

1 **Assessment of potentially toxic elements in vegetables cultivated in urban and peri-urban sites in**  
2 **the Kurdistan region of Iraq and implications for human health**

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9 assessment

10 **Abstract**

11 Vegetable fields in and around urban areas in the Kurdistan region of Iraq may have higher than  
12 background concentrations of potentially toxic elements (PTEs) from contamination sources including  
13 municipal waste disposal and waste water used for irrigation. The purpose of this study was to assess PTE  
14 concentrations in soils and the edible parts of field-grown vegetables to quantify potential health risks to  
15 the local population. In this survey, 174 soils and 26 different vegetable and fruit types were sampled from  
16 15 areas around Sulaymaniyah and Halabja cities. Sampling was undertaken from fields in urban, peri-  
17 urban and rural locations including sites close to areas of waste disposal.

18 The soils are calcareous (pH 7.67 - 8.21) and classified as silty loam, sandy or silty clay with organic matter  
19 content between 6.62 and 11.4%. Concentrations of PTEs were typically higher in waste disposal areas  
20 compared with urban, peri-urban and rural areas. Pollution load indices (PLI) suggested that agricultural  
21 soils near waste disposal sites were contaminated with some trace elements. Potentially toxic element  
22 concentrations in vegetables were highly variable. Higher total concentrations of PTEs were measured in  
23 vegetables from the waste areas with decreasing concentrations in urban, peri-urban and rural areas. Risks  
24 to human health were assessed using hazard quotients (HQ). Vegetable consumption poses no risk for  
25 adults whereas children might be exposed to Ni, As and Cd. Although HQs suggest elevated risk for  
26 children from consumption of some vegetables, these risks are likely to be lower when realistic dietary  
27 consumption levels are considered.

28

29 **Introduction**

30 Urban agriculture is a primary source of income and nutrition for some populations and, used strategically,  
31 can increase urban sustainability, particularly in developing countries. Expansion of urban areas can result  
32 in considerable alteration of the natural environment, affecting water quality and quantity and increasing  
33 the accumulation of waste materials. In developed countries, urban and peri-urban agriculture (UPA) may  
34 be practiced in specific areas, including allotments and community vegetable gardens, while in developing  
35 countries UPA is mainly practiced in informal areas such as, rear and front gardens, road verges and on  
36 waste ground. UPA can be important for food production, strengthening communities, and helping to  
37 reduce both socio-economic and environmental problems. It can also pose health risks if cultivation takes  
38 place on sites contaminated with potentially toxic elements (PTEs) and organic contaminants (Alloway,  
39 2004, Clark et al., 2006). Exposure pathways include direct ingestion, dermal contact and inhalation (Wang  
40 et al., 2011, Gebrekidan et al., 2013). In countries where water is scarce, wastewater (sometimes diluted  
41 by freshwater sources) is often used for irrigation.

42 Vegetables play an important role in providing essential dietary nutrients. However, vegetables and crops  
43 grown in urban and peri-urban areas generally contain higher concentrations of PTEs and other pollutants  
44 compared with crops and vegetables grown in rural areas (Gupta et al., 2019). Although some trace metals  
45 act as micro-nutrients to maintain normal body function, exposure to elements such as Cr, Cu, As and Zn  
46 can result in neurologic disorders, headaches, renal and liver diseases when they exceed safe limits (Lin et  
47 al., 2013). There is also evidence that long-term exposure to low doses of some PTEs may increase the  
48 incidence of cancer. Itoh et al. (2014) found that dietary Cd intake through ingestion of contaminated rice  
49 and other vegetables was directly related to the incidence of postmenopausal breast cancer. Increased risk  
50 of lung cancer has been observed as a result of occupational exposure to mists and dusts containing  
51 hexavalent chromium (Liu et al., 2013). Zhao et al. (2014) report a statistically significant correlation  
52 between top soil Pb concentration and gastric cancer, and between Hg in grain and liver cancer. In the  
53 Sulaymaniyah province of northern Iraq it is estimated that fresh fruits, grains and vegetables are grown in  
54 as many as 2000 locations including rural, urban and peri-urban areas, some of which are contaminated  
55 with PTEs and other substances. The produce grown at these sites is sold as a component of daily meals  
56 eaten by more than 500,000 urban residents.

57 The objective of this study was to provide a first comprehensive survey of soil, water and vegetation  
58 concentrations of major and trace elements in Sulaymaniyah province and to establish whether elevated  
59 concentrations are present in food crops that might pose a risk to human health. Soil, water and produce  
60 samples were collected from sites across the province from agricultural fields in urban, peri-urban and rural  
61 areas and close to sites of municipal waste disposal.

## 62 **Materials and methods**

### 63 *Sampling*

64 The Kurdistan region of Iraq is mountainous with calcareous soils. It borders Syria to the west, Iran to the  
65 east and Turkey to the north. The climate is semi-arid continental with hot and dry summers (35-48°C) and  
66 cold and wet winters (7-13°C). Soil and water samples were initially collected from areas located in or  
67 around the cities of Sulaymaniyah, Halabja, Kalar, Sirwan and Khurmal (Table 1, Figure 1). Subsequently,  
68 paired plant and soil samples were collected around the cities of Sulaymaniyah and Halabja where  
69 cultivation for subsistence and/or commercial consumption is practiced.

70 Composite top soil (1-15 cm) samples (c.1 kg) were collected to be representative of different fields within  
71 each sampling area. Each sample was collected using a clean stainless steel trowel and placed in a plastic  
72 bag for transport to the laboratory before being air dried. Water pH and electrical conductivity (EC) were  
73 measured in the field at the time of sampling. Water samples for trace element analysis were immediately  
74 filtered (< 0.45 µm) before being preserved by acidification to 2% HNO<sub>3</sub>. Plant samples were placed in  
75 paper bags and transported to the laboratory on the day of sampling.

### 76 *Sample Processing*

77 Dry soil samples were gently disaggregated using a pestle and mortar and sieved to obtain the <2 mm  
78 fraction. Soil pH was determined using 5 g of <2 mm sieved, air-dried soil after suspension in 12.5 mL of  
79 distilled water and shaking at 40 rpm for 30 minutes. Measurements were made using a Hanna pH-209 pH  
80 meter with combined glass electrode (Ag/AgCl; PHE 1004) calibrated at pH 7.0 and 4.01, allowing 5 minutes  
81 for the reading to stabilize. Loss on ignition (%LOI) was used to estimate the percentage of organic matter  
82 in the samples. A known weight of <2 mm oven-dried soil in a pre-weighed ceramic crucible was placed in  
83 a muffle furnace (Gallenkamp) overnight at 550°C, to combust organic matter. The crucibles and  
84 combusted soil were then placed in a desiccator to cool before weighing and calculation of %LOI. A  
85 portion of <2 mm sieved and homogenised soil was finely ground using an agate ball mill (Retsch Model  
86 PM400, Germany) before digestion with 70% hydrofluoric acid, nitric acid and perchloric acid (Trace  
87 Element Grade (TEG), Fisher Scientific, UK) in a teflon-coated graphite block digester (Analysco, UK) using  
88 PFA digestion vessels. The digested samples were diluted to 50 mL using MilliQ water and stored in PTFE  
89 bottles (5% HNO<sub>3</sub>) pending elemental analysis. All digests were diluted 1:10 with MilliQ water immediately  
90 prior to analysis.

91 The fresh weight (FW) of plant samples was recorded as soon as possible after sampling before  
92 approximately half of each sample was washed with tap water and then thoroughly rinsed in distilled water  
93 to remove surface soil contamination. The remaining plant material was left unwashed. Washed and  
94 unwashed portions were oven dried at 70°C for 72 h and re-weighed to determine dry weight (DW).

95 Samples were finely ground in an ultra-centrifugal mill (Retsch Model ZM200, Germany) fitted with a 0.5  
96 mm titanium screen. Ground material (200 mg) was digested in pressurised PFA vessels in 6.0 mL of 70%  
97 Trace Analysis Grade (TAG, Thermo-Fisher Scientific, UK) HNO<sub>3</sub> with microwave heating (Anton Paar  
98 'Multiwave'). Digested samples were diluted to 20 mL using MilliQ water and stored pending elemental  
99 analysis. Immediately before analysis, samples were diluted 1:10 with MilliQ water.

#### 100 *Elemental Analysis*

101 Elemental analysis was undertaken using an ICP-MS (Model X-Series II, Thermo-Fisher Scientific) operated  
102 in 'collision cell mode' (7% hydrogen in helium) to reduce polyatomic interferences. Samples were  
103 introduced from an autosampler (Cetac ASX-520) through a concentric glass venturi nebuliser (flowrate c.  
104 0.8 mL min<sup>-1</sup>). Internal standards were introduced to the sample stream via a T-piece and included Rh (20  
105 ng mL<sup>-1</sup>) and Ir (10 ng mL<sup>-1</sup>) in 2% TAG HNO<sub>3</sub>. External multi-element calibration standards (Claritas-PPT  
106 grade CLMS-2, Certiprep/Fisher) were used for calibration. Sample processing was undertaken using  
107 Plasma lab software (version 2.5.4; Thermo-Fisher Scientific) configured to employ separate calibration  
108 blocks and internal cross-calibration where required.

#### 109 *Quality Control*

110 Quality control was assessed by digestion of NIST Standard Reference Material (SRM 2711 Montana soil)  
111 and (SRM 1573a tomato leaf) alongside soil and plant samples to confirm data quality. Recoveries (%) were  
112 within the acceptable range of uncertainty for each element.

#### 113 *Pollution load index (PLI)*

114 To assess the degree of contamination around sampling sites a pollution load index (PLI) was calculated  
115 (Jorfi et al., 2017) according to:

$$116 \quad PLI = \sqrt[n]{CF_1 \times CF_2 \times CF_3 \times CF_n} \quad (1)$$

117 Where CF<sub>n</sub> is the concentration factor for an individual element, defined as C<sub>s</sub>/C<sub>b</sub> where C<sub>s</sub> was the  
118 concentration of the element in the sample compared to the background, C<sub>b</sub>. Values of C<sub>b</sub> were the mean  
119 concentrations of individual PTEs observed in samples from rural areas. PLI ≤1 indicates PTE loads close the  
120 rural baseline while PLI >1 indicates PTE contamination.

