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Assessment of potentially toxic trace element contamination in urban allotment soils and their uptake by onions: A preliminary case study from Sheffield, England

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Abstract

Toxic trace element (TTE) contamination in urban soils may pose potential health risks, especially in cities with previous industrial activities. This study aimed to investigate soil contamination in urban allotments in Sheffield, the uptake of TTEs in autumn and spring sown onions (Allium cepa), and their potential risks on human health via consumption of the crops. Paired soil and plant samples were taken in triplicates from four private allotments to assess potentially elevated levels of lead (Pb), zinc (Zn), copper (Cu), arsenic (As), and chromium (Cr). These elements in soils exceeded the ambient background levels for England. Both Pb and As exceeded some UK and EU soil tolerable limits. Concentration factors (CF) were calculated as the ratio of trace element in the plant as compared to that in the soil, and uptake rates were in the order Zn>Cu>Cr>Pb>As. Concentrations were higher for most TTEs in spring sown onions (SSO), and had significantly higher CF ($p<0.05$) for Pb and Cr than autumn sown onions (ASO), whereas the opposite was true for As. Toxic elements in plants did not exceed FAO/WHO intake limits when considering TTE content per plant and consumption rates. Human health risk assessment calculations using target hazard quotients (THQ) and hazard indexes (HI) indicated that consuming onions alone did not pose an immediate health risk.

Keywords

Urban agriculture; Allotment soils; Toxic trace elements; Plant uptake; Health risk assessment

1. Introduction

Nearly 50% of the global population now live in cities, and it is expected to rise to 70% by 2050 (Malik et al., 2013; United Nations, 2008). At the same time, it is estimated that approximately 800 million people across the world are engaged in some sort of agricultural activity, contributing to 15-20% of the world's food production (Lorenz et al. 2015). Recently urban agriculture is receiving significant momentum around major cities in the world as such practices are closely associated with human health and wellbeing (Perez-Vazquez et al., 2005; Sustainable Development Commission, 2008). This is a common technique to revamp unused plots of land in urban areas for both the aesthetic appeal and to build neighbourhood cohesion (Palmer, 2018). Some of these plots of land, however, may have been left unused for reasons such as previous soil contamination.

One of the major problems facing urban food production is toxic trace elements (TTEs) found in soil and produce (Alfaro et al., 2017; Antisari et al., 2015; Hu et al., 2013; Laidlaw et al., 2018; Mitchell et al., 2014). Although there are various pathways for the intake of trace elements, the transfer of elevated amounts of these TTEs into the food chain may adversely affect the health conditions of local population where the crops are consumed (Dehghani et al., 2017; Islam et al., 2007; Qing et al., 2015; Tchounwou et al., 2012). Exposure assessments of these potentially harmful heavy metals through vegetable consumption is well documented, especially in areas with a history of smelting and mining activity (Augustsson et al., 2015; Beccaloni et al., 2013; Chen et al., 2014; Intawongse and Dean, 2006; Pelfrêne et al., 2013; Wang et al., 2012). Given many health and well-being benefits achieved through urban agriculture, it is absolutely vital to adopt appropriate management of these urban soils and monitor produce grown thereon for the presence of contaminants including TTEs. Public health risk must be assessed to better understand exposure to TTEs via urban agricultural activities, especially due to the growing trend of own-grown food consumption by urban dwellers (Ngumbi, 2017; Palmer, 2018).

In the UK, there are an estimated 300,000 allotments, and 87 percent of households have their own garden (Buck et al., 2016). Increased urbanisation and a history of industrial activities and environmental pollution, however, have led to many UK cities reporting high levels of heavy metals in gardens and allotments (Giusti, 2011; Knight, 2004; Moir and Thornton, 1989). In many cases the sites were remediated, and allotment holders were advised not to consume crops from the previously contaminated lands. No allotment holders demonstrated signs of toxic metal poisoning, and blood level concentrations were within the normal range (Hough et al., 2004; Knight, 2004; Prasad and Nazareth, 2000). However, there is a serious lack of information about the risk of TTE exposure in populations who consume foods, especially vegetable crops, grown in allotment soils. In addition, some allotments might be located on a previously declared contaminated site but did not receive any real remediation treatment and now host agricultural activity without any risk assessment for TTEs.

TTEs may also accumulate at higher than ambient background levels due to anthropogenic activities. This may occur due to atmospheric deposition throughout urban areas from fossil fuel combustion and dust from contaminated sites. The most significant source of lead contamination in vegetables derives from the aerial deposition of particulates (Giusti, 2011; Hough et al., 2004). Other areas especially vulnerable to contamination are those with a history of waste and sewage sludge dumping, metalliferous mining and smelting, and metallurgical industries (Alloway, 2004, 1995; Culbard et al., 1988; Douay et al., 2013). Thus, many previous industrial sites now used for gardening purposes may pose a significant risk to human health.

Human exposure to potentially toxic metals by ingestion depends largely on their concentrations in consumed crops. The amount of metal taken up by plants in relation to the amount of that present in the soil can be represented by the concentration factor (CF), defined as the ratio of the plant concentration of a metal (as dry weight) to its concentration in the soil (Noli and Tsamos, 2016). Many factors regulate this ratio as not all metal ions in the soil are bioavailable to plants. Metal concentrations in plants are influenced by physicochemical properties of the soil and levels of metal concentrations in the soil. Soil pH is an especially important physicochemical characteristic in assessing the mobility of metal cations. Generally, TTE cations are most mobile under acidic soil conditions, and decrease in bioavailability with increasing pH (Gebrekidan et al., 2013; Jung, 2008; Malik et al., 2013; Sauvé et al., 2000). However, it is important to note that this relationship can be confounded as the effect of changing metal ion reactivity is highly variable (Caporale and Violante, 2016; Hough et al., 2003). Some plants have also demonstrated metal tolerance mechanisms due to various traits such as selective uptake of ions, the decreased permeability of membranes and localisation of metals in certain areas of the plant (Jitendra Kumar et al., 2015; Viehweger, 2014). Typically, the highest concentrations of pollutants are found in plant roots and the lowest in plant seeds (Sharma and Dubey, 2005). Such defence mechanisms also depend on the type of metal, as the same plant may take up different quantities depending on the element itself (Fytianos et al., 2001; Stasinos et al., 2014a, 2014b).

Both soil and plant factors as discussed above could potentially alter the TTE chemistry and mobility in the soil plant system. As a result, a contaminated allotment which was declared safe several years ago for growing crops might become unsafe soil today. Therefore, the overarching aim of this investigation is to evaluate the potential risks to human health through consumption of allotment-grown vegetables, which will directly feed into urban soil ecosystem services including food security, health and well-being of urban population. This investigation focuses on onions, which are one of the most widely grown and consumed vegetables in the UK. Specifically, this study aims to: determine the concentrations and spatial variation of Pb, Zn, Cu, As, and Cr in allotment soils in Sheffield, UK, assess the uptake of above metals by spring and autumn sown onions (A. cepa), and estimate the risk to humans based on the onion consumption rates.

