Imagine that the government sets out a policy of reducing the number of smokers by 20 %. This is equivalent to a reduction in the proportion of smokers from 19 % to 15.2 %. By changing the prior information in the smoking node, the BN uses the law of total probability to instantly calculate the new likelihood of lung cancer for the population (Equation 6.4) (Frank 2015).

$$P(x) = \sum_n P(x|U_1 \dots U_n) * P(U_1) * P(U_2) \dots P(U_n) \quad 6.4$$

Where $P(x)$ is the probability of a state in a node occurring and U represents the probabilities of 'n' number of other variables on which x is conditionally dependent (adapted from Frank, 2015).

This gives the following:

$$P(LC=y) = P(LC=y|s=y)*P(S=y) + P(LC=y|S=n)*P(S=n)$$

$$P(LC=y) = 0.317*0.152 + 0.012*0.848 = 0.058 \text{ or } 5.8 \%$$
6.5

Therefore, a reduction in the number of smokers by 20 % would reduce the risk of lung cancer in the UK population from 7 % to 5.8 % (Figure 6.3).



Figure 6.3: The DAG for smoking and cancer with the finding $P(S=y) = 0.152$.

Bottom-up modelling

Bayesian networks can also operate in reverse. If it was assumed that the number of individuals contracting lung cancer remained the same despite the reduction in smoking described earlier, the BN can be used to determine the new likelihood someone was a smoker given that they contracted lung cancer within their lifetime. Bottom-up modelling is derived by the BN using Bayes Theorem (Troldborg *et al.* 2013; Frank 2015) (Equation 6.1) in the same way as was used to derive the CPT (Equation 6.2 and 6.3) (Figure 6.4):

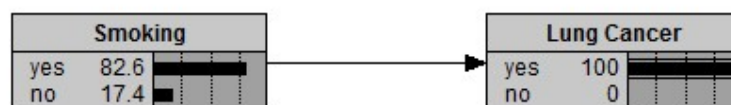


Figure 6.4: The updated likelihood of someone being a smoker given that they have contracted lung cancer in their lifetime.

In this example, the likelihood of someone having smoked given that they have contracted lung cancer in their lifetime, reduced from 84 % to 82.6 %.

6.2.4 Types of nodes and networks

BNs have been developed to be used for a number of purposes so different types can be useful in different circumstances. BNs are normally comprised of nature nodes (see Figure 6.1), and constant nodes - represented by an oval in NETICA which can only take on one value. Constant nodes do not need to be connected to other nodes but can be used in equations.

Other types of network include:

- Bayesian Decision networks (BDNs), which have been developed for the purpose of e.g. cost-benefit and decision analysis (Frank 2015). These consist of 'decision' and 'utility' nodes in addition to the usual nature and constant nodes. Decision nodes represent choices or management options and utility nodes represent decision outcomes.
- Object orientated BNs (OoBNs) consist of several BNs joined together, which can be useful for repetitive or hierarchical models (Chen and Pollino 2012).
- Dynamic BNs (DBNs) consist of several iterations of the same model for different points in time (Frank 2015) a solution to the limitations of modelling dynamic systems with BNs.

6.2.5 Deriving probability tables

Probability tables and CPTs do not have to be populated manually as in Figure 6.2. In many BN software, including NETICA, CPTs can be learnt from existing data sets and equations. Learning probabilities from case data can be achieved in various ways and many BN software provide built-in algorithms (such as counting-learning in the NETICA software) (Frank 2015). Conditional probabilities can also be derived from equations using NETICA software which are calculated by random sampling from the range of data each state represents (Norsys 2017). The number of random samples can be defined and uncertainty from the randomisation process can be included (Norsys 2017).

In NETICA and other software, continuous data has to be discretised into states that occupy separate ranges within the node. The size of a conditional probability tables is the product of the number of states in the node and the number of states in each parent (Norsys 2017). Therefore, using a large number of states to represent a variable (node) increases the precision of the model, but also computation, so a compromise has to be made (Liu *et al.* 2012). For CPTs which are manually derived using expert elicitation, a maximum number of 5 states per node has been suggested (Marcot *et al.* 2006).

6.2.6 Model design and validation

Bayesian Networks are strongly influenced by the model design as both the number of states used and the number of interconnecting nodes have an influence on final outcomes. The first step of the model development, is to define the aims and purposes of the model and identify model components which will form the basis of the conceptual model which forms the DAG.

Principles of good BN model design include:

- Creating nodes which are quantifiable and able to be validated with existing or collected data (Marcot *et al.* 2006).
- Combining nodes if they are not predictable and observable at the scale of the model to reduce complexity (Chen and Pollino 2012).
- Creating the simplest possible model at a minimum cost to precision (Marcot *et al.* 2006).
- Transparent reporting of the model design process (Chen and Pollino 2012).
- Continued updating and evaluation of the model even once it is accepted for general use (Pollino *et al.* 2007).
- Model validation through both testing with and without data, and review by experts (Marcot *et al.* 2006).

Validation and testing of the model is an important part of the development of an effective BN. Marcot *et al.* (2006) suggest several stages of development from a model suitable for internal use and development (α - level model), through to a peer-reviewed model (β - level model) which is then rigorously tested and validated with external data sets (γ - level model). At the α - level, the model is defined and tested experimentally to check if its performance matches expectations (Marcot *et al.* 2006).

Sensitivity analysis is one tool which can be used to test model performance. Sensitivity analysis can help identify child nodes which have the greatest influence on model outputs (Frank 2015), knowledge gaps and to determine whether the model performed as expected (Marcot *et al.* 2006; Pollino *et al.* 2007; Chen and Pollino 2012). Sensitivity to parameters is a further form of sensitivity analysis involving incrementally changing states within each node to observe the effect on surrounding nodes. This can help to identify weaknesses in the model design, but can be very time-consuming (Frank 2015). The γ - level model should be a peer-reviewed model which is calibrated with case-data (Marcot *et al.* 2006).

6.3 MODEL DEVELOPMENT

The BN was designed in three sections; a section describing the dose of HMs attributable to gardening activities, a section describing the health benefits of gardening, and a section combining both benefits and risks. Tables describing each of the nodes, the states and the relationship with child and parent nodes for each section are given below. A copy of the model, and the case data used to derive it, can be accessed through Appendix VI. Due to time constraints the model was limited to exposure to Cd and Pb. The model was also limited to health effects among adults as only adult data was acquired in the gardener's survey in chapter 5.

Selecting nodes, states and obtaining data

The design of the model and the number of states in each node can influence its operation (Marcot *et al.* 2006). Parent nodes in the BN were informed by case data for allotment gardeners collected in chapters 2 and 5, so the number of states in these nodes was set according to the source data. Child nodes were parameterised using equations or case data, and were usually designed to have fewer than 5 states to reduce model complexity.

6.3.1 Health risks

The structure of the exposure model to Cd and Pb from allotment soils was derived using the CLEA model in the UK (EA 2009d; Section 1.3.4 of this thesis). Nodes used in the calculation of exposure to Cd and Pb from

gardening are presented in Table 6.2. The daily dose of Cd and Pb attributable to exposure through urban allotment gardening, are calculated by summation of the dose from each exposure pathway (Equation 1.1 – Chapter 1). The relevant exposure pathways to HMs for an allotment scenario included in the CLEA model are: exposure through direct soil ingestion, consumption of home grown produce, consumption of soil adhering to produce, skin contact with soils and outdoor inhalation of dusts and vapours (EA 2009d). Of these pathways, skin contact with soils is irrelevant for Cd and Pb, for which dermal absorption of inorganic Cd and Pb is negligible (DEFRA 2013b; e) and neither Cd nor Pb is volatile in the environment so inhalation of vapours is irrelevant (DEFRA 2013b; e). The inhalation of soil dusts was calculated initially, but was found to have a negligible influence on dose; consistent with a sensitivity analysis of the factors influencing dose for allotment gardeners performed using the CLEA model (DEFRA 2013a).

Produce Consumption

Plant uptake regression models developed in Chapter 3 were used to estimate transfer of contaminant from soils into produce instead of concentration factors used by the CLEA model (see Equation 1.2). Grouped produce models were used instead of individual produce models (Table 3.17) (see Chapters 3 and 4), as regression models have not yet been developed to cover all of the types of produce grown. The volume of produce consumed by gardeners was informed by a survey of allotment gardeners in chapter 5 (Table 5.1), rather than the CLEA approach of

using national survey data for produce consumption adapted through the use of a derived 'home-grown fraction'.

Direct and indirect soil ingestion

Direct ingestion, and indirect ingestion, of soil adhered to produce are considered to be two separate pathways in the CLEA model. However, soil tracer studies used by the CLEA model to estimate soil ingestion, do not distinguish between ingestion of soil adhered to produce and direct ingestion of soil through incidental hand-to-mouth contact (Hough et al 2004). Direct and indirect soil ingestion were grouped together and represented by the *soil ingestion rate* node. The effect of indirect soil ingestion on the overall soil ingestion rate was modelled using Equation 1.4 (Chapter 1) with the states in the *produce consumption* node used to estimate the range of consumption volume (g fw), and an assumed soil loading factor of 0.001 (dimensionless) (EA 2009d). The influence of produce preparation was not considered. It was determined that if *produce consumption* was in the 'medium' or 'high' categories, the *soil ingestion rate* node must also be 'medium' or 'high' based on the contribution from indirect soil ingestion (Table 6.1).

Table 6.1: Soil ingestion rate (SIR) as a function of produce consumption.

State in produce consumption node (Kg Fw/ day)	Low (0 - < 0.085)	Medium (0.085 - < 0.167)	High (0.167 - < 0.215)
Calculated increase in SIR (mg / day)	0 - < 8.5	8.5 - < 16.7	16.7 - < 21.5

Background dose

The inclusion of a background dose of contaminants in the CLEA model has been subject to some debate (DEFRA 2013a). Given that background dose is still included in the CLEA model, there is an option to include background dose from ingestion of Cd or Pb in the BN. Background dose is unlikely to be a major component to the overall Pb dose for allotment gardeners, but smoking is a major contributor to the background dose of Cd, so the background dose of Cd can be a significant contributor to exposure for some gardeners (DEFRA 2013a).

Combining exposure pathways

To reduce model complexity and the need for intermediary nodes, the nodes 'Cd Dose' and 'Pb dose' are populated by a single equation which calculates the total exposure across all pathways for allotment gardeners in $\mu\text{g kg}^{-1} \text{ BW day}^{-1}$. This is a conservative estimate of the systemic dose as the bioavailability of Cd and Pb from soil derivatives to the human body is assumed to be 100% - consistent with the CLEA model (EA 2009d).

