Contamination of staple crops by heavy metals in Sibaté, Colombia

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1. Introduction

With environmental pollution in the 21st century, air quality, water resources and farmland soil availability for agricultural activities have declined progressively becoming a major public health issue (Mazari, 2014; He et al., 2019; Ahmad et al., 2019).

The term Heavy metal applies to the group of metals and metalloids with an atomic density greater than 4 g/ml or at least 5 times greater than the density of water. They can be present in the environment by natural and anthropogenic sources. Human exposure to heavy metals such as arsenic include drinking water, crops, processed food items, vegetables, mushrooms, animal products etc (Onakpa et al., 2018). Heavy metal mean ± standard error (SE) were as follows (mg/kg) As 2.36 ± 0.185, Cd 0.16 ± 0.009, Co 0.43 ± 0.019, Cr 12.1 ± 0.453, Cu 13.1 ± 1.68, Ni 0.00, Pb 7.07 ± 0.482 and Zn 3.976 ± 0.332. Cd, Cr, As, Co and Ni showed high transfer factor in Cynara scolymus. Moreover, heavy Pb, Cu and Zn transfer factor were present in Petroselinum crispum. Except for Daucus carota roots, there was a high metal transfer specifically in Petroselinum crispum leaves and other different plant parts, with high transfer factor for Cr, As, Co, Pb, Cu and Zn.

Among the most important causes related to this problem is the use of contaminated water for crop irrigation. Thus, it has drawn the attention of control and regulation entities due to the impact on safety, a concept that involves the monitoring of hazards associated with products for human consumption (Li et al., 2015; Fernández, 2012).

In emerging countries, several of the regularly consumed agricultural products are not safe for eating, mainly due to the use of pest control techniques and irrigation practices. These practices use water sources close to reservoirs that carry contaminants such as pesticides, fungicides and herbicides utilized for biological control (Bejarano-Roncancio et al., 2016; Del Puerto Rodríguez et al., 2014). Pesticides, fungicides and herbicides contain different types of HMs, including arsenic (As), cadmium (Cd), cobalt (Co), chromium (Cr), copper (Cu), mercury (Hg), nickel (Ni), lead (Pb), tin (Sn) and zinc (Zn). In humans, high levels of HMs are toxic, producing complications from chronic poisoning to cancer
Sibaté is a municipality surrounded by an industrial complex producing intermediate and capital goods. Specifically, it is one of the main agricultural producers in Cundinamarca, where it uses water from the Bogotá River basins. In regularly consumed vegetables, produced in municipalities near the middle basin of the Bogotá River, including Sibaté phytopathogens have been identified (Corrales et al., 2018). However, the Regional Autonomous Corporation of Cundinamarca (CAR) has denounced the use of untreated water sources and possibly contaminated with heavy metals to irrigate crops. In addition to the problem associated with unhealthy crop irrigation practices, in Colombia there is little research on biocarbonized products (Biochars). Moreover, biocarbonized products application is not mandatory. Nevertheless, sufficient evidence exists regarding at least three fundamental aspects of its application: 1. It improves physicochemical properties of the soil, 2. It reduces heavy metal bioavailability, including Pb, As, Co and molybdenum (Mo) and last it increases biological soil fertility and crop productivity (El-Naggar et al., 2019; El-Naggar et al. 2020).

This research aimed to determine if [HMs] in vegetables in the typical Colombian diet, which were produced and collected in a crop near a water basin, was higher than the established threshold for known vegetables. Many emerging countries base their economy on agricultural activities, supply vegetables to the local market and export some products to other countries, generating risk for consumers, turning this problem into a global context.

2. Materials and methods

2.1. Area of study

Samples were gathered from the location La Union in the municipality of Sibaté, Cundinamarca (Colombia) at 4° 30’03”N 74° 15’52”W; 2570 m.a.s.l. Samples were collected from soils and parts of the following plants *Cynara scolymus* (Artichoke), *Daucus carota* (Carrot) and *Petroselinum crispum* (Parsley). The municipality has an approximate extension of 12,560 hectares, where 10,870 are rural and 10.1% are utilized for agricultural production. The main crops grown are potatoes, strawberries and peas, followed by other vegetables. The average temperature is 13.5 °C (6 °C–18 °C) and the average rainfall is 723 mm. The driest month (24 mm) is January in contrast to October, the month with the most rainfall (102 mm).

