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

3

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21 **Abstract**

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

37 **Keywords**

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

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45 **1. Introduction**

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

56 One of the major problems facing urban food production is toxic trace elements (TTEs)
57 found in soil and produce (Alfaro et al., 2017; Antisari et al., 2015; Hu et al., 2013; Laidlaw et
58 al., 2018; Mitchell et al., 2014). Although there are various pathways for the intake of trace
59 elements, the transfer of elevated amounts of these TTEs into the food chain may adversely
60 affect the health conditions of local population where the crops are consumed (Dehghani et al.,
61 2017; Islam et al., 2007; Qing et al., 2015; Tchounwou et al., 2012). Exposure assessments of
62 these potentially harmful heavy metals through vegetable consumption is well documented,
63 especially in areas with a history of smelting and mining activity (Augustsson et al., 2015;
64 Beccaloni et al., 2013; Chen et al., 2014; Intawongse and Dean, 2006; Pelfrêne et al., 2013;
65 Wang et al., 2012). Given many health and well-being benefits achieved through urban
66 agriculture, it is absolutely vital to adopt appropriate management of these urban soils and
67 monitor produce grown thereon for the presence of contaminants including TTEs. Public health
68 risk must be assessed to better understand exposure to TTEs via urban agricultural activities,

69 especially due to the growing trend of own-grown food consumption by urban dwellers
70 (Ngumbi, 2017; Palmer, 2018).

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

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

91 Human exposure to potentially toxic metals by ingestion depends largely on their
92 concentrations in consumed crops. The amount of metal taken up by plants in relation to the
93 amount of that present in the soil can be represented by the concentration factor (CF), defined as

94 the ratio of the plant concentration of a metal (as dry weight) to its concentration in the soil (Noli
95 and Tsamos, 2016). Many factors regulate this ratio as not all metal ions in the soil are
96 bioavailable to plants. Metal concentrations in plants are influenced by physicochemical
97 properties of the soil and levels of metal concentrations in the soil. Soil pH is an especially
98 important physicochemical characteristic in assessing the mobility of metal cations. Generally,
99 TTE cations are most mobile under acidic soil conditions, and decrease in bioavailability with
100 increasing pH (Gebrekidan et al., 2013; Jung, 2008; Malik et al., 2013; Sauvé et al., 2000).
101 However, it is important to note that this relationship can be confounded as the effect of
102 changing metal ion reactivity is highly variable (Caporale and Violante, 2016; Hough et al.,
103 2003). Some plants have also demonstrated metal tolerance mechanisms due to various traits
104 such as selective uptake of ions, the decreased permeability of membranes and localisation of
105 metals in certain areas of the plant (Jitendra Kumar et al., 2015; Viehweger, 2014). Typically, the
106 highest concentrations of pollutants are found in plant roots and the lowest in plant seeds
107 (Sharma and Dubey, 2005). Such defence mechanisms also depend on the type of metal, as the
108 same plant may take up different quantities depending on the element itself (Fytianos et al.,
109 2001; Stasinou et al., 2014a, 2014b).

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

119 autumn sown onions (*A. cepa*), and estimate the risk to humans based on the onion consumption
120 rates.

121

122 **2. Materials and methods**

123 *2.1. Study area*

124 This investigation was carried out in the city of Sheffield in South Yorkshire, England,
125 UK, which is home to centuries of industrial activities with an international reputation in the
126 steel industry. High levels of Pb were previously reported in Sheffield, where levels of Pb over
127 11,000 mg kg⁻¹ in the top 50 cm of soil were discovered in domestic gardens, while the Soil
128 Guideline Value for residential land use with plant uptake is 450 mg Pb kg⁻¹ (DEFRA and
129 Environment Agency, 2002; Knight, 2004). Investigation into the area's history revealed there
130 had been a Pb rolling mill and smelter in operation until the late 19th century in the location
131 where homes now stand. As the homes were built before any contamination assessment
132 development controls, residents were not aware of the high levels of Pb in the area. Since
133 concentrations of Pb in these domestic gardens were well above the UK trigger levels,
134 remediation was later undertaken. Such findings raise the concern of other possible contaminated
135 sites in Sheffield, especially where there is the potential for ingestion of elevated TTEs via own-
136 grown foods. In a geochemical survey of Sheffield to identify metal pollution across the city,
137 where gardens were tested, all TTE concentrations exceeded their Soil Guideline Value (SGVs)
138 for residential land use with plant uptake (Rawlins et al., 2005). In Sheffield alone, there are
139 more than 70 allotment sites with over 3,000 plots (Sheffield City Council, 2017).