2. Materials and methods

2.1. Study area

This investigation was carried out in the city of Sheffield in South Yorkshire, England, UK, which is home to centuries of industrial activities with an international reputation in the steel industry. High levels of Pb were previously reported in Sheffield, where levels of Pb over 11,000 mg kg^{-1} in the top 50 cm of soil were discovered in domestic gardens, while the Soil Guideline Value for residential land use with plant uptake is 450 mg Pb kg^{-1} (DEFRA and Environment Agency, 2002; Knight, 2004). Investigation into the area's history revealed there had been a Pb rolling mill and smelter in operation until the late 19th century in the location where homes now stand. As the homes were built before any contamination assessment development controls, residents were not aware of the high levels of Pb in the area. Since concentrations of Pb in these domestic gardens were well above the UK trigger levels, remediation was later undertaken. Such findings raise the concern of other possible contaminated sites in Sheffield, especially where there is the potential for ingestion of elevated TTEs via own-grown foods. In a geochemical survey of Sheffield to identify metal pollution across the city, where gardens were tested, all TTE concentrations exceeded their Soil Guideline Value (SGVs) for residential land use with plant uptake (Rawlins et al., 2005). In Sheffield alone, there are more than 70 allotment sites with over 3,000 plots (Sheffield City Council, 2017).

2.2. Allotment site selection

Initially the project was designed to sample the allotments owned by Sheffield City Council, however, access to these sites was denied. Privately owned allotment sites were then contacted for testing, which were only six in number, as compared to the 70 sites owned by the city council. Out of these six site managers contacted, only four agreed to participate in the study and provided access to their plots. The locations of these allotments is depicted in the Supplementary Information (SI: Figure 1). Allotments identified were: Well Community Allotments (WCA), Brightside Gardens (BSG), Oughtibridge Allotments (OBA), and Handsworth & Richmond Allotments (HRA). All allotments were within ~5 miles from the Sheffield city centre. An investigation into the previous land use patterns of these allotment sites was performed using Digimaps (EDINA Historic Digimap Service, 1890). Archived maps dating back to the year 1890 indicated no sign of previous industrial activity on these allotment sites. WCA, OBA, and HRA were documented allotments dating back at least 100 years. BSG also has a long history of allotment use, though some major Sheffield industrial works neighboured the site. In a questionnaire about plot maintenance, all participants answered that they watered their plots exclusively with rainwater collected at allotment sites. Questionnaires about fertiliser and compost use also were completed by each participant to identify possible confounding factors.

2.3. Soil sampling and analysis

A total of 10 plots were tested, with soil and onion samples taken in triplicate from each plot (total soil samples n=30, total onion samples n=30). One plot was tested at WCA, and three plots were tested at BSG, OBA, and HRA. All soil and plant samples were taken between June and July 2017.

Core samples were collected from each plot, from a depth of 0 to 20 cm using a small hand auger, avoiding the edges of individual plots. Soil samples were prepared and analysed using standard procedures for soil bulk density, soil texture, pH (deionized H_2O and $CaCl_2$ extracts), electrical conductivity (EC), and total C and N. Total C and N were measured by an elemental analyser (Vario El CubeCN, Elementar, Germany). A Delta-50 X-ray fluorescence spectrometer (XRF) was used in the benchtop workstation to provide a rapid simultaneous measurement of TTE concentrations in soil samples. Each sample was analysed using three beams (50 kV, 40 kV, and 15 kV); each beam was run for 60 seconds. XRF was performed in triplicate, with a total of 9-minute run per sample, and the average metal concentration values of each beam were used for the analysis.

2.4. Plant sampling and analysis

Onions (Allium cepa) were chosen for this study as they represent a common own-grown vegetable and were cultivated by all participants. As selection was based on plot holder participation, rather than requiring all participants to cultivate under the same conditions, different types of onions were likely obtained. This was considered appropriate as the emphasis of this study was on the variability of metal uptake in onions and its relation to soil concentrations. The onions were divided into two categories according to their planting time, as their edible parts differed:

- "Autumn sown onions (ASO)" (n=6) were planted in October/November 2016, and were collected from two plots at HRA. Being fully grown onions, the inner bulb (outer skin removed) was considered edible.
- "Spring sown onions (SSO)" (n=24) were planted in March/April 2017, and were collected from WCA, BSG, OBA, and one plot at HRA. Having the maturity of a spring onion, the entire bulb and 10 cm of the stem were considered edible.

Onions selected for analysis were chosen away from the borders of the plot to avoid samples with potential contamination from factors outside the plot. The entire plant was removed carefully with a hand trowel. Each onion sample was washed with tap water as to simulate household cleaning practices. Any visible soil particles were washed away. Samples were first air dried, and then at 70 °C in a hot-air oven for at least one week. Once dried, non-edible parts of the plants were removed according to their classification as mentioned above (ASO or SSO), and the remaining edible parts of each sample was milled using a ball mill, creating a homogenous mixture. Acid digestion was performed using EPA Method 3050B (SW-846) (EPA, 1996), and the extracts were analysed for TTEs using inductively coupled plasma mass spectrometry (ICP-MS, Model DRC 11, Pelkin Elmer, USA).

All ICP-MS concentrations of TTEs in plants were generated as dry weight (dw) basis. To assess soil-plant relationships and TTE uptake by plants, concentration factors (CF) were determined. This was calculated as the ratio of TTE concentrations detected in the plants (dry weight basis) over its concentration in the corresponding soil (dry weight basis).

2.5. Human health risk assessment

To evaluate the impact of onion consumption with potentially elevated levels of TTEs on human health, a risk assessment was performed. Exposure to TTE depends on the concentration of the element in the food and the daily food consumption rate. Estimated daily intake (EDI) can therefore be calculated using Eq. 1 (Chamannejadian et al., 2013; Hang et al., 2009; Zheng et al., 2007):

$$EDI = \frac{C \times Con}{Bw} \qquad \text{(Eq. 1)}$$

Where, EDI (μg kg^{-1} Bw^{-1} day^{-1}) is the amount of TTE consumed; C (μg g^{-1}) is the concentration of TTE in onion; Con (g $person^{-1}$ day^{-1}) is the average daily consumption of vegetables in the UK, assuming worst case scenario that all vegetables consumed were raw onions (20 g $person^{-1}$ day^{-1} for males, 38 g $person^{-1}$ day^{-1} for females (Bates et al., 2014); Bw is the average body weight (83.6 kg for males and 70.2 kg for females, Office for National Statistics, 2010).