121 *Assessment of Soil Contamination of Plant Samples*

122 Unwashed plant samples were assessed for extraneous soil contamination using the approach of Mwesigye  
123 et al (2016). The percentage of soil contamination was estimated for each element by comparison with soil  
124 and plant vanadium (V) concentrations:

$$125 \quad P(\%) = \frac{(V_p \times M_s) \times 100}{V_s \times M_p} \quad (2)$$

126 where P (%) is the percentage contamination of the plant with soil particles for a given element (M),  $V_p$  and  
127  $V_s$  are the vanadium concentrations in the plant and soil samples, respectively, and  $M_p$  and  $M_s$  are the  
128 concentrations of a given element in the plant and soil samples, respectively. This approach provides an  
129 approximate percentage of plant contamination with soil particles based on the assumption that there is  
130 no systematic uptake of V by the plant. Although a labile form of V can be present in calcareous soil  
131 ( $HVO_4^{2-}$ ), vanadate ( $VO_4^{3-}$ ) ions are unlikely to be taken up by plants (Joy et al., 2015).

132 *Risk assessment from PTEs in vegetables*

133 Hazard quotients to assess risks to human health from consumption of locally grown produce were  
134 calculated using the USEPA approach (USEPA, 2000):

$$135 \quad HQ_M = \frac{ADD_M}{RfD} \quad (3)$$

136 where  $HQ_M$  is the hazard quotient for a given element,  $RfD$  ( $mg\ kg^{-1}\ d^{-1}$ ) is the reference dose, defined as  
137 the maximum tolerable daily ingestion of an element that has no adverse health impact.  $ADD_M$  ( $mg\ kg^{-1}\ d^{-1}$ )  
138 is the average daily intake of the element:

$$139 \quad ADD_M = \frac{(DI \times C_{FW})}{W_B} \quad (4)$$

140 where DI represents the daily intake of fruit and vegetables ( $kg\ d^{-1}$ ),  $C_{FW}$  is the concentration of an element  
141 in the edible plant material ( $mg\ kg^{-1}\ FW$ ) and  $W_B$  is human body weight (kg). Assumed daily intakes (DI)  
142 were  $0.342\ kg\ d^{-1}\ FW$  for adults and  $0.232\ kg\ d^{-1}\ FW$  for children with body weights of 70 and 16.2 kg,  
143 respectively (Hamad et al., 2014). Reference doses were 0.003, 0.012, 0.04, 0.3, 0.0003, 0.00036 and

144 0.0035 mg kg<sup>-1</sup> day<sup>-1</sup> for Cr, Ni, Cu, Zn, As, Cd and Pb, respectively (WHO, 1982; US EPA Iris Database, 2009;  
145 Environment Agency, 2009a, Environment Agency, 2009b).

#### 146 *Statistical analysis*

147 Descriptive statistics are presented as means, medians, standard errors, differences between means,  
148 minima and maxima. A two way ANOVA was used to analyse concentration differences between sampling  
149 sites. Pearson correlation coefficients and linear regressions were calculated to determine the relationships  
150 between total trace elements in soil and edible parts of vegetables. The data were statistically analysed  
151 using the statistical packages SPSS 17.0 and MINITAB 16.0. A probability (p) <0.05 was considered to be  
152 statistically significant when testing null hypotheses.

### 153 **Result and discussion**

#### 154 ***Soil characterization***

155 Soil properties are given in Table 2. Soil pH typically ranged from 7.67 to 8.21 confirming the calcareous  
156 nature of the soils which have previously been classified as silty loams, sandy and silty clays (Rashid, 2010).  
157 The highest pH values were recorded at sites irrigated with groundwater, springs and drilled wells in the  
158 Anab area of Halabja. Long-term irrigation with waste water has been suggested to decrease soil pH and to  
159 increase the organic matter of top soils (Rattan et al., 2005, Xu et al., 2010) although this pH difference  
160 probably reflects the underlying geology and variation in soil type. The lowest soil organic matter (SOM)  
161 (6.62 and 6.8% LOI) was recorded in fields irrigated with clean water and diluted waste water in the Kalar  
162 river side and Anab area; the highest SOM (11.4% LOI) was in fields near the municipal waste disposal site  
163 in Sulaymaniyah city, which is irrigated with diluted domestic waste water. Major element chemistry  
164 reflects the calcareous geology of the area; calcium is the dominant cation present at significantly greater  
165 concentrations (45900 – 127000 mg kg<sup>-1</sup>) than other cations. Studies in areas of calcareous geology have  
166 shown similar major element profiles (Iqbal and Shah, 2011, Nazif et al., 2015).

167 No local environmental standards exist for trace elements in soils in Iraq therefore concentrations were  
168 compared to UK Soil Guideline Values (SGV), European and WHO guidelines (EU/WHO) (Table 2). Large  
169 standard deviations for some elements demonstrate heterogeneity between soils within the same  
170 sampling area. Total concentrations of individual elements also varied significantly (p<0.01) between study  
171 areas suggesting localized contamination sources. Figure 2 shows trace elements in the Halabja area  
172 compared based on land use (waste, urban, peri-urban and rural areas). Data for Sulaymaniyah City and  
173 other sites are given in Figures S1 & S2. Concentrations of Cu, Zn, Cd and Pb were significantly greater and  
174 more variable in areas subject to waste disposal compared with other urban or rural areas. No obvious  
175 increase was observed for Cr, Ni and As at waste disposal areas. The lowest concentrations of all elements

176 were recorded in the rural areas. Waste disposal and urban activity therefore appears to be increasing the  
177 concentrations of some PTEs in nearby agricultural soils.

178 Concentrations of Cr were frequently above SGVs and EU/WHO guidelines in most soil samples, including  
179 those collected in waste disposal area, but all fell within the typical range reported for calcareous soils of 5  
180 -150 mg kg<sup>-1</sup> (Kabata-Pendias and Mukherjee, 2007). Soils from Anab (n=9) had significantly lower Cr  
181 concentrations which may reflect lower geological background concentrations. Chromium concentrations  
182 were similar to those in a study conducted in Damascus (Syria) by Möller et al. (2005) who observed that  
183 the main sources of Cr in soils at their sampling sites were geogenic as concentrations were close to  
184 “pedogeochemical” background values.

185 Nickel concentrations varied from c.75 mg kg<sup>-1</sup> in the waste area in Halabja (n=3) to c.185 mg kg<sup>-1</sup> at Kalar  
186 Riverside (peri-urban area, n=11) with all samples exceeding typical concentrations for calcareous soils of  
187 5-20 mg kg<sup>-1</sup> (Kabata-Pendias and Mukherjee, 2007). Concentrations also all exceeded the EU standard of  
188 50 mg kg<sup>-1</sup>. Although greater than typical concentrations in calcareous soils elevated Ni concentrations  
189 were observed by Habib et al. (2012) who investigated the trace element content of 25 calcareous soil  
190 samples in Baghdad city (Iraq). They reported Ni concentrations ranging between 105 - 210 mg kg<sup>-1</sup>, similar  
191 to those observed in this study, which may suggest that geogenic Ni concentrations in Iraqi soils are greater  
192 than in most calcareous soils. Copper and Zn concentrations were lower than the permitted UK sludge  
193 limits for agricultural soil (50 and 300 mg kg<sup>-1</sup>) and EU standards (100 and 300 mg kg<sup>-1</sup>), respectively, except  
194 at the waste disposal site in Halabja (Figure 2) where higher mean concentrations were recorded (263 and  
195 773 mg kg<sup>-1</sup>, respectively). Mean concentrations of As, Cd and Pb were below SGVs and EU limits except for  
196 Cd and Pb in the waste disposal area of Halabja. Concentrations of these elements in rural areas were  
197 generally lower than those in urban, peri-urban and waste.

198 Pollution load indices were used to assess overall trace elemental contamination in the study areas. Values  
199 for Sulaymaniyah and Halabja and soils from other sites are shown in Figure 3. The PLI values in Halabja city  
200 were higher than for Sulaymaniyah city under all land uses but especially at the waste disposal area which  
201 was clearly more polluted than the other sampling areas. Values of PLI in Sulaymaniyah city are centred  
202 around ‘safe’ limits except for soils collected around the waste area. Both cities had greater PLI values than  
203 for soils collected in other areas of the province. Overall the data also indicate some PTE contamination at  
204 waste disposal sites in both cities and in agricultural fields near waste disposal sites. Urban and peri-urban  
205 areas also had slightly elevated soil PTE concentrations compared with rural sites.

206 ***Trace element concentrations in fresh and waste waters***

207 Physico-chemical properties of water samples are given in Table 3. All samples are within the safe pH range  
208 recommended for irrigation waters (pH 6-9). The EC of the waters was typically in the range  
209 500-600  $\mu\text{S cm}^{-1}$  with the exception of waste water in the Halabja area (c. 950  $\mu\text{S cm}^{-1}$ ). Trace element (e.g.  
210 Co, Ni, Cu) concentrations are slightly greater in the waste waters compared to the fresh waters but are  
211 well below the maximum concentrations recommended for irrigation water (Ayers, 1994) indicating that  
212 the use of these waters for irrigation purposes is not likely to result in elevated toxic element  
213 concentrations in the soils or plants.

#### 214 ***Accumulation of trace elements in vegetables***

215 The mean concentrations of PTEs in 26 types of unwashed and washed fruits and vegetables are presented  
216 in Table 4. A significant variation between PTEs in all sample types was clear ( $p < 0.05$ ). The greatest  
217 concentrations of Al, V, Cr, Fe, Ni, Se, Cd, Pb and U were 4653, 9.43, 14.7, 3392, 10.6, 0.196, 0.892, 1.91  
218 and 0.062  $\text{mg kg}^{-1}$  (dry weight), respectively, in unwashed radish leaves while the highest concentration of  
219 Mn was 1745  $\text{mg kg}^{-1}$  in vine leaves. Cobalt was highest (2.88  $\text{mg kg}^{-1}$ ) in washed cress, Cu was highest  
220 (17.0  $\text{mg kg}^{-1}$ ) in unwashed tarragon and Zn was highest (53.0  $\text{mg kg}^{-1}$ ) in unwashed okra. These  
221 concentrations are likely due to external contamination of the samples with soil particles although some  
222 trace element uptake by plants is possible.