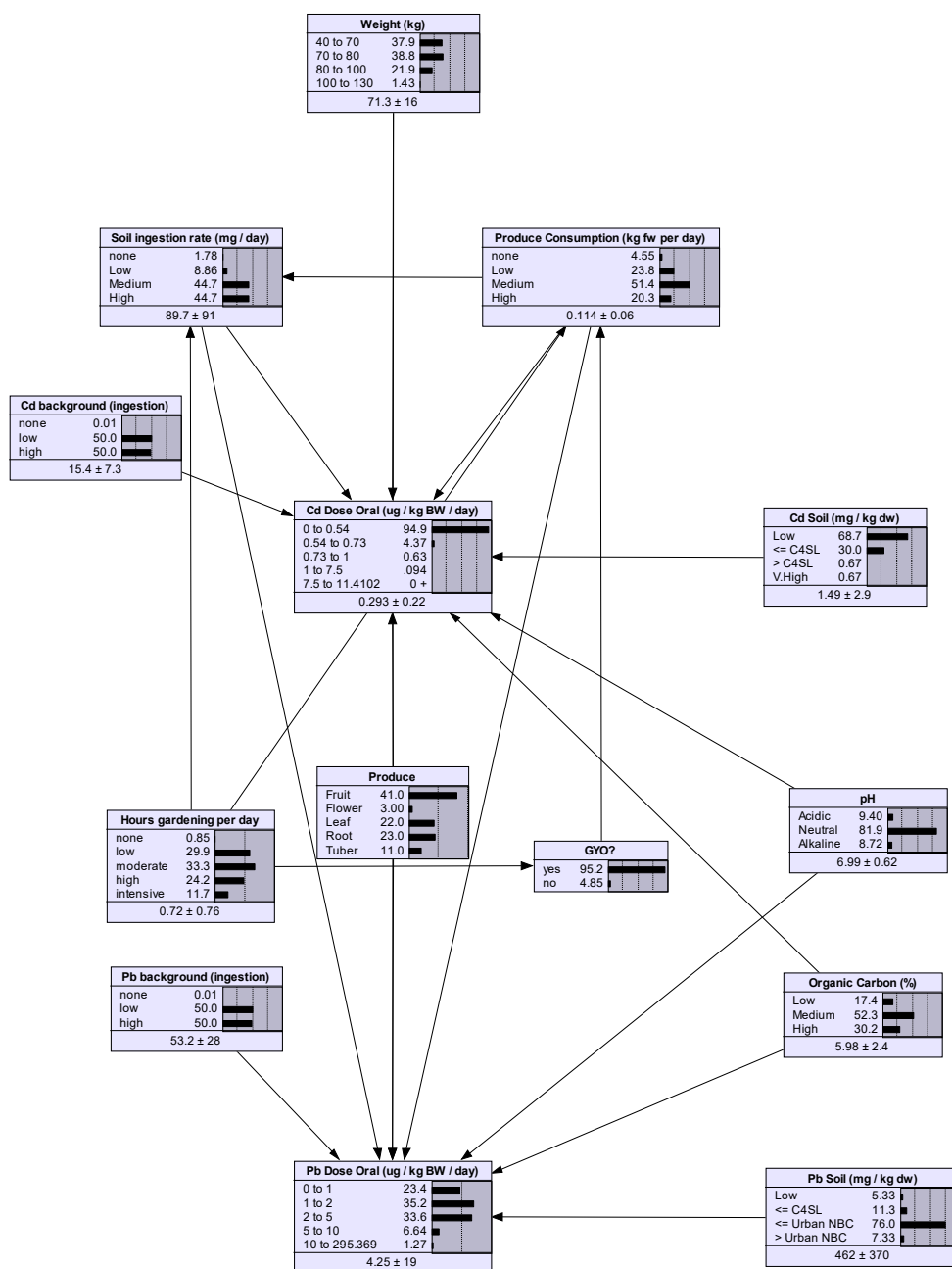


Figure 6.5: The structure of the health risks section of the BN.

Table 6.2: A description of the nodes in the health risks section of the BN to calculate Cd and Pb exposure for urban gardeners.

Node	Parent/Child nodes (nodes in red refer to other sections of the BN)	Definition	State:			Description
			Title (if applicable)	Value	Probability (initial)	
Cd Soil (mg kg ⁻¹ dw)	Child: <i>Cd dose oral</i>	Concentration of Cd in allotment soil (mg kg ⁻¹ dw)	Low ≤ C4SL > C4SL V. High	0- <1 1- <5 5- <10 10- <50	0.68 0.30 0.01 0.01	'Low' state assumed to be below C4SL of 4.9 mg kg ⁻¹ dw Cd which equates to minimal risk (DEFRA 2013a). Other states estimated from expert knowledge. Parameterised with allotment data from Chapter 2.
Pb Soil (mg kg ⁻¹ dw)	Child: <i>Pb dose oral</i>	Concentration of Pb in allotment soil (mg kg ⁻¹ dw)	Low ≤ C4SL ≤ NBC V.High	0- <40 40- <90 90- <830 830- <2000	0.05 0.11 0.76 0.07	Low' state assumed to be below C4SL of 84 mg kg ⁻¹ dw Pb which equates to minimal risk (DEFRA 2013a). Above the NBC of 830 mg kg ⁻¹ dw Pb, Pb concentrations can be considered abnormal for urban environments (Ander <i>et al.</i> 2013). Parameterised with allotment data from Chapter 2.
Hours gardening	Children: <i>GYO?</i> , <i>Soil ingestion rate</i> , <i>Produce consumption</i> , <i>exercise, fruit and vegetable consumption</i>	The mean number of hours spent gardening (hr day ⁻¹)	None Low Moderate High Intensive	0 0- <0.2 0.2- <0.7 0.7- <1.4 1.4- <3.5	0.01 0.30 0.33 0.24 0.12	States and probabilities were determined from a survey of gardeners' routines by estimating the average amount of time spent in hr day ⁻¹ , from the average number of hours and days gardeners spent on their plots (Chapter 5).
Body weight	Children: <i>Cd dose oral</i> , <i>Pb dose oral</i>	Body weight of the receptor (kg)	- - - -	40- <70 70- <80 80- <100 100- <180	0.37 0.38 0.22 0.03	States were distributed according to approximate weight quartiles of the UK population (HSCIC 2012b) Probabilities for gardeners were informed by gardener' responses in chapter 5.
pH	Children: <i>Cd dose oral</i> , <i>Pb dose oral</i>	Soil pH	Acidic Neutral Alkaline	5- <6.5 6.5- <7.5 7.5- <9	0.09 0.82 0.09	Soil pH range tends to be between 5 and 9 for allotments (Alloway 2004). State probabilities were informed by results from Chapter 2

Table 6.2 (continued)

<u>Node</u>	<u>Parent/child nodes</u>	<u>Definition</u>	<u>Title:</u>	<u>Value:</u>	<u>Probability</u>	<u>Description</u>
Organic Carbon (%)	Children: <i>Cd dose oral, Pb dose oral</i>	Soil organic carbon content (% of soil mass)	Low Medium High	1- <4 4- <7 7- <10	0.18 0.52 0.30	Soil carbon content tends to be between 0-10 % for allotments (Alloway 2004). State probabilities informed by soil survey of allotments (Chapter 2).
Produce	Children: <i>Cd dose oral, Pb dose oral</i>	Types of produce consumed grouped according to edible tissues	Fruit Flower Leaf Root Tuber	- - - - -	0.41 0.03 0.22 0.23 0.11	States informed by produce categories used for the regression models in Chapter 3. Probabilities informed by estimates of produce consumption rates using answers to the gardening and health survey (Chapter 5).
GYO?	Child: <i>Produce consumption</i>	Do gardeners use their garden to grow produce?	yes no	- -	94 6	Informed using case data from chapter 5. Related to hours gardening as those who spent no time gardening will not be able to grow their own produce.
Cd background (ingestion)	Child: <i>Cd dose oral</i>	Background dose (mg day ⁻¹)	None Low High	0 >0 - <18.4 18.4- <24.7	<0.01 0.50 0.50	The mean background Cd ingestion is 18.4 ug / day with a 95 th percentile of 24.7 ug / day in Europe (EFSA 2009).
Pb background (ingestion)	Child: <i>Pb dose oral</i>	Background dose (mg day ⁻¹)	None Low High	0 >0- <58.2 58.2- <6.6	<0.01 0.50 0.50	The mean background Pb ingestion is 58.2 ug / day with a 95 th percentile of 96.6 ug / day in Europe (EFSA 2009).
Produce Consumption	Parents: <i>Hours gardening, GYO?</i> Children: <i>Cd dose oral, Pb dose oral, soil ingestion rate</i>	Produce consumption volume (kg fw day ⁻¹)	None Low Medium High	0 >0- <0.085 0.085- <0.167 0.167- <0.215	0.05 0.24 0.51 0.20	Informed from produce consumption estimates from a survey of gardeners (Table 5.1). The 'low' and 'high' consumption states represent approximately the lower and upper quartiles of consumption.

Table 6.2 (continued)

Node	Parent/child nodes	Definition	Title:	Value:	Probability	Description
Cd dose oral	Parents: <i>Cd soil, pH, Organic Carbon, Cd background, produce, produce consumption, soil ingestion rate, Weight</i> <i>Children: see Table 6.3</i>	Cd dose for allotment gardeners ($\mu\text{g kg}^{-1}\text{ BW day}^{-1}$)	-	0- <0.54	0.95	The Cd and Pb oral dose nodes were populated using equation 6.6: $D = [(R_m \times P_c \times C_r + S_r / 10^6 \times M_s] \times 1000 + B) / \text{BW} \quad 6.6$ Where R_m is the output from produce- metal specific regression models (Table 3.17), P_c produce consumption (kg fw), C_r Fresh to dry weight conversion factor (adapted from EA, 2009a), S_r the soil ingestion rate (mg day^{-1}), M_s the concentration of the metal in soil ($\text{mg kg}^{-1}\text{ dw}$) B the background dose ($\mu\text{g / day}$) and BW bodyweight (kg). Five states were selected to represent each node based on toxicological outcomes – the lowest is below the respective LLTC for Cd and Pb.
			-	0.54- <0.73	0.04	
			-	0.73- <1.0	0.01	
			-	1.0- <7.5	<0.01	
			-	7.5- <11	<0.01	
Pb dose oral	Parents: <i>Pb soil, pH, Organic Carbon, Pb background, produce, produce consumption, soil ingestion rate, Weight</i> <i>Children: see Table 6.4</i>	Pb dose for allotment gardeners ($\mu\text{g kg}^{-1}\text{ BW day}^{-1}$)	-	0- <1	0.23	
			-	1- <2	0.35	
			-	2- <5	0.34	
			-	5- <10	0.07	
			-	10- <300	0.01	

6.3.2 Health benefits

There are many possible and wide-ranging health benefits that have been attributed to gardening and GYO (Leake *et al.* 2009; Buck 2016). To try and model every lifestyle change and possible health outcome from gardening would be near impossible, so the first step in creating the health benefits model was to reduce the number of outcomes being considered. Three main lifestyle changes related to gardening appear to be discussed more prominently than others in the literature, and have also been studied widely in epidemiology. These are: increased fruit and vegetable consumption, increased physical activity and improved mental health. The first two were observed in a survey of gardeners in chapter 5, whilst improvements in the mental health component of the SF 36 v 2 were insignificant ($P > 0.05$), relative to national surveys. The effect of changes in these lifestyle factors on the following selected health outcomes was considered: cancer and CVD.

Creating a model

Models of the occurrence of CVD and cancer are rare in the literature – most likely due to the complexity of reliably predicting the occurrence of either disease. An odds model developed by Martin *et al.* (2008) considered the influence of several risk factors on chronic heart disease (CHD) morbidity, including (low) fruit and vegetable consumption, exercise and mental health. The model combined odds ratios (ORs) from epidemiological studies for the occurrence of CHD as a result of the risk

factors (see Section 1.4.2 for an explanation of risk ratios). There was no precedent that the researchers could find for similarly predicting the risk of cancer. The Martin *et al.* (2008) approach was adapted for use in the present study to predict the reduced likelihood of CVD and cancer with increased fruit and vegetable consumption, physical activity and improved mental health. Given that it would be difficult to obtain the medical information used in Martin *et al.* as part of a risk assessment, the health benefits model was limited to lifestyle factors (LFs) including confounders such as alcohol consumption, BMI and smoking status. Parent nodes for each LF were populated using data obtained in a survey of gardeners (chapter 5).

