Environment International 122 (2019) 130–141

Contents lists available at ScienceDirect

Environment International

journal homepage: www.elsevier.com/locate/envint

An apple a day? Assessing gardeners' lead exposure in urban agriculture sites to improve the derivation of soil assessment criteria

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ARTICLE INFO

Handling Editor: Olga Kalantzi

Keywords:
Urban soil
Urban agriculture sites
Lead
Human health risk assessment
Blood lead
Crop lead

ABSTRACT

Globally, many of our urban agriculture sites (UAS) contain high levels of lead (Pb), a contaminant of toxicological concern to humans. To improve the derivation of soil assessment criteria at UAS, and avoid inappropriate closure of these valuable community spaces, we sampled nearly 280 paired soil and crop samples across 31 UAS gardens. This sampling was coupled with an exposure and food frequency questionnaire and participants blood Pb levels (BLL), (43 gardeners and 29 non-gardening neighbours). In 98% of the sampled soils, Pb concentrations were above the current UK soil guideline for UAS (80 mg/kg), however despite the high soil Pb (geometric mean: 324 mg/kg), and high soil bioaccessible Pb (geometric mean: 58.7%), all participants BLL were < 4.1 μg/dL (range: 0.6–4.1 μg/dL). Indeed, there was no statistically significant difference between the BLL of the UAS gardeners and those of their non-gardening neighbours ($p = 0.569$).

Pb uptake, however, varied with crop type and our study highlights the suitability of certain crops for growing at UAS with elevated Pb (e.g. tubers, shrub and tree fruit), whilst limiting the consumption of others (selected root vegetables, such as rhubarb, beetroot, parsnips and carrots, with observed Pb concentrations > 0.1 mg/kg FW).

The importance of defining the exposure scenario of a specific sub-population (i.e. UAS gardeners) is highlighted. Our preferred models predict site specific assessment criteria (SSAC) of 722–1634 mg/kg. We found fruit and vegetable consumption rates by all participants, and not just the UAS gardeners, to be considerably higher than those currently used to derive the UK's category 4 screening levels (C4SLs). Furthermore, the soil to plant concentration factors (SPCFs) used to derive the UAS C4SL significantly over predict Pb uptake. Our study indicates it may be appropriate to develop a distinct exposure dataset for UAS. In particular we recommend the derivation of SPCFs that are reflective of urban soils, both in terms of the range of soil Pb concentrations typically observed, but also the sources (and hence human oral bioaccessibility and plant-availability) of this Pb.

1. Introduction

Increasing urbanisation, growing concerns over food security and a greater attention on healthy eating, healthy lifestyles and green infrastructure in cities, have all led to a greater focus on our urban soils. Not only does urban agriculture provide ready access to affordable fresh crops, but community gardening is seen as a health promoting activity (Leake et al., 2009; Van Den Berg and Custers, 2011; Alaimo et al., 2008; Litt et al., 2011), as well as providing opportunities for outdoor learning, community development and improved social cohesion (Wakefield et al., 2007; Armstrong, 2000).

Urban agriculture sites (UAS) and community gardens, known as allotments in the UK, are frequently reported with high concentrations of a range of potentially harmful elements (e.g. Clark et al., 2006; Mitchell et al., 2014; Rouillon et al., 2017). Legacy lead (Pb) is one of the most common contaminants in our urban soils with Pb-based paints,

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https://doi.org/10.1016/j.envint.2018.10.054
Received 21 June 2018; Received in revised form 10 October 2018; Accepted 26 October 2018
Available online 16 November 2018
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automotive Pb emissions, ashes from residential fires and industrial (often aerosol) emissions the main reported sources (Sterrett et al., 1996; BGS London Earth). Acute and chronic exposure to Pb can result in a range of systemic effects in humans including, immunologic, reproductive, cardiovascular, renal, neurologic, and developmental effects (ATSDR, 2007; PHE, 2017). Children are most at risk and even low levels of environmental Pb exposure have been linked with a range of behavioural and cognition deficits (Aguiar et al., 2010; Chandramouli et al., 2009; Grandjean, 2010; Lanphear et al., 2005). Low levels of environmental Pb exposure are also of concern in adults, particularly pregnant and lactating mothers as Pb stored in bone can be mobilized during pregnancy (Gomaa et al., 2002), and the placenta and mammary glands allow for the transfer of Pb between the mother and the baby or foetus (Filippelli and Laidlaw, 2010). Studies suggest environmental Pb exposure in adults may contribute to chronic kidney disease (Ekong et al., 2006; Navas-Acien et al., 2009), hypertension (the most sensitive effect in adults; Navas-Acien et al., 2007; Vupputuri et al., 2003), cardiovascular disease (Lanphear et al., 2018) and even spontaneous abortion in females (Gidlow, 2015).

Growing food in contaminated urban areas has the potential to increase our exposure to Pb. The transfer of Pb to humans can occur via both direct exposure to soil (e.g. soil and dust ingestion via outdoor and indoor exposure, respectively; inhalation of particles containing Pb) and indirect exposure pathways (e.g. consumption of food containing Pb and Pb-contaminated water). The relative importance of each pathway to overall risk will vary depending on the contaminant, the human receptor and the land-use. Understanding the relative importance of different exposure pathways at UAS requires information across a range of variables, including homegrown crop consumption rates, Pb concentration in the crops, as well as knowledge of behavioural aspects of the gardeners. For our study, this necessitated a community-research partnership involving the UAS gardeners, the local city council, and researchers across a range of academic and government-funded agencies. A steering group was established and included representation from Northumbria and Newcastle Universities, the Allotments Working Group, UK Health and Safety Executive, Environment Agency, Food Standards Agency, WCA Environment and a number of independent experts.

A key exposure pathway for urban gardeners is via the ingestion of homegrown crops; both through direct plant contaminant uptake, but also soil remaining attached to crops at the point of ingestion. Typically, the inhaled fraction is only a relatively minor exposure pathway for metals such as Pb in urban agriculture (Hough et al., 2004). As such, we did not include specific modelling of the inhalation pathway in this study.

To pose a human health risk, a fraction of the ingested (or inhaled) contaminant must be bioavailable (i.e. available for absorption into the systemic circulation). Soil oral bioaccessibility refers to the portion of total soil contaminant that may be extracted using an in vitro protocol, essentially as a surrogate measure of human bioavailability. In vitro bioaccessibility studies, that quantify the fraction of a contaminant in soil released during passage through the gastro-intestinal tract, have been widely reported in the literature (e.g. Boisa et al., 2013; Denys et al., 2012; Palmer et al., 2015; Pelfrene et al., 2015). Given growing usage and acceptance of soil oral bioaccessibility testing as part human health risk assessments, we included such testing as part of our Newcastle Allotments Biomonitoring Study (NABS).

An important element of the project was to provide detailed information to support UK regulators who must decide if sites are suitable for urban agriculture, and thus the use of the Contaminated Land Exposure Assessment (CLEA) model was deemed the most appropriate model in this context as it is in common use (EA, 2009a). The current UK soil screening values (known as category 4 screening levels; C4SLs) are used to screen out low risk sites. No national study has specifically targeted UAS gardeners and the UAS C4SL is based on a range of national data sets (e.g. for crop consumption rates and % homegrown fraction in the diet), other generic assumptions (e.g. data for soil to plant correction factors, SPCFs), as well as rather dated UAS specific data on frequency and duration of visits. Results of sensitivity analyses using the CLEA model have highlighted that SPCFs and consumption of homegrown crops are key parameters/assumptions that cause uncertainty in the derivation of soil assessment criteria at UAS (CL:AIRE consortium, 2014a). By better constraining a range of selected input parameters our study aims to give greater confidence to regulators who must decide if sites are suitable for use as UAS by adult gardeners. Our specific objectives were thus to (i) assess the uptake of soil Pb across a range of typical UAS crops, (ii) to investigate the total and oral bioaccessible Pb in the soil, (iii) to estimate the gardeners' indirect Pb exposure via homegrown fruit and vegetable consumption patterns, and (iv) characterise the risks associated with UAS gardening through modelling. For a number of pragmatic reasons, our study focussed on adult gardeners: few children regularly visit two of the three UAS investigated as part of this study and ethical considerations led to a focus on sampling adult blood Pb. However, we acknowledge that children may still be exposed via the consumption of homegrown crops and also by contamination of house dust; both form the basis of current on-going work. Improving our understanding of the relative importance of specific exposure pathways for UAS gardeners will also allow regulators, the gardeners themselves, site owners and developers to better target advice on minimising exposure to Pb.