223 Mean concentrations of PTEs determined in washed and unwashed vegetable samples are compared in  
224 Figure 4. This suggests that, with a few exceptions, there is little difference between PTE concentrations in  
225 washed and unwashed samples. The greatest deviation from the 1:1 line was found for Zn, suggesting a  
226 contribution from particles containing only Zn, especially at greater total concentrations. Many authors  
227 have reported that washing vegetables plays an important role in minimizing trace element concentrations  
228 in produce grown in and around cities and near road verges (Qadir et al., 2000, Itanna, 2002, Sharma et al.,  
229 2008, Nabulo et al., 2010, Ali and Al-Qahtani, 2012, Kumar, 2013) but washing has not reduced trace  
230 element concentrations here and therefore concentrations likely reflect the trace element concentrations  
231 of the vegetables as consumed.

232 Mean concentrations in the most commonly consumed vegetables such as eggplant, pepper, okra, tomato,  
233 cowpea and chard are presented in Figure 4, grouped according to land use. Most of the vegetables can be  
234 consumed raw but some are cooked before eating, which may alter the PTE concentrations. Greatest  
235 concentrations were typically observed in vegetables grown in or around waste disposal sites with  
236 progressively reduced concentrations in urban, peri-urban and rural areas.

237 Concentrations of Cr were all well below the FAO/WHO 'safe limit' of 2.3  $\text{mg kg}^{-1}$  FW based (FAO/WHO,  
238 2001). Nickel and Cu in all vegetable types, in both cities, were greater than UK recommended values of 0.2



239 and 0.5 mg kg<sup>-1</sup> FW, respectively (UK Food Standard Agency, 2009) with the exception of Ni in eggplant,  
240 tomato and cucumber. Nickel and Cu concentrations in this study are similar concentrations in the edible  
241 parts of vegetables observed in a Libyan study (Elbagermi et al., 2013). Concentrations of As, Cd and Pb  
242 also exceeded UK and FAO/WHO standards in selected fruits and vegetables (Figure 5). Arsenic  
243 concentrations in the vegetable samples exceeded the UK limit of 0.005 mg kg<sup>-1</sup> FW in the waste area and  
244 also exceeded this limit in other land use areas for okra, cucumber and chard. Concentrations of As were  
245 however lower than values reported by Nazemi (2012) in vegetables grown in long term waste water  
246 irrigated fields in Iran. Cadmium concentrations exceeded UK limits for many vegetables for all land uses.  
247 Cadmium concentrations in okra in the waste and peri-urban areas were also higher than EU standards for  
248 non-leafy vegetables of 0.05 mg kg<sup>-1</sup> FW (European Commission, 2004). Greatest Cd accumulation was  
249 observed in a sample of celery (0.11 mg kg<sup>-1</sup> FW) and the lowest in samples of cowpea (0.002 mg kg<sup>-1</sup> FW).  
250 Yang et al. (2009) undertook a field trial to investigate Cd accumulation in leek, pak choi, carrot, radish,  
251 tomato and cucumber grown on Cd-contaminated soils. They found that Cd concentrations varied from  
252 0.01 to 0.1 mg kg<sup>-1</sup> FW, which is a similar range to that observed in this study. In general, Pb  
253 concentrations were significantly above the UK limit of 0.01 mg kg<sup>-1</sup> FW in chard collected in all areas of  
254 Sulaymaniyah and Halabja cities. Lower concentrations of trace elements were typically detected in fruits  
255 compared with leafy vegetables, which is in agreement with other studies. For example, Gebrekidan et al.  
256 (2013) observed higher accumulation of Ni and Cd in chard and lettuce compared with fruits of tomato and  
257 green pepper.

258 Ranges of element concentrations within and between vegetable types indicate that trace element  
259 accumulation varies between different species of plants and/or among different plants of the same species  
260 (Säumel et al., 2012, Xu et al., 2013). For example in an assessment of exposure risks associated with urban  
261 horticulture in the city of Berlin, Säumel et al. (2012) observed that leafy vegetables had greater  
262 concentrations of Cu, Zn, Ni compared to fruits, stem and root vegetables. Leaf type and morphology is  
263 likely to play a significant role in determining the extent of trapping of soil particles and the ability to  
264 remove these particles with washing.

### 265 ***Soil contamination of vegetable samples***

266 The extent of soil contamination of the vegetables samples was estimated from Equation 2. Strong  
267 correlations between V and Fe in plants indicated contamination of vegetables with soil particles (Figure 6,  
268  $r = 0.97$  and  $0.93$  for leafy vegetables and fruits, respectively). The greatest contamination by soil particles  
269 was in the unwashed leafy vegetables (leek, celery, purslane, spring onion, tarragon and chard), Table 5.  
270 For example mean proportions of Cr and Co derived from soil particles in washed samples were >40% for  
271 purslane and tarragon, >60% in leek, celery and chard, and >80% in spring onion. Nickel concentrations  
272 attributable to soil particles were typically >30% in washed vegetables. Washing was therefore insufficient

273 to remove all adhering soil particles for many vegetables but probably reflects concentrations a consumer  
274 might be exposed to after washing vegetables in the home.

### 275 ***Health risk assessment***

276 Non-carcinogenic risk from consumption of vegetables was assessed according to Equation 3. Element-  
277 specific HQs were calculated for the most commonly-consumed leaf and fruit vegetables (eggplant,  
278 cowpea, okra, tomato, cucumber and chard) sampled from waste, urban, peri-urban and rural areas for  
279 both adults and children. Results for children are presented in Figure 7. Hazard quotients exceeded 1.0 for  
280 one or more element in pepper, okra, cowpea and chard in samples collected across all land use types. This  
281 suggests that consumption of these vegetables regularly and in large amounts may exposure children to  
282 risk. Calculated HQs for adults were typically below 1.0 with the exception of Cd in okra and chard from  
283 urban areas, Ni in cowpea (excluding rural sites) and Cr chard from waste disposal areas (Figure S3)  
284 suggesting that some specific combinations of metals and crop types may be potentially problematic.

285 Element-specific HQs were calculated for seven commonly-consumed vegetables assuming an equal  
286 dietary contribution from each vegetable. Results for land use types for both adults and children are  
287 shown in Figure 8. A clear influence of municipal waste and urban pollution on the agricultural fields was  
288 observed with HQs typically greater than for peri-urban and rural areas. HQs calculated for adults for this  
289 mixture of commonly consumed vegetables were all < 1.0 suggesting no risk. Values for children suggested  
290 a slight risk from Ni and Cd for city dwellers and possible risks from Cr, Ni, As and Cd if the vegetables were  
291 grown close to waste disposal sites. To refine these findings further a full dietary survey would be  
292 required to understand the relative contributions of specific vegetable types and other foods to the diets of  
293 adults and children in the Kurdistan region of Iraq.

### 294 **Conclusions**

295 Soil contamination as a consequence of municipal waste disposal and urban activities in Sulaymaniyah and  
296 Halabja cities has been clearly identified as a source of PTEs to surrounding areas. Vegetable crops grown  
297 close to waste areas and urban fields contained higher concentrations of PTEs than crops grown in peri-  
298 urban and rural areas. For vegetables in rural areas, most PTEs were found to be below accepted  
299 regulatory limits. A significant proportion of PTE content of vegetables appeared to be derived from soil  
300 dust rather than systemic uptake despite washing of samples. Consumption of vegetables grown close to  
301 municipal waste disposal sites may pose a potential health risk to children but this requires confirmation  
302 following a comprehensive dietary survey that includes other food types.

303

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**Table 1:** Sampling site locations and descriptions.

City	Area	GPS	Total no. sites	Classification	Plant samples	Water samples	Description
<i>Sulaymaniyah</i>	Waste site	35°28'47.51"N 45°25'28.36"E	5	Waste	Y		Municipal waste disposal site located on the outskirts of the city close to agricultural fields. Leachates from the site flows across the surrounding fields. Waste disposed includes, glass, metals, batteries, plastics and organic material.
	Tanjaro	35°48'01.54"N 45°42'07.19"E	11	Peri-urban	Y		Located c. 10 km from Sulaymaniyah city centre. Most vegetables grown in this area are sold in Sulaymaniyah grocery markets. The Tanjaro river is the main source of irrigation water. Domestic waste water channels from the city flow in to this river.
	Kana sura	35.540020"N 45.391216"E	8	Urban	Y		Located in a built-up area, waste water channel flows through the site that is used to grow vegetables.
	Kani goma	35.541090°N 45.370417"E	6	Peri-urban	Y		Close to the conurbation. A waste water channel from Sulaymaniyah city runs through this site. Numerous orchards exist and farmers grow vegetables between the trees.
	Industrial	35.486328°N 45.428052"E	27	Waste	N	4	Located to the south of Sulaymaniyah close to the municipal waste disposal site with a wide range of agricultural produce. Small scale industry e.g. metal workshops and scrap dealers are nearby. Domestic waste water discharges into the Tanjaro river.
<i>Halabja</i>	Kani kolka	35.187476°N 45.987567"E	13	Urban	Y		Located between domestic houses and a hospital. A waste water channel from Halabja city runs through this site and is used as a source of irrigation.
	Kani spi	35.204790°N 45.978693"E	5	Peri-urban	Y		Close to the city. Two waste water channels from Halabja run through the site and are used for irrigation.
	Anab	35.206296°N 46.008673"E	9	Rural	Y		A village c.10 miles east of Halabja. Situated on a wide plain mostly used for growing wheat and barley, but vegetables are also grown in this area. The main source of irrigation is spring and ground water.
	Northwest	35.190031°N 45.979305"E	9	Peri-urban	N	0	Northwest of Halabja. Agricultural fields surrounding waste site
	Waste area	35°12'28.08"N, 45°56'57.65"E	3	Waste	N	0	Northwest of Halabja. Waste disposal site in close proximity to agricultural fields
	North	35°11'25.75"N, 45°59'13.34"E	24	Urban	N	7	North of Halabja, waste water channel collect discharge from industrial and domestic areas including the hospital.
<i>Kalar</i>	Grdagozina	34°35'02.44"N, 45°16'55.18"E	11	Peri-urban	N	4	Southwest of Kalar in a residential area. Agricultural area irrigated with fresh water.
	Riverside	34°36'21.63"N, 45°19'04.63"E	11	Peri-urban	N	4	Agricultural area in the south of Kalar, irrigated with water from the Sirwan river.
<i>Sirwan</i>		35°15'12.74"N, 45°51'31.72"E	24	Peri-urban	N	4	Agricultural fields frequently flooded by the river which receives a combination of waste and fresh water.
<i>Khurmal</i>		35°18'15.83"N, 46° 1'29.88"E	8	Peri-urban	N	3	Northwest of the city in an area where pomegranate orchards predominate. Irrigation is freshwater in some locations and sewerage in others





**Table 3:** Water properties and mean elemental concentrations ( $\mu\text{g L}^{-1}$ ) in fresh and waste waters used for irrigation.