The standard EPA method for risk assessment states that the risk of non-carcinogenic effects is determined as the ratio of the dose from exposure to site media as compared to a dose that is thought to be of no risk (USEPA, 2001). This is the target hazard quotient (THQ). A quotient value less than one indicates no significant risk of non-carcinogenic effects. THQ can be determined by Eq. 2 (Zheng et al., 2007):

$$\text{THQ} = \frac{EDI}{RfD} \quad \text{(Eq. 2)}$$

Where, RfD is the reference oral dose (μg kg^{-1} day^{-1}). RfD values used for Pd, Zn, Cu, Cr, As were 3.5, 300, 40, 1500, 50 μg kg^{-1} day^{-1} (FAO/WHO, 1997; UNEP/FAO/WHO, 1992; US EPA IRIS, 2018). In many cases, however, exposure may result from two or more pollutants, creating an additive effect. To calculate the additive effect, hazard index (HI) is generated as the sum of a mixture of toxic elements (Eq. 3) (Hang et al., 2009; Zheng et al., 2007):

$$HI = \sum_{n=1}^{i} THQ_n \quad \text{(Eq. 3)}$$

2.6. Data analysis

All statistical analyses were performed using Excel 2016 and SPSS Statistics 23 software packages, and plots were made using GraphPad Prism (7.03) software.

3. Results

3.1. Soil physiochemical properties

The bulk density, pH, EC and C:N ratio are presented in Table 1. Soil textures were either silty loam or sandy loam. Bulk densities were generally low as crops were grown with soil-garden compost mixture. All soils were between the pH ranges of 5.6-6.9 (pH with $CaCl_2$), the lowest mean pH was found in OBA (mean pH=5.6). The soil with the highest EC was in plot 1 in BSG (808.3 µS cm^{-1}), and the lowest in plot 1 in OBA (419.3 µS cm^{-1}). The average percent C contents was 11.04 %, but varied from 6.18 to 11.88 %. The C:N ratio varied from 14.85 to 22.25.

3.2. TTE concentrations in soils

Three soil samples from each plot were averaged to determine mean TTE concentrations. The soil Pb, Zn, Cu, Cr and As concentrations are represented in Figure 1 (a-e) for comparative purposes and to identify outliers, as the data was not symmetrically distributed. The data from each plot are also presented as Supplementary Information (SI: Table 1), which includes background pH and TTE concentrations in England, the UK Soil Guidelines Values (SGV), and the EU tolerable limits.

Table 1. Soil characteristics, means and standard deviations of three samples from each plot and allotment means where more than one plot was sampled.

Allotment	Plot	Bulk Density (g/cm^3)	pH (H_20)	pH ($CaCl_2$)	Conductivity (µS/cm)	% N	% C	C:N
Well Community Allotments (WCA)	1	0.69 ± 0.07	6.2 ± 0.20	5.9 ± 0.20	442.7 ± 47.7	0.35 ± 0.01	6.18 ± 0.40	17.60 ± 1.0
Brightside Gardens (BSG)	1	0.88 ± 0.13	7.0 ± 0.20	6.8 ± 0.10	808.3 ± 237.0	0.33 ± 0.01	6.91 ± 0.40	21.13 ± 1.3
	2	0.74 ± 0.08	7.0 ± 0.40	6.8 ± 0.20	508.3 ± 51.1	0.39 ± 0.03	7.63 ± 1.0	19.25 ± 1.0
	3	0.64 ± 0.03	7.1 ± 0.10	6.8 ± 0.10	505.0 ± 66.2	0.41 ± 0	8.98 ± 0.10	21.77 ± 0.50
Mean		0.75 ± 0.08	7.0 ± 0.20	6.8 ± 0.10	607.2 ± 118.1	0.38 ± 0.02	7.84 ± 0.5	20.71 ± 0.90
Oughtibridge Allotments (OBA)	1	0.46 ± 0.03	5.8 ± 0.10	5.6 ± 0.10	419.3 ± 73.9	0.56 ± 0.06	9.23 ± 1.2	16.33 ± 0.40
	2	0.46 ± 0.05	6.0 ± 0.30	5.7 ± 0.30	421.3 ± 99.0	0.41 ± 0.04	6.73 ± 0.7	16.21 ± 0.30
	3	0.48 ± 0.02	5.8 ± 0.10	5.6 ± 0.10	660.3 ± 138.1	0.52 ± 0.05	7.71 ± 0.8	14.85 ± 0.60
Mean		0.46 ± 0.03	5.9 ± 0.20	5.6 ± 0.20	500.3 ± 103.7	0.50 ± 0.05	7.89 ± 0.9	15.80 ± 0.40
Handsworth & Richmond Allotments (HRA)	1	0.51 ± 0.07	6.4 ± 0.10	6.2 ± 0.10	641.3 ± 128.0	0.53 ± 0.04	11.88 ± 1.2	22.25 ± 0.60
	2	0.55 ± 0.10	7.2 ± 0.10	6.9 ± 0.10	732.0 ± 136.5	0.59 ± 0.02	10.94 ± 0.7	18.65 ± 0.60
	3	0.58 ± 0.07	6.1 ± 0.10	5.8 ± 0.10	525.0 ± 56.0	0.60 ± 0.02	10.83 ± 0.8	17.99 ± 0.39
Mean		0.54 ± 0.08	6.6 ± 0.10	6.3 ± 0.10	632.8 ± 106.9	0.57 ± 0.03	11.21 ± 0.9	19.63 ± 1.87