Obtaining risk ratios

It was important to select an epidemiological metric that could be compared across health outcomes. Relative risks (RRs) and hazard ratios (HRs) are derived from cohort studies which are more common in the literature for studies of cancer and cardiovascular incidence (Case-control studies leading to the derivation of ORs are useful when health outcomes are rare in the population: Gerstman 2003). A literature review with a number of criteria was undertaken to obtain risk ratios comparing lifestyle factors with the risk of CVD or cancer morbidity.

Publications containing risk ratios were preferred if they:

- Contained RRs (HRs or ORs were used if RRs not available) published for the association between different levels of a LF and morbidity of cancer or CVD.
- Contained RRs published for cancer or CVD in general as opposed to for one specific type of cancer or CVD.
- Were based on a meta-analysis of available literature (preferred) OR a prospective cohort study of a large population group containing a mix of age and genders.
- Were based on a similar population to the study cohort in a developed country (e.g. UK, USA or Western Europe).
- Contained RRs calculated such that a positive influence on health gave an $RR < 1$ and a negative influence on health, an $RR > 1$.
- Contained RRs presented for different age / gender groups (preferred) OR RRs and HRs were adjusted for age and gender.
- Contained RRs adjusted for other confounding factors including smoking, socio-economic status, alcohol consumption, medical history etc.

The organisation of the nodes describing the relative risk of disease for exposure to each LF, were determined by the amount of information available from each publication and the source data (Table 6.3). Conditional probability tables in each of these nodes were derived by hand by calculating the upper and lower bounds of the relative risk using the source literature, for each state in the parent node(s).

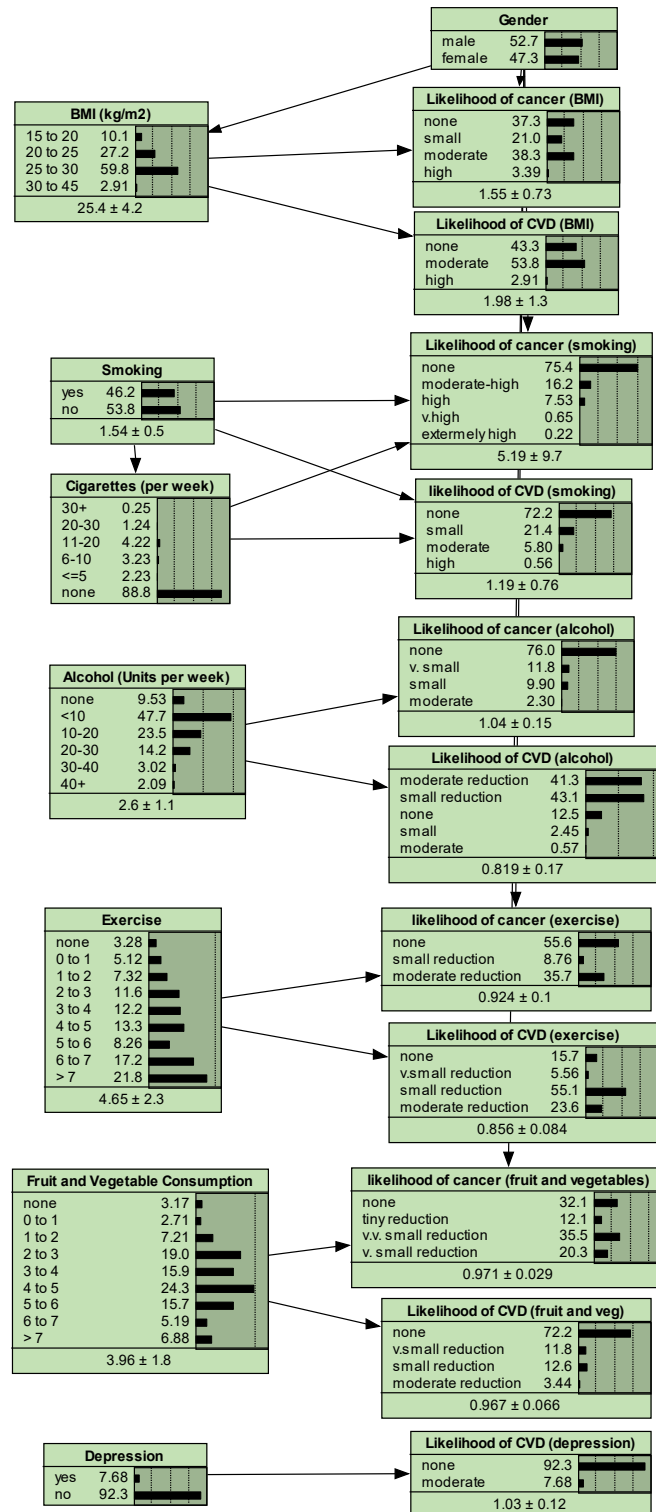


Figure 6.6: The structure of the health benefits section of the BN.

Table 6.3: A description of the nodes in the BN involved in the calculation of health benefits from urban gardening.

Node	Parent/Child nodes (nodes in red refer to other sections of the BN)	Definition	State:			Description/explanation
			Title (if applicable)	Value	Probability (initial)	
Gender	Children: <i>Smoking, BMI, likelihood of cancer (smoking), likelihood of cancer (fruit and vegetables), likelihood of cancer (exercise), weight</i>	Gender (male:female split)	Male Female	- -	0.53 0.47	Gender is important confounder in body weight used to estimate health risks and BMI among gardeners in the study population. RR's are also reported for specific genders in some epidemiological studies – other studies are adjusted for differences between genders.
Smoking	Children: <i>Cigarettes, likelihood of cancer (smoking)</i>	Has the individual ever smoked tobacco?	Yes No	- -	0.46 0.54	Smoking, even if the individual does not currently smoke can have an effect on the likelihood of disease. This is accounted for in Freedman et al for the relationship between smoking and cancer and included in the BN.
Cigarettes (per week)	Children: <i>likelihood of cancer (smoking), likelihood of CVD (smoking)</i>	The number of cigarettes smoked in the last 7 days	- - - - - -	30+ 20-30 11-20 6-10 ≤5 None	<0.01 0.01 0.04 0.03 0.02 0.89	Increased cigarette consumption is linked to both increased risk of cancer and CVD (Freedman <i>et al.</i> 2005; Teo <i>et al.</i> 2006). States derived from Ch 5 gardening survey.
Alcohol (units per week)	Children: <i>Likelihood of cancer (alcohol), likelihood of CVD (alcohol)</i>	The number of units of alcohol consumed in the last 7 days	- - - - - -	40+ 30-40 20-30 10-20 <10 None	0.02 0.03 0.14 0.24 0.48 0.10	Increased alcohol consumption is linked to an increase in cancer (Bagnardi <i>et al.</i> 2015) However, small quantities of alcohol may be protective against CVD but the mechanisms for this are unclear and should be treated with caution (Ronksley <i>et al.</i> 2011).

Table 6.3 (continued)

<u>Node</u>	<u>Parent/child nodes</u>	<u>Definition</u>	<u>Title:</u>	<u>Value:</u>	<u>Probability</u>	<u>Description</u>
Depression	Child: Likelihood of CVD (depression)	Diagnosis of depression informed using MCS score from SF 36 v2. (Ch 5)	Yes No	- -	0.08 0.92	Mental health is linked with Cardiovascular disease (Van der Kooy <i>et al.</i> 2007). The association between cancer and psychological health has been found to be weak or non-existent by a couple of authors (McGee <i>et al.</i> 1994; Dalton <i>et al.</i> 2002).
Fruit and Vegetable Consumption	Parent: <i>Hours gardening per day</i> Children: <i>likelihood of cancer (fruit and vegetables), likelihood of CVD (fruit and vegetables)</i>	The number of portions of fruit and vegetables consumed on the previous day	None <1 1-<2 2-<3 3-<4 4-<5 5-<6 6-<7 ≥7	None 0- <1 1- <2 2- <3 3- <4 4- <5 5- <6 6- <7 7+	0.03 0.03 0.07 0.19 0.16 0.24 0.16 0.05 0.07	The link between increased fruit and vegetable consumption and cancer is subject to some debate (Willett 2010). However, there are some casual mechanisms and increased fruit and vegetable consumption may reduce consumption of more harmful foodstuffs in the diet. Fruit and vegetable consumption is linked more strongly with a reduction in CVD (Dauchet <i>et al.</i> 2006).
Exercise	Parent: <i>Hours gardening per day</i> Children: <i>likelihood of cancer (exercise), likelihood of CVD (exercise)</i>	The number of occasions 30 minutes or more moderate or intensive exercise was achieved in the last 7 days.	None <1 1-<2 2-<3 3-<4 4-<5 5-<6 6-<7 ≥7	None 0- <1 1- <2 2- <3 3- <4 4- <5 5- <6 6- <7 7+	0.03 0.05 0.07 0.12 0.12 0.13 0.08 0.17 0.22	Exercise has a role in reducing the risk of certain types of cancer and CVD (Warburton 2006).

Table 6.3 (continued)

Node	Parent/child nodes	Definition	Title:	Value:	Probability	Description
BMI	Parents: <i>Gender</i>	Weight (kg) divided by height squared (m ²)	-	15- <20	0.10	Body mass index (BMI) is commonly used as a measure of obesity and overweight. It is easy to measure and a risk factor for both cancer and CVD (Meigs <i>et al.</i> 2006; Larsson and Wolk 2007)
	Children: <i>Likelihood of cancer (BMI), likelihood of CVD (BMI)</i>		-	20- <25	0.27	
			-	25- <30	0.60	
			-	30- <45	0.03	
Likelihood of cancer (smoking)	Parents: <i>Smoking, Cigarette consumption, gender.</i>	Relative risk of cancer smoking	None	1	0.75	Relative risks informed by large American (> 400k adults) prospective cohort study (Freedman <i>et al.</i> 2008)Hazard ratios (proxy for RRs) published for the relationship between lung cancer and smoking alone, for specific genders and cigarette volume. No distinction in Freedman et al 2008) between RRs for ≤5 and 6-10 cigarettes. Follow-up period = 8 years.
	Child: Cancer likelihood		Moderate-high	1- <20	0.16	
			High	20- <40	0.08	
			V.high	40- <60	0.01	
extremely high	60- <80	<0.01				
Likelihood of cancer (alcohol)	Parent: <i>Alcohol</i>	Relative risk of cancer alcohol consumption	None	1.0	0.76	Relative risks informed by meta analysis of relative risks of cancer alcohol consumption (Bagnardi <i>et al.</i> 2001). RR's presented in categories of g ethanol per day converted to units using 1unit = 10ml ethanol = 10g.
	Child: Cancer likelihood		V.small	1.0- <1.1	0.12	
			Small	1.1- <1.2	0.10	
			Moderate	1.2- <2.6	0.02	
Likelihood of cancer (BMI)	Parents: <i>BMI, gender</i>	Relative risk of cancer BMI	None	1.0	0.37	Relative risks informed by meta analysis (Larsson and Wolk 2007) for colon cancer alone. RRs presented for a 5 unit increase in BMI with baseline < 25 kg m ² according to gender.
	Child: Cancer likelihood		Small	1.0- <1.5	0.21	
			Moderate	1.5- <2.5	0.38	
			High	2.5- <6	0.03	
Likelihood of cancer (fruit and vegetables)	Parents: <i>Fruit and vegetable consumption, gender</i>	Relative risk of cancer fruit and vegetable consumption	None	1.00	0.32	Relative risks informed by large (> 400k adults) European prospective cohort study (Boffetta <i>et al.</i> 2010). Conservative hazard ratios (proxy for RRs) published for quintiles of consumption (g day ⁻¹) according to gender. Conversion: 80g ≈ one portion.
	Child: Cancer likelihood		tiny reduction	<1-0.98	0.12	
			V small red.	<0.98-0.95	0.36	
			Small red.	<0.95-0.90	0.20	