140

141 *2.2. Allotment site selection*

142 Initially the project was designed to sample the allotments owned by Sheffield City
143 Council, however, access to these sites was denied. Privately owned allotment sites were then

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

158

159 *2.3. Soil sampling and analysis*

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

164 Core samples were collected from each plot, from a depth of 0 to 20 cm using a small
165 hand auger, avoiding the edges of individual plots. Soil samples were prepared and analysed
166 using standard procedures for soil bulk density, soil texture, pH (deionized H₂O and CaCl₂
167 extracts), electrical conductivity (EC), and total C and N. Total C and N were measured by an
168 elemental analyser (Vario El CubeCN, Elementar, Germany). A Delta-50 X-ray fluorescence

169 spectrometer (XRF) was used in the benchtop workstation to provide a rapid simultaneous
170 measurement of TTE concentrations in soil samples. Each sample was analysed using three
171 beams (50 kV, 40 kV, and 15 kV); each beam was run for 60 seconds. XRF was performed in
172 triplicate, with a total of 9-minute run per sample, and the average metal concentration values of
173 each beam were used for the analysis.

174

175 *2.4. Plant sampling and analysis*

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

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

189

190 Onions selected for analysis were chosen away from the borders of the plot to avoid
191 samples with potential contamination from factors outside the plot. The entire plant was removed
192 carefully with a hand trowel. Each onion sample was washed with tap water as to simulate

193 household cleaning practices. Any visible soil particles were washed away. Samples were first
194 air dried, and then at 70 °C in a hot-air oven for at least one week. Once dried, non-edible parts
195 of the plants were removed according to their classification as mentioned above (ASO or SSO),
196 and the remaining edible parts of each sample was milled using a ball mill, creating a
197 homogenous mixture. Acid digestion was performed using EPA Method 3050B (SW-846) (EPA,
198 1996), and the extracts were analysed for TTEs using inductively coupled plasma mass
199 spectrometry (ICP-MS, Model DRC 11, Pelkin Elmer, USA).

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

204

205

206 2.5. Human health risk assessment

207 To evaluate the impact of onion consumption with potentially elevated levels of TTEs on
208 human health, a risk assessment was performed. Exposure to TTE depends on the concentration
209 of the element in the food and the daily food consumption rate. Estimated daily intake (*EDI*) can
210 therefore be calculated using Eq. 1 (Chamannejadian et al., 2013; Hang et al., 2009; Zheng et al.,
211 2007):

$$212 \quad EDI = \frac{C \times Con}{Bw} \quad (\text{Eq. 1})$$

213 Where, *EDI* ($\mu\text{g kg}^{-1} \text{Bw}^{-1} \text{day}^{-1}$) is the amount of TTE consumed; *C* ($\mu\text{g g}^{-1}$) is the
214 concentration of TTE in onion; *Con* ($\text{g person}^{-1} \text{day}^{-1}$) is the average daily consumption of
215 vegetables in the UK, assuming worst case scenario that all vegetables consumed were raw
216 onions ($20 \text{ g person}^{-1} \text{day}^{-1}$ for males, $38 \text{ g person}^{-1} \text{day}^{-1}$ for females (Bates et al., 2014); *Bw* is

217 the average body weight (83.6 kg for males and 70.2 kg for females, Office for National
218 Statistics, 2010).

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

$$224 \quad THQ = \frac{EDI}{RfD} \quad (Eq. 2)$$

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

$$230 \quad HI = \sum_{n=1}^i THQ_n \quad (Eq. 3)$$

231

232

233 2.6. Data analysis

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

236

237 3. Results

238 3.1. Soil physiochemical properties

239 The bulk density, pH, EC and C:N ratio are presented in Table 1. Soil textures were
240 either silty loam or sandy loam. Bulk densities were generally low as crops were grown with

241 soil-garden compost mixture. All soils were between the pH ranges of 5.6-6.9 (pH with CaCl₂),
242 the lowest mean pH was found in OBA (mean pH=5.6). The soil with the highest EC was in plot
243 1 in BSG (808.3 $\mu\text{S cm}^{-1}$), and the lowest in plot 1 in OBA (419.3 $\mu\text{S cm}^{-1}$). The average percent
244 C contents was 11.04 %, but varied from 6.18 to 11.88 %. The C:N ratio varied from 14.85 to
245 22.25.