To irrigate crops, farmers illegally use water from the Bogotá River and Muña dam without previous treatment (Figure 1) (Güiza-Su á rez et al., 2015; Dí az-Granados and Camacho Botero, 2012).

2.2. Sample collection

Sampling of plant and soil material was performed randomly using a zeta figure in order to make a general scan of area of study.

2.3. Preparation of plant and soil samples

Acid digestion was carried out using 10 g of fresh material with HCl:HNO₃ at a 1:1 ratio. Material was incubated for 15 days in chilled conditions. The resulting solution was filtered and 50 ml water type T1 was added and capped.

2.4. Determination of heavy metals

Concentrations of Cu, As, Pb, Cr, Zn, Co, Cd and Ni were determined using sample solutions obtained from plants and soil (n = 4 for each plant and soil sample, respectively). This process was carried-out employing atomic absorption instruments (Varian AA-140 and Shimadzu AA-7000). The atomic absorption instrument was operated in the flame mode with air and acetylene. For each metal, gas flow conditions and burner height were established as recommended by the manufacturer according to the instruments’ default program. The stock solutions for performing the calibration curve were prepared from an analytical certified standard at 1000 mg/L for each metal (Merck & Co Ins. USA). To obtain calibration curves calculations were performed with 1% nitric acid. Wavelengths and detection limits used in the atomic absorption instrument are shown in Table 1:

2.5. Statistical analysis

Data was expressed as a mean ± SE and all estimations subtracted the value obtained in the soil. Transfer of metals from the soil to the vegetable was calculated using the following equation:

\[
\text{Transfer} = \frac{\text{Amount in vegetable}}{\text{Amount in soil}} \times 100
\]
Table 1. Wavelengths and detection limit of the calibrated instrument used.

| Element | Wavelength (nm) | Detection limit (LoD) |
|---------|----------------|----------------------|
| Cd      | 228.8          | 0.500                |
| Co      | 304.4          | 0.500                |
| As      | 193.7          | 0.500                |
| Zn      | 213.9          | 0.005                |
| Pb      | 261.4          | 0.500                |
| Ni      | 232.0          | 0.500                |
| Cu      | 324.8          | 0.005                |
| Cr      | 357.9          | 0.200                |

% transfer = \( \left( \frac{\sum_{i} C_{\text{mo}}}{C_{\text{ms}}} \right) \times 100 \), (n: number of organs)

Cmo: metal concentration in organ; Cms: metal concentration in the soil.

Correlation between [HMs] determined in soil (Spearman_Rho) or in vegetables (Pearson_r) were established. To compare mean concentrations of a specific metal among plants or among plant parts a One-way ANOVA was performed. To determine the interaction between plants and their parts related to the mean of each HM, two-way ANOVA was performed. A p value of <0.05 was considered significant. JASP® Version 0.11.1 and Minitab® 19.2 statistical packages were used to analyze the information.

3. Results and discussion

3.1. General characteristics

To identify eight type of metal in four repeated samples of leaves, stems and roots for the Daucus carota, Cynara scolymus and Petroselinum crispum vegetable species 416 measurements were made. Additionally, the concentration of the metal in the flowers of Cynara scolymus and sample of the surrounding soil of each plant was determined.

Among the metals evaluated, the average concentrations of As (2.36 mg/kg), Pb (7.07 mg/kg) and Cr (12.1 mg/kg) in vegetables were higher than the internationally established maximum standards 0.5 mg/kg, 0.1 mg/Kg and 0.5 mg/kg, respectively (t-Test, p<0.001) (FAO, 2018; CODEX STAN 193, 1995; CODEX, 2019). The minimum and maximum concentrations of metals (mg/kg) for As ranged between 0.11-4.74, Cd between 0.03-0.37, Co between 0.16-0.64, Cr between 6.5-17.7, Cu between 3.41-34.18, Ni 0.00, Pb between 0.84-12.5 and Zn between 1.21-9.31. The average concentrations of metals are shown in Table 2.