Sites		pH	EC	Na	Mg	K	Ca	Al	V	Cr	Mn	Fe	Co	Ni	Cu	Zn	As	Se	Sr	Cd	Cs	Ba	Pb	U
<i>Sulaymaniyah</i>	Fresh	7.70	557	2460	10100	1560	59700	28.3	5.86	2.35	20.4	31.5	0.25	4.16	1.27	37.1	0.85	1.08	481	0.02	0.01	86.3	0.076	1.02
	Waste	8.00	652	23500	8220	5780	56300	13.8	7.82	2.30	276	26.7	1.51	10.0	1.25	20.8	2.79	0.37	400	0.02	0.01	73.8	0.15	0.44
<i>Halabja</i>	Fresh	7.70	604	3830	11200	890	72900	20.1	3.74	2.49	13.2	26.6	0.13	2.58	0.79	65.0	0.51	0.91	528	0.04	0.00	125	0.23	0.98
	Waste	8.00	946	66800	7200	10110	58800	40.5	2.53	3.07	124	57.5	0.62	5.78	2.66	60.8	1.06	0.73	364	0.07	0.05	106	0.87	0.44
<i>Kalar</i>	Fresh	7.80	603	15500	15200	900	65400	11.2	7.99	3.92	0.90	10.6	0.03	0.85	0.34	15.1	0.39	0.58	1400	0.01	0.00	57.1	0.07	1.82
	Waste	7.70	453	10400	9690	1860	50300	56.1	3.00	2.94	6.76	82.5	0.14	2.91	0.81	19.9	1.19	0.48	607	0.02	0.01	74.5	0.24	0.48
<i>Sirwan</i>	Fresh	7.80	520	15000	13700	950	60900	21.9	8.80	1.02	2.03	18.9	0.04	1.23	0.60	15.5	0.40	0.49	1320	0.02	0.01	50.3	0.14	1.60
	Waste	7.90	620	12600	12600	2160	74000	11.4	5.56	1.59	37.6	19.4	0.39	4.11	0.65	17.4	1.85	0.56	584	0.02	0.01	94.2	0.09	0.64
<i>Khurmal</i>	Fresh	8.20	427	4470	6010	910	58000	13.0	1.38	5.52	1.34	33.0	0.06	4.00	4.02	27.5	5.87	0.16	234	0.08	0.05	32.2	1.36	0.34
	Waste	7.80	505	24500	3880	4490	49700	18.2	1.21	6.86	70.4	49.4	0.36	4.91	1.47	37.9	0.66	0.18	138	0.03	0.02	28.1	0.30	0.24
<i>FAO/WHO*</i>								5000	100	100	200	5000	50	200	200	2000	100		20	10			5000	

\* Recommended maximum concentration in irrigation water applied at a rate of 10, 000 m<sup>3</sup> ha<sup>-1</sup> yr<sup>-1</sup>, FAO/WHO, <http://www.fao.org/3/T0234E/T0234E06.htm>.







<b>Plant</b>		<b>n</b>		<b>Cr</b>	<b>Co</b>	<b>Ni</b>	<b>Cu</b>	<b>Zn</b>	<b>Se</b>	<b>Cd</b>	<b>Ba</b>	<b>Pb</b>
				<b>%</b>	<b>%</b>	<b>%</b>	<b>%</b>	<b>%</b>	<b>%</b>	<b>%</b>	<b>%</b>	<b>%</b>
Dill	Unwashed	1		20.9	41.2	15.7	1.98	1.11	0.98	1.66	6.27	11.1
	Washed	1		50.3	52.5	32.8	3.60	2.16	2.24	3.29	12.7	28.3
Quince	Unwashed	4	Mean:	11.3	12.8	8.4	1.61	1.17	4.43	0.84	5.42	33.4
			SD:	6.4	3.7	2.6	0.44	0.73	2.84	0.28	4.57	24.9
Pomegranate	Peeled	14	Mean:	41.4	54.7	13.9	0.53	0.96	3.39	7.75	41.0	46.7
			SD:	40.9	39.1	10.9	0.38	0.57	2.58	10.23	28.4	38.0
Fig	Unwashed	11	Mean:	22.9	11.3	2.6	0.48	0.63	3.28	1.66	2.98	31.4
			SD:	9.9	5.5	1.1	0.27	0.24	2.43	1.08	2.00	17.0
Eggplant	Unwashed	11	Mean:	12.9	13.0	6.0	0.19	0.32	0.85	0.19	3.71	30.0
			SD:	10.0	13.5	4.9	0.32	0.62	1.09	0.13	3.13	43.3
Courgette	Unwashed	4	Mean:	38.8	13.2	4.99	0.58	0.51	1.49	2.17	19.9	39.2
			SD:	26.5	6.9	3.34	0.61	0.43	1.51	1.66	15.1	37.1
Water Melon	Peeled	4	Mean:	31.2	24.5	8.9	3.86	2.05	1.11	0.73	13.4	37.7
			SD:	26.7	20.7	9.0	6.54	3.50	1.41	1.04	19.1	50.4
Melon	Peeled	3	Mean:	53.6	38.9	16.3	5.79	3.15	2.33	1.08	15.8	39.6
			SD:	20.2	16.3	9.1	5.80	3.27	2.97	0.96	7.7	44.0
Onion	Peeled	2	Mean:	25.1	26.3	4.27	0.303	0.307	0.441	0.434	3.25	19.6
			SD:	6.95	18.6	1.49	0.102	0.015	0.401	0.174	0.339	15.4

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**Figure 3:** Elemental PLI (Cr, Ni, Cu, Zn, As, Cd, Pb, Cd) calculated for the cities of Sulaymaniyah and Halabja according to land use compared to the rural area (Anab) which was used to define background concentrations of PTEs in the soil of the area.

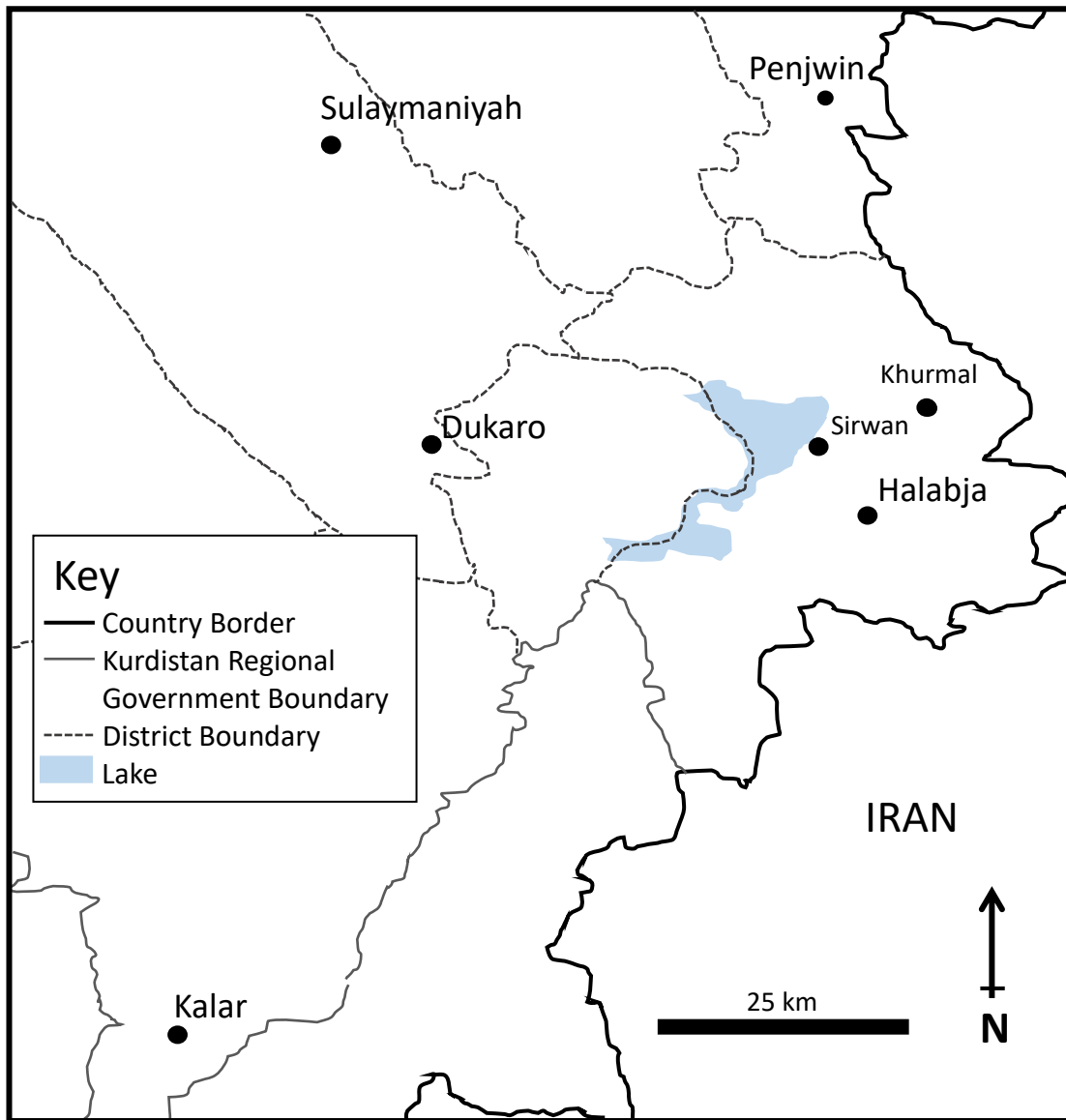
**Figure 4:** A comparison of concentrations of PTE in washed and unwashed vegetables.