246

247 *3.2. TTE concentrations in soils*

248 Three soil samples from each plot were averaged to determine mean TTE concentrations.
249 The soil Pb, Zn, Cu, Cr and As concentrations are represented in Figure 1 (a-e) for comparative
250 purposes and to identify outliers, as the data was not symmetrically distributed. The data from
251 each plot are also presented as Supplementary Information (SI: Table 1), which includes
252 background pH and TTE concentrations in England, the UK Soil Guidelines Values (SGV), and
253 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 ₂ O)	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

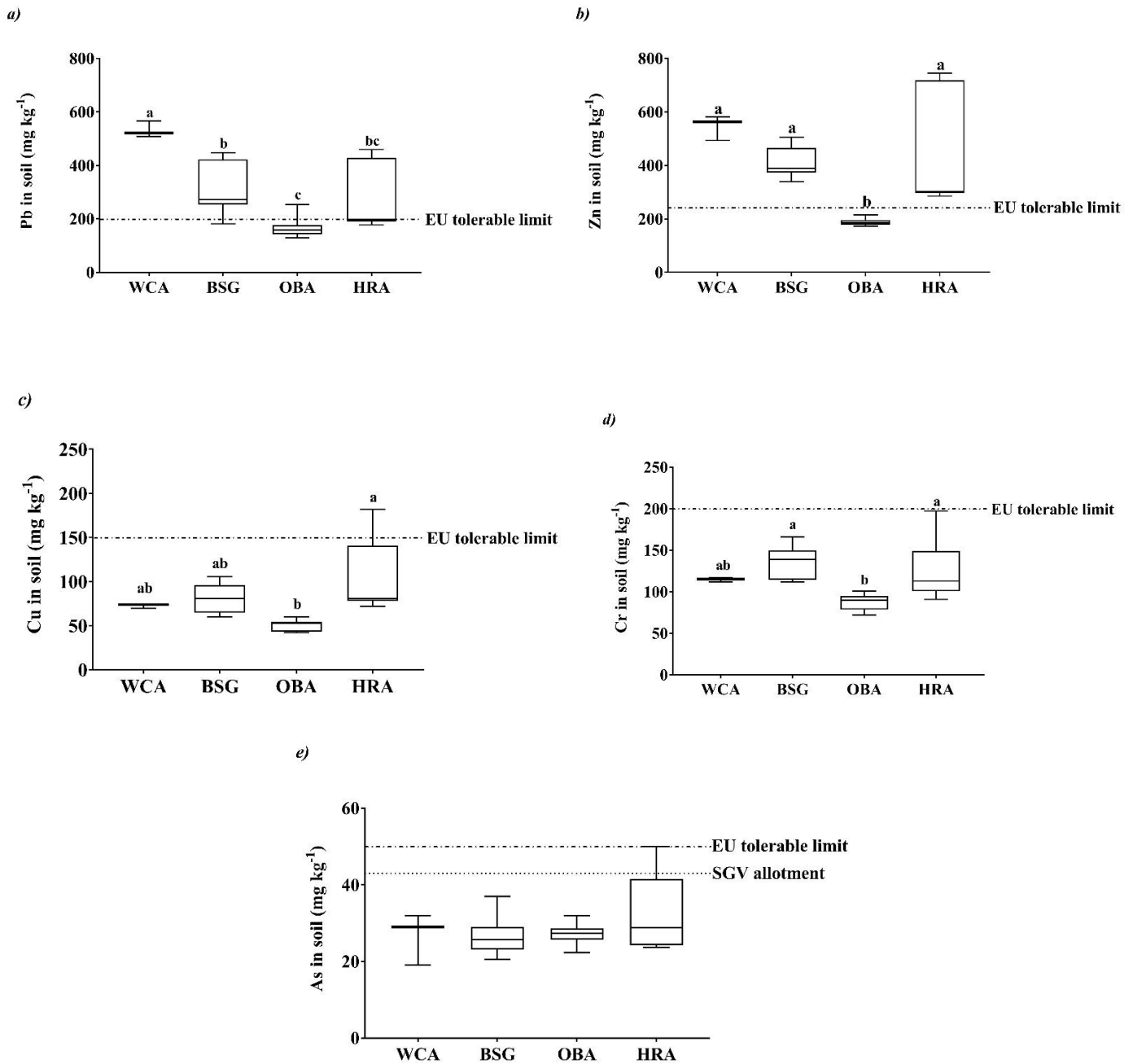


Figure 1a-e. Boxplots showing median, 25th and 75th percentiles of TTE in soil (mg kg⁻¹). Allotments are WCA Well Community Allotments, BSG Brightside Gardens, OBA Oughtibridge Allotments, HRA Handsworth & Richmond Allotments. Note the Y-axis scale differs between graphs. Significant differences ($p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