2. Materials and methods

2.1. Study sites and sampling

Three UAS were selected, all located in Newcastle-upon-Tyne, in the county of Tyne and Wear. Newcastle is the regional capital of NE England (population 280,200; ONS, 2016) with a history of coal mining and heavy industry, including the Pb industry. Lead in the topsoils of the county typically fall within the upper 25th percentile of concentrations in English soils (i.e. > 99.5 mg/kg Pb, DEFRA, 2012; Rimmer et al., 2006) and previous sampling by the city council had identified each of the three selected UAS has having raised Pb levels (Bramwell et al., 2008). Whilst none of the sampled sites are on formerly industrial land, each has a long history of additions of coal ash from domestic hearths being added to the soil to improve the drainage of naturally clay-rich soils in the region. In addition, other known sources of Pb include the use, and occasional burning, of Pb-painted wood on the sites, as well as legacy atmospheric sources from the burning of fossil fuels. From each individual UAS garden plot, three different crop types were collected, in duplicate or triplicate, in addition to the soil from around the plant roots. Nearly 280 paired soil and crop samples were collected across 31 UAS garden plots, with an exposure and food frequency questionnaire going to each UAS garden plot holder and a non-gardening neighbour (control participant), and selected home sampling (tap-water, indoor and exterior dust, although these data are not reported here). In addition, 43 of the gardeners and 29 of their non-gardening neighbours had a venous blood sample collected on-site by a state registered nurse, and sent for analysis to the UK Health and Safety Executive laboratory in Buxton, a UKAS accredited laboratory for blood analysis (for further details of sampling and analysis method see Bramwell et al., 2018). The study was approved by the Newcastle University Ethics Committee; study number 00804. Participants provided informed consent prior to participation.

2.2. Soil analyses

The pH, organic matter content and pseudo-total Pb content of the soil were determined by Derwentside Environmental Testing Services (DETS), Consett, UK (an MCERTS accredited laboratory for these analyses). Soil samples were dried at < 50 °C for a minimum of 16 h. A subsample of each dried soil was then ground using a mechanical mixer mill and sieved through a 250 μm sieve to ensure homogeneity. All sieves comply with BS 410–1 and calibration checks were undertaken using 250 μm NIST Traceable Sieve Calibration Standard. The pH value of a soil-deionized water suspension was determined electrometrically using a glass electrode (in-house method No. DETCS 2008). The organic matter content of the soils was determined using in-house method No. DETSC 2002, based upon the Walkley and Black (1934) method. The soils were analysed for their pseudo-total Pb concentration by aqua-regia hot-block digestion and ICP-OES, using in-house method No. DETSC 2301, based on their UKAS accredited Standing Committee of Analysts (2006) method. A range of additional elements were also determined and selected characteristics of the soils at the three UAS are presented in Supplementary information (SI) Table SI_1. Method limits of detection (LOD, calculated as 3 times the SD measured in a set of preparation blanks handled as samples) are also given in Table SI_1.

In order to evaluate the oral bioaccessibility of the Pb following soil ingestion, the bioaccessible fraction was determined using the Unified BARGE Method (UBM), developed and adopted by the Bioaccessibility Research Group of Europe (BARGE; Wragg et al., 2009). As the < 250 μm particle size fraction is more likely to adhere to hands or food produce and be transferred by ingestion through hand-to-mouth contact (Duggan et al., 1985) a fraction of the dried soil samples was gently disaggregated with a mortar and pestle before sieving though a < 2 mm nylon sieve. The < 2 mm fraction was then screened through a < 250 μm sieve to produce the smaller particle size fraction used for bioaccessibility testing. The UBM is an in vitro method for simulating the human digestive system and uses synthetic digestive fluids, 0.6 g of the < 250 μm soil fraction, and a gastric pH of 1.2 for 1 h at 37 °C. Information on the preparation of the simulated saliva fluid/gastric fluid, and subsequent procedure are detailed in Wragg et al. (2009). Selected soil samples were prepared and analysed in duplicate, with procedural blanks and BGS guidance reference material (BGS 102, British Geological Survey, Keyworth, UK) included for quality assurance. Analyses were undertaken on a ICP-OES (PerkinElmer Optima-8000, Beaconsfield, UK). The instrument was calibrated with standard solutions, and to counter matrix interferences, an internal standard (scandium) was added to all samples and calibration standards (with a recovery of 80–120%). Our bioaccessibility protocol produced a method LOD of 0.01 mg/l Pb.

2.3. Plant analyses

Plant samples were washed and prepared as though for eating and then frozen. Whilst we acknowledge that there is variation between individuals' preparation of crops for eating, for the purpose of this study crop types that are commonly peeled were peeled prior the freezing. The determined Pb concentration thus refers to Pb in the typical edible portion of the plant. Sample preparation techniques have been shown to influence the Pb concentration, especially where specific surfactants have been used in the laboratory that may remove surface entrained soil particles (e.g. Attanayake et al., 2014; Defoe et al., 2014). As we were keen to mirror actual intake by the consumer we used typical kitchen-style cleaning, peeling and chopping equipment, although we were careful to avoid cross-contamination. We used stainless steel blades where relevant, and Milli-Q water (Millipore, Bedford, MA, USA) for the final rinsing.

A sub-set of crop samples (n = 147) was selected for analysis. Selection reflected representation across the six crop categories identified in the UK CLEA model (green, root and tuber vegetables, and herbaceous, shrub and tree fruit; EA, 2009b), allowing also for duplicate and triplicate samples sampled from two to three different adjacent plants, respectively. The frozen crop samples were chopped/sliced using a stainless steel knife and packed in polyethylene bags then dry-ice for shipping to ALS Global laboratories in Sweden.

Upon arrival at the laboratory, samples were weighed into 50-ml polyethylene vessels and Suprapur grade nitric acid (HNO_3, Sigma-Aldrich Chemie Gmbh, Munich, Germany) was added at ratio of 10 ml acid per 1 g of sample. Vessels were capped and placed in a heated (120 °C) graphite hot-block digestion system for 2 h. After cooling to room temperature, transparent digests were diluted to 50 ml by addition of de-ionized Milli-Q water. All laboratory ware in contact with the samples or sample digests was soaked in 0.7 M HNO_3 (> 24 h at room temperature) and rinsed with Milli-Q water prior to use. All sample manipulations were performed in clean laboratory areas (Class 10,000) by personnel wearing clean room gear and following all general precautions to reduce contamination (Rodushkin et al., 2010).

Lead concentration was determined using an ELEMENT XR (Thermo Scientific) double-focusing sector field ICP-MS instrument using a combination of internal standardization (indium added to all measurement solutions) and external calibration. Details of the operating conditions and measurement parameters can be found elsewhere (Rodushkin et al., 2008). Method LOD was 0.0004 Pb mg/kg fresh weight (FW). Selected plant samples were analysed in duplicate and a set of certified reference materials (CRM 1549 Non-Fat Milk Powder, CRM 1567a Wheat Flour, CRM 1547 Peach leaves, all from National Institute of Standards and Technology, Gaithersburg, MD, USA) was included as quality assurance/quality control in every preparation batch.