These results are correlated with previous studies in the water quality of the Bogota River in which the high grade of contamination of the river has been demonstrated, with heavy elements such as arsenic (As), cadmium (Cd), cobalt (Co), chromium. (Cr), copper (Cu), iron (Fe), mercury (Hg), manganese (Mn), nickel (Ni), lead (Pb), tin (Sn), zinc (Zn), and a wide variety of substances potentially toxic organic and inorganic (Miranda et al., 2010). At the beginning when Bogota River water stat to be use for crops, the mobility of heavy metals was greater in uncontaminated soils. During the initial application period, the organic matter improves the properties of the soil and controls the solubility of the metals, protecting the plant from possible accumulation. However, over time these benefits decrease with the mineralization of the organic fraction. This effect can be established due to the prolonged use of Bogota River waters which for years has been added to the soil during the irrigation of crops. The acidity of the soil and its texture increased the solubility of Pb, but its mobilization, in the case of the soil in the region, could be due to the fact that the organic matter of the soil favors its accumulation.

3.2. Transfer of metals from the soil to the vegetable

Physicochemical properties for each specific heavy metal and soil type determine the possible transfer of metal from the soil to the vegetable. This phenomenon alters the concentration of metals in the soil and has been indirectly determined by a high correlation and collinearity between the concentrations of measured metals (Gutierrez et al., 2016). In general, soils of the Bogotá River basin are acidic, frank to clay loam and medium fertility (Lora-Silva and Bonilla-Gutierrez, 2010). Such characteristics, added to the high saturation of aluminum, give the soil the ability to carry out cationic exchange (CAR, 2006). In soil collected samples a strong and direct correlation between Ni and Cu was identified (r: 0.978), Pb and Cu (r: 1) and Pb and Ni (r: 0.978). Likewise, a strong and indirect correlation was identified between Co and Cu (r: -0.937) and Pb and Co (r: -0.937).

The investigators took into consideration Cu is considered the most mobile of heavy metals. It is a very versatile trace cation and, in the soil, it shows a great capacity to chemically interact with other mineral components (Méndez-Romero et al., 2003). In contrast to what was identified in the soil, strong and direct correlation was determined in plants only between Zn and Co (r: 0.911) and moderate between Cu and Cr (r: 0.744), suggesting the presence of other factors related to the transfer from the ground to the plant, specifically the type of vegetable and each of its parts. It was observed that Cd and Cu were the metals with greater and lesser transfer proportions from the soil to the vegetables, respectively.

Although Cu in ionic form is highly mobile, it easily precipitates with anions such as sulfates, carbonates, oxides and oxyhydroxides, which causes it to be fixed in the soil (Kabata-Pendias et al., 2001). Cd, Cr, As,

Table 2. Average concentrations of metals (mg/kg).

| Vegetable species       | Cd    | Co    | As    | Zn    | Pb    | Ni    | Cu    | Cr    |
|------------------------|-------|-------|-------|-------|-------|-------|-------|-------|
| Daucus carota          | 0.15  | 0.40  | 1.82  | 2.87  | 6.96  | 0.00  | 6.80  | 8.80  |
| Root                   | 0.11  | 0.27  | 0.36  | 1.22  | 7.52  | 0.00  | 4.79  | 9.53  |
| Stem                   | 0.13  | 0.36  | 2.79  | 2.90  | 6.68  | 0.00  | 4.28  | 7.27  |
| Leaves                 | 0.19  | 0.57  | 2.51  | 4.48  | 6.68  | 0.00  | 11.3  | 9.60  |
| Cynara scolymus        | 0.16  | 0.41  | 2.58  | 3.31  | 5.69  | 0.00  | 19.4  | 12.8  |
| Flowers                | 0.08  | 0.22  | 3.50  | 2.97  | 4.18  | 0.00  | 5.36  | 13.5  |
| Root                   | 0.16  | 0.47  | 2.00  | 3.06  | 7.51  | 0.00  | 32.7  | 13.6  |
| Stem                   | 0.18  | 0.34  | 2.54  | 2.32  | 6.89  | 0.00  | 7.94  | 12.1  |
| Leaves                 | 0.22  | 0.48  | 2.85  | 4.16  | 3.55  | 0.00  | 8.45  | 10.7  |
| Petroselinum crispum   | 0.16  | 0.52  | 2.43  | 6.20  | 9.25  | 0.00  | 19.4  | 15.0  |
| Root                   | 0.18  | 0.47  | 2.85  | 4.31  | 11.6  | 0.00  | 25.7  | 14.6  |
| Stem                   | 0.16  | 0.50  | 1.86  | 5.03  | 4.17  | 0.00  | 5.42  | 13.0  |
| Leaves                 | 0.15  | 0.61  | 2.59  | 9.27  | 11.9  | 0.00  | 27.2  | 17.4  |

*SE is not shown, but it can be found in the supplementary material.
Co and Ni metals had the highest transfer rates to *Cynara scolymus*, and Pb. Moreover, Cu and Zn metals were transferred to *Petroselinum crispum*. In general, the proportion of metal transfer to *Daucus carota* was intermediate compared to the other vegetables (Figure 2, left).