254

255

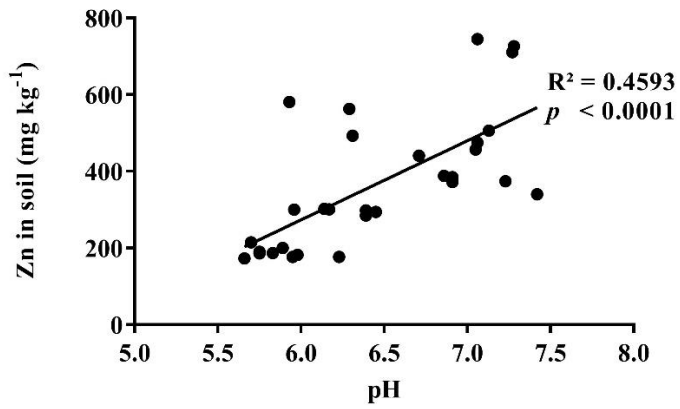
256 The SGVs are set by the UK to assess the risk to human health from contaminated soil exposure. SGVs differ
257 based on land usage, with the SGVs for allotments used here for comparison. The previous SGV for As has
258 been modified from 20 mg kg⁻¹ to 43 mg kg⁻¹ in the current guidelines (DEFRA and Environment Agency,
259 2002; Environment Agency, 2009). Only plot 2 in HRA had As concentration above the current SGV (43 mg
260 kg⁻¹). All allotments had concentrations above the previous As SGV (20 mg kg⁻¹). The previous Pb SGV was
261 450 mg kg⁻¹, of which plot 1 at WCA had concentrations above this limit. Plot 2 at HRA, and plots 2 and 3 at
262 BSG exceeded the previous SGV for Cr (130 mg kg⁻¹). No new guidelines were issued, and these previous
263 SGVs have now been withdrawn (DEFRA and Environment Agency, 2002).

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

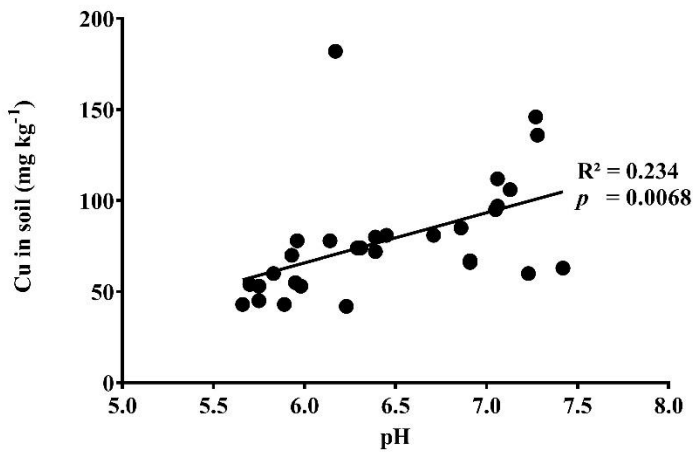
274 All data sets were confirmed to be normally distributed. Differences in mean metal concentrations
275 between the four allotments were analysed using SPSS one-way ANOVA. Concentrations of Pb between
276 allotments were significantly different ($p < 0.05$), the same was true for Zn, Cu, and Cr between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

278 The data showed a positive relationship between soil pH (in H₂O) and concentrations of Zn, Cu and
279 Cr. This is illustrated in Figure 2a-c, regression coefficients are statistically significant ($p < 0.05$). Multiple
280 regression

a)



b)



c)

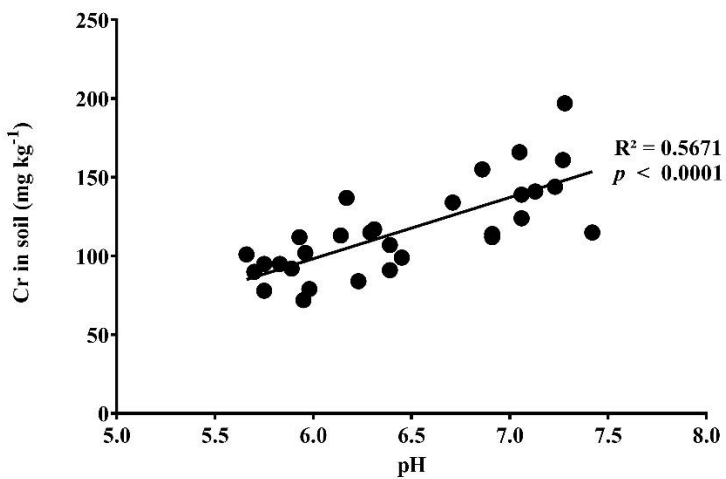


Figure 2a-c. Statistically significant relationships ($p < 0.05$) between the soil concentration of Zn (a), Cu (b) and Cr (c) and soil pH (in H₂O) ($n=30$).