Table 6.3 (continued)

<u>Node</u>	<u>Parent/child nodes</u>	<u>Definition</u>	<u>Title:</u>	<u>Value:</u>	<u>Probability</u>	<u>Description</u>
Likelihood of cancer (exercise)	Parents: <i>Exercise</i> Child: Cancer likelihood	Relative risk of cancer exercise	None Small red. Moderate red.	1.0 <1.0-0.9 <0.9-0.7	0.56 0.09 0.36	Relative risks informed by (12k adults) prospective cohort study (Albanes <i>et al.</i> 1989) for cancer. RRs published according to gender for qualitative categories of recreational exercise. 'Much exercise' - 5+ occasions week ⁻¹ ; 'moderate' - >2-<5 occasions; 'little or none' - ≤2 occasions. 'Much exercise' used as baseline so RRs reversed so that 'little or none' considered baseline. Follow up ≈ 10 years.
Likelihood of CVD (smoking)	Parents: <i>Cigarettes, smoking</i> Child: CVD likelihood	Relative risk of CVD cigarette consumption	None Small Moderate High	1.0 >1.0- <1.1 1.1- <1.2 1.2- <1.4	0.88 0.06 0.05 <0.01	Relative risks informed by INTERHEART cohort study (27k adults) (Teo <i>et al.</i> 2006). Publication gives ORs (used as proxy for RRs) for increased risk of acute myocardial infarction with cigarette use (proxy for CVD).
Likelihood of CVD (alcohol)	Parents: <i>alcohol consumption</i> Child: CVD likelihood	Relative risk of CVD alcohol consumption	Moderate red. Small red. None Small (risk) Moderate (rk)	0.5-<0.8 0.8- <1.0 1.0 1.0- <1.3 1.3- <1.9	0.41 0.43 0.12 0.02 <0.01	Estimated RRs informed by meta-analysis (Ronksley <i>et al.</i> 2011). RRs given for increased ethanol consumption in g day ⁻¹ converted to units: 1unit = 10ml = 10g. The relationship between CHD and stroke incidence with alcohol consumption is mixed with a possible reduction in CHD risk with small quantities of alcohol consumption.
Likelihood of CVD (BMI)	Parents: <i>BMI</i> Child: CVD likelihood	Relative risk of CVD alcohol consumption	None Moderate High	1 1- <4 4- <10	0.43 0.54 0.03	RRs informed by a cohort study (~3k adults) (Meigs <i>et al.</i> 2006) RRs given for incidence of CVD per 5 unit gain > 25 units BMI. Follow up < 11 yrs.
Likelihood of CVD (fruit and vegetable consumption)	Parents: <i>Fruit and vegetable consumption</i> Child: CVD likelihood	Relative risk of CVD Fruit and vegetable consumption	None v.small red. Small red. Moderate red.	1.0 <1.0-0.9 <0.9-0.8 <0.8-0.7	0.72 0.12 0.13 0.03	RRs informed by meta-analysis of cohort studies for CHD (incidence or mortality) (Dauchet <i>et al.</i> 2006). RRs published for each additional portion of fruit and vegetables consumed day ⁻¹ .

Table 6.3 (continued)

<u>Node</u>	<u>Parent/child nodes</u>	<u>Definition</u>	<u>Title:</u>	<u>Value:</u>	<u>Probability</u>	<u>Description</u>
Likelihood of CVD (exercise)	Parents: <i>Exercise</i> Child: <i>CVD likelihood</i>	Relative risk of CVD exercise	None V.small red. Small red. Moderate red.	1 <1-0.9 <0.9-0.8 <0.8-0.7	0.16 0.06 0.55 0.24	RRs informed by meta-analysis (Sattelmair <i>et al.</i> 2011) for incidence of CHD. RRs published according to US leisure activity guidelines: 'advanced guidelines'- 5+ occasions week ⁻¹ ; 'basic' - >2-<5 occasions; 'no LTPA' - ≤2 occasions.
Likelihood of CVD (depression)	Parents: <i>Depression</i> Child: <i>CVD likelihood</i>	Relative risk of CVD depression	None Moderate	1 >1-< 1.8	0.92 0.08	RRs informed by meta-analysis (Van der Kooy <i>et al.</i> 2007) for incidence of CVD. RRs reported on a binary scale for incidence of CVD depression.

6.3.3 Combining risks and benefits

Several approaches were discussed in chapter 1 for the combination of health benefits and risks from urban gardening; including the use of risk ratios, disability-adjusted life years (DALYs) and expert elicitation. There are significant limitations to each of these methods discussed in Chapter 1. The approach developed here involves combining risk ratios for the incidence of disease from both the health benefits and health risks sections of the model.

Relative risks of disease from HM exposure

Several major health outcomes of exposure to Pb and Cd were considered in the development of the model. Human exposure to Cd can result in renal failure – identified by symptoms of tubular dysfunction; osteoporosis/osteomalacia - which may result in an increased risk of bone fractures; and an increased risk of some cancers (Järup and Åkesson 2009). Endpoints of greatest concern from human exposure to Pb at low concentrations are increased risk of hypertension (leading to cardiovascular outcomes) and renal dysfunction (ATSDR 2007).

Relative risks for the occurrence of each endpoint for toxicity given exposure to Cd and Pb expressed as concentrations in urine and blood respectively, were identified from the literature. Data from toxicokinetic models (ATSDR 2007- Pb; EFSA 2011- Cd) were then used to relate concentrations of Cd and Pb in urine and blood with an intake dose.

Combining relative risks

To estimate the combined relative risk of disease, Equation 6.7 was used:

$$RR_{combined} = \prod_{i=1}^{i=x} (RR_i)^{\left(\frac{1}{x}\right)} \quad 6.7$$

Where $RR_{combined}$ represents the overall relative risk of disease given x number of relative risks (RR). The RRs of cancer and CVD attributable to Cd and Pb exposure, were combined with RRs for each LF, to estimate the overall risk (RR) of cancer or CVD. To estimate the overall risk from all diseases in the model, RRs for cancer, CVD, and other diseases resulting from potential exposure to Cd and Pb, were combined together.

There is no precedent in the literature for combining risk ratios other than Martin *et al.* (2008) in which the log-transformed product was used. Equation 6.7 was developed for the BN as an improvement on Martin *et al.* (2008), with the power function ensuring that extreme values are controlled in the model and enabling the combined RR to be presented on the same scale with benefits (<1), and risks (>1). Estimates of combined RRs in the model are untested and should be used cautiously. For this reason, the overall risk node consists of just two states: one describing the probability of the inputs in the BN resulting in an overall benefit ($RR < 1$), and the other the probability of the inputs in the BN resulting in an overall risk ($RR > 1$).

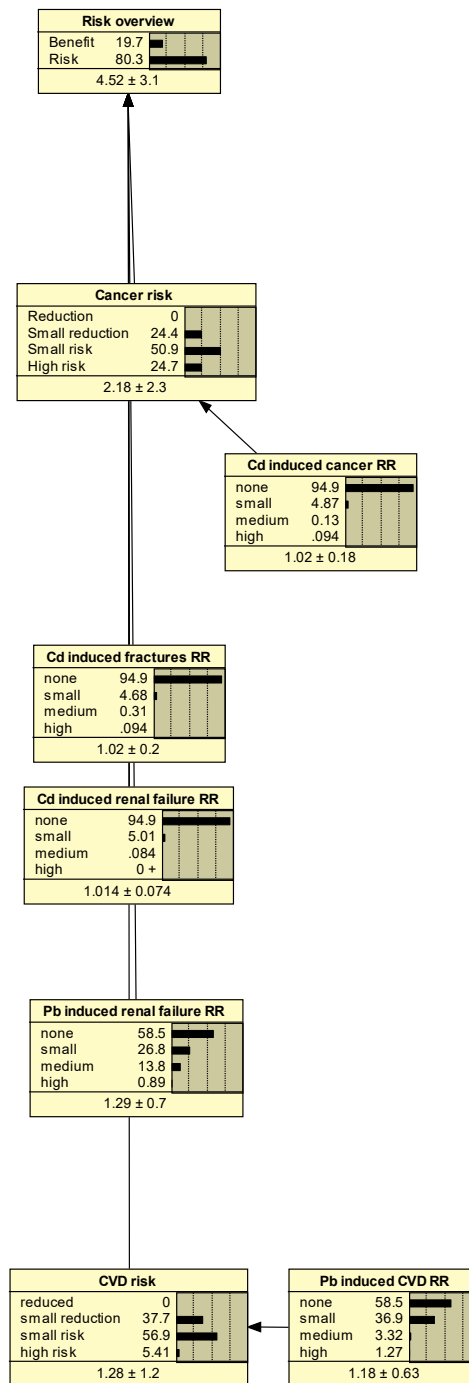


Figure 6.7: The structure of the section of the BN combining risks and benefits. The relative risks from each of the nodes are combined in the overall risk node at the top of the diagram.

Table 6.4: A description of the nodes involved in combining risks and benefits in the BN.