2.4. Questionnaire data and exposure modelling

NABS participants were asked to provide a range of personal and behavioural information. This included details of time spent at the UAS and a food frequency questionnaire to allow fruit and vegetable consumption patterns to be assessed, along with the proportion of home-grown crops in the diet. Consumption rates were calculated as follows:

$$\text{Consumption Rate (g food weight per kg body weight per day)} = (\text{Median portion size} \times \text{daily portions of individual foods}) / \text{average UK body weight}$$

Average adult portion sizes were obtained from various sources and average crop weight per week consumed (g) calculated (see Table SI_2). Average UK adult (male and female) body weight (bw) was obtained from the UK Office for National Statistics (ONS, 2009). The same food groups were then grouped together to produce consumption rates for each crop group.

The homegrown fraction (HF) is the fraction of consumed crop that is grown in the UAS. We used the HF information from our food frequency questionnaire to determine the 50th percentile (P50) and the 90th percentile (P90) HF for each individual fruit or vegetable for all participants, then gardeners and controls separately. To estimate the HF for each crop group, first we calculated the weight of each crop group consumed by each participant (the median portion size was used here, based on a variety of information sources; see Table S1_2), then the weight of homegrown crops consumed for each participant, and then finally we calculated the HF fraction (by weight) based on these two datasets.

When considering the most appropriate exposure assessment model for modelling gardeners blood Pb level (BLL), the authors note that whilst the IEUBK model is perhaps the most widely used, the model is for children and not adults (USEPA, 2007). As an important element of the NABS project was to provide detailed information to support UK regulators, the CLEA model (software version 1.071; EA, 2009a) was deemed the most appropriate model in this context. CLEA is a deterministic model where the conceptual exposure model is represented using a series of equations and associated input parameter values (EA, 2009a). CLEA allows almost all of the parameter values to be adjusted by the user and so the results of the NABS study can be input directly to generate site specific soil assessment criteria (SSAC) for adult UAS gardeners.

2.5. Statistical analyses

Summary statistics describing selected soil properties, trace and major elemental concentrations were prepared and presented in Table SI_1. Where datasets were larger than n = 5 we used the geometric mean (where the product, instead of the sum, of values is used and the nth root obtained) to describe the central tendency rather than the arithmetic mean. The geometric mean is typically the preferred method where datasets are skewed. A range of non-parametric statistical techniques were employed to further support data analysis. Spearman's rank correlation coefficients were calculated to investigate the relationships between selected soil properties and crop Pb concentration. Mann-Whitney and Kruskal-Wallis tests were used to compare the difference in crop consumption rates between our UAS gardeners and their non-gardening neighbours (our control participants), and crop Pb concentrations between the different crop groups and different crop types, respectively. All non-parametric statistical testing was undertaken using Minitab® 18 software (Minitab Ltd., Coventry, UK).

3. Results and discussion

Each of the empirically-derived input parameters from our study are systematically presented and discussed. The data are then modelled, with implications for UAS gardening discussed.

3.1. Selected soil properties and crop Pb data

The geometric mean pH of the soils across the three sites was neutral (pH 7.1 ± 0.37, pH range 5.7–8.2), whilst the soil organic matter content indicated percentages ranging from 5 to 25%, with a geometric mean of 17% ± 4.64%. The geometric mean pseudo-total Pb concentration in the soil was 324 mg/kg ± 156 mg/kg, calculated following the removal of one extreme outlier at 6700 mg/kg (Fig. 1). Ninety-eight % of the soil samples were over the UK UAS C4SL of 80 mg/kg (used to screen out low risk sites; DEFRA, 2014).

No association was observed between crop Pb and soil Pb concentrations (Fig. 2, Table SI_3, p = 0.085), even after stratifying by crop group (e.g. Fig. SI_1 A and B), and by crop plant type (e.g. Fig. SI_1 C and D). Such an observation has been reported at other UAS (Attanayake et al., 2014; Defoe et al., 2014; Spliethoff et al., 2014). This lack of correlation is not totally unexpected due to the range of confounding variables influencing this relationship and our study

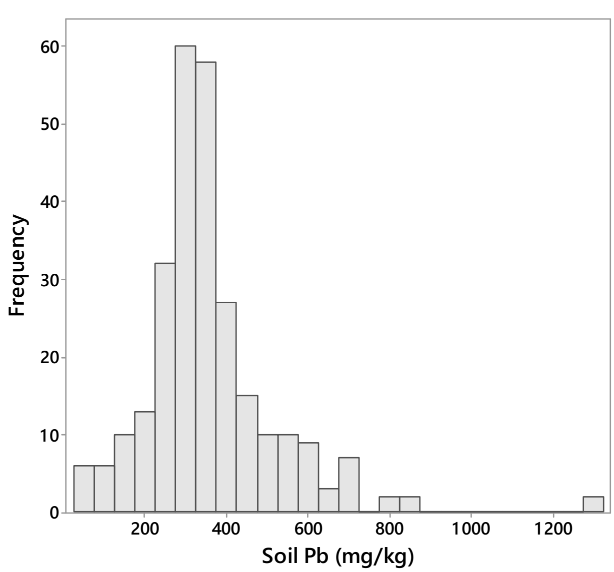

Fig. 1. Pseudo-total Pb concentration in the soils (mg/kg, n = 279), excluding one outlier (6300 mg/kg).

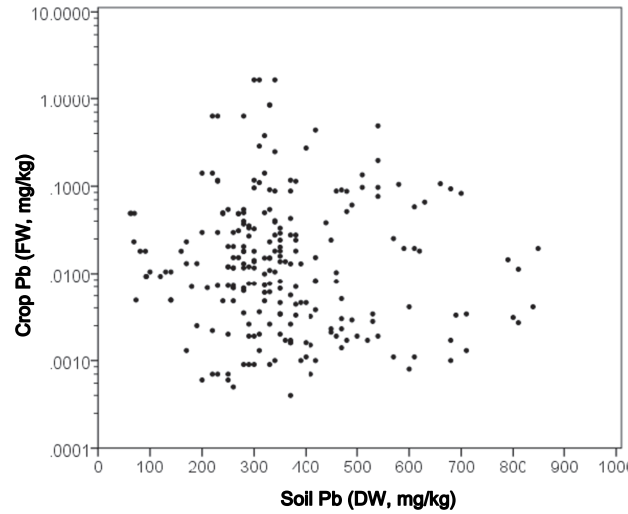

Fig. 2. Association between soil Pb and crop Pb concentrations (mg/kg). (Spearman's $r = -0.112$, p = 0.085). Note: two soil outliers have been removed for presentation purposes (one at 1200 mg/kg and one at 6300 mg/kg).

highlights the need to be critical of the use of total soil Pb to predict crop Pb. Metal accumulation in plants is influenced by a range of factors, such as the mobility of the metal in the plant phloem and/or xylem, the plant genotype, and a range of soil physico-chemical conditions that influence the bioavailability of the metal in soil (e.g. soil pH, texture, organic matter content, and in the case of Pb, the available phosphorus concentration), (Alexander et al., 2006; Brown et al., 2015; Ma et al., 2007; McBride et al., 2013; Reid et al., 2003). In addition, crops can become contaminated with Pb through aerial deposition/rainsplash onto the above ground parts of the plant, which then becomes entrained into the plant as it grows, or embedded in the plant-surface cuticle (Attanayake et al., 2014). Indeed, for many urban garden soils atmospheric deposition of Pb onto plants is thought to be of equal if not greater importance to root uptake of Pb (Alexander et al., 2006). This may be especially relevant in relation to above ground crops and their growth stage at collection. Literature on the contribution of aerial Pb deposition to the overall Pb contamination in crops is limited, although aerial sources of Pb have been shown to increase plant Pb concentrations (Schreck et al., 2014; Voutsa et al., 1996). In our study, we were unable to differentiate between Pb present in soil particles adhering to the crops and that taken up into in the plant, although all crops were washed and prepared as for eating using a standard kitchen approach (i.e. carrots peeled; main crop potatoes peeled; 'new'-type potatoes scrubbed).