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

284

285 3.3. TTE concentrations in plants

286 The Joint Food and Agriculture Organisation/World Health Organisation (FAO/WHO) Expert
287 Committee on Food Additives (JECFA) identify maximum levels for contaminants and toxins in foods for Pb
288 and As (FAO/WHO, 2011a), and are presented for comparison in SI: Table 2. Figure 3 depicts TTE
289 concentrations in plants. Assessment of WCA TTE concentrations in relation to the other allotments is
290 limited as only one plot at this allotment was tested. HRA plots 1 and 3 are ASO, while all others are SSO.
291 All metals, except As, were lower in ASO than SSO. BSG had the highest mean concentrations of Pb in
292 plants (0.23 mg kg^{-1}). Mean Zn concentrations in plants were similar between allotments and ranged from
293 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
294 detected in WCA nor in plot 2 at OBA and mean As concentration was the highest in plants at HRA.
295 Concentrations of Cr ranged from 0.02 mg kg^{-1} to 0.34 mg kg^{-1} with concentrations 15 times higher in SSO
296 than ASO. Six plots had plants with Pb levels higher than the FAO/WHO guidelines of 0.10 mg kg^{-1} . No
297 plants had levels higher than the FAO/WHO guidelines for As (0.10 mg kg^{-1}).

298

299 3.4. Concentration Factor (CF)

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

305 TTE uptake between ASO and SSO was also compared (Figure 4a-c). This was done to interpret
306 metal uptake in relation to time spent in the ground, as well as to examine localisation of metals in the plant,
307 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