Node	Parent/Child nodes (nodes written in red relate to other parts of the BN)	Definition	State:			Description/explanation
			Title (if applicable)	Value	Probability (initial)	
<u>Explanation to the approach of estimating RRs for endpoints of Cd toxicity:</u>						
The relationship between dose and urinary excretion of Cd is displayed in figure 2 of (EFSA 2011). Urinary excretion (g Cd g creatinine) can be observed in this figure to approximately double with a doubling of intake dose. Each doubling of intake dose in Cd dose node is therefore expected to result in a doubling of urinary Cd excretion. RRs were given for the increased risk of bone fractures and cancer as a result of a doubling of urinary Cd excretion in Staesson <i>et al.</i> (1999) and Nawrot <i>et al.</i> (2006) respectively. In both papers, the baseline for exposure is approximately 1 µg L ⁻¹ urine which can be equated to a dose of ~ 0.54 µg Cd kg BW ⁻¹ : the minimum dose in the Cd dose node. Relative risks of kidney failure as a result of Cd exposure were determined using Hellstrom <i>et al.</i> (2001). There was no recorded urinary excretion from participants in the study by Hellstrom <i>et al.</i> (2001) so high exposure was equated to a dose > 7.5 µg Cd kg BW ⁻¹ ; moderate exposure – 1-7.5 µg Cd kg BW ⁻¹ and low exposure – 0.54-1 µg Cd kg BW ⁻¹ . For all toxicity endpoints, an upper limit of RR=10 was assigned to doses of Cd in excess of 7.5 µg Cd kg BW ⁻¹ .						
Likelihood of Cd induced cancer	Parent: <i>Cd Dose</i>	Estimated relative risk of cancer exposure to Cd	None	1.0	0.95	RRs obtained from Nawrot et al (2006) for a doubling of Cd urinary excretion (HRs used as a proxy for RRs). Cohort study population = 994 adults.
	Small		>1.0- <1.5	0.05		
	Medium		1.5- <2	<0.01		
	High		2- <10	<0.01		
Likelihood of Cd induced fractures	Parent: <i>Cd Dose</i>	Estimated relative risk of fractures exposure to Cd	None	1.0	0.95	RRs obtained from Staesson et al (1999) for a doubling of Cd urinary excretion. Cohort study population = 1107 adults
	Small		>1.0- <1.5	0.05		
	Medium		1.5- <2	<0.01		
	High		2- <10	<0.01		
Likelihood of Cd induced renal failure	Parent: <i>Cd Dose</i>	Estimated relative risk of renal failure exposure to Cd	None	1.0	0.95	RRs obtained from Hellstrom et al (2001) for the likelihood of renal replacement therapy administered by clinicians in response to Cd exposure.
	Small		>1.0- <1.5	0.05		
	Medium		1.5- <3	<0.01		
	High		3- <10	<0.01		

Table 6.4 (continued)

Node	Parent/Child	Definition	Title	Value	Probability	Description/explanation
<u>Explanation to the approach of estimating RRs for endpoints of Pb toxicity:</u> Slope factors for the relationship between blood Pb and intake dose were obtained from (ATSDR 2007). The range of slope factors given in (ATSDR 2007) is 0.4 – 0.65 ug Pb dl-1 blood per ug Pb day-1. From this, the corresponding range of Pb blood concentrations to the states in the Pb dose node were calculated. For each increment of Pb dose, corresponding RRs were obtained. ORs published in Muntner <i>et al.</i> (2003) for the NHANES III study were used to estimate RRs of renal failure identified as a reduction in glomular filtration rate from oral exposures to Pb. HRs from Menke <i>et al.</i> (2006) were used to estimate risks of CVD from low-level exposure to Pb (blood Pb < 10 ug dl-1) and RRs from Lusteberg and Silbergeld (2002) were used to estimate risks of CVD from high-level exposure to Pb (blood Pb > 10 ug dl-1). The upper limit of the RR for extremely high exposures (10-295 µg Pb kg BW-1) was assigned a value of 10 reflecting the potential damage of Pb toxicity on the kidney and the circulatory system.						
Likelihood of Pb induced CVD	Parent: <i>Pb Dose</i> Child: <i>CVD Risk</i>	Relative risk of CVD exposure to Pb	None	1.0	0.56	Menke et al (2006) - US cohort study: NHANES III (> 18 k adults) - HRs used as a proxy for RRs Lusteberg and Silbergeld (2002) - US cohort study: NHANES II (> 20 k adults) RRs from Lusteberg and Silbergeld (2002) were equated to Menke et al (2006) by multiplying RRs by HRs from the highest category in Menke <i>et al.</i>
			Small	>1.0-1.5	0.37	
			Medium	1.5- <2	0.03	
			High	2- <10	0.01	
Likelihood of Pb induced renal failure	Parent: <i>Pb Dose</i> Child: <i>Risk Overview</i>	Relative risk of renal failure exposure to Pb	None	1.0	0.59	RRs obtained from Muntner <i>et al.</i> (2003) (ORs used as a proxy for RRs) for the risk of reduced glomular filtration rate (defined as < 60 mL min ⁻¹ per 1.73m ² body surface area. Study cohort NHANES III (> 15 k adults).
			Small	>1.0- <1.5	0.27	
			Medium	1.5- <3	0.14	
			High	3- <10	<0.01	

Table 6.4 (continued)

<u>Node</u>	<u>Parent/Child</u>	<u>Definition</u>	<u>Title</u>	<u>Value</u>	<u>Probability</u>	<u>Description/explanation</u>
Cancer risk	Parent: <i>Likelihood of: Cd induced cancer, cancer (smoking), cancer (alcohol), cancer (BMI), cancer (Fruit and vegetables), cancer (exercise)</i>	Combined RR of cancer lifestyle factors and Cd exposure	Reduction	0.00 - <0.80	0	The combined RR of cancer calculated from RRs describing the likelihood of cancer for LFs and Cd exposure. The CPT for the node was populated using equation 6.7
			Small red.	0.80- <1.00	0.24	
			Small risk	1.00- <1.25	0.51	
			High risk	1.25- <10.00	0.25	
	Child: <i>Risk overview</i>					
CVD risk	Parent: <i>Likelihood of: Pb induced CVD, CVD (smoking), CVD (alcohol), CVD (BMI), CVD (Fruit and vegetables), CVD (exercise)</i>	Combined RR of CVD lifestyle factors and Cd exposure	Reduction	0.00- <0.80	0	The combined RR of CVD calculated from RRs describing the likelihood of CVD for LFs and Pb exposure. The CPT for the node was populated using equation 6.7
			Small red.	0.80- <1.00	0.38	
			Small risk	1.00- <1.25	0.57	
			High risk	1.25- <10.00	0.05	
	Child: <i>Risk overview</i>					
Risk overview	Parent: <i>Cancer Risk, CVD risk, Likelihood of Cd induced fractures, Likelihood of Pb induced renal failure, Likelihood of Pb induced CVD</i>	Combined RR displaying overall risks and benefits from LFs, exposure to Cd and Pb.	Benefit	0- <1	0.20	The combined RR for all risks relating to LFs and Cd and Pb exposure from gardening. The CPT for the node was populated using equation 6.7
			Risk	1- <10	0.80	

6.4 MODEL APPLICATION

The BN can be applied to assess benefit and risk in several ways. The overall benefit/risk score for gardeners is presented in the *risk overview* node as a probability of the combination of model inputs resulting in a benefit or a risk. This 'score' includes the effect of confounding factors such as smoking and cigarette consumption which may be removed from the study if desired, by selecting the state 'none' in the relevant RR nodes (e.g. *likelihood of cancer (smoking)*). Other nodes such as the *Cd dose* and the *Pb dose* node may also be useful outputs for the purposes of risk assessment. Nodes for which the states are unknown can be represented using a uniform distribution. The BN may be applied to bottom-up (starting with model inputs) or top-down modelling (starting with model outputs):

- Using a bottom-up approach, the BN can be applied to site specific risk assessment as well as scenario modelling. When used for site-specific assessment, the BN is useful for studying high- (or low) risk individuals or groups. Scenario modelling can be used to determine the outcome of changing model inputs such as soil pH, organic carbon content and produce consumption. The representation of variables as states with probabilities, allows 'realistic worst-case scenarios' to be determined and tested against expectations.

- Using a top-down approach, the BN can be applied to determine ‘safe’ concentrations of HMs in soils, and as a tool to aid risk management decisions. The effect on the rest of the network of maximising benefits and minimising risks, can help determine the most effective methods to maximise overall health. It may be necessary to constrain the effect on some variables (e.g. gender) that cannot be altered, whilst exploring the impact of changing others which can be (e.g. soil pH and organic carbon content).

6.5 MODEL TESTING AND VALIDATION

It was not possible at this stage to validate the BN with an independent data set, so the model should be considered to be an α -level model: suitable only for internal assessment and review (Marcot *et al.* 2006). Initially, BNs can be tested by modelling logical assumptions known as predictive inference, and through sensitivity analysis (Troldborg *et al.* 2013). Sensitivity analysis is used to create a ranking which indicates how much the variation in one node is influenced by the variation in other nodes in the network (Frank 2015). Sensitivity analysis is also useful to ensure that independent nodes are appropriately separated from each other in the BN (Frank, 2015).

6.5.1 Sensitivity analysis.

Sensitivity analyses were carried out for selected key nodes in the BN (including *risk overview*, *Cd dose*, and *Pb dose*). The *risk overview* node was

mostly informed by the *cancer* and *CVD* RR nodes. *Body Mass Index* was most influential as a predictor of risk, followed by *Pb dose* and *cigarette use* (Figure 6.8). This may reflect the low use of cigarette consumption among the cohort of allotment gardeners and relatively high Pb concentrations in the soil of some of the allotments the gardeners were using. *Soil Pb* was also a more important factor in the analysis than fruit and vegetable consumption and exercise.

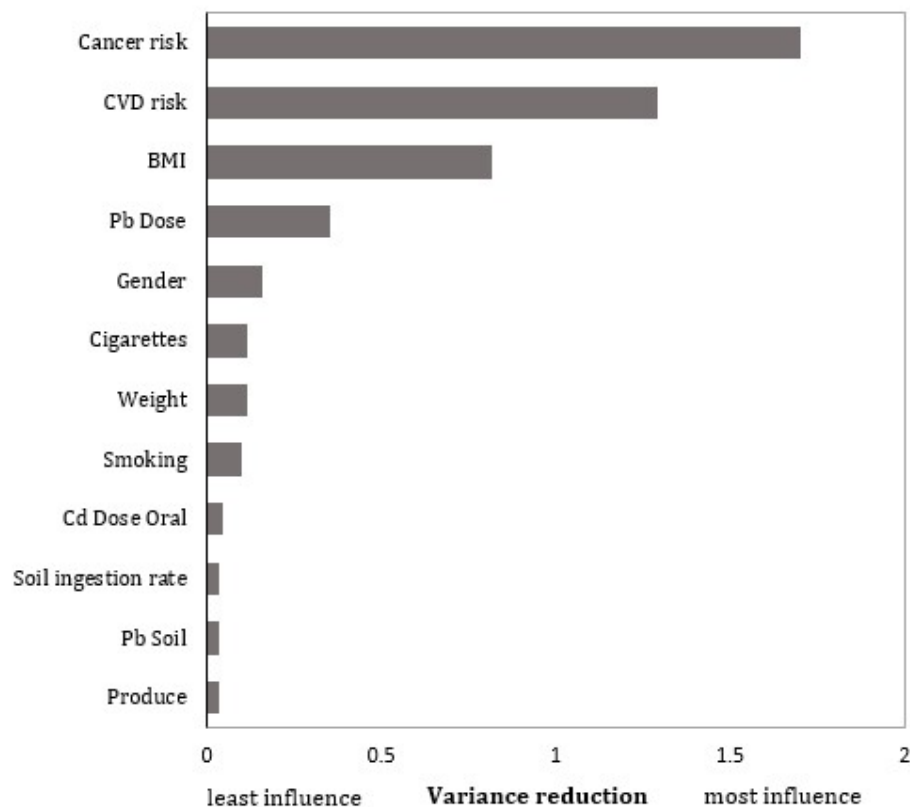


Figure 6.8: Results of sensitivity analysis on risk overview node. Selected nodes only – intermediate nodes and nodes accounting for less than 0.3 of the variance in the risk overview node removed.

Sensitivity analysis of the *Pb dose* node showed that this node was mainly influenced by the *CVD risk* and *Pb induced renal failure* nodes (Figure 6.9). A surprising result of the sensitivity analysis of the *Pb dose* node was the

similarity in the influence of *produce consumption* and *soil ingestion rate*, as Pb is not very bioavailable so it is unlikely to be readily taken up by plants (Chapters 3 & 4). Produce consumption does contribute to indirect ingestion of soil in the network as increased produce consumption will result in increased exposure to soil adhered to produce surfaces (EA 2009d). Furthermore, the Pb regression models used to estimate plant uptake may over-estimate Pb uptake at higher concentrations (seen in lettuces in Chapter 4). The *Pb dose* node was unrelated to nodes such as *Cd soil* and *alcohol consumption* showing appropriate division of the network to these nodes.