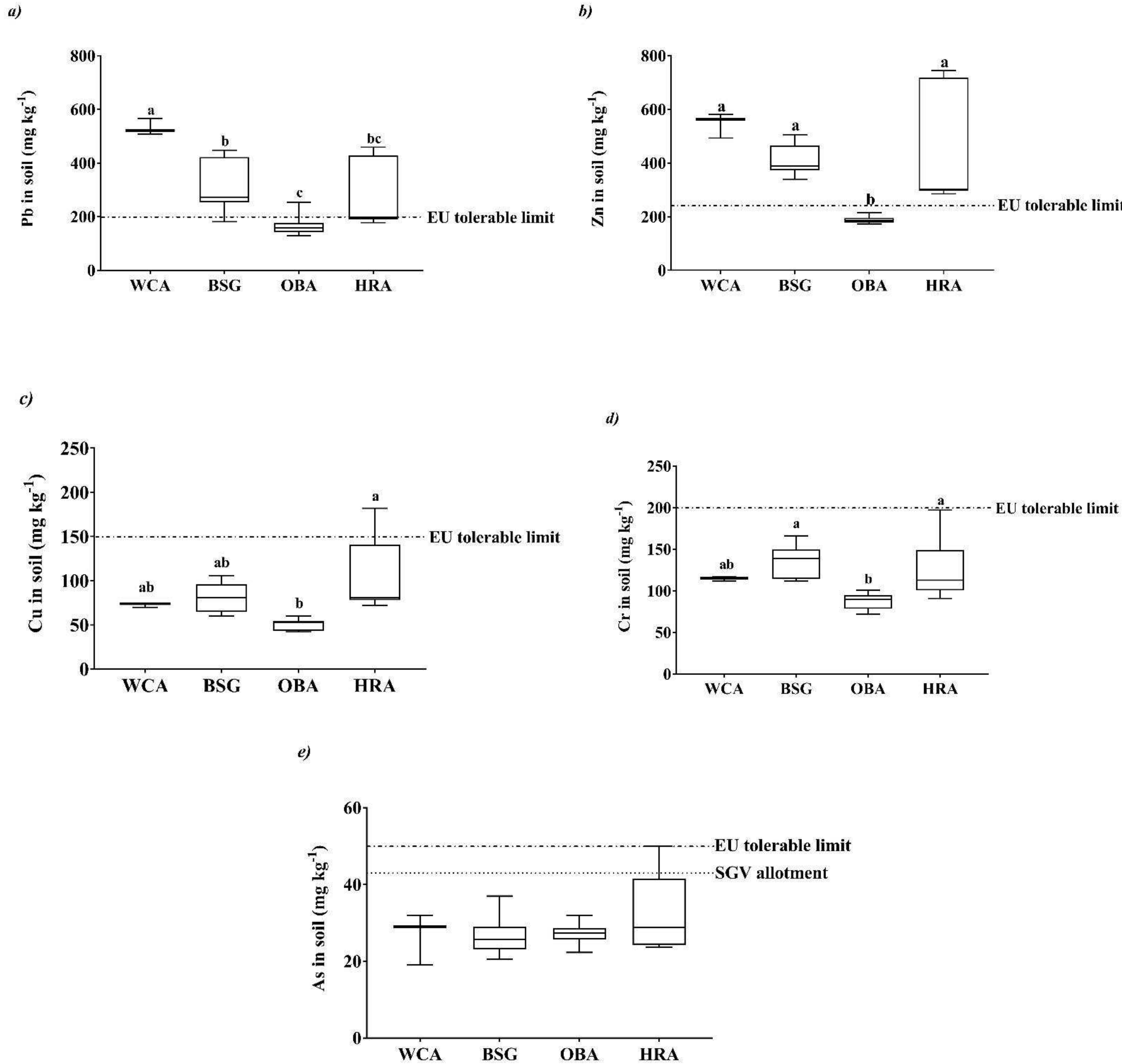

Figure 1a-e. Boxplots showing median, 25th and 75th percentiles of TTE in soil (mg kg^{-1}). Allotments are WCA Well Community Allotments, BSG Brightside Gardens, OBA Oughtibridge Allotments, HRA Handsworth & Richmond Allotments. Note the Y-axis scale differs between graphs. Significant differences ($p<0.05$) between the different groups are shown using lower case letters.

The SGVs are set by the UK to assess the risk to human health from contaminated soil exposure. SGVs differ based on land usage, with the SGVs for allotments used here for comparison. The previous SGV for As has been modified from 20 mg kg^{-1} to 43 mg kg^{-1} in the current guidelines (DEFRA and Environment Agency, 2002; Environment Agency, 2009). Only plot 2 in HRA had As concentration above the current SGV (43 mg kg^{-1}). All allotments had concentrations above the previous As SGV (20 mg kg^{-1}). The previous Pb SGV was 450 mg kg^{-1}, of which plot 1 at WCA had concentrations above this limit. Plot 2 at HRA, and plots 2 and 3 at BSG exceeded the previous SGV for Cr (130 mg kg^{-1}). No new guidelines were issued, and these previous SGVs have now been withdrawn (DEFRA and Environment Agency, 2002).

European countries have a variety of methods to determine human/environmental risk levels associated with TTE concentrations in soil. Most recently, Finnish legislation has been applied internationally as it provides an appropriate representation of mean values used by different national systems within Europe (Ministry of the Environment, 2007; van der Voet et al., 2013), and are referred hereto as the 'EU tolerable limits'. The EU tolerable limit values presented in SI: Table 1 are guideline values, where if they are exceeded, the area poses health/ecological risks. These EU tolerable limits are higher than the UK's SGVs for all metals except for Pb. Almost all plots sampled in this study had concentrations higher than this EU tolerable level of Pb (200 mg kg^{-1}). Most plots also had Zn levels higher than the EU tolerable limit (250 mg kg^{-1}), with plot 2 at HRA almost tripled the limit (727.33 mg kg^{-1}). Soil concentrations of Cu, As, and Cr did not exceed the EU tolerable limits.

All data sets were confirmed to be normally distributed. Differences in mean metal concentrations between the four allotments were analysed using SPSS one-way ANOVA. Concentrations of Pb between allotments were significantly different ($p<0.05$), the same was true for Zn, Cu, and Cr between allotments. However, there was no significant difference in As levels between allotments ($p>0.05$).

The data showed a positive relationship between soil pH (in H_2O) and concentrations of Zn, Cu and Cr. This is illustrated in Figure 2a-c, regression coefficients are statistically significant ($p<0.05$). Multiple regression