To explore differences in the Pb concentrations between the different crop groups (Fig. 3; Table SI_4) we employed the Kruskal-Wallis test. The test determines whether statistically significant differences occur between the median Pb concentration of each crop group. The output (p value 0.001; Table SI_5 A and B) indicated that the median Pb concentration differs significantly for at least one of the crop groups. As the chi-square approximation may not be accurate in groups with less than n = 4 (Minitab® 18), the Kruskal-Wallis test was re-run again, but this time without the shrub, tree fruit and tuber groups. The resultant p-value (p = 0.007; Table SI_5 C and D) again indicated that at least one of the crop groups had a statistically different median Pb concentration. We concluded that the root crops had a significantly higher median Pb concentration compared to those of the green vegetables and the herbaceous fruit. The differences in crop Pb concentrations was further explored using the Mann-Whitney test. Statistically significant differences were observed between the median Pb concentration of the root crops and each of tree fruit, herbaceous fruit, tubers and green vegetables (Table SI_6). Only the shrub fruit failed to show a significantly different median Pb concentration to root crops. In addition, the green

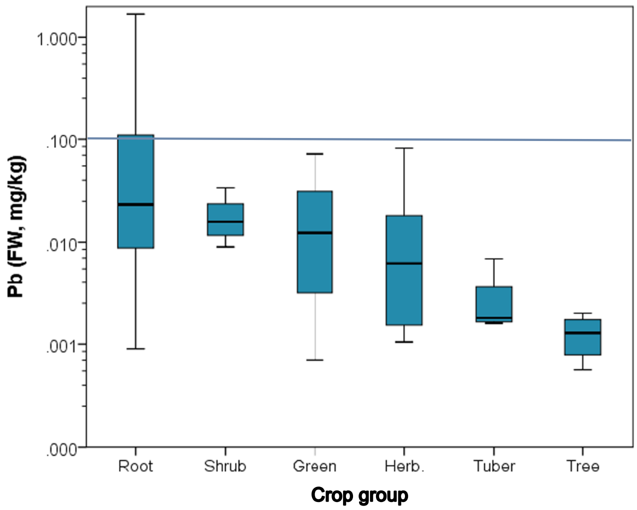

Fig. 3. Plant Pb concentration (FW, mg/kg) according to crop group; includes a 0.1 mg/kg reference line (the WHO/FAO limit in certain foodstuffs, including root vegetables). The solid black bar within each box is the arithmetic mean.

vegetables also indicated significantly different median Pb concentrations to tubers and tree fruit, and between tree fruit and shrub fruit. Indeed, as highlighted in Fig. 3, tree fruit and tubers (i.e. potatoes) tended to have the lowest Pb concentrations, with the highest Pb concentrations in the root vegetables (e.g. beetroot, carrots, parsnips, and rhubarb - although classed as a 'stalk or stem vegetable' by the FAO/WHO-CODEX (2010), rhubarb is grouped with the root vegetables in CLEA, EA, 2009a). These findings accord well with the observations of others across a range of different Pb sources (Chaney et al., 2010; Codling et al., 2016; Zhuang et al., 2009). Tubers are typically reported to take up very little Pb, even in Pb rich soils, whilst root vegetables, and especially those with expanded hypocotyls (e.g. carrot, beetroot, radish, turnips), have been reported to consistently show higher Pb concentrations than other crop groups (e.g. Codling et al., 2015; Defoe et al., 2014; Finster et al., 2004). In potatoes, unlike root vegetables with expanded hypocotyls where xylem runs through the edible part of the plant, the xylem connections between basal roots and tubers are thought to be non-functional (Reid et al., 2003). Chaney et al. (2010) used X-ray absorption fine structure to show how the Pb in carrots is largely associated with the xylem. Potatoes (tubers) are essentially 'phloem-fed'. For tubers, as with fruits, with low rates of transpiration, the flow through the xylem is reduced and so less residual Pb is accumulated (Reid et al., 2003). Furthermore, studies investigating the Pb concentration of various parts of the plant suggest that Pb is largely retained within the root, with limited translocation to plant shoots and fruits (Finster et al., 2004). This is clearly of significance to root crops, where higher Pb concentrations might be anticipated, and corroborated by our statistical testing indicating the root crops to have significantly higher median Pb concentrations compared to the majority of other crop groups. Pb levels for a number of the root crops exceeded the FAO/WHO-CODEX permissible levels in food guidelines; the rhubarb geometric mean Pb at 0.173 mg/kg (n = 7) was nearly 2× over the permissible level (Fig. SI_2). Whilst the Pb concentration in the rhubarb samples is elevated, this needs to be contextualised by the intake (consumption) rate which is limited with on average 1.4 portions eaten per week over the growing season (135 g mean weight consumed per week, Table SI_2). Root crops, with expanded hypocotyls like carrots, have also been shown to accumulate more Pb in the inner xylem tissues rather than the outer peel (Codling et al., 2015) and so differences in exposure can also result from differences in vegetable preparation behaviours such as peeling/not peeling. Of the potato samples analysed (n = 6), the variety with the skin typically left on, and so just well-scrubbed in terms of our preparation, did have a higher Pb concentration (arithmetic mean of 0.0108 mg/kg, n = 3) compared to the peeled varieties (arithmetic mean of 0.0017 mg/kg, n = 3), although we had insufficient potato samples to statistically test this observation.

For the majority of crops analysed, the concentrations of Pb were found to be within the permissible levels in food set by the FAO/WHO guidelines (FAO/WHO-CODEX 1995, 2010 amendment; 0.3 mg/kg FW for green vegetables and 0.1 mg/kg for all other fruits and vegetables), Table SI_4 and Fig. SI_2. Indeed, one of the reasons for the generally low uptake of Pb could be the moderately high soil pH, typical of many urban soils (i.e. geometric mean pH 7.1, Table SI_1). Statistical exploration of the differences between individual crop types within a crop group was also undertaken. In practice, this exploration was restricted to onions, rhubarb, leeks and beetroot from the root crops, and beans and cabbage from the green vegetables as only these two groups had sufficient Pb concentration data available (i.e. > 4 samples) for more than one crop type. For the green vegetables, the Mann-Whitney test indicated that the median Pb concentration of the beans was significantly different to that of the cabbages ($p = 0.000$; Fig. SI_3A); the box and whisker plot highlighting cabbages had higher Pb concentration than the beans. Of the root crops, the Mann-Whitney test indicated that only the onions had a significantly different Pb concentration to the other crop types (Fig. SI_3B). This was corroborated by the Kruskal-Wallis test (p value = 0.008; Table SI_5E and F) which indicated the median onion Pb concentration was far lower than the overall median, with rhubarb the highest. The occurrence of statistically significant within and between crop group variations in Pb concentrations, with no to minimal association between crop Pb and soil Pb concentration, raises concerns about the uncritical application of SPCFs in exposure modelling. The presence of a relationship is implicit in the use of SPCFs to estimate the amount of metal taken up by plants relative to the concentration in the soil, and SPCFs are used in CLEA in common with other generic exposure models (EA, 2009a). Clearly further work is required to explore whether specific sub-groupings can be identified within the broader 6 crop groups, and if the use of such sub-groups would help reduce uncertainty in the application of SPCFs in exposure modelling.

3.1.1. Soil to plant concentration factors

As alluded to above, SPCFs are a key area of uncertainty in exposure modelling Pb at UAS (Augustsson et al., 2015; CL:AIRE consortium, 2014a). SPCFs are based on the Pb concentration (FW) in the edible portion of the plant, divided by the total Pb concentration in the soil (dry weight, DW). A high degree of variability in Pb SPCFs is reported in the literature, which typically range across several orders of magnitude for the same crop group (Table 1 and Table SI_7 for individual studies across the literature; see also Augustsson et al., 2015). This variability is highlighted, for example, in the NABS SPCF data for the different root vegetables (Fig. 4), which range from 8.85×10^{-5} to 5.31×10^{-3}. Given the range of SPCFs across our data, we have used geometric mean as an estimate of central tendency for each crop group. The CL:AIRE consortium (2014b) used empirically derived geometric mean SPCF to derive soil assessment criteria for DEFRA. These values have been incorporated as the default data within the CLEA model (and are given in Table 1). All of the NABS calculated SPCFs are an order of magnitude, or more, lower than the default data used by the CL:AIRE consortium (2014b) to derive the C4SLs. However, in comparison to the wider literature, our SPCFs for shrub and herbaceous fruit, green and root vegetables are in keeping with the reported ranges, with only our tubers and tree fruits an order of magnitude lower. The SPCF order in the default data within the CLEA model is as follows: Tuber > Green > Root > Herbaceous > Tree > Shrub. For NABS the SPCF order is: Root > Shrub > Herbaceous > Green > Tuber > Tree. This same pattern was also evidenced in the Pb concentration across the crop groups (Fig. 3). For the default data within the CLEA model, the highest geometric mean SPCF is for tubers, which is at odds with our

Table 1
Soil to plant concentration factors (SPCF) (plant FW mg kg/soil DW mg kg) used to derived the C4SLs in CLEA, empirically determined in NABS, and indicative global range observed at other urban allotment sites, residential gardens and pot experiments on non-artificially spiked soil.