**Figure 5:** Mean concentrations of PTEs ( $\text{mg kg}^{-1}$  FW) in unwashed eggplant, pepper, okra, tomato, cowpea and chard grown in waste, urban, peri-urban and remote areas in both Halabja and Sulaymaniyah cities. Vertical bars related with each histogram show standard errors of the means ( $n=3$ ). The solid and dashed lines denote UK and WHO/EU standards, respectively.

**Figure 6:** Correlations between Fe and V concentrations in (a) leafy vegetables and (b) fruiting vegetables.

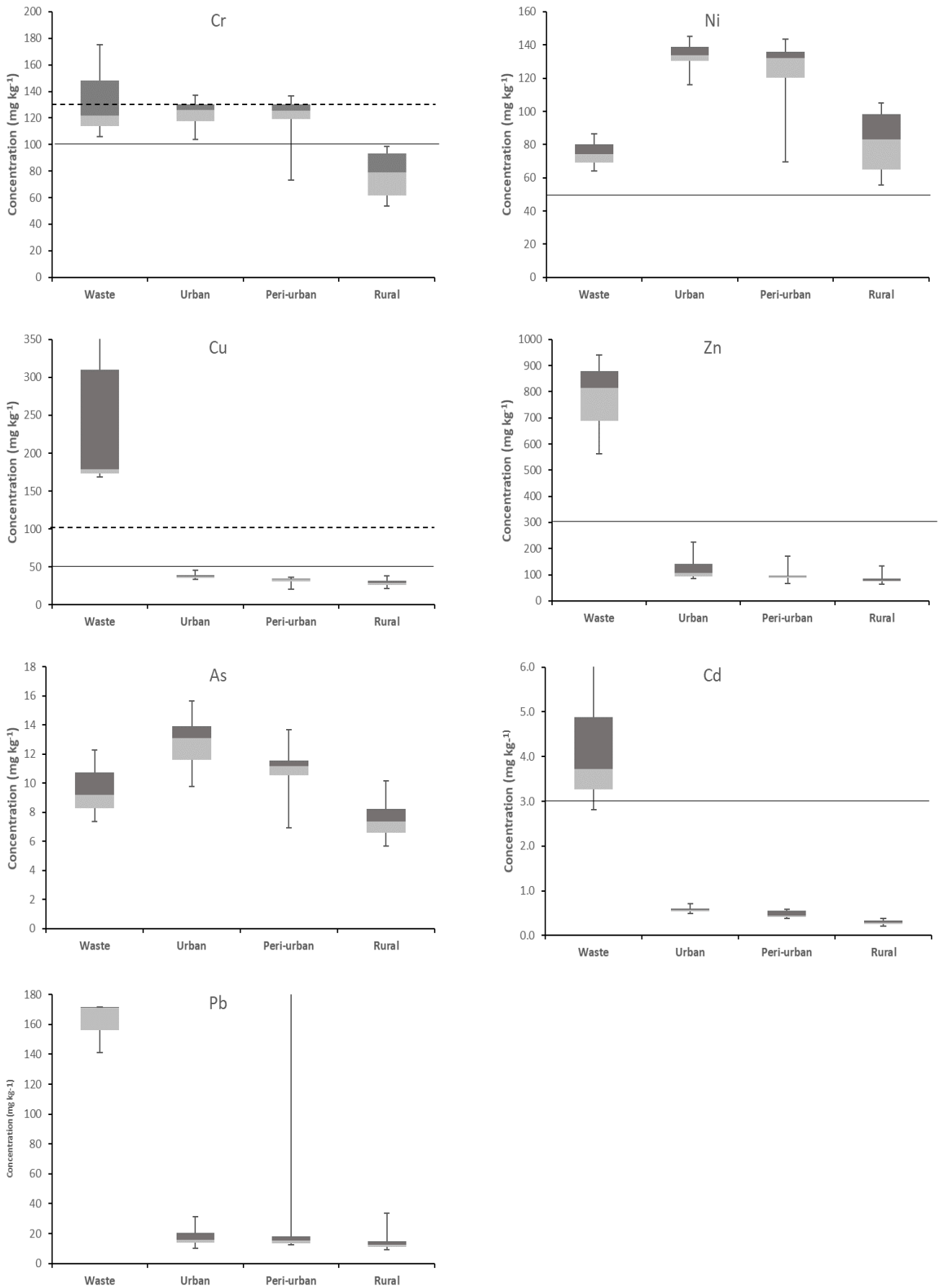
**Figure 7:** Mean hazard quotients (HQ) for children for consumption of different vegetable. A  $\text{HQ} \geq 1$ , implies a potential risk.

**Figure 8:** Mean hazard quotients (HQ) for (a) adults and (b) children, assuming equal dietary contributions of seven common consumed vegetables (eggplant, pepper, tomato, okra, cow pea, cucumber and chard). A  $\text{HQ} \geq 1$ , implies a potential hazard to the population.

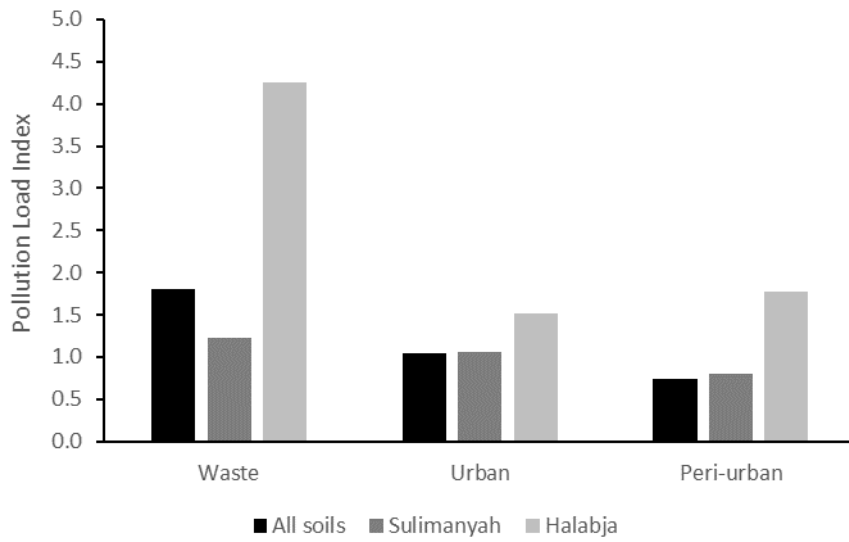


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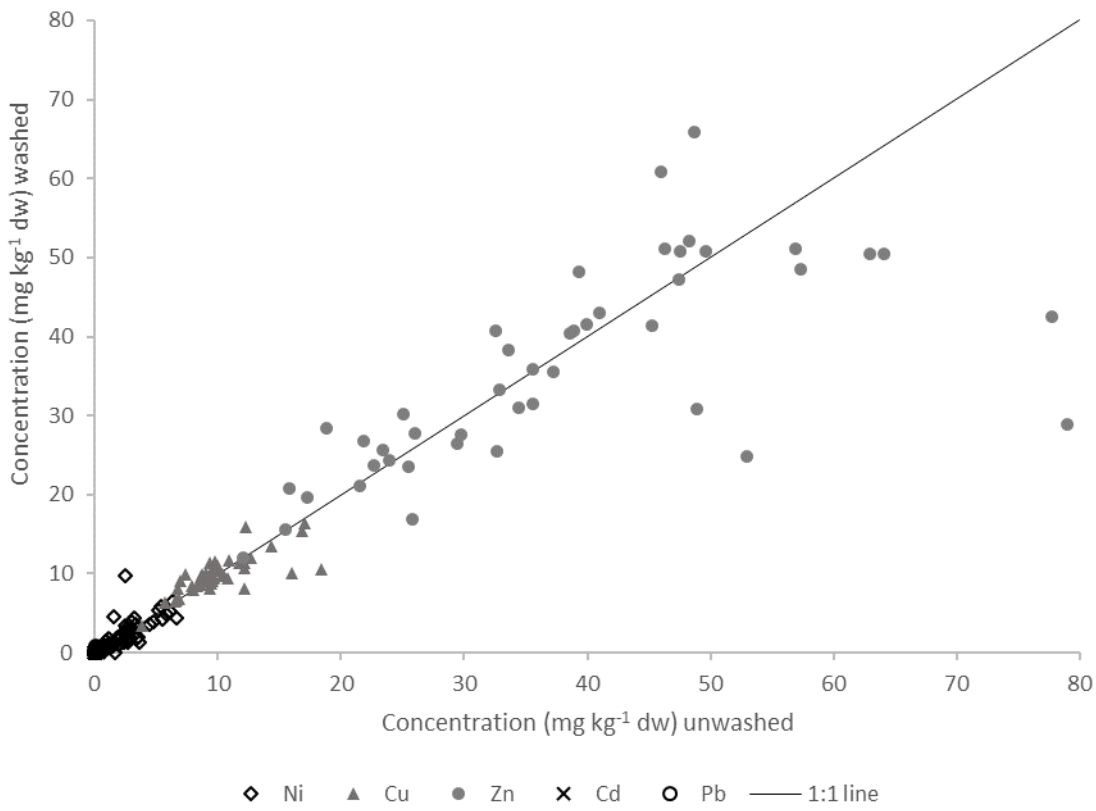