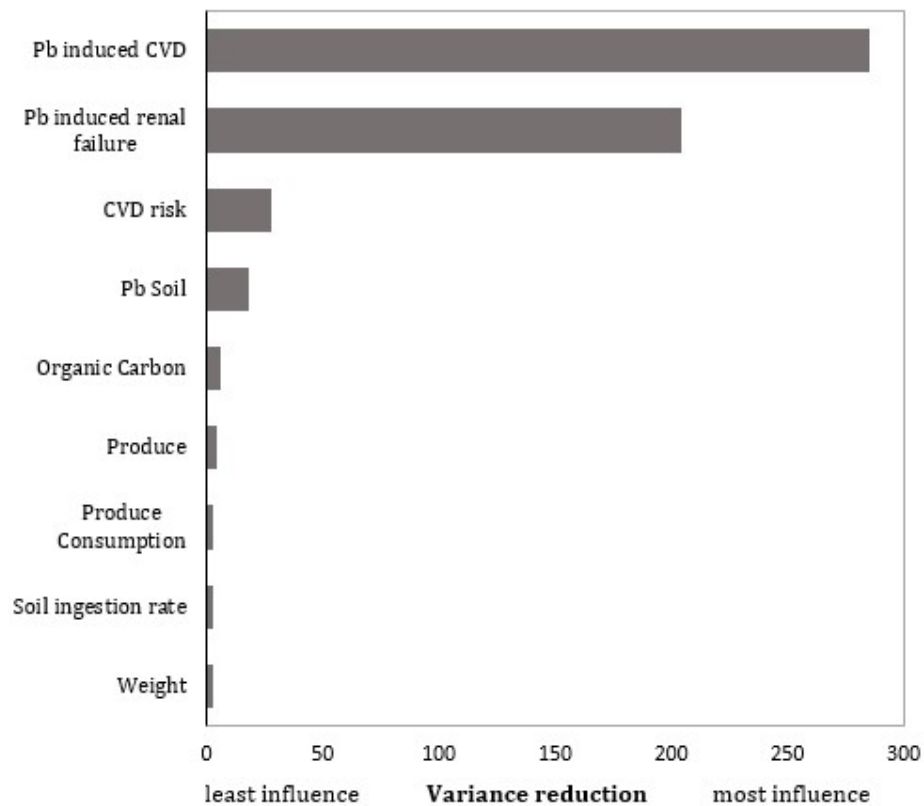


Figure 6.9: Results of sensitivity analysis on Pb dose node. The first nine most influential nodes are included.

Sensitivity analysis of the *Cd dose* node showed that it is influenced by *Pb dose* more than it is influenced by *Cd soil* (Figure 6.10). This is slightly counter-intuitive; however, the low Cd concentrations in the allotment soils studied are unlikely to result in a dose above the lowest state in the *Cd dose* node. Therefore, this node is more strongly influenced by other factors such as produce consumption and soil ingestion, which also have a combined influence on Pb dose.

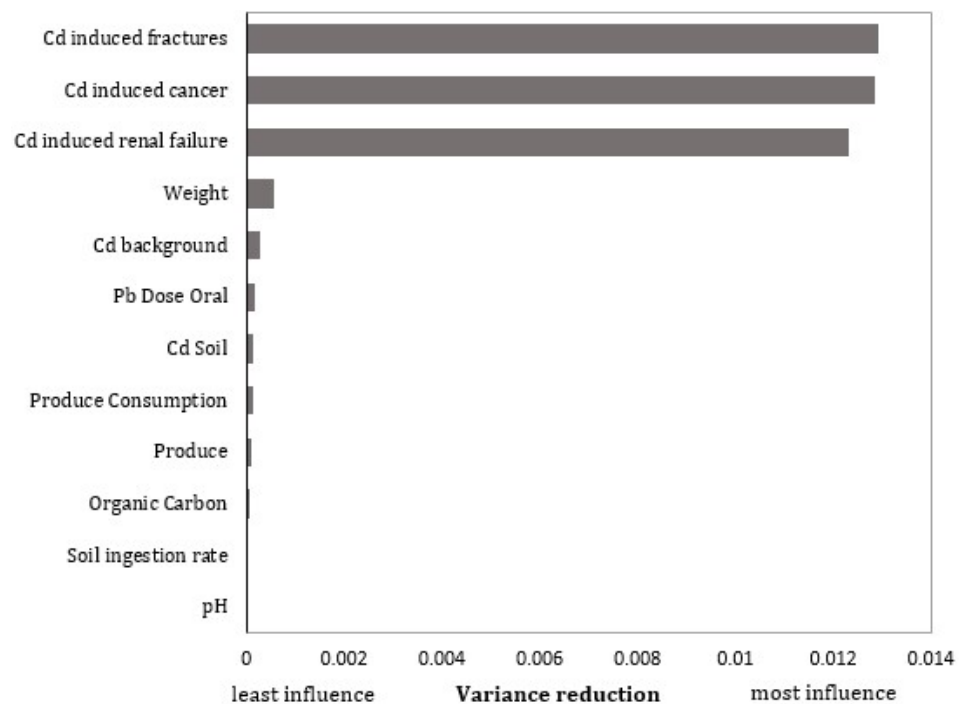


Figure 6.10: Sensitivity analysis for the Cd dose node. Only selected nodes shown which had the greatest influence and were of greatest interest.

6.5.2 Testing logical assumptions

The model outputs performed as expected with respect to some simple logical assumptions. The model predicted a higher probability of risk in the *risk overview* node with increased cigarette consumption, for increased alcohol consumption (above 20 units per week), for increased BMI (above 25 kg m⁻²), for increased doses of Cd and Pb, and increased concentrations of Cd and Pb in the soil (above the C4SL). The model predicted a lower probability of risk with increased fruit and vegetable consumption, exercise, and improved mental health (Figure 6.11). The model also predicted a higher probability of risk with increased produce consumption and increased ingestion of soil. The highest risk produce to consume, according to the model, was leafy produce (e.g. lettuces) and the lowest risk produce was tubers (e.g. potatoes). This corresponds well to known trends in produce uptake of heavy metals, especially Cd, which is known to accumulate more in leaves than in tubers (Chapters 3 & 4; Clemens *et al.* 2006).

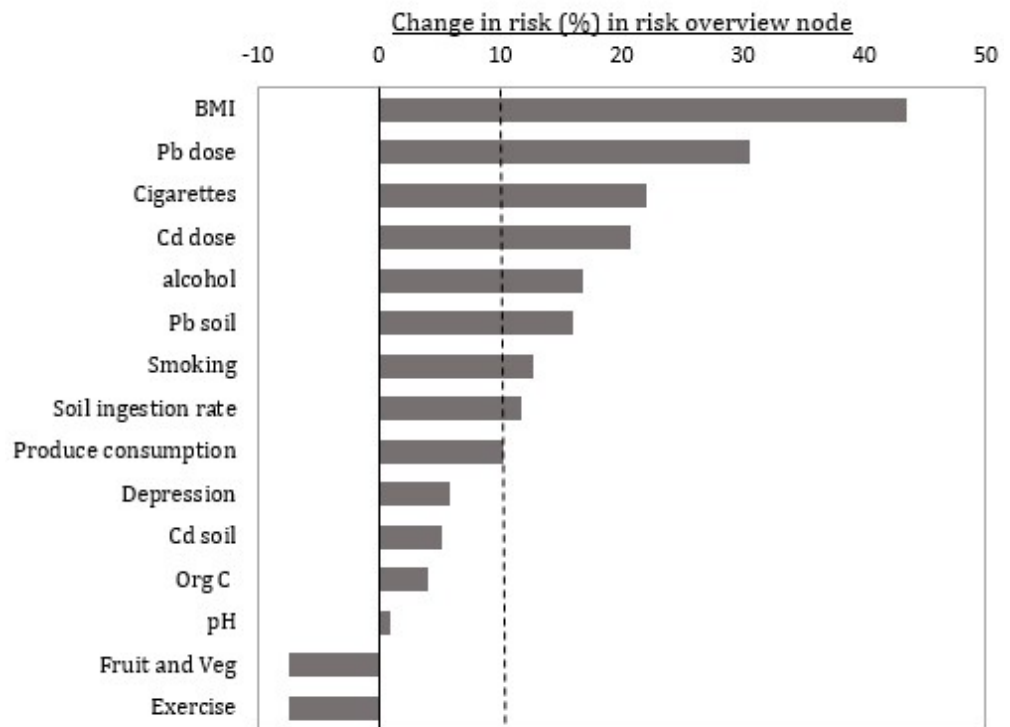


Figure 6.11: Change in the probability of the state 'risk' in the risk overview node, resulting from a change from the highest to lowest risk state in each parent node (a lower pH was considered to be higher risk as low pH is expected to result in a higher mobility of HMs into plants).

6.5.3 Testing model predictions of benefits and risks

The model was tested in its prediction of benefits and risks using the cohort of gardeners studied in chapters 2 and 5. Whilst some of the allotments studied did contain elevated concentrations of soil Pb, it was expected that the benefits of gardening would exceed the risk from Cd and Pb exposure for this cohort, because the soil of the allotments studied is not considered to constitute contaminated land. However, the influence of gardening on the study cohort was found to have a mixed result on the overall risk score.

The initial risk score for the study cohort of gardeners was 80% in the *risk overview* node. The effect of gardening on this risk can be determined by removing gardening from the model. By selecting 'none' in the *hours gardening* node, the risk from exposure to metals becomes zero and the benefits from fruit and vegetable consumption and exercise are reduced to a level corresponding to the general UK population. It was found that doing any amount of gardening, even just 0-0.2 hours day⁻¹, was found to increase the overall risk score in the model more than is compensated by the benefits of fruit and vegetable consumption according to the model predictions. Interestingly, the risk score did not increase with increased hours spent gardening from none to intensive (1 ½ - 3 ½ hours day⁻¹), suggesting that the increased risk from Pb exposure as a result of doing more gardening, may be compensated to some extent by the increased benefits from fruit and vegetable consumption and exercise (Figure 6.12). Consuming low amounts of fruit and vegetables (< 3 portions) and

exercising infrequently (< 2 times per week), was found to result in a higher risk score (89%) compared to doing any amount of gardening for the study cohort.

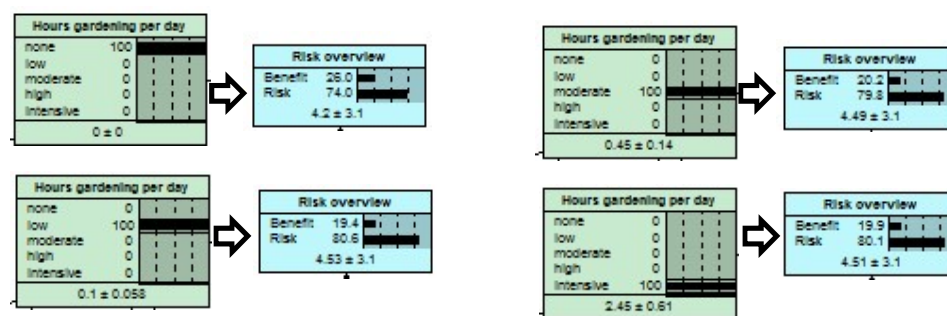


Figure 6.12: The change in overall probability of risks and benefits with incremental changes to the hours gardening node.