Crop category	SPCF (geometric mean)		Indicative global range[a]
	C4SL derived data in CLEA[b]	NABS	
Shrub fruit	2.05×10^{-4}	5.34×10^{-5} (n = 4)	8.5×10^{-5}–1.1×10^{-4}
Green veg	4.19×10^{-3}	3.37×10^{-5} (n = 33)	7.3×10^{-6}–1.1×10^{-2}
Tuber	7.31×10^{-3}	4.88×10^{-6} (n = 4)	4.9×10^{-5}–6.8×10^{-3}
Root veg	4.02×10^{-3}	8.85×10^{-5} (n = 35)	8.2×10^{-6}–1.1×10^{-1}
Herbaceous fruit	7.49×10^{-4}	1.83×10^{-5} (n = 10)	3.5×10^{-5}–5×10^{-3}
Tree fruit	2.29×10^{-4}	3.72×10^{-6} (n = 4)	7.6×10^{-6}–1.2×10^{-4}

[a] Studies do not include artificially spiked soils or soils where the source of Pb is principally from water, effluent or sewage sludge; where reported as plant DW in the literature this has been converted to FW (FW corrected data supplied by Ian Martin (EA) pers. comm., 2017). Source references in Table SI_3.
[b] CL:AIRE consortium, 2014b.

findings and with the literature which typically reports limited Pb uptake by tubers in urban soils (Table SI_7).

SPCFs are influenced by a variety of physical, chemical and biological factors, such as bioavailability/speciation of the Pb in the soil, physico-chemical properties of the soil (such as pH, organic matter content and soil moisture content), geographical factors (such as slope, aspect), and a range of plant factors (e.g. plant nutrient status, plant growth stage, the effect of other element uptake and differences across plant families, species and even between different cultivars of the same species; (Alexander et al., 2006). In addition, the metal concentration in the edible fraction is also influenced by how the crops are prepared and cooked (e.g. Pelfrene et al., 2015). A key assumption underpinning the use of SPCFs is that there is a relationship between the soil Pb concentration and that determined in the plant. Pb uptake by plants is reported to be a passive process, and thus one might anticipate greater uptake in higher Pb soils (Kabata-Pendias, 2001). A positive relationship between soil Pb and plant Pb has been reported in a number of studies of urban soils (e.g. Demirezen and Aksoy, 2006; Finster et al., 2004; Jorhem et al., 2000) and on experimental plots (Samsoe-Peterson et al., 2002). Caution, however, needs to be applied about comparisons being drawn with studies where artificial spiking with metal salts has been used, rather than 'real' soils where soil ageing can lead to the metal becoming more tightly bound in the soil and thus less plant-available over time. A number of recent studies on urban soils and community gardens have reported a poor association between soil Pb and plant Pb (McBride et al., 2014; Spliethoff et al., 2016; Warming et al., 2015), in keeping with the weak correlations highlighted in our NABS data (Fig. 2, Fig. SI_1 and Table SI_31, indicating no statistically significant correlations between soil Pb and crop Pb)., McGrath and Zhao (2015), in their review of metal uptake by food crops in the UK concluded that the Pb concentrations of food plants cannot be predicted reliably from soil measurements. Whilst Augustsson et al. (2015), observed a decrease in SPCF with increasing levels of soil contamination, as plant concentrations did not increase to the same degree as the soil concentrations highlighting the importance of selecting appropriate SPCF for the level of soil contamination encountered. Clearly the relationship is complex, and even with site-specific determinations of SPCFs a high degree of uncertainty remains, especially where contamination levels cover several orders of magnitude and the risk assessor needs to be cognisant of this in their professional judgement of the sites risks.

3.2. Oral soil Pb bioaccessibility

Lead bioaccessibilities (n = 21) ranged from 32% - 76% (Fig. SI_4i), with bioaccessible concentrations ranging from 58 to 705 mg/kg (Fig. Si_4ii; Table SI_8). A large body of research has highlighted the dependence of Pb bioaccessibility on the mineral and chemical forms of Pb in the soil. Lead from gasoline, car-batteries and Pb-based paints (Pb-oxides and Pb-carbonates) are typically reported to have higher bioaccessibility in soils (Hunt, 2016; Walraven et al., 2015), whilst a lower bioaccessibility is reported in the literature attributed to certain primary and secondary mineral phases. Soils with higher organic matter

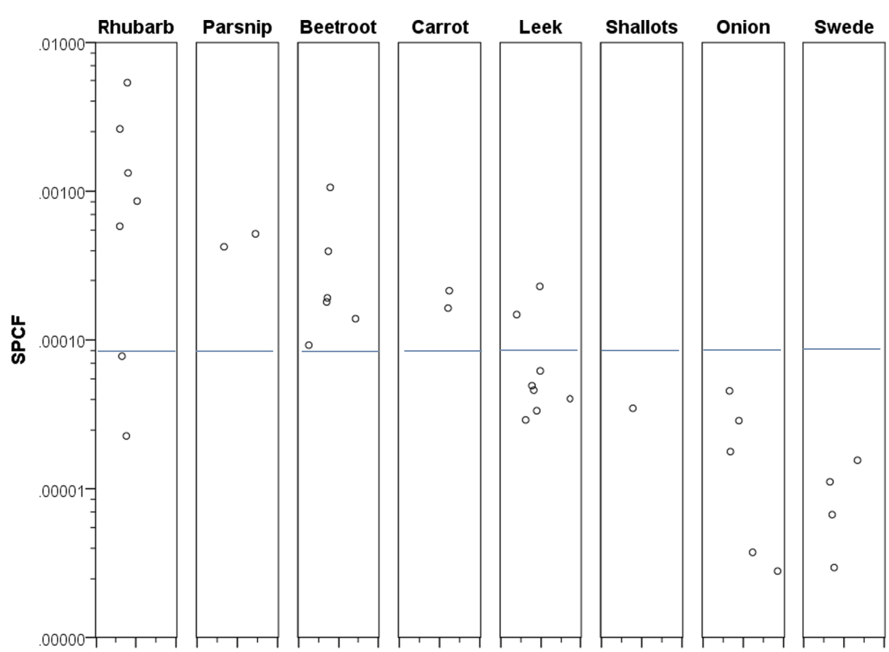

Fig. 4. Calculated soil to plant concentrations factors (SPCF) for each plant type analysed in the root vegetable group. The dashed line represents the root vegetable group geometric mean. The X axis is soil Pb concentration (0 to 800 mg/kg).

Table 2
CLEA defaults used to derive the urban agriculture site generic screening level (C4SL), and the NABS modelled parameters, used to derive the site specific assessment criteria (SSAC).