^aND, None Detected

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

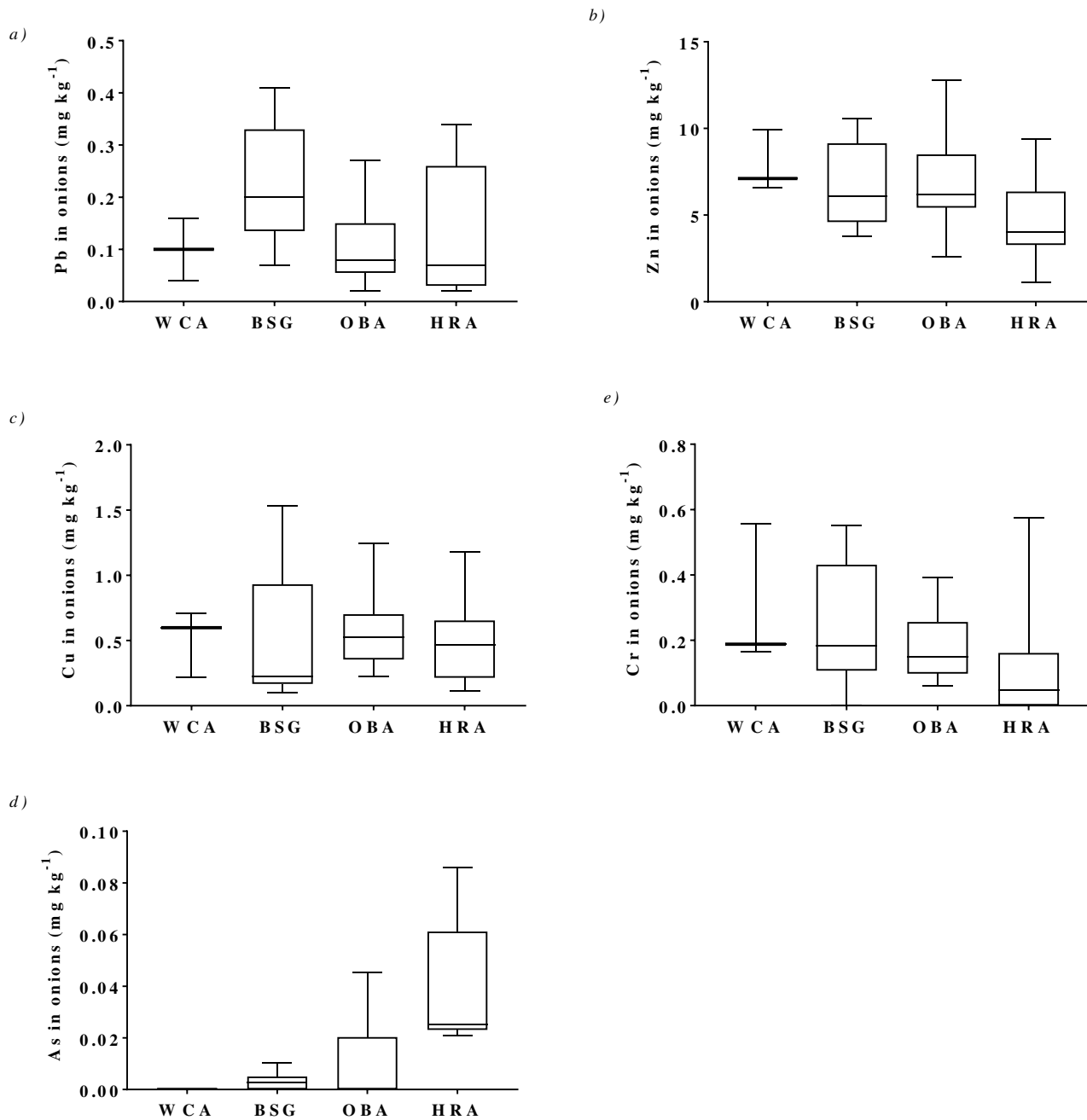


Figure 3a-e. Boxplots showing median, 25th and 75th percentiles of TTE concentrations in plants (mg kg⁻¹, fw). Allotments are *WCA* Well Community Allotments, *BSG* Brightside Gardens, *OBA* Oughtibridge Allotments, *HRA* Handsworth & Richmond Allotments. HRA values include ASO and SSO data grouped together.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