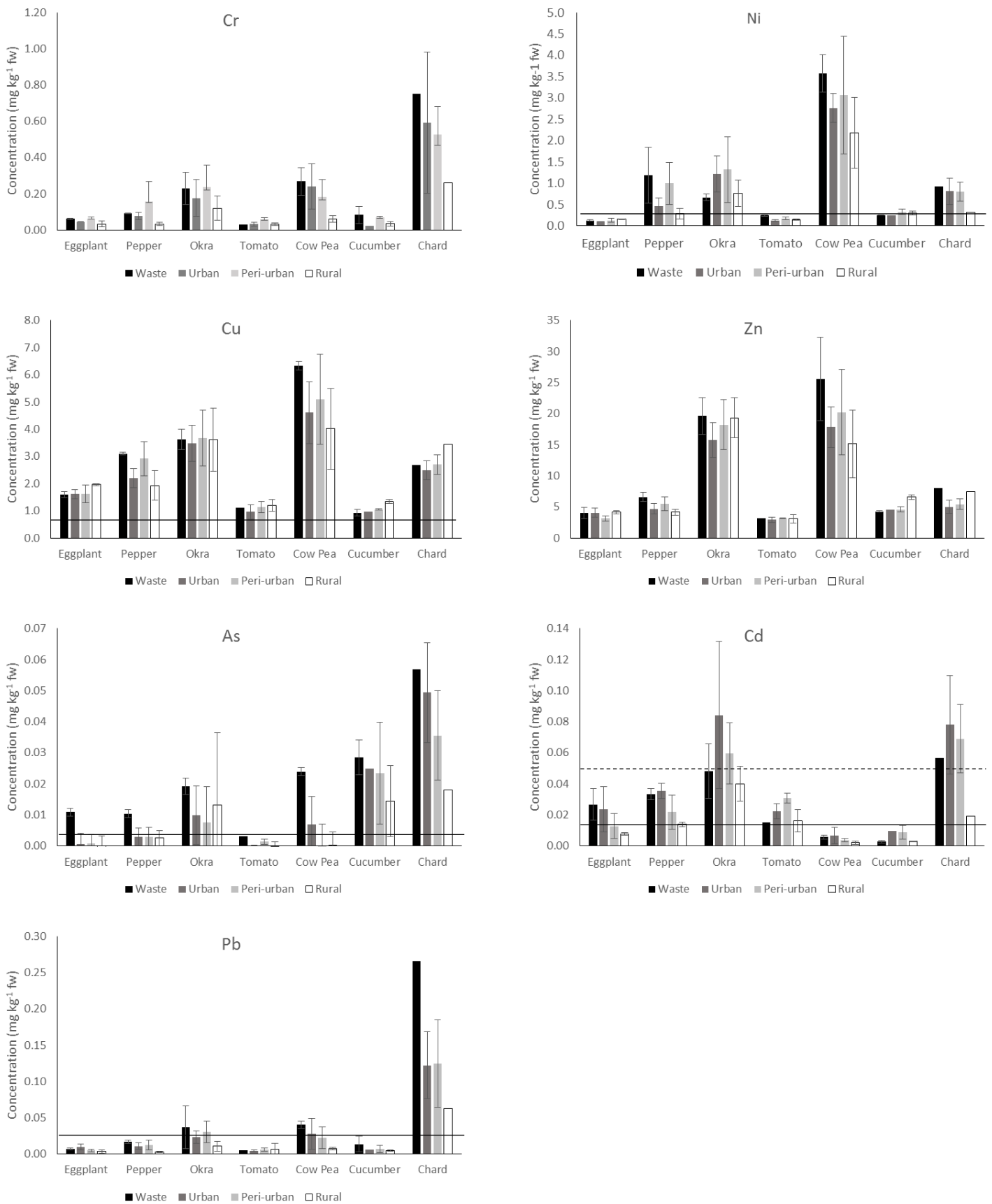
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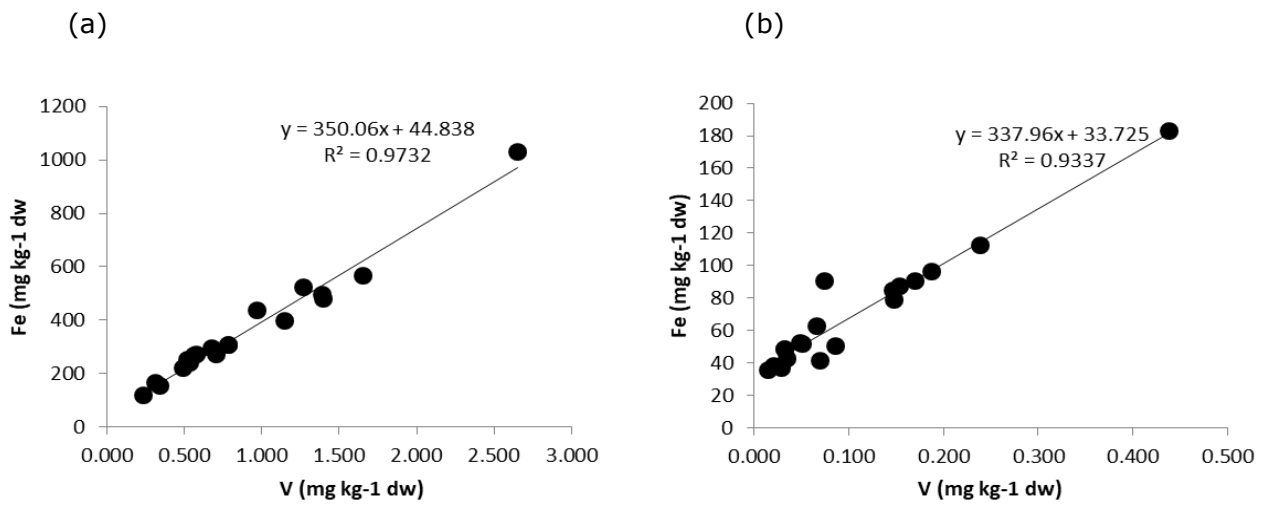
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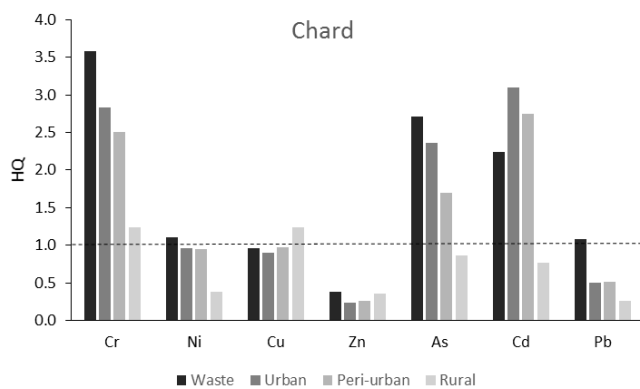
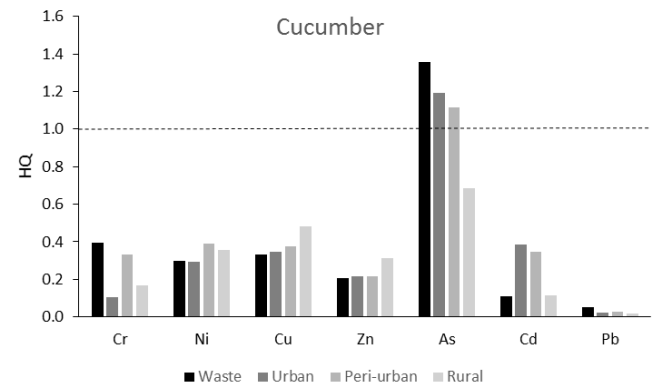
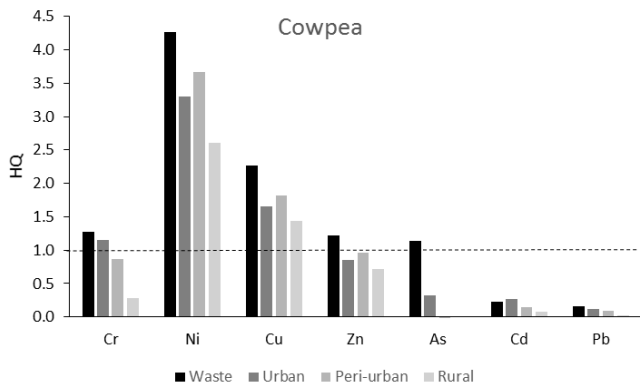
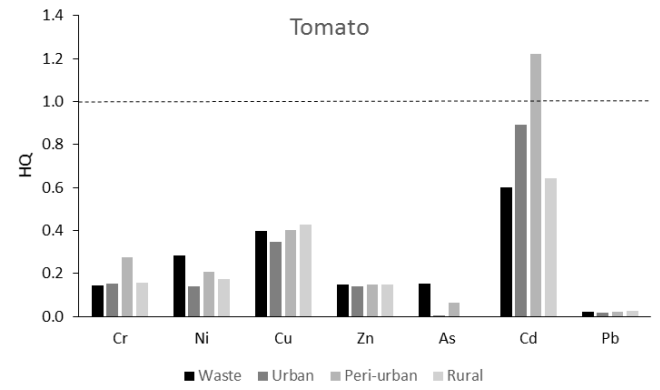
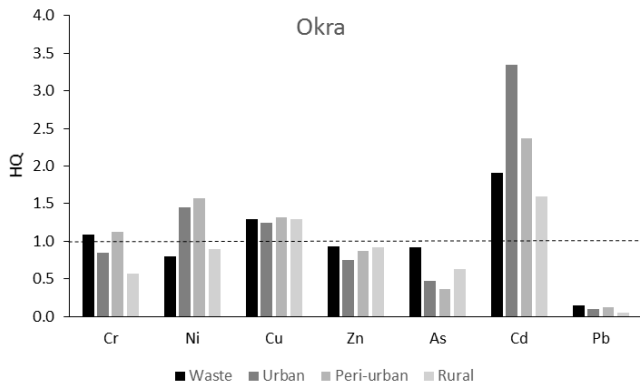
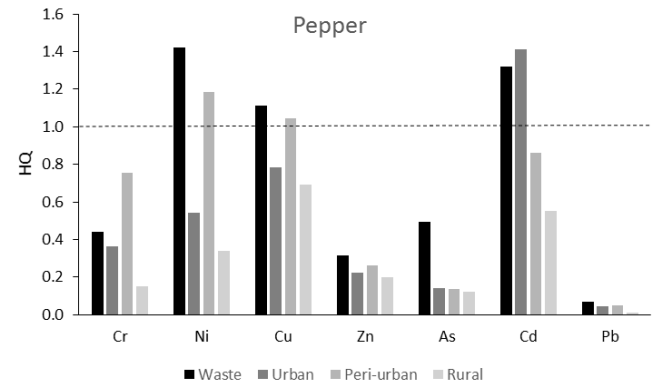
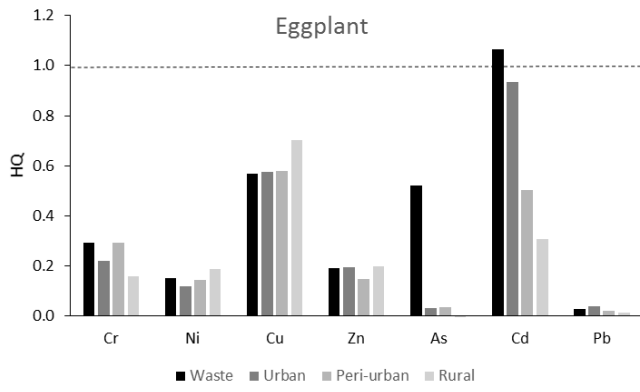
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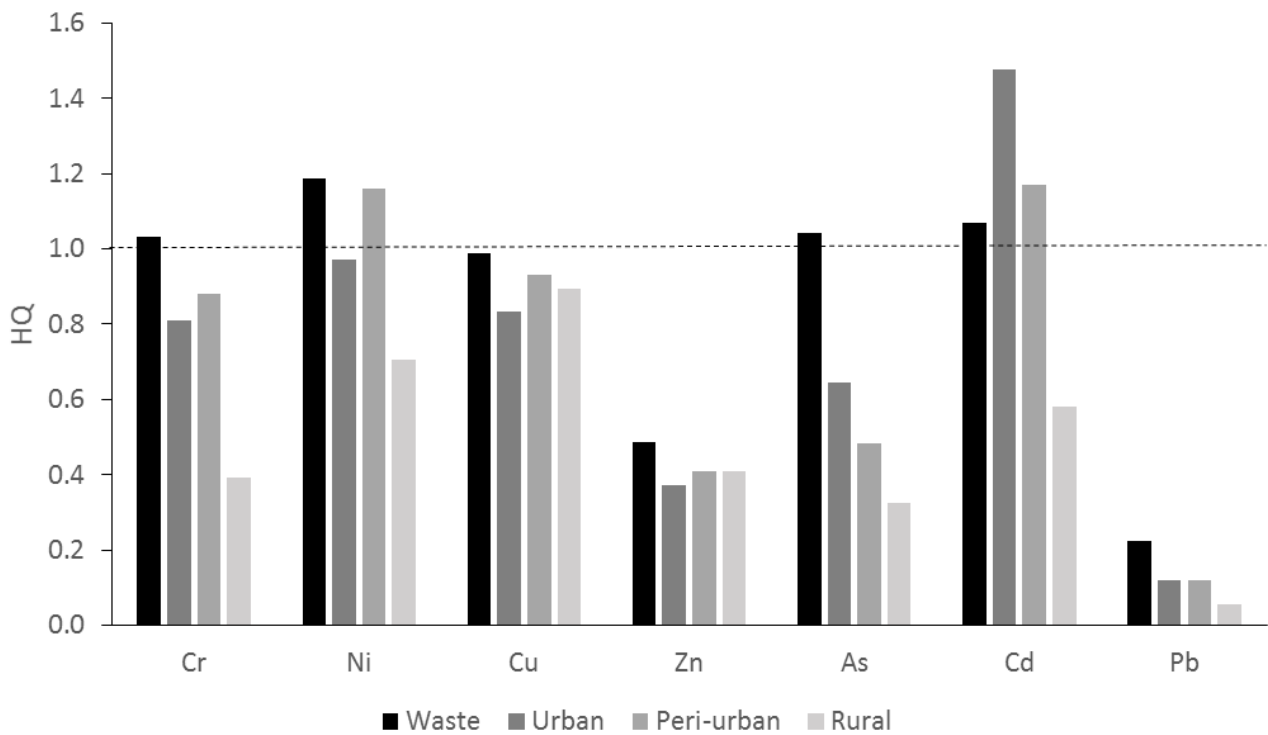
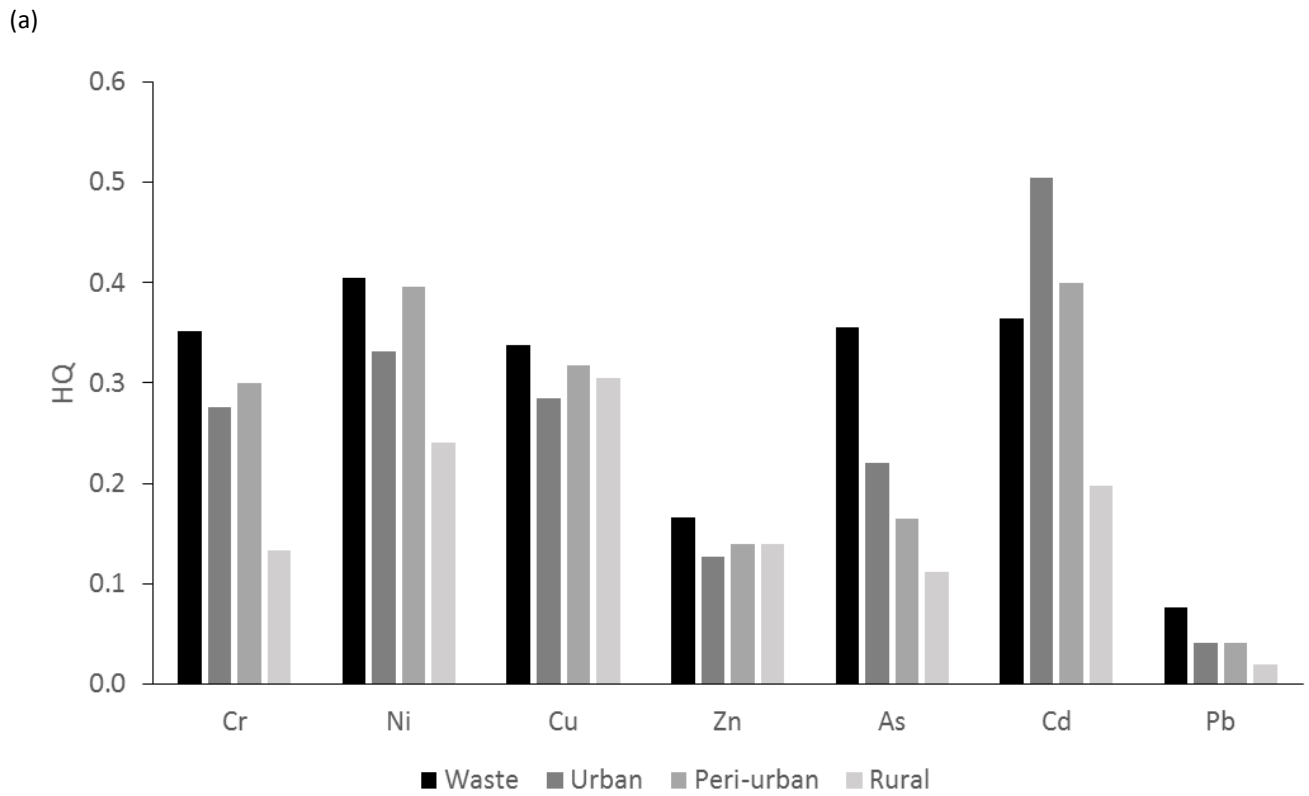
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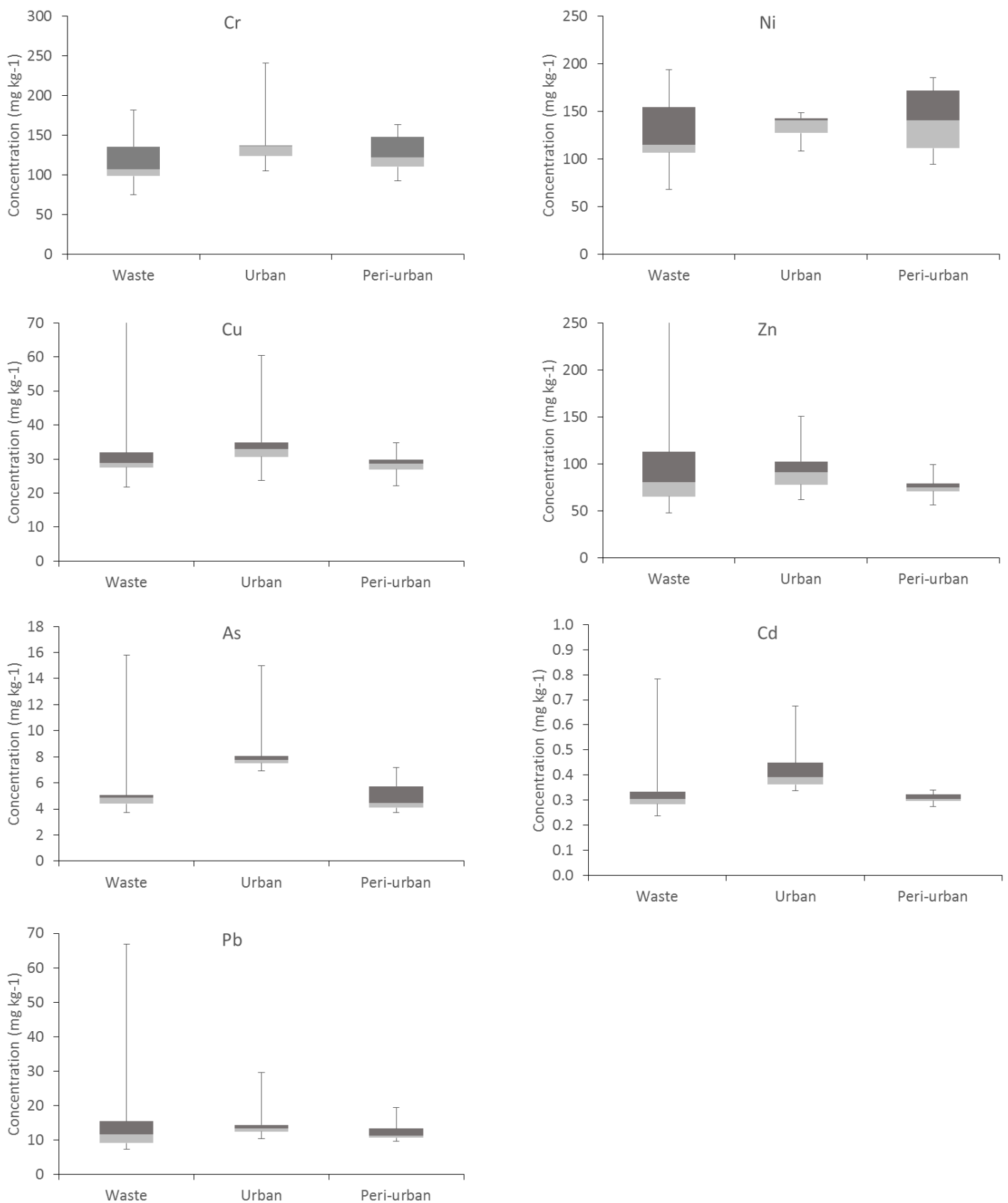
**Figure 7:** Mean hazard quotients (HQ) for children for consumption of different vegetable. A HQ  $\geq 1$ , implies a potential risk.