Parameter	CLEA default	Modelled scenarios (S)							
Receptor	Child (CLEA ages classes 1–6)	Female 16–65 (CLEA age class 17)							
Exposure factors									
Soil and dust ingestion	25–130 days/yr	258 days/yr[a]							
Consumption of homegrown produce	180–365 days/yr	365 days/yr							
Skin contact outdoor	25–130 days/yr	258 days/yr[a]							
Inhalation of dust and vapour outdoor	25–130 days/yr	258 days/yr[a]							
Occupancy period	3 h/day[a]	3 h/day[a]							
Soil to skin adherence factor	1 mg/cm/day	0.3 mg/cm/day[a]							
Soil and dust ingestion rate	0.1 g/day	0.05 g/day[a]							
Receptor									
Body weight	5.6–19.70 kg	70 kg[a]							
Body height	0.7–1.10 m	1.6 m[a]							
Inhalation rate	10.30–24.90 m^3/day	31.6 m^3/day[c]							
Max exposed skin fraction (outdoor)	0.26–0.26 m^2/m	0.27 m^2/m[a]							
Chemical/plant									
Relative bioavailability soil	0.6	0.6							
Adopted threshold daily intake value^ based on a lower limit of toxicological concern of 3.5 μg dL^{-1} blood lead level	1.4 μg kg bw^{-1} day^{-1} (IEUBK model, USEPA, 2007)	1.3 μg kg bw^{-1} day^{-1}[b] (Carlisle and Wade, 1992)							
		S1				S2			
Soil to plant concentration factor (SPCF) (Produce Pb FW/soil Pb DW)	C4SL data in CLEA (Table 1)	C4SL data in CLEA (Table 1)				NABS geometric mean SPCF data (Table 1)			
Consumption rate (CR) data	C4SL CR data in CLEA[e] (Table 3)	S1-A C4SL CR data[e]		S1-B NABS CR data[f]		S2-A C4SL CR data[e]		S2-B NABS CR data[f]	
Consumption rate (CR) approach	top two[e]	Top two[e]	Not top two[e]	Top two[f]	Not top two[f]	Top two[e]	Not top two[e]	Top two[f]	Not top two[f]
C4SL/SSAC: modelled with C4SL P90 HF[d] mg/kg	80 mg/kg	182	157	88	64	2877	2516	1949	1292
SSAC: modelled with NABS All P90 HF[d] mg/kg		55	51	31	26	1773	1609	1125	821
SSAC: modelled with NABS gardener P90 HF[d] mg/kg		49	46	27	23	1634	1454	1016	722

[a] EA (2009a, 2009b).
[b] As reported by CL:AIRE consortium (2014b).
[c] Data from Ian Martin (EA) pers. comm. 2017. Figure reflects updated inhalation dataset, recalculated to reflect the changes to short-term rates for UAS gardeners.
[d] HF (homegrown fraction) reported in Table 4.
[e] Default data in CLEA reported in Table 3.
[f] NABS data reported in Table 3.

are also reported to have reduced bioaccessible Pb where the formation of stable Pb-humic complexes are postulated (Cai et al., 2016). Indeed, in keeping with this literature, we observed a statistically significant negative correlation (Spearman's rho of −0.775) between bioaccessibility and organic matter content (Table SI_3).

Denys et al. (2012) reported UBM in vitro – in vivo comparisons and concluded UBM bioaccessibility data to be a reliable predictor of in vivo relative bioavailability (RBA) for Pb in soils contaminated by mining slags and fly-ash across a range of concentrations and bioaccessibilities (Denys et al., 2012). However, given the complexity of translating in vitro oral bioaccessibility to RBA for use in CLEA we considered the most pragmatic approach, at this time, was to utilise the current default RBA for Pb of 0.6 in our exposure modelling (Table 2). The CLEA default RBA of 0.6 is predicated on an assumption of a dietary bioaccessibility of 100% and 'typical' urban soil Pb bioaccessibilities of *c.* 60% (CL:AIRE consortium, 2014b). Given the median oral bioaccessibility at each of our 3 sites were 62.8%, 57.3% and 64.6%, respectively (Fig. SI_4i), we considered the current default RBA in CLEA to be suitably reflective of our UAS.

3.3. Questionnaire: food frequency questionnaire and time spent on the UAS

Whilst the major exposure pathway for Pb in a residential setting is through soil and dust ingestion, for urban gardeners exposure from food intake, and importantly the consumption of homegrown crops, is considered the dominant pathway (CL:AIRE consortium, 2014a). Both the consumption rate of fruit and vegetables, and the % of homegrown produce in the diet, are uncertainties identified in the sensitivity analyses of the CLEA model. Modelling Pb exposure via food intake at UAS is challenging. Not only do we need to model the consumption of a range of homegrown fruit and vegetables, but the plant Pb concentrations will vary according to soil properties, plant physiologic factors and other environmental conditions (such as seasonality, amount of plant biomass, atmospheric deposition in dry or windy conditions, rainsplash in wet conditions, and soil moisture content). Ideally, UAS gardeners need to be viewed as a subset of the general population (about which we have national survey data, such as that used to derive the C4SLs using the CLEA model), with their own set of behaviours. They are an important subset, but seldom studied in their own right.

Through our food frequency questionnaire we had sufficient dietary data to calculate consumption rates for each of the different crop groups, and in Table 3 we compare the NABS data (UAS gardeners, controls and all participants) with the data used to derive the C4SLs using the CLEA model. One of the recent modifications proposed and adopted in the derivation of the UK C4SL by DEFRA (CL:AIRE consortium, 2014a) is the use of central tendency values (50th percentile or P50) for fruit and vegetable consumption rates rather than 90th percentile values (P90); with the exception of the 'top two' crop groups expected to give the highest exposure for that contaminant (CL:AIRE consortium, 2014a). For Pb, the 'top two' crop groups are reported to be green vegetables and tubers. For these two crop groups the 90th percentile consumption rates are retained (Table 3). The default consumption rate data in the CLEA model are based on the National Diet and Nutrition Surveys (2008–2011) and reflect the behaviours of the general population (EA, 2002). Homegrowers are reported to be amongst the highest consumers of fruit and vegetables (CL:AIRE consortium, 2014b), and the NABS data supports this assertion for all but

Table 3
Vegetable and fruit consumption rates for each of the crop categories, for the Newcastle Allotments Biomonitoring Study (NABS; for gardeners, controls and all participants) and the C4SL derived data in CLEA for age class 17 (AC17; 16–65 years old). CLEA and NABS consumption rates used in the 'top 2' approach for modelling are given in bold.

NABS		Vegetable consumption rate (g fw kg^{-1} bw day^{-1})			Fruit consumption rate (g fw kg^{-1} bw day^{-1})		
		Green	Root	Tuber	Herb.	Shrub	Tree
All participants	P90	**4.82**	4.57	**3.64**	5.95	1.58	5.77
Gardeners only		4.98	4.94	4.07	6.33	1.60	5.56
Controls only		4.80	4.09	3.19	5.23	1.52	9.25
All participants	P50	2.19	**2.02**	2.10	**2.42**	**0.5**	**2.26**
Gardeners only		2.45	2.17	2.43	2.50	0.70	2.28
Controls only		1.95	1.62	1.69	1.97	0.33	2.21
C4SL derived data in CLEA (AC17)	P90	**2.36**	1.12	**2.35**	1.29	0.18	2.38
	P50	1.26	**0.6**	1.18	**0.69**	**0.09**	**1.27**

tree fruit (Table 3) however the difference in consumption rate between our UAS gardeners and our controls was not statistically significantly different (Mann-Whitney test: green vegetables $p = 0.6088$; herbaceous fruit $p = 0.1414$; root vegetables $p = 0.2287$; shrub fruit $p = 0.1303$; tree fruit $p = 0.5524$; tubers $p = 0.0864$). As such, we refer only to the NABS 'all participant' consumption rates in our subsequently modelling.