6.6 DISCUSSION

6.6.1 Advantages of the BN model

There are number of advantages of the BN presented here for the risk assessment of urban gardens. Firstly, the flexibility of the BN allows risk assessors to model many aspects of urban gardening. The BN can be used to calculate the impact of exposures to contaminants in the soil on different sub-groups of the population (e.g. vegans or vegetarians); to suggest effective remediation strategies, to carry out scenario modelling, and to derive 'safe' limits of contaminants in soils. Expanding or revising the network is straightforward (although each change should be tested and validated before it is applied), allowing risk assessors to implement changes and test the effect of other variables (such as the effect of produce preparation and changes to soil texture). The BN can also be easily adapted to new data sets (Pollino *et al.* 2007; Tighe *et al.* 2013),

which means the model should be improved as more data (e.g. on health benefits) is made available. Finally, BNs are able to cope with different types of data and relationships between variables that are uncertain or unknown (see: Chen and Pollino 2012; Troldborg *et al.* 2013). This makes the BN well suited to modelling health benefits from gardening which are less well categorised than health risks.

Dealing with uncertainty

A further advantage of the probabilistic, BN approach, is that uncertainty is represented graphically within the model design. It has been argued previously that probabilistic outputs can confuse the decision making process (Lindley 2001). As a result, many existing risk assessment models calculate a realistic, worst – possible outcome for exposure to contaminants (National Academy of Sciences 2013). However, as the actual risk falls somewhere between zero and this upper bound, in most cases this is an over-estimate (National Academy of Sciences 2013). Because BNs display the probability of the full range of possible outcomes, they provide a fairer representation compared to existing models of the distribution of risks, which is important for situations (including gardening, but also other scenario's such as the consumption of fish contaminated with methyl mercury: Mozaffarian and Rimm 2006) where there may be a benefit from the activity that results in exposure. Using a BN approach for the assessment of urban gardens may result in fewer, unwarranted closures of sites for which adverse effects on human health have not been observed and are unlikely to occur (see: Leake *et al.*

2009). This could result in more urban spaces being made available to gardeners which may also have a number of societal and economic benefits (Section 1.1).

6.6.2 Limitations of the BN approach

Whilst there are several advantages to the BN approach for assessing the impact on health of urban gardening, there are also some clear disadvantages. Firstly, it is not possible in most commercial BN software to represent continuous data except through the use of discrete intervals. This reduces the accuracy of model predictions, and will further increase uncertainty in the outcomes of a risk assessment (Aguilera *et al.* 2011). The design of the BN is also an important factor (Chen and Pollino 2012). In the BN presented here, the choices of lifestyle factors (benefits) and endpoints of toxicity (risks), whilst fully considered in the model design, are likely to bias estimates of risks and benefits. Furthermore, the model complexity and the number of nodes between model inputs and outputs is also an important factor and can add uncertainty to the model. Uncertainty in the model design is not represented by the BN so does have to be considered separately by stakeholders using the model (Chen and Pollino 2012).

Limitations in predicting benefits and risks

Application of the BN to predict the risks and benefits of the study cohort suggested that the risks from exposure to Pb in the soil would outweigh

the benefits of gardening. Whilst this finding could be accurate, it goes against the general expectation in the literature that the benefits of gardening are likely to outweigh the risks from low level exposure to Pb or other HMs (Leake *et al.* 2009; Brown *et al.* 2016). There are a number of limitations specific to the BN presented here which may have had an influence on the depiction of benefits and risks. To start with, there was a limited amount of data available to the author to describe Pb and Cd toxicity at concentrations greater than the reference dose (RfD). The dose-response relationships of these chemicals beyond the RfD has a large bearing on the risk to any individuals exposed to these contaminants in the soil, and could have been modelled incorrectly using the limited data available. Epidemiological evidence for the effect of gardening on disease is also absent in the literature (Buck 2016), which means establishing a relationship between gardening and disease was difficult. Furthermore, the method used to combine risk ratios is unvalidated and could add considerable uncertainty to the model outcomes. Finally, plant uptake models (including the regression models developed here) are likely to over-estimate Pb-uptake into produce (see Chapter 4) which may have resulted in higher estimates of Pb exposure for the gardeners studied here than would be expected. A few authors have suggested that low exposure to Cd and Pb from soils could result in adverse health effects in the general population which would be borne out by the predictions of the BN model in its current form (Nawrot *et al.* 2006; Zahran *et al.* 2013).

6.6.3 Future model development

There are a few suggested changes to the BN presented here that were outside the scope of the current project which might improve its ability to be used to accurately predict health benefits and risks from urban gardening. These include:

- Developing a framework based on DALYs or expert knowledge to compare the severity of health outcomes (e.g. cancer, renal failure) in the model. The derivation of a DALY would allow a score to be obtained in terms of the estimated number of years of life lost as a result of practising, or not practising, urban gardening. However, there are some limitations with this approach including deriving disability weights for exposures to Cd (Section 1.4.3).
- Incorporating toxicokinetic models which estimate the dose-response relationship for Cd and Pb. Physiologically-based pharmacokinetic (PBPK) models can be used to predict dose-response patterns for doses at concentrations greater than the RfD (Swartjes and Cornelis 2011).
- Testing and validation of the approach to combine RRs from different parts of the BN.

- Testing and validation of the BN with epidemiological data and testing of the produce consumption estimates using more accurate survey techniques such as food diaries or meal replication.

6.7 CONCLUSIONS

To the author's knowledge, this is the first study to attempt to model the benefits and risks from urban gardening, and to present a framework which could be developed to include the health benefits of gardening within risk assessment. There are few studies presently in the literature that take such a holistic view on health outcomes for urban gardeners. Furthermore, there are no epidemiological studies at present that have investigated the influence of gardening on the prevalence of disease in the population (Buck 2016). Gardening is already recognised for its role in therapy for a number of illnesses (Simson and Straus 1997) and there is an ongoing debate as to whether gardening therapy should be funded on the NHS (BBC Radio 4 2017). In the BN produced here, it can be demonstrated that gardening (excluding exposure to contaminants in soils), does have a positive effect on health as a reduction in disease. Further research is required to validate this finding and epidemiological studies, in particular, are required to investigate the overall influence and value of gardening on human health: especially for sub-populations such as the elderly and infirmed, for whom it is likely to be of most benefit (Wang and MacMillan 2013; Buck 2016).

The BN presented here provides a framework which could be developed to predict the trade-off between the health benefits and risks of urban gardening. However, there is still some work to be done to create a model that would be appropriate for use in risk assessment. Whilst BNs do offer some great advantages for the purpose of risk assessment, there are still some limitations, including the discretisation of continuous data by most commercial software, which should be resolved before BNs become the preferred choice of model for this purpose. Future work should focus on the development of BNs for risk assessment, for which they have so much potential (Pollino *et al.* 2007; See also: Aguilera *et al.* 2011) and the validation and development of the BN presented here, so that it could be adapted for use in mainstream risk assessment. Based on the evidence presented in this chapter, it is strongly encouraged that health benefits should be incorporated into risk assessment decisions concerning urban gardens and allotments, for the protection and promotion of human health.

7 CONCLUSIONS

The main aims of this thesis were to improve risk assessments for urban gardeners exposed to contaminants in soil, and to develop a framework which could be used to estimate the trade-off between health benefits and risks attributable to urban gardening. These aims were achieved through an investigation of a cohort of urban gardeners, the creation of Freundlich regression models describing plant uptake of HMs, and the development of a BN to combine estimates of both health benefits and risks. The study has also highlighted a number of points worthy of further discussion and research.

7.1 DO THE HEALTH BENEFITS OF URBAN GARDENING OUTWEIGH THE RISKS?

Ultimately this question depends on a number of factors specific to an individual situation, such as the concentration of contaminants in the soil, and the relevant routes of exposure (produce consumption, soil ingestion etc.). The BN model developed here (Section 6.3.3) supports the view that below the C4SL of 4.9 mg Cd kg⁻¹ soil and the C4SL of 84 mg Pb kg⁻¹ soil, these metals are highly unlikely to result in adverse health effects to the adult population (health effects on children were not studied) and therefore any gardening on these soils will likely be beneficial to health. Beyond the respective C4SLs for Cd and Pb in soils, it is likely that a concentration will be reached at which the health benefits are balanced, and then exceeded by the health risks from metal toxicity. Further research should aim to review C4SLs to account for the health benefits of

gardening, or to provide local authorities with a means to consider health benefits when making decisions over the future use of urban gardens and allotments.

7.1.1 Are gardeners at risk from lead exposure?

On the basis of the BN model, it is possible that the cohort of allotment gardeners studied in this thesis may be at low-level risk from exposure to Pb. Concentrations of Pb in the allotment soils studied, were frequently found to be greater than the C4SL of 84 mg Pb kg⁻¹ soil, and infrequently found to exceed the NBC of 830 mg Pb kg⁻¹ soil. These concentrations are not uncommon for soils in urban gardens and allotments in the UK (Davies 1978; Hursthouse *et al.* 2004; Mulloli *et al.* 2004; Giusti 2011) and the disparity between the NBC in urban areas for Pb, and the C4SL, gives some cause for concern. Remediation of Pb below the C4SL is likely to be impractical due to the possibility of recontamination with air-borne Pb (Brown *et al.* 2016). Lead exposure may be over-estimated by risk assessment models, as the bioavailability of Pb is likely to be over-estimated by plant uptake models (Section 3.3.4), and 'black carbon soots' present in urban soils may also bind to Pb reducing its bioavailability for absorption into the blood stream (Leake *et al.* 2009).

7.1.2 Future and ongoing work to determine Pb exposure

From the discovery of the Pb concentrations in the soils of allotments in Nottingham, several strands of work to further investigate this issue have

been initiated. The source of Pb in the allotment soils studied here is being further investigated using a novel, analytical technique developed as part of a separate PhD, and an MSc project (unpublished) has already studied the proportion of Pb in the allotments that is derived from petrol using Pb isotopes. These studies will help determine where the Pb in the soils of these allotments originated and to reduce further inputs in the future. Another study, initiated by the University of Sheffield (Edmundson *et al.*), is using the allotment soils collected in this study to develop a technique to investigate the proportion of soil Pb bound to black carbon soots. This project also aims to carry out a more detailed investigation of exposure to contaminants across a number of urban allotments in the UK.

7.2 IS IT POSSIBLE TO DO MORE TO MANAGE RISK?

7.2.1 Increasing public awareness

Increasing awareness among gardeners of the risk of exposure to HMs and how to manage it, can help to reduce risks, and increasing knowledge may also reduce the perceived severity of the risks (Barnett and Breakwell 2001). The awareness of a cohort of gardeners of contaminants in the soil in an industrial area in Baltimore (USA), was studied by Kim *et al.* (2014). The survey of 70 gardeners found that most were aware of the issue of contamination with Pb. However, relatively few respondents (3%) were aware of the risks from contamination of soils with Cd and other HMs (Figure 7.1).