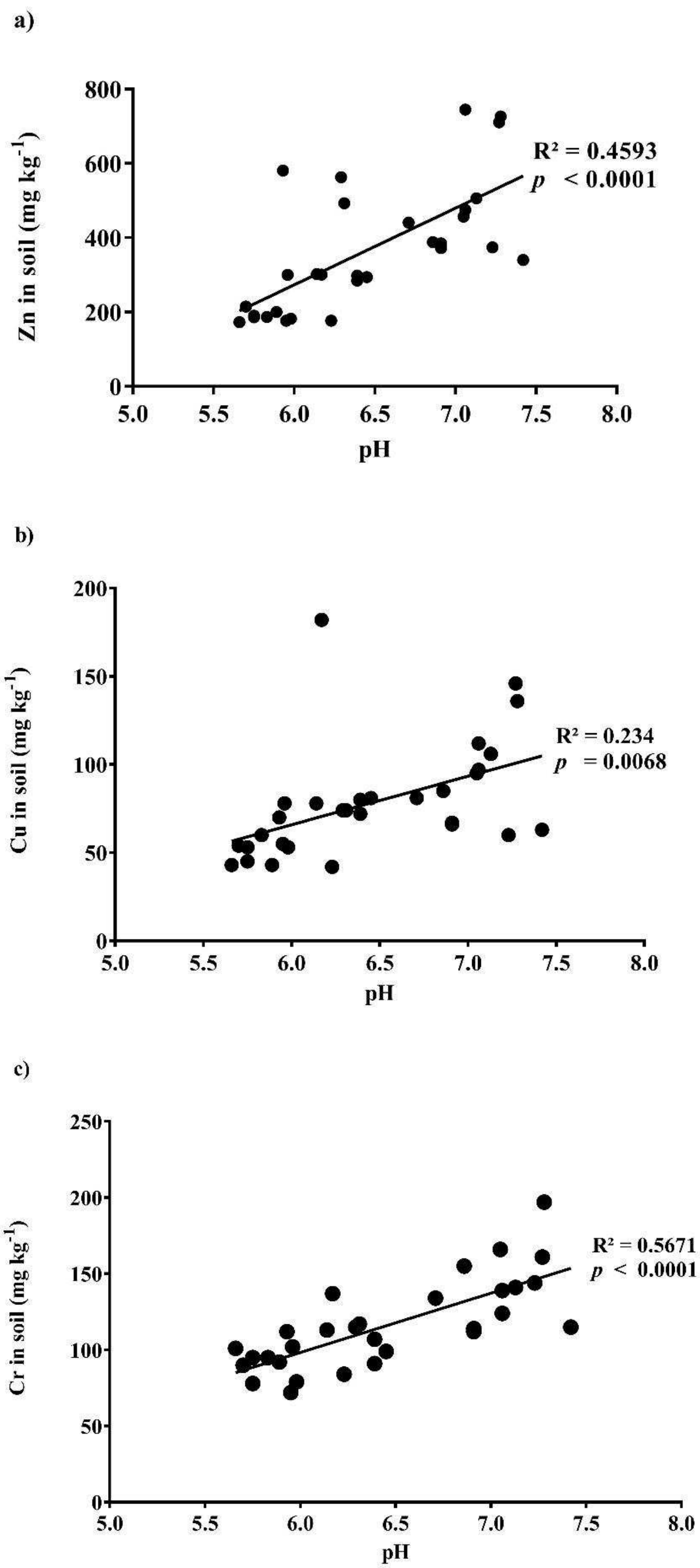

Figure 2a-c. Statistically significant relationships ($p<0.05$) between the soil concentration of Zn (a), Cu (b) and Cr (c) and soil pH (in H_2O) (n=30).

analysis was employed to identify any relationship between soil physicochemical properties and concentrations of TTE in soils. Significant regression ($p<0.05$) were found between Zn vs pH (R^2=0.4593), Cu vs pH (R^2=0.234), and Cr vs pH (R^2=0.5671)

3.3. TTE concentrations in plants

The Joint Food and Agriculture Organisation/World Health Organisation (FAO/WHO) Expert Committee on Food Additives (JECFA) identify maximum levels for contaminants and toxins in foods for Pb and As (FAO/WHO, 2011a), and are presented for comparison in SI: Table 2. Figure 3 depicts TTE concentrations in plants. Assessment of WCA TTE concentrations in relation to the other allotments is limited as only one plot at this allotment was tested. HRA plots 1 and 3 are ASO, while all others are SSO. All metals, except As, were lower in ASO than SSO. BSG had the highest mean concentrations of Pb in plants (0.23 mg kg^{-1}). Mean Zn concentrations in plants were similar between allotments and ranged from 3.17 mg kg^{-1} to 8.55 mg kg^{-1}. Cu concentration ranged from 0.35 mg kg^{-1} to 0.85 mg kg^{-1}. No As was detected in WCA nor in plot 2 at OBA and mean As concentration was the highest in plants at HRA. Concentrations of Cr ranged from 0.02 mg kg^{-1} to 0.34 mg kg^{-1} with concentrations 15 times higher in SSO than ASO. Six plots had plants with Pb levels higher than the FAO/WHO guidelines of 0.10 mg kg^{-1}. No plants had levels higher than the FAO/WHO guidelines for As (0.10 mg kg^{-1}).

3.4. Concentration Factor (CF)

A higher CF indicates more mobile/bioavailable metal ions in the soil. The order of metal uptake/transfer from soil to plant was Zn>Cu>Cr>Pb>As. These were calculated using means of all onions. The order of metal uptake from soil to plant was calculated as an average CF of each metal. No levels of As were detected in WCA nor in plot 2 at OBA, and thus CF could not be calculated for these two plots. Results are presented in Table 2.

TTE uptake between ASO and SSO was also compared (Figure 4a-c). This was done to interpret metal uptake in relation to time spent in the ground, as well as to examine localisation of metals in the plant, as ASO and SSO differed in edible parts. ASO and SSO were grouped into CF_{Autumn} and CF_{Spring},

Table 2. Concentration factor (CF), ratio of metal in plant (mg kg^{-1}, dry weight) to metal in soil (mg kg^{-1}, dry weight).

Allotment	Plot	Pb	Zn	Cu	Cr	As
Well Community Allotments (WCA)	1	1.88E-03	1.43E-01	7.06E-02	2.67E-02	ND[a]
Brightside Gardens (BSG)	1	6.20E-03	1.84E-01	7.89E-02	2.17E-02	1.06E-03
	2	9.90E-03	2.19E-01	1.04E-01	2.42E-02	1.08E-03
	3	6.18E-03	9.09E-02	1.72E-02	8.16E-03	1.18E-03
Oughtibridge Allotments (OBA)	1	7.29E-03	4.64E-01	7.31E-02	1.91E-02	8.63E-04
	2	9.37E-03	3.09E-01	1.70E-01	1.47E-02	ND[a]
	3	4.52E-03	3.56E-01	1.24E-01	2.93E-01	1.03E-02
Handsworth & Richmond Allotments (HRA)	1*	1.97E-03	1.61E-01	4.83E-02	3.03E-03	8.58E-03
	2	6.62E-03	9.06E-02	5.71E-02	2.14E-02	1.57E-02
	3*	3.41E-03	1.05E-01	3.62E-02	1.36E-03	1.03E-02

[a]ND, None Detected

*Denotes Autumn sown onions (ASO), all others are Spring sown onions (SSO).