Our NABS 'all participant' P90 consumption rates are significantly greater than the P90 default data in the CLEA model (with green vegetables 2 times; root vegetables 4 times; tubers 1.5 times; herbaceous fruit 4.6 times; shrub fruit 8.7 times; and tree fruit 2.4 times greater, respectively), Table 3. Indeed, our NABS P50 'all participant' data is largely comparable to the P90 data in the CLEA model (Table 3). Not only do our NABS UAS gardeners consume more fruit and vegetables that the default general population data used to derive the C4SLs in CLEA, but so do the NABS control participants. As such, the current approach of using a combination of P50 and P90 data (but only for the 'top two') to derive the C4SL may not be sufficiently protective, especially of UAS gardeners. Indeed, over the last decade or so we have seen a growth in the promotion of healthy eating campaigns (e.g. Public Health England's Change4Life campaign), grown your own campaigns (including in-school activities) and the promotion of healthier lifestyles through, for example, 'be active' campaigns (e.g. the UK's NHS Fit4Life campaign). As such, it does not seem surprising that the more up-to-date NABS consumption rate estimates, both for the controls as well as the gardeners, are all higher that the rather dated 2008–2011 data on which the current C4SLs are derived.

As noted above, the HF is another key uncertainty identified by the sensitivity analyses in CLEA (CLAIRE consortium, 2014b). The CLEA default HF data is based on data drawn from the general population 2004/5 Expenditure & Food Survey (ONS, 2009). Our NABS 'all participant' P90 data shows a greater HF than the CLEA P90 HF, with the majority of NABS crop categories 1.5 to 2 times greater than in CLEA, but with tubers 7 times higher and tree fruit 0.7 times lower (Table 4). Given the small size of many of the UAS in our study (typically < 12.5 m × 5 m) very few plots had fruit trees, and this may explain our NABS underestimate of the HF compared to the CLEA default data.

In this context, it is interesting to note that the CL:AIRE team working on the C4SLs assumed that gardeners (with a focus here more on residential gardens rather than UAS) would not be able to grow more than two food types to the extent that they were P90 consumers (E. Stutt, WCA Environment, pers. comm. 2018). Indeed, our NABS data suggest that many UAS gardeners have both P90 consumption rates and high HFs across several crop groups and not just a 'top-two'. It may thus be appropriate to develop distinct exposure datasets for the two types of gardener; residential 'yards' and UAS.

Data, used in the CLEA model to derive the C4SL, for frequency and duration of time spent on the UAS is from Saunders (1993). Unfortunately, the categories used in the NABS questionnaire did not match exactly the CLEA categories. In addition we only gathered information on visits during the growing season (Spring–Autumn). There is however a good correspondence between the Saunders (1993) data and the obtained NABS visit frequency and time spent on the UAS (Table SI_9). The CLEA default data assumes a visit frequency (adult) of 285 days a year (daily visit in summer and 3 times a week in winter), and a visit duration of 3 h. Based on the available NABS data we have no reason to disagree with these assumptions.

Table 4
50th percentile (P50) and 90th percentile (P90) homegrown fraction for the C4SL derived data in CLEA and the Newcastle Allotments Biomonitoring Study (NABS).

		Homegrown fraction (%)							
		C4SL data in CLEA		NABS data					
				Gardener		Control		All	
		P50	P90	P50	P90	P50	P90	P50	P90
Vegetables	Green	5	33	35	69	0	3	16	59
	Root	6	40	35	55	0	10	13	49
	Tuber	2	13	30	100	0	1	0	92
Fruit	Herbaceous	6	40	21	73	0	7	7	63
	Shrub	9	60	54	99	0	12	0	94
	Tree	4	27	0	42	0	1	0	18

3.4. Exposure assessment modelling

For exposure modelling, the selection of an appropriate heath criteria value (HCV) is critical. A HCV is used to describe a level of exposure (i.e. the lower level of toxicological concern, tolerable daily intake or index dose) which is considered protective of human health. The CL:AIRE consortium (2014b) used the Carlisle and Wade method to estimate the daily Pb intake that would lead to a geometric mean blood lead concentration of 3.5 μg/dL, and derived an estimated intake of 1.3 μg kg bw^{-1} day^{-1} (Table 3). EFSA (2010) also used the Carlisle and Wade method in their evaluation of lead dietary exposure to adults.

Using the UK CLEA model, and the CL:AIRE consortium (2014b) intake dose estimate of 1.3 μg kg bw^{-1} day^{-1} (derived to stay below a blood lead concentration of 3.5 μg/dL), we determined SSAC for UAS soil Pb using a number of scenarios. Scenarios (S) 1 and 2 differ in the SPCFs applied (Table 2); S1 uses the SPCF dataset used to derive the Pb C4SL, whilst S2 uses the NABS derived SPCF (as reported in Table 1). The modelling was further split according to the consumption rate data. Scenarios with the letter A were modelled using the consumption rate data used to derive the Pb C4SL and scenarios with the letter B were modelled using NABS 'all participant' consumption rate data (as reported in Table 3). Here, each model was also run with, and without, the ‘top-two’ approach (i.e. use of central tendency values (P50) for fruit and vegetable consumption rates except for the ‘top two’ crop groups expected to give the highest exposure for that contaminant, when the 90th percentile values (P90) are used; for Pb the ‘top-two’ are tubers and green vegetables, CL:AIRE consortium, 2014a). Finally, the role of HF was incorporated into the modelling, with SSAC generated using the high-end (P90) data used to derive the Pb C4SL, the P90 for all of the NABS participants (gardeners and controls), and also the NABS gardeners P90 data (as reported in Table 4).

The sensitivity of changing the SPCFs is clearly highlighted (Table 2) with the C4SL default SPCFs (S1) generating SSAC > 15 times lower than the NABS SPCFs (S2). Whilst we acknowledge the shortcomings of determining SPCFs (as highlighted and discussed in Section 3.1), given our NABS SPCFs are empirically derived based on on-site sampling we consider the NABS data is preferable to the generic data used to derive the Pb C4SL.

The NABS derived changes to the consumption rates (the B scenarios in Table 2) and the NABS HF variations have a more modest impact on the predicted SSAC, in comparison to the changes due to the SPCFs. It is evident, however, that the more fruit and vegetables consumed, and the greater the HF, then the lower the SSAC needs to be to be protective of the UAS gardener. As to the use of the ‘top two’ approach, given people growing their own fruit and vegetables are unlikely to grow sufficient to enable them to eat all crop types at P90 rates, then there is clearly some merit in the approach. However, the impact on the SSAC is relatively modest. For the consumption rate data used to derive the C4SL the impact of using the ‘top two’ approach on the SSAC is on average around 10% (range 6–14%), but larger for the NABS consumption rate data, with a range of impact of 11–34%. A site-specific selection of the ‘top two’ may be the pragmatic way forward, indeed, our empirical evidence indicates root vegetables to be the clear ‘top-one’ in Newcastle, instead of green vegetables and tubers.

Using both the higher NABS all participant P90 consumption rates and the higher NABS gardeners P90 HF then a SSAC of 722 mg/kg is predicted (model: S2-B NABS CR, not top two, Gardener P90 HF), compared to the consumption rate data used to derive the Pb C4SL (model: S2-A, C4SL CR, top two, C4SL P90 HF) which predicts a SSAC of 2877 mg/kg (Table 2). The C4SL derived P90 HF data results in SSAC that are nearly double the NABS P90 HF derived SSACs (Table 2). We consider it a suitably precautionary approach to go with the NABS gardeners P90 HF, resulting in a range of SSAC across our studied Newcastle UAS of 722–1634 mg/kg. No soil samples exceeded the higher SSAC. Only eight individual soil samples exceeded the lower screening value (across 5 different UAS garden plots, encompassing all three UAS), representing just 3% of the investigated soil samples. Whilst acknowledging the uncertainties in the treatment of the NABS dietary data, these exceedances do suggest that the dissemination of risk management advice based on good practice guidelines is still relevant and appropriate.

The NABS blood Pb data are reported in detail elsewhere (Bramwell et al., 2018) but it is relevant to note that all of the gardeners, with the exception of one outlier at 11.4 μg/dL (thought to be linked to the recent renovation of leaded windows) had BLLs below 4.1 μg/dL (range 0.6–4.1 μg/dL). Furthermore, there was no statistically significant difference between the BLL of the UAS gardeners and those of their non-gardening neighbours ($p = 0.569$), (Bramwell et al., 2018). This adds further evidence to suggest that the current C4SL of 80 mg/kg is overly conservative, and SSAC across our studied Newcastle UAS of 722–1634 mg/kg, based on our selected model parameters, would provide more pragmatic and appropriate values for adults using UAS.