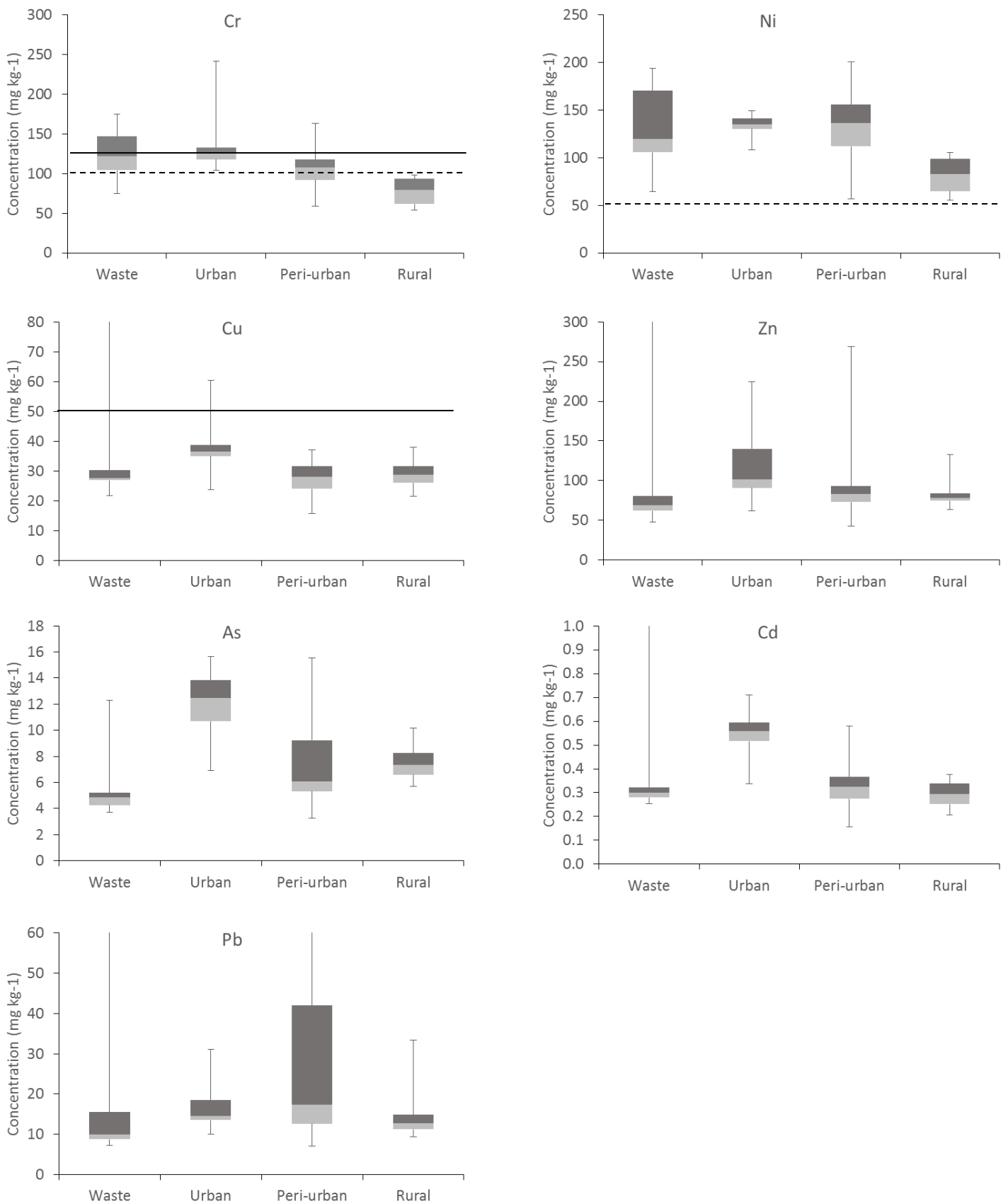
**Figure 8:** Mean hazard quotients (HQ) for (a) adults and (b) children, assuming equal dietary contributions of seven common consumed vegetables (eggplant, pepper, tomato, okra, cow pea, cucumber and chard). A  $HQ \geq 1$ , implies a potential hazard to the population.