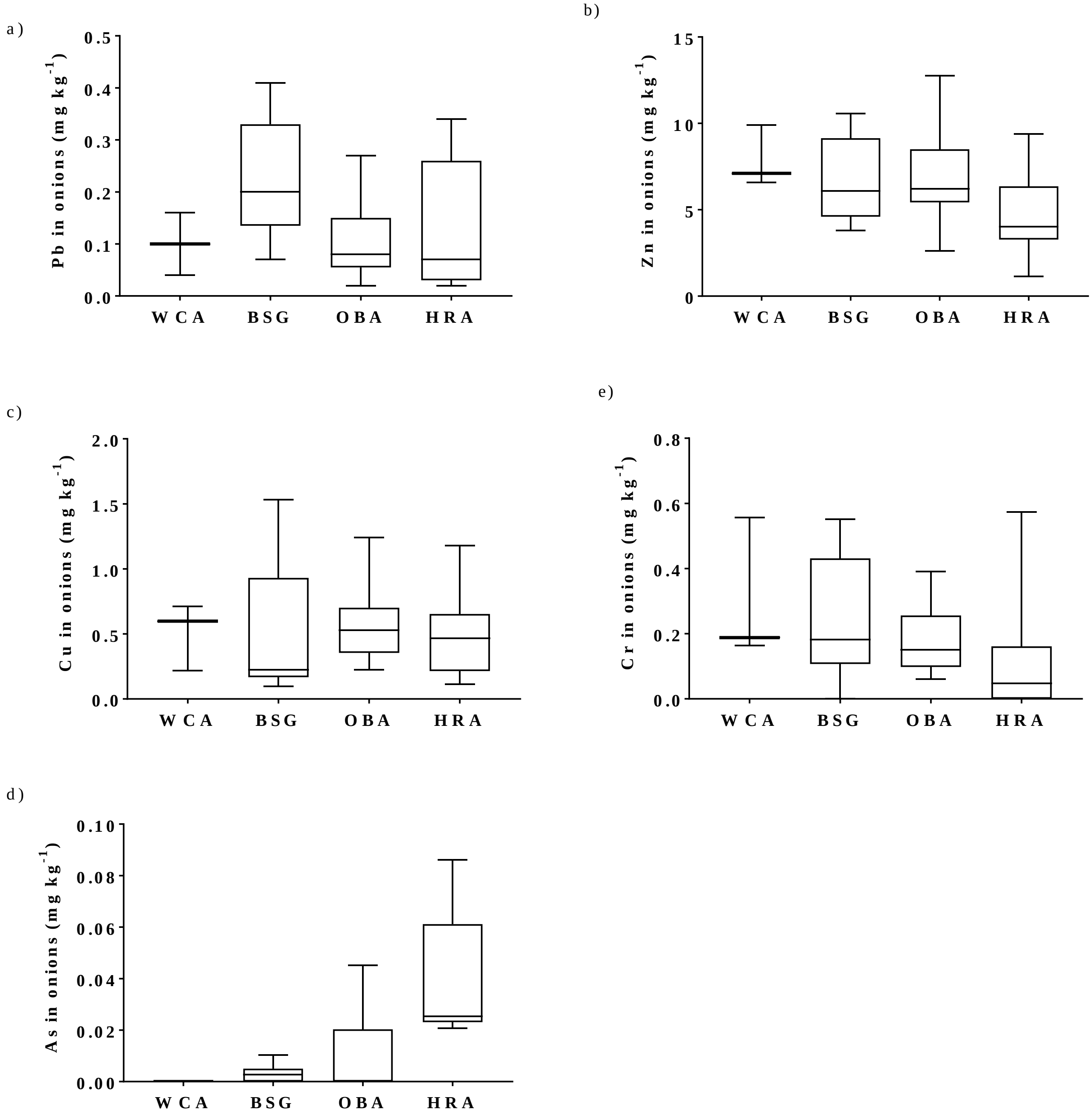

Figure 3a-e. Boxplots showing median, 25th and 75th percentiles of TTE concentrations in plants (mg kg^{-1}, fw). Allotments are WCA Well Community Allotments, BSG Brightside Gardens, OBA Oughtibridge Allotments, HRA Handsworth & Richmond Allotments. HRA values include ASO and SSO data grouped together. Note the Y-axis scale differs between graphs. No As was detected in WCA.

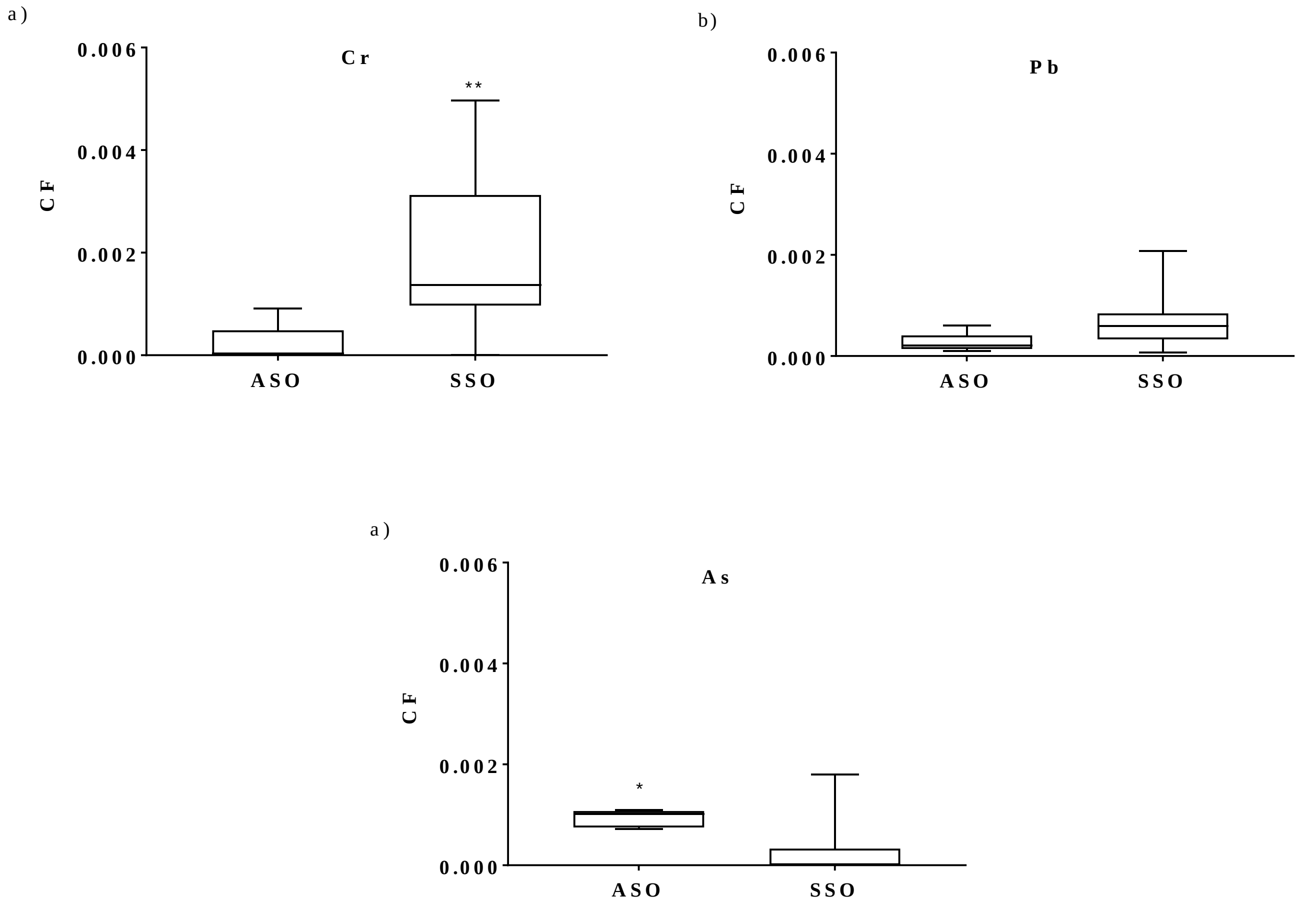

Figure 4a-c. Boxplots showing median, 25th and 75th percentiles of significant different CF for Pb, As, and Cr, in ASO compared to SSO. Pb and Cr were significantly higher in SSO ($p<0.05$), while As was significantly higher in ASO ($p<0.05$). Significant differences are indicated using symbols (* $p<0.05$ and ** $p<0.01$). Note the Y-axis scale differs between graphs.

respectively, to compare uptakes. Mean CFs were determined for CF_{Autumn} and CF_{Spring} for each metal. Uptakes of metals between ASO and SSO were compared using non-parametric Mann-Whitney U tests due to the small ASO sample size (n=6), and because the data were not normally distributed. CF_{Spring} had significantly higher Pb (p=0.021) and Cr (p=0.00) than CF_{Autumn}. CF_{Autumn}, however, had significantly higher As (p=0.029) than CF_{Spring}.