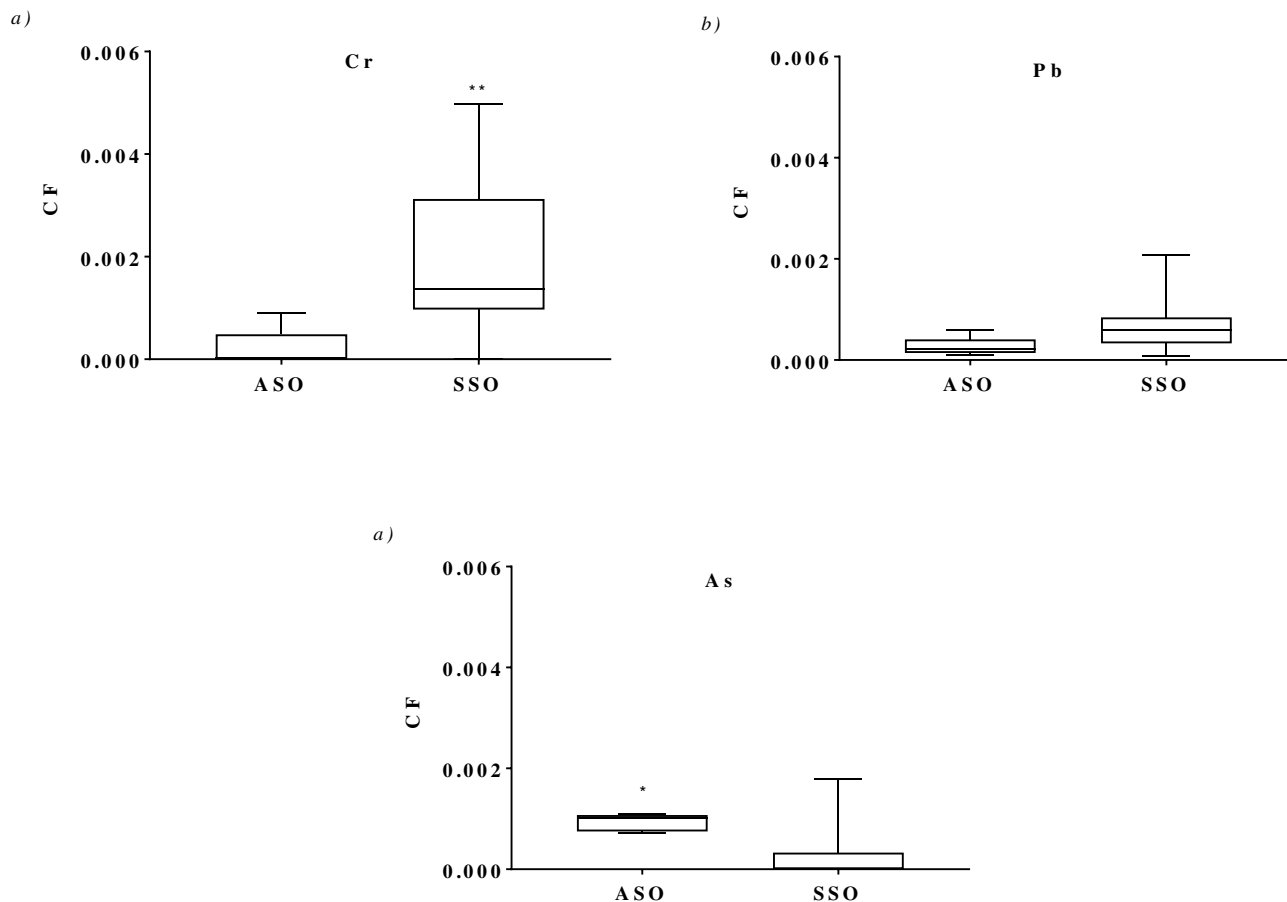


Figure 4a-c. Boxplots showing median, 25th and 75th percentiles of significant different CF for Pb, As, and Cr, in ASO compared to SSO. Pb and Cr were significantly higher in SSO ($p < 0.05$), while As was significantly higher in ASO ($p < 0.05$). Significant differences are indicated using symbols (*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

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

314

315 3.5. Potential health risk

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

322

323 4. Discussion

324 4.1. Soil metal contamination in allotments

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

331 The EU tolerable limits set regulations on concentration levels allowed for TTEs depending on land
332 usage and states the need for action if surpassed. The EU tolerable limits for all non-industrial land use are
333 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	

^aReference oral dose ($\mu\text{g kg}^{-1} \text{day}^{-1}$)

^bEstimated daily intake ($\mu\text{g kg}^{-1} \text{day}^{-1}$)

^cTarget Hazard Quotient (EDI/RfD)

^dHazard Index when multiple metals are present ($\sum \text{THQ}_n$; $n = 1 \text{ to } i$)

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

336 Other UK allotment TTE concentrations are inconsistent and vary depending on location. A study of
337 Pb in allotment soils in a London borough revealed concentrations that ranged from 513 to 2,910 mg kg⁻¹
338 (Prasad and Nazareth, 2000), whereas in Bristol the median Pb concentration was 210 mg kg⁻¹ (Giusti,
339 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
340 23.1 mg kg⁻¹ Cr (Giusti, 2011). Such results are consistent with concentrations found in this Sheffield study,
341 however Cr concentrations in the Sheffield allotments were more than three times greater than those found in
342 Bristol.

343 The largest survey of garden soils carried out by Culbard et al. (1988) analysed 4,650 garden soils
344 around Great Britain. Results indicated levels of Pb, Cu and Zn were elevated, with a mean Pb concentration
345 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
346 mining history. A soil geochemical survey of Sheffield determined median Pb levels to be 161 mg kg⁻¹, and
347 where samples were from the domestic garden, all Pb concentrations were above the SGV of 450 mg kg⁻¹
348 (Rawlins et al., 2005). As previously discussed, concerning levels of Pb, as high as 14,863 mg kg⁻¹, were
349 found in domestic gardens in a Sheffield neighbourhood that was previously a Pb rolling mill (Knight, 2004).
350 Such findings indicate the need for site specific risk assessment.

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

356

357

358

359 4.2. Plant uptake and CF

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

371 Though it was not shown here, a significant correlation was detected in As in soil to As in plant
372 ($R^2=0.4143$). This is similar to other results especially those found in the derivation of SGVs for arsenic in
373 relation to root vegetables (Environment Agency, 2009; Zandsalimi et al., 2011). No other soil-plant
374 concentration correlations were significant. Lack of linearity between plant and soil concentrations of the
375 other metals can be explained by many factors influencing metal uptake by plants and complexity of ion
376 transfer. Variables such as soil physicochemical properties, applied fertilisers, the type of plant species, etc.,
377 can influence these uptake rates (Chen et al., 2016; Chojnacka et al., 2005; Liu et al., 2015). Furthermore,
378 uptake of metals may result from sources other than soil, such as polluted air and water. Air contaminants are
379 less likely to affect root vegetables such as the ASO but may contribute to SSO contamination as the stems
380 are part of the edible vegetable. Contaminated particulates in the air have been documented to increase levels
381 of TTEs in plants (Antisari et al., 2015). This is especially important when examining Pb contamination as
382 combustion of leaded petrol in vehicles and coal combustion has led to increased atmospheric deposition and
383 remains the most significant source of Pb contamination in vegetables (Barraclough, 2007; Hough et al.,

384 2004). Hand washing of vegetables also does not remove all soil particles and may contribute TTE
385 concentrations of the onions. The maturity levels of the vegetable and different consumed parts also may
386 explain the different observed concentrations of TTEs between the two groups. A noted limitation was also
387 the lack of certainty on the onion cultivars used in the study and the small sample size of ASO. The soil TTE
388 concentrations could have been different in these two plots as compared to the other SSO plots and could
389 have had an influence on the results.