In terms of importance for describing the exposure scenario of this specific sub-population (i.e. UAS gardeners) then our NABS data needs to be seen alongside the generalised ‘national’ datasets, which although based on a larger number of participants (compared to our study cohort of n = 73 participants), do not target specific sub-populations such as UAS gardeners. We do however urge caution, given the localised nature of our dataset (Newcastle), and the inherent difficulties in obtaining meaningful food frequency data, but until additional up-to-date national data becomes available such targeted studies provide more realistic SSAC based on site-specific data. Here, the highlighting of site-specific, or sub-population specific, data is important as it leads to very different results than the generic C4SL. Indeed, the use of site-specific data is recommended by the CLEA methodology, although the relevance and importance of sub-population specific datasets is not as considered as the NABS study suggests it needs to be going forward. The C4SL values are after all conservative screening values, and in this respect our findings are reassuring that they reaffirm that they are on the precautionary side, rather than indicating that they are not conservative enough.

Finally, this initial focus on adult gardeners as part of the first phase of the NABS, does not diminish the need to also consider the exposure of children to Pb in UAS. Children are the group that is the most sensitive to Pb health risk, and, even though typically they are not actively participating in gardening in the urban allotment to any great extent, they are subjected to other pathways of exposure and are more likely to have incidental ingestion (e.g., putting fingers in the mouth). Modelling child exposure using the CLEA defaults, but including the NABS SPCF generates, a SSAC of 883 mg/kg Pb and current work is underway to establish the ways in which children interact with such sites and their homegrown fruit and vegetable consumption patterns.

Given the sensitivity of children to Pb exposure and our inadequate knowledge of these exposure parameters, until we have further data, backed up by childrens' blood Pb data, we have only considered SSAC for adult gardeners at this time.

3.5. Implications for UAS gardening

Modelling the NABS data highlights the sensitivity of exposure estimates to SPCF for UAS compared with the generic values used to derive the C4SL. However, the NABS data does not show a significantly elevated exposure as measured by blood Pb samples. Typical mitigation approaches focus on reducing the soil Pb concentration and/or the soil-to-plant transfer rate. Importing clean soil is often impractical and not economically viable in the context of UAS when compared with the higher priority need to remediate residential backyards/gardens. Furthermore, we need to set any mitigation approach within the context that all of the gardeners' blood Pb levels (with the exception of the accounted for outlier) in NABS were < 4.1 μg dL. Soil amendments like organic matter (animal manure, compost, peat), phosphate compounds, liming and biochar have been shown to reduce the plant-Pb uptake rate, but with varying degrees of success (Attanayake et al., 2014; Defoe et al., 2014; Mahar et al., 2015; Murray et al., 2011; Nanthi et al., 2014; Sterrett et al.,1996). Recent studies using nanoparticles derived from waste water residuals potentially offer an innovative sustainable solution for the immobilisation of Pb in soil (Elkhatib et al., 2018). Indeed, in this context, the often poor soil quality of many urban soils can be seen as a positive driver, requiring the incorporation of compost and other soil amendments to improve the fertility and/or soil workability and in so doing dilute the Pb concentration. Organic matter, in the form of animal manure and/or other types of compost is often supplied centrally to UAS and in so doing provides a positive management strategy for reducing exposure risk.

The variations of Pb uptake across the different crop groups highlights a role for focussing on the types of crop grown as a widely applicable way forward towards mitigating any raised exposure at UAS. Minimising the growing and consumption of certain commonly grown root vegetables, in particular rhubarb, beetroot, parsnips and carrots, whilst encouraging the growth of tubers, shrub and tree fruit would seem a sensible management practice in contexts such as those investigated in our UAS. Coupled with ‘good management practices’, aimed at reducing the potential ingestion of soil-bound Pb, especially where we know the Pb is relatively bioaccessible (such as in our study) but where we also know gardeners BLLs are < 5 μg/dL. Such management practices would include: keeping soil moist during dry periods and in windy conditions; peeling and thoroughly washing all crops, and hands, before eating; taking care to reduce the back-tracking of soil into the home (after Brown et al., 2015).

4. Conclusions

A key aim of the Newcastle Allotments Biomonitoring Study (NABS) was to improve the derivation of SSAC for UAS, to give greater reassurance to the general public and greater confidence to regulators who must decide if sites are suitable for use to avoid inappropriate closure of these valuable community spaces, preventing unnecessary stress and concern. This detailed study demonstrates the benefits of site-specific and sub-population specific data as conservative assumptions used in generic screening criteria can give a different impression of the risk. A major outcome of NABS has been to build an exposure dataset specifically for adult UAS gardeners. The major findings of our study are as follows:

1. Pb concentrations in 98% of the soil samples across the three UAS were above the generic UK C4SL for UAS (80 mg/kg), although the majority of crop samples had Pb levels < 0.1 mg/kg FW and our gardeners had BLLs < 4.1 μg/dL.
2. Pb uptake varied with crop group and crop type and our study highlights the suitability of certain crops for growing at UAS with elevated soil Pb (e.g. tubers, shrub and tree fruit), whilst we recommend limiting the consumption of others (selected root vegetables such as rhubarb, beetroot, parsnips and carrots). Although we note that where skins are left on (such as for certain root and tuber vegetables) this has the potential to significantly increase the determined crop Pb levels.
3. Findings of the NABS suggest the need for SPCFs that are site-specific, or at least sub-group specific, rather than those that have been developed for generalised national screening of contaminated sites. All of the NABS geometric mean SPCFs were one to two orders of magnitude lower than the dataset used to derive the UAS C4SL, and changing these parameters in the CLEA model to reflect our empirically determined geometric means led to a > 10-fold increase in the SSAC. This large difference in SPCFs is believed to reflect a number of inter-related mechanisms, including the high soil Pb concentration and the nature of the Pb contamination in these UAS that differed to many of the previous studies upon which the C4SL default data are based. We recommend the derivation of SPCFs that are reflective of urban soils, both in terms of the range of soil Pb concentrations typically observed, but also the sources (and hence human oral bioaccessibility and plant-availability) of this Pb.
4. The consumption rate data on which the C4SL are derived underestimates the quantity of fruit and vegetables consumed by our UAS gardeners. Using a conservative approach to the NABS dietary data, our preferred modelling predicts SSAC of 722–1634 mg/kg.

Findings of this study could trigger (i) the greater use of site-specific data (although this is more costly to collect), (ii) targeted use of the NABS data at similar UAS to inform assessment and management. Clearly this has advantages, but there may be sites such as former orchards where Pb is more available and we need to be careful that sites are not screened out where they should not be, and (iii) revision of the screening value for all scenarios. Here the importance of studies such as ours, for describing the exposure scenario of a specific sub-population (i.e. UAS gardeners) clearly emphases the role of such targeted datasets if we are to derive more appropriate soil assessment criteria for all scenarios.

Acknowledgments

This paper does not reflect the organisational opinions or recommendations of Public Health England (PHE) or the Health and Safety Executive (HSE). The authors wish to thank all of the many colleagues and students from Northumbria and Newcastle Universities and Newcastle City Council who assisted with the sampling programme (Phil Hartley, Nicole Houghton, Lauren Holden, Isabella Entwistle, Josh Hui, Nicola Hestlehurst, Tracy Kelstrup, Tomos Robinson), and to Ed Stutt from WCA Environment and Ian Martin from the Environment Agency for invaluable comments on the draft manuscript. Support for laboratory analyses was provided by Derwentside Environmental Testing Services and ALS Global (many thanks to Geraint Williams). Bramwell and Entwistle kindly acknowledge funding from the Institute for Sustainability and Social Renewal, Newcastle University, Society of Brownfield Risk Assessment (SoBRA) and the Niger Delta Development Commission (NDDC), Nigeria (funding for P. Amaibi).

Appendix A. Supplementary data

Supplementary data to this article can be found online at https://doi.org/10.1016/j.envint.2018.10.054.

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