3.5. Potential health risk

The THQs of each TTE through onion consumption are listed in Table 3. All of the calculated THQ values were < 1, indicating health risks associated with TTE exposure for men and women are insignificant if residents only ingest one type of TTE from foodstuff. The potential health risk of the TTEs examined in the study combined or HI also was < 1 for both men and women. This suggests that there is no significant potential health risk for men or women consuming onions from these allotments when considering the collective effect of the levels of the five TTEs analysed in this study.

4. Discussion

4.1. Soil metal contamination in allotments

All allotment TTE concentrations were well above the ambient background levels for England, however, they were similar to ambient pH for England (Barraclough, 2007). This demonstrates that there are other external factors that have led to these increased levels of contaminants. A few plots exceeded the SGVs for Pb, As, and Cr. These SGVs indicate levels of metals in allotments below which there are minimal long-term health risks, concentrations above these limits should be further assessed to determine if remediation is necessary.

The EU tolerable limits set regulations on concentration levels allowed for TTEs depending on land usage and states the need for action if surpassed. The EU tolerable limits for all non-industrial land use are higher as compared to the UK's SGVs for all metals except for Pb. Such variation highlights the dependency

Table 3. Estimated daily intake by male and females and potential health risk due to onion consumption.

Individuals	Element	RfD[a]	EDI[b]	THQ[c]	HI[d]
Males	Pb	3.5	0.034	0.010	0.018
	Zn	300	1.513	0.005	
	Cu	40	0.125	0.003	
	Cr	1500	0.036	0.000	
	As	50	0.005	0.000	
Females	Pb	3.5	0.076	0.022	0.041
	Zn	300	3.423	0.011	
	Cu	40	0.283	0.007	
	Cr	1500	0.081	0.000	
	As	50	0.011	0.000	

[a]Reference oral dose (μg kg^{-1} day^{-1})

[b]Estimated daily intake (μg kg^{-1} day^{-1})

[c]Target Hazard Quotient (EDI/RfD)

[d]Hazard Index when multiple metals are present (ΣTHQ_n ; n = 1 to i)

of risk assessment on toxicological references employed. Most metal concentrations in allotment soils were below the EU limits, except for Pb and some Zn concentrations.

Other UK allotment TTE concentrations are inconsistent and vary depending on location. A study of Pb in allotment soils in a London borough revealed concentrations that ranged from 513 to 2,910 mg kg^{-1} (Prasad and Nazareth, 2000), whereas in Bristol the median Pb concentration was 210 mg kg^{-1} (Giusti, 2011). Other metal median values in Bristol were 272.6 mg kg^{-1} Zn, 60.1 mg kg^{-1} Cu, 21.7 mg kg^{-1} As, and 23.1 mg kg^{-1} Cr (Giusti, 2011). Such results are consistent with concentrations found in this Sheffield study, however Cr concentrations in the Sheffield allotments were more than three times greater than those found in Bristol.

The largest survey of garden soils carried out by Culbard et al. (1988) analysed 4,650 garden soils around Great Britain. Results indicated levels of Pb, Cu and Zn were elevated, with a mean Pb concentration in garden soils of 298 mg kg^{-1}. Levels of Pb as high as 1,870 mg kg^{-1} were found in areas with previous mining history. A soil geochemical survey of Sheffield determined median Pb levels to be 161 mg kg^{-1}, and where samples were from the domestic garden, all Pb concentrations were above the SGV of 450 mg kg^{-1} (Rawlins et al., 2005). As previously discussed, concerning levels of Pb, as high as 14,863 mg kg^{-1}, were found in domestic gardens in a Sheffield neighbourhood that was previously a Pb rolling mill (Knight, 2004). Such findings indicate the need for site specific risk assessment.

As found in this study, similar correlations between soil physiochemical properties and TTE concentrations were seen in the UK Soil and Herbage pollutant Survey (UKSHS), where significant positive correlations were found between Zn and pH (p=0.01) in the overall UK soil dataset (Barraclough, 2007). Consistent with results from this Sheffield study, Sauvé et al. (2000) also found a low correlation coefficient between Pb and pH, as compared to the correlation coefficient for Zn and pH.

4.2. Plant uptake and CF

Relatively neutral soil pH could explain low bioavailability of metals in soils (Ming et al., 2016). There was a positive correlation seen when comparing Pb in plants vs soil pH (R^2=0.2905); however, no correlation was significant for the other soil physiochemical characteristics (e.g., total C) and plant metal concentrations. One of the most predictive factors in metal uptake by plants is pH, with low soil pH known to increase metal mobilisation and bioavailable concentrations (Golia et al., 2008; Puga et al., 2015). All soils tested had relatively neutral pH, indicating TTEs may be less mobile in these soil conditions as compared to soil with lower pH values. This could explain the lack of correlation between soil-plant metal concentrations. Though soil mixtures were homogenous upon the use of XRF, some metal distribution within allotments may not have been uniformly distributed and may have produced a seemingly high level of metal in comparison to what onion fibrous roots were exposed to. This could also give a reason for the lack of correlation between soil-plant concentrations of metals.

Though it was not shown here, a significant correlation was detected in As in soil to As in plant (R^2=0.4143). This is similar to other results especially those found in the derivation of SGVs for arsenic in relation to root vegetables (Environment Agency, 2009; Zandsalimi et al., 2011). No other soil-plant concentration correlations were significant. Lack of linearity between plant and soil concentrations of the other metals can be explained by many factors influencing metal uptake by plants and complexity of ion transfer. Variables such as soil physicochemical properties, applied fertilisers, the type of plant species, etc., can influence these uptake rates (Chen et al., 2016; Chojnacka et al., 2005; Liu et al., 2015). Furthermore, uptake of metals may result from sources other than soil, such as polluted air and water. Air contaminants are less likely to affect root vegetables such as the ASO but may contribute to SSO contamination as the stems are part of the edible vegetable. Contaminated particulates in the air have been documented to increase levels of TTEs in plants (Antisari et al., 2015). This is especially important when examining Pb contamination as combustion of leaded petrol in vehicles and coal combustion has led to increased atmospheric deposition and remains the most significant source of Pb contamination in vegetables (Barraclough, 2007; Hough et al., 2004). Hand washing of vegetables also does not remove all soil particles and may contribute TTE concentrations of the onions. The maturity levels of the vegetable and different consumed parts also may explain the different observed concentrations of TTEs between the two groups. A noted limitation was also the lack of certainty on the onion cultivars used in the study and the small sample size of ASO. The soil TTE concentrations could have been different in these two plots as compared to the other SSO plots and could have had an influence on the results.