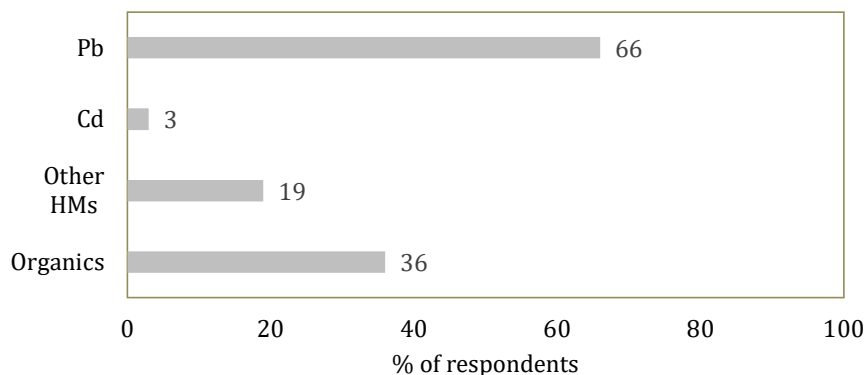


Figure 7.1: The percent of respondents (n=70) who were aware of each type of contaminant in a survey of gardeners in Baltimore, USA. Data was obtained from Kim *et al.* (2014).

The same survey also investigated gardeners' awareness of methods to manage their own exposure. Half of the gardeners were aware that they could reduce their exposure to contaminants by reducing their produce consumption (GYO), but a smaller proportion were aware of other methods to reduce their exposure, and nearly a quarter did not know how to reduce their exposure at all (Figure 7.2).

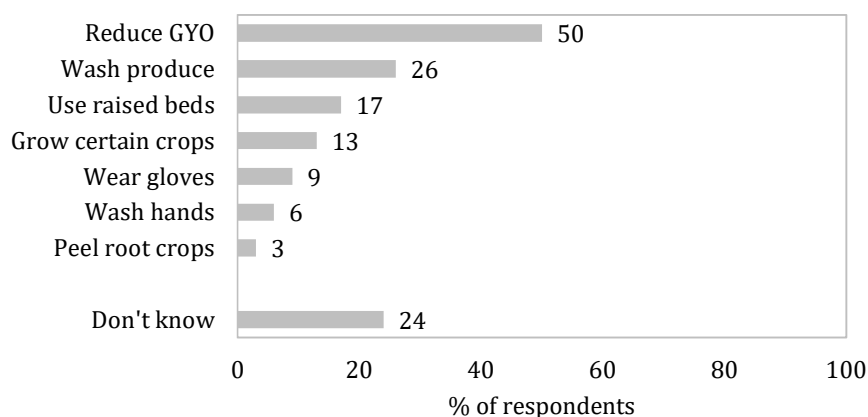


Figure 7.2: The percent of respondents (n=70) who could suggest methods to reduce their exposure to soil contaminants in a survey of gardeners from Baltimore, USA. Data was obtained from Kim *et al.* (2014).

7.2.2 Effectiveness of different methods to reduce exposure

It was possible to use the BN to investigate the effect of different management techniques on the overall risk/benefit score for the cohort of allotment gardeners (Figure 7.3).

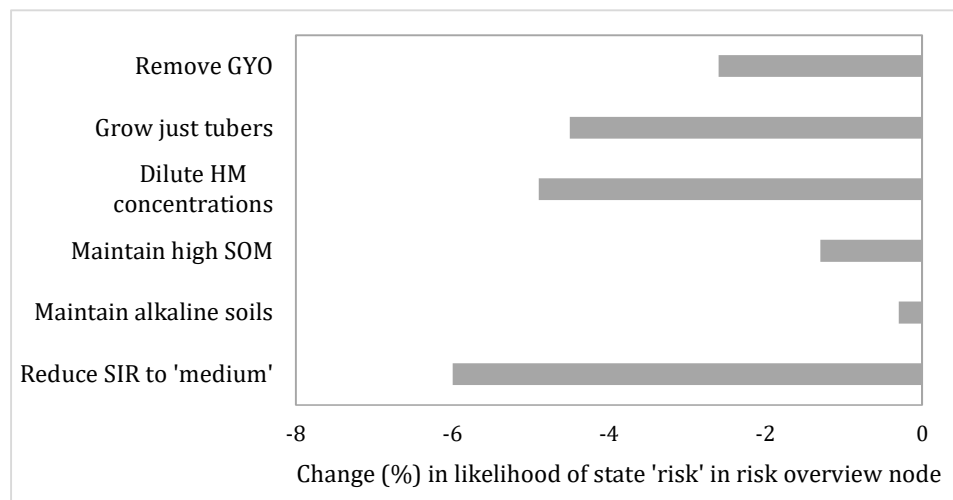


Figure 7.3: The effect of changing model inputs in the BN to reflect exposure management techniques, on the likelihood of the 'risk' state being selected by the BN.

Changing produce consumption:

Reducing or removing produce consumption, reduces exposure to Cd and Pb but also reduces the health benefits that could be gained from consumption of fruit and vegetables. It is therefore likely to be more beneficial to strategically grow low accumulating crops such as tubers, in preference to high accumulators such as leafy produce (Figure 7.3). Changing produce consumption is likely to be more effective to reduce exposures to bioavailable elements such as Cd and Zn. Plant uptake of Pb

is small (e.g. Section 4.3), but low accumulating root and tuber vegetables may be more likely to contain adhered soil Pb due to their proximity to the soil (differences in produce preparation are not currently accounted for by the BN).

Soil management:

Using raised beds or adding compost is likely to dilute the concentrations of HMs present in the soil and reduce exposure (e.g. Mitchell *et al.* 2014). The influence of reducing the HM concentrations in the allotments studied by approximately half was estimated to reduce health risks using the BN (Figure 7.3). Other soil management techniques such as liming to raise the soil pH and increasing the amount of SOM, had a small effect on the overall risk according to the model. Gardens are generally maintained to have a high proportion of SOM in the soil and a neutral pH (Alloway 2004) so increasing the amount of SOM and soil pH is likely to have a limited effect. Raised beds may become re-contaminated with dust from surrounding soils over time and require continued maintenance to reduce exposure in the long-term (Clark *et al.* 2008).

Personal hygiene:

The effect of personal hygiene measures such as wearing gloves, washing hands and produce, was determined by studying the effect of selecting a 'medium' soil ingestion rate in BN. Reducing soil ingestion through personal hygiene may possibly have a profound effect on the overall

exposure, especially to HMs such as Pb for which soil ingestion is a more important exposure pathway than produce consumption (Clark *et al.* 2008; DEFRA 2013a).

7.2.3 Educating gardeners from the present study

Whilst many gardeners in this study did report washing and peeling produce prior to consumption, there were some types of produce (especially berries) which were not uncommonly consumed raw prior to washing (Section 5.3.4). It is the intention of the author, following the conclusion of this project, to discuss and further educate the gardeners from the study cohort on methods to reduce their personal exposure to contaminants in the soils of their gardens. Future work should also research the influence of personal hygiene measures on reducing exposure to HMs.

7.3 UNCERTAINTY IN RISK ASSESSMENT MODELS

A crucial issue for risk assessment for scenario's involving gardening, remains dealing with uncertainty in model predictions. In this thesis, it was demonstrated that predictions by plant uptake models can vary by up to three orders of magnitude (Section 3.3). Whilst there have been improvements in recent years, large uncertainty still remains in the exposure assessment and toxicity assessment components of risk assessment models (Swartjes and Cornelis 2011). Furthermore, it is

important to consider how this uncertainty is calculated and represented in model outputs.

7.3.1 Produce consumption

Modelling exposure through produce consumption is likely to pose one of the greatest challenges to be able to accurately predict gardeners' exposure to HMs. This thesis has provided both improved estimates of plant uptake through the development of Freundlich regression models (Section 3.3) and improved estimates of produce consumption, by estimating the latter directly from the responses of gardeners (Section 5.3.4). Further research is still required to reduce uncertainty in this area. Produce consumption estimates provided in this thesis should be validated by using more robust methods of dietary measurement. Methods such as meal duplication appear to be less biased than other dietary recall methods, although are better suited to predicting short-term consumption patterns (Lee-Han *et al.* 1989). Future work is also required to develop more reliable models of plant uptake and especially models which consider foliar uptake – a contributor to Pb concentrations in internal plant tissues (Schreck, Bonnard, *et al.* 2012; section 3.3.4).

7.3.2 Toxicity assessment

A major limitation to the development of a health benefits and risks model for gardeners in this thesis, was the scarcity of the knowledge of chemical toxicity beyond well- established toxicological reference values

(e.g. RfDs). The significance of exceeding a reference value is specific to an individual case, and will depend upon the incline of the dose-response curve beyond the reference value (Swartjes and Cornelis 2011). Physiologically-based pharmacokinetic (PBPK) models which can be used for this purpose, have been developed for some chemicals, but more research needs to be done in order to fully understand the fate and transport of these chemicals in the human body and to adapt risk assessment models accordingly (Swartjes and Cornelis 2011).

7.3.3 Propagation of uncertainty

Representing uncertainty also remains an important challenge in risk assessment. As demonstrated by the BN presented in chapter 6, it is not adequate to consider uncertainty in risk assessments arising from just one parameter, but also how uncertainty in this parameter will interact with other parameters and components throughout the model. Whilst still early in development for risk assessment, BNs allow the propagation of uncertainty to be visually represented throughout the model and can be easily understood by stakeholders. The variation discovered between existing exposure models (Swartjes 2007), demonstrates the uncertainty arising from model predictions as a result of differences in the default input parameters and the model designs. Whilst the outputs from probabilistic models can be confusing (Lindley 2001), the uncertainty in any model prediction should be represented, even if this is only alongside a deterministic outcome so that good risk management decisions can be made.

7.4 CONCLUDING REMARKS

From the work achieved in this thesis and previously, it can be concluded that gardening is a very beneficial activity. 'Grow-Your-Own' activities help individuals from a wide range of backgrounds and ages to live a healthy lifestyle, supported by access to fresh fruit and vegetables. Community gardens and allotments also provide opportunities for different community groups to meet, and therefore they also have a wider societal benefit. In urban areas, garden spaces provide corridors for wildlife and a much needed break from the surrounding metropolis. Well-managed gardens also reduce health risks in the surrounding area from contaminants, as garden soils are typically fertile and contain lots of organic matter (Brown *et al.* 2016). With society becoming increasingly urban, gardening spaces in the city deserve more protection from sources of contamination arising from local industry and vehicular emissions, and more attention in terms of resources and funding from local authorities.

This study highlights the need to consider the benefits of gardening alongside health risks in risk assessment, and in the management and future use of urban gardens. Removing access to urban gardens which are slightly or moderately contaminated, may reduce health risks from exposure to contaminants in the soil, but may have a net negative effect on human health and well-being. Future research is needed to further quantify the effect of urban gardening on disease and to develop the model framework presented here for risk assessment of urban gardens.

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9 APPENDICES

Due to the size and format of some of the files included in the appendices, it was decided to include these separately in a USB attached to this thesis. These are summarized below:

Appendix I. **Survey communications and ethical approval**

- Contains a copy of the ethical approval form and supporting documents which explain how contact was made with plot holders, the NCC and allotment organisations.

Appendix II. **Questionnaire material**

- Contains a copy of the gardening and health questionnaire relevant to chapter 5, and the soil amendment questions, relevant to chapter 2.

Appendix III. **Plant uptake model database.**

- Contains a copy, including references, of the Hough and Nguyen databases in a Microsoft access database.

Appendix IV. **Soil sampling data from allotment survey**

- Contains an anonymised copy of the data acquired from soil sampling of allotments. This data is presented in the format that was passed on to the NCC according to requirements for ethical approval.

Appendix V. **Blocking structure for beans and lettuces**

- Contains a copy of the blocking structure developed for the pot experiment presented in chapter 4.