The proportion of metal transfer was highly variable when explored by specific part of the vegetable. The transfers of six out of eight metals (Cr, As, Co, Pb, Cu, Zn) were elevated in *Petroselinum crispum* leaves. Several studies have shown that heavy metal accumulation is higher in leafy vegetables. These types of vegetables generally grow faster with a higher rate of perspiration than leafless vegetables. Additionally, they are more exposed to air pollutants (Taiyang et al., 2019; Chen et al., 2013).

Additionally, except for the roots of *Daucus carota*, there was a high proportion of metal transfer to all parts of the vegetable (Figure 2, right).

In the Bogotá savanna, the soils are of varied textures, tend to acidity (pH 4 to 6.5), and generally have low concentrations for most of the elements, except some sites in the middle basin (Sibate) where the accumulation of some metals occurs. This is particularly important to explain the origin and geochemical behavior of the elements in soils, because in addition to the fact that the parental materials provide trace elements, the conditions of pH and redox potential existing in the medium, determine to a great extent the mobility and accumulation of contributed elements. Under basic pH conditions, the predominant species of Cu, Ni, Zn, and Co are geochemically immobile, since they can be co-precipitated as carbonates or adsorbed by these or by iron-oxy-hydroxides or clays and concentrated in soils, such as in the lower basin of the Bogotá River. As and Cd are moderately mobile under different pH conditions, however, the presence of carbonates, iron oxy-hydroxides (favored with the increase in pH) or clays, favor their fixation and accumulation in alkaline soils. Mn and Pb are very poorly mobile under different pH conditions and remain accumulated near the source that originates them. In addition to the above, Mn can precipitate as hydroxide and remain adsorbed together with iron oxy-hydroxides and in the case of Pb, it can be adsorbed and fixed in the clay material of the soils. The mobility is high and increases with pH, however, the presence of clays and carbonate ions favors its adsorption and accumulation in the soils of the lower basin. The predominance of acidic pH in the soils of the upper basin, mainly in the flat part of the Bogotá savanna, may favor the existence of mobile phases of Cu, Ni, Zn, Cd and Co, a fact that facilitates their migration in the aqueous phase and may partially explain the lower accumulation (Prieto et al., 2018).

### 3.3. Metal concentration in vegetable parts

The variability of the transfer of metals from the soil to the vegetable and its parts directly affects the concentration of both. Interaction between the type of metal and the vegetables was identified (two-way ANOVA; df: 14, F-statistic: 4.305, p-value: < 0.001; Figure S3). In addition, the interaction between the type of metal and the vegetable part was determined (two-way ANOVA; df: 21, F-statistic: 7.968, p-value: < 0.001; Figure S3).

The average concentration of Co, Cr, Cu and Zn was different among vegetables. Likewise, the average concentration of As, Co, Cu, Ni and Zn was different among the parts of each vegetable (Figure 3, Table 2.1 and Table 2.2).

A sensitivity analysis that included the flowers in the one-way ANOVA did not change the results presented in Table 2.1.

Heavy metals are natural elements that are distributed throughout the Earth’s crust. The presence of high levels of some of them is mainly due to anthropogenic activities. Such activities produce alterations in the geochemical cycles of metals, producing accumulation and environmental pollution (Weissmannova and Pavlovska, 2017).

Urbanization in rural areas has grown in parallel with industrialization near water basins, with the annual volume of wastewater produced and the volume of water reused, with or without treatment, often for agricultural production and human consumption (Patnaik, 2018).

The use of wastewater is a legal and viable alternative to the extent of applied pretreatment. Countries with high economic income (HIC) have optimized wastewater treatment processes managing to reuse more than 90% of those generated in industrial processes (Khalid et al., 2018).

In Colombia, the department that reuses the highest volume of contaminated water is Cundinamarca, followed by Antioquia. However, as in other countries with low economic income (LIC), the volume of treated wastewater is not comparable among main Capital cities and the municipal level.