## Supplementary Information





**Figure S1:** Mean concentrations of Cr, Ni, Cu, Zn, As, Cd and Pb for all land uses in Sulaymaniyah city. Grey boxes denote the data between the 25<sup>th</sup> and 50<sup>th</sup> percentile and black boxes the data falling between the 25<sup>th</sup> and 75<sup>th</sup> percentile. Maximum and minimum values and shown by the upper and lower error bars respectively. Solid line and dashed lines denote SGV and EU/WHO standards respectively.



**Figure S2:** Mean concentrations of Cr, Ni, Cu, Zn, As, Cd and Pb for all land uses for all sites. Grey boxes denote the data between the 25<sup>th</sup> and 50<sup>th</sup> percentile and black boxes the data falling between the 25<sup>th</sup> and 75<sup>th</sup> percentile. Maximum and minimum values and shown by the upper and lower error bars respectively. Solid line and dashed lines denote SGV and EU/WHO standards respectively.

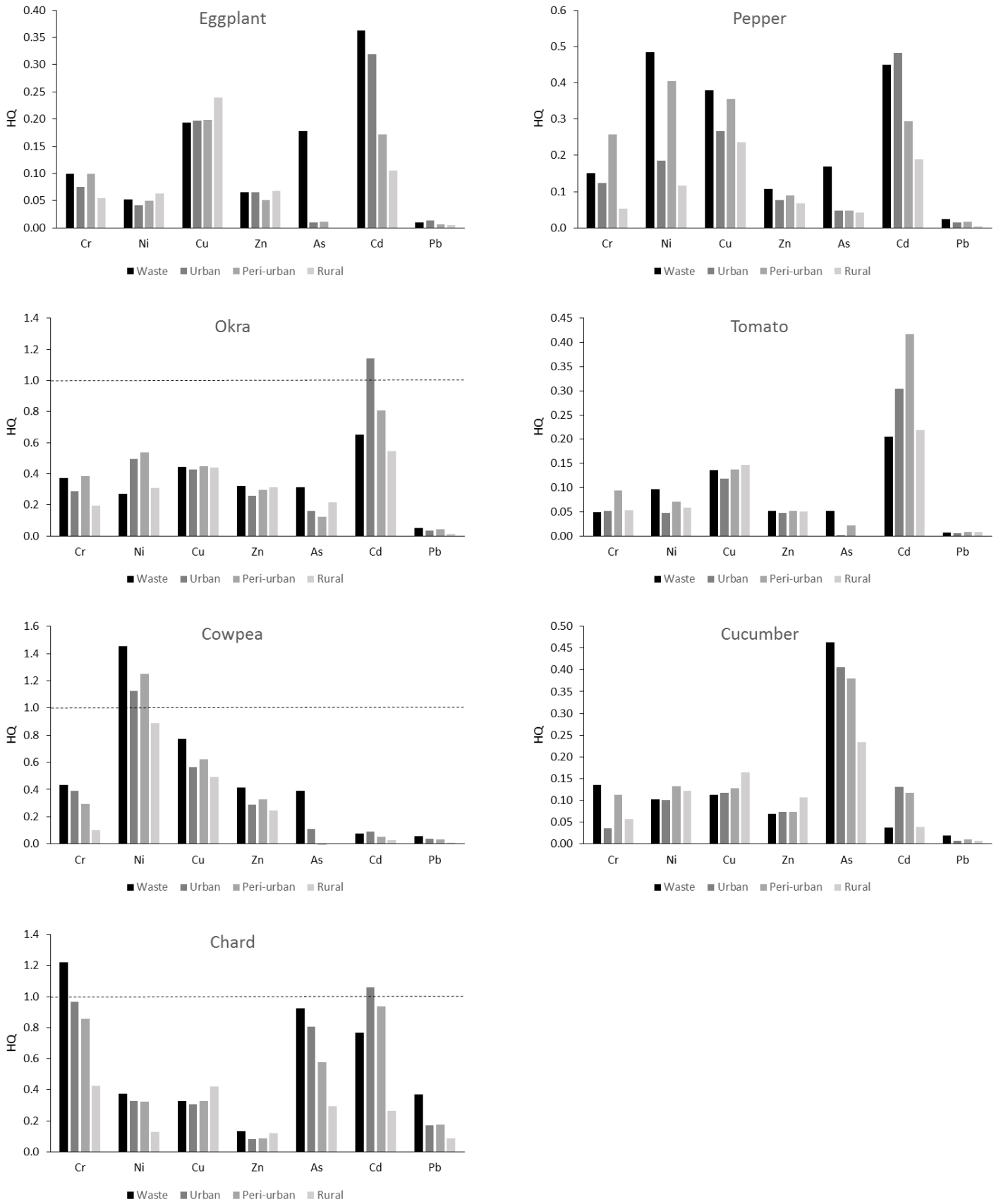


Figure S3: Mean hazard quotients (HQ) for adults.