Additionally, defence mechanisms by plants may localise TTEs in different regions of the plant. Selection of tissue analysed may greatly vary results of apparent metal concentrations in plants (Prasad and Nazareth, 2000). In onions, Pb has been found to localise in the fibrous root tips, while the root base, closest to the bulb of the onion, has been found to have the lowest concentrations of Pb (Wierzbicka, 1987). Other studies examining the uptake of Pb by onions, have reported Pb concentrations in leaf and shoots to be more than double than that found in the bulb, with the largest amount of Pb found in the basal part of leaves. This above-below soil pattern of metal distribution was seen in multiple varieties of onions, and demonstrated a great capacity to translocate Pb in its tissues in the presence of highly contaminated soils (Michalak and Wierzbicka, 1998). It was seen in this study that SSO was higher in concentrations of almost all metals. This could be explained by the localisation of TTEs in the basal portion of the leaves which were included for sample analysis in SSO (as they were spring onions), but was removed in the ASO (as they were mature onions). Though concentrations of As were higher in ASO than SSO, this could be due to the relatively high levels of As in the soil at HRA (where the ASO were grown), as the SSO at HRA had even higher levels of As.

The TTE concentrations in plants in this study are consistent with other reports of metal transfer capabilities by plants in contaminated soils (Augustsson et al., 2015; Chojnacka et al., 2005; Dinelli and Lombini, 1996). The CF order of metal uptake from soil to plant was Zn>Cu>Cr>Pb>As, which is well documented (Alexander et al., 2006; Chen et al., 2014; Intawongse and Dean, 2006; Islam et al., 2015; Wang et al., 2012; Xian, 1989). Fytianos et al. (2001) found much greater CFs, for example, CF in onions grown in contaminated soils were 0.35, 0.31, and 0.21 for Zn, Cu and Pb, respectively. However, this is still consistent with previous results that Pb is especially known to not be readily taken up by plants, due in part to its stronger adhesion to the soil matrix (Barraclough, 2007; Gebrekidan et al., 2013; Holmgren et al., 1993). Such relationships were seen in this study where the WCA, which had the highest levels of Pb in the soil, had some of the lowest concentrations of Pb in the plants. Augustsson et al. (2015) similarly found that root vegetable metal concentrations were only moderately elevated, despite high concentrations in the soil where they were grown, and most vegetables were below food contaminant legislation levels.

4.3. Consumption and health risk assessment

TTE concentrations, dry weight, in plants were converted to fresh weight (fw) basis using recorded moisture contents (US EPA, 1997) for better comparison to FAO/WHO food standards (FAO/WHO, 2011a). Six plots had onions that exceeded the FAO/WHO maximum level of Pb in root vegetables of 0.10 mg Pb kg^{-1} fw (FAO/WHO, 2011a). FAO/WHO also previously suggested a provisional tolerable weekly intake (PTWI) for adults of 0.025 mg Pb kg^{-1} body weight (FAO/WHO, 2011b). In considering the amount of Pb per plant and consumption rates, an unreasonable quantity of onions would need to be consumed per week to reach this PTWI. This PTWI has now been withdrawn as recent dose-response analysis does not indicate any threshold to be health protective for effects of Pb, and FAO/WHO have not established a new PTWI. Levels of As concentrations in plants were well below the FAO/WHO maximum level for contaminants in foods of 0.10 mg As kg^{-1} (FAO/WHO, 2011a). The highest concentration of As was the SSO at HRA (0.069 mg kg^{-1}). This is consistent with other studies in which contaminated sites, even with significant soil pollution, did not yield vegetables with intakes above tolerable daily intakes (Augustsson et al., 2015; Beccaloni et al., 2013; Chen et al., 2014; Fytianos et al., 2001; Pelfrêne et al., 2013; Wang et al., 2012).

Bioavailability and human exposure are often hard to assess as metals may be present in forms not harmful to the environmental or to human health. It is difficult to assess total exposure as the pathways of TTE intake may also include contaminated drinking water, oral soil ingestion, and consumption of other contaminated food source. Within the body, many factors such as absorption in the gastrointestinal tract and stomach acidity also affect exposure. Uptake from the gastrointestinal tract may differ according to element species and to plant type (Augustsson et al., 2015; Intawongse and Dean, 2006). The actual harm posed to allotment holders growing crops in soils with elevated levels of TTEs is likely to be minimal. Allotment holders typically do not solely consume food grown in their allotments, which makes risk assessment more difficult. Other variables may likewise affect contamination and metal exposure, for instance, vegetable preparation and type of vegetable consumed vary in washing and peeling procedures to remove soil particles, and absorb different amounts of metals (Alexander et al., 2006; Intawongse and Dean, 2006).

It is also important to note that though toxic exposure is ubiquitous among populations, some subpopulations will be disproportionately affected. Especially vulnerable groups such as children, elderly adults, pregnant women or those with lowered immune systems due to chronic illness may experience more serious effects from TTE exposure. For example, an adolescent that consumes that same quantity of contaminated foodstuffs, but with half the bodyweight of an adult, is more at risk for exposure. Furthermore, prenatal exposure to certain TTEs has been documented to increase risk of cancer in childhood and interfere with crucial developmental stages that can lead to adverse birth outcomes and increased risks of disease (Balk et al., 2011; Bergman et al., 2012; Grandjean et al., 2007; Hines et al., 2010; Woodruff et al., 2008).

5. Conclusions

Evaluation of allotments in Sheffield revealed that concentrations of Pb, Zn, As, and Cr exceeded some UK and EU soil limit guidelines. Such findings do not necessarily lead to high levels of metals in plants grown in these soils, as little correlation was evident between soil and plant metal contents. This is due to a variety of factors governing the bioavailability and uptake of metals by plants. Because of this, onions grown in these soils did not exceed foodstuff regulations.

Following this initial screening, further detailed assessment of these areas can be completed in the future, especially in those soils that exceeded limit guidelines. Also, as soil metal concentrations vary greatly according to previous land use, site-specific risk assessments should be conducted, especially in zones with past industrial history that are now used for gardening purposes.

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Appendix A. Supplementary material

Supplementary information can be found in the online version.

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