Although there are many scientific publications and grey literature on this subject, most arise from HIC, where management of contaminated wastewater is optimal compared with the LIC countries. To address this issue the main problems of LICs include lack of fresh water for irrigation of crops and resources for wastewater treatment. Additionally, lack of education and human awareness campaigns on problems, such as crop risk and regulatory updating and implementation of laws aimed at wastewater management (Khalid et al., 2018; Cossio et al., 2019). Among the negative effects of irrigating with contaminated water in agricultural crops include increased concentration of HMs. It is the most relevant effect with the greatest impact on human health (Banerjee and Gupta, 2013).

Several problems arise with HMs: these include the non-degradable and cumulative character, progressive deterioration of soils, and the hyper-accumulation of plants. This can cause parts of the plant such as roots to accumulate >90% of the HMs without transferring them to other parts (Rascio and Navari-Izzo, 2011; Khalid et al. 2018).

![Figure 2. Transfer fraction from the ground. Left half. The global fraction of metal transferred from the soil to each vegetable. Right half.标准化 proportion of metal transfer was identified for each vegetable part. Dc (*Daucus carota*), Cs (*Cynara scolymus*), Pc (*Petroselinum crispum*).](image-url)
Figure 3. Average concentration of metals per plant and plant part. To visualize minimal differences in the concentration of metals per vegetable species and vegetable part, averages were transformed with the Box-Cox method ($\lambda$: 0.0275661). A transformed mean range between 0 – 1.10225 was obtained.

### Table 2.1. Differences in the transformed concentrations of metals.

| Metal | One-way ANOVA; p-value (F-statistic) | Vegetable organs |
|-------|------------------------------------|------------------|
| As    | 0.180 (1.81)                       | 0.021 (4.38)     |
| Cd    | 0.121 (2.26)                       | 0.073 (2.84)     |
| Co    | 0.006 (5.97)                       | 0.000 (11.14)    |
| Cr    | 0.000 (54.58)                      | 0.225 (1.56)     |
| Cu    | 0.003 (6.79)                       | 0.000 (9.77)     |
| Ni    | 0.310 (1.21)                       | 0.003 (6.91)     |
| Pb    | 0.421 (0.89)                       | 0.055 (3.18)     |
| Zn    | 0.000 (14.16)                      | 0.000 (10.45)    |

Flowers were left-out from the ANOVA analysis. The bold is very important to highlight the values that present significant differences.

### Table 2.2. Characteristics related to metal concentration.

| Metal | Tukey’s Post-hoc test |
|-------|-----------------------|
|       | Vegetable | | Vegetable part |
|       | Dc | Cs | Pc | Roots | Stems | Leaves |
| As    | - | - | - | + | ++ | +++ |
| Cd    | - | - | - | - | - | - |
| Co    | + | +++ | +++ | + | + | +++ |
| Cr    | + | +++ | +++ | - | - | - |
| Cu    | + | +++ | +++ | - | +++ | - |
| Ni    | - | - | - | + | +++ | + |
| Pb    | - | - | - | - | - | - |
| Zn    | + | + | +++ | + | + | +++ |

The bold is representative of the different groups. The number of crosses represents average metal concentration in each group [(Dc (Daucus carota), Cs (Cynara scolymus), Pc (Petroselinum crispum)]. Tables S1 and S2 show the marginal averages with Box-Cox transformation.
The vegetables with more capacity for accumulation of HMs and danger for human consumption are those whose edible parts grow below ground. In many Colombian agricultural products, the part of the plant consumed is the root. Examples of this are carrots, potatoes, beets, and cassava, among others. This is fundamental in our economy because the new generations have a tendency towards a healthier diet and give relevance to the inclusion of roots and other parts of vegetables as part of their diet (Herforth et al., 2019; Gómez et al., 2019).

Cereals are a fundamental part of the diet due to their positive effect on the gastrointestinal tract and the immune system. An Iranian study identified higher than allowed for human consumption (Cr, Ni, Zn and Cu) heavy metal concentrations in cereals and other agricultural products sold in supermarkets. However, when potential intake of heavy metals was estimated adjusted for weekly average cereal consumption, no levels exceeded the tolerable toxic metal consumption per week (Meghdad et al., 2016).

In agreement with the previous finding, a systematic revision of the literature including 50 articles to determine toxic metal concentrations in frequently consumed cereals by humans by an Iranian population, established 95% of the commercialized Iranian cereals in the past two decades was reported to have inappropriate As, Cd and Pb concentrations (Sharafi et al., 2019).

Even though results from Pirsaheb et al. demonstrated expected consumed metal consumption per week would be in a tolerable range, it