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

404 The TTE concentrations in plants in this study are consistent with other reports of metal transfer
405 capabilities by plants in contaminated soils (Augustsson et al., 2015; Chojnacka et al., 2005; Dinelli and
406 Lombini, 1996). The CF order of metal uptake from soil to plant was Zn>Cu>Cr>Pb>As, which is well
407 documented (Alexander et al., 2006; Chen et al., 2014; Intawongse and Dean, 2006; Islam et al., 2015; Wang
408 et al., 2012; Xian, 1989). Fytianos et al. (2001) found much greater CFs, for example, CF in onions grown in

409 contaminated soils were 0.35, 0.31, and 0.21 for Zn, Cu and Pb, respectively. However, this is still consistent
410 with previous results that Pb is especially known to not be readily taken up by plants, due in part to its
411 stronger adhesion to the soil matrix (Barraclough, 2007; Gebrekidan et al., 2013; Holmgren et al., 1993).
412 Such relationships were seen in this study where the WCA, which had the highest levels of Pb in the soil, had
413 some of the lowest concentrations of Pb in the plants. Augustsson et al. (2015) similarly found that root
414 vegetable metal concentrations were only moderately elevated, despite high concentrations in the soil where
415 they were grown, and most vegetables were below food contaminant legislation levels.

416 417 *4.3. Consumption and health risk assessment*

418 TTE concentrations, dry weight, in plants were converted to fresh weight (fw) basis using recorded
419 moisture contents (US EPA, 1997) for better comparison to FAO/WHO food standards (FAO/WHO, 2011a).
420 Six plots had onions that exceeded the FAO/WHO maximum level of Pb in root vegetables of 0.10 mg Pb kg⁻¹
421 fw (FAO/WHO, 2011a). FAO/WHO also previously suggested a provisional tolerable weekly intake
422 (PTWI) for adults of 0.025 mg Pb kg⁻¹ body weight (FAO/WHO, 2011b). In considering the amount of Pb
423 per plant and consumption rates, an unreasonable quantity of onions would need to be consumed per week to
424 reach this PTWI. This PTWI has now been withdrawn as recent dose-response analysis does not indicate any
425 threshold to be health protective for effects of Pb, and FAO/WHO have not established a new PTWI. Levels
426 of As concentrations in plants were well below the FAO/WHO maximum level for contaminants in foods of
427 0.10 mg As kg⁻¹ (FAO/WHO, 2011a). The highest concentration of As was the SSO at HRA (0.069 mg kg⁻¹).
428 This is consistent with other studies in which contaminated sites, even with significant soil pollution, did not
429 yield vegetables with intakes above tolerable daily intakes (Augustsson et al., 2015; Beccaloni et al., 2013;
430 Chen et al., 2014; Fytianos et al., 2001; Pelfrène et al., 2013; Wang et al., 2012).

431 Bioavailability and human exposure are often hard to assess as metals may be present in forms not
432 harmful to the environmental or to human health. It is difficult to assess total exposure as the pathways of
433 TTE intake may also include contaminated drinking water, oral soil ingestion, and consumption of other

434 contaminated food source. Within the body, many factors such as absorption in the gastrointestinal tract and
435 stomach acidity also affect exposure. Uptake from the gastrointestinal tract may differ according to element
436 species and to plant type (Augustsson et al., 2015; Intawongse and Dean, 2006). The actual harm posed to
437 allotment holders growing crops in soils with elevated levels of TTEs is likely to be minimal. Allotment
438 holders typically do not solely consume food grown in their allotments, which makes risk assessment more
439 difficult. Other variables may likewise affect contamination and metal exposure, for instance, vegetable
440 preparation and type of vegetable consumed vary in washing and peeling procedures to remove soil particles,
441 and absorb different amounts of metals (Alexander et al., 2006; Intawongse and Dean, 2006).

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

450

451 **5. Conclusions**

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

457 Following this initial screening, further detailed assessment of these areas can be completed in the
458 future, especially in those soils that exceeded limit guidelines. Also, as soil metal concentrations vary greatly

459 according to previous land use, site-specific risk assessments should be conducted, especially in zones with
460 past industrial history that are now used for gardening purposes.

461

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468

469 **Appendix A. Supplementary material**

470 Supplementary information can be found in the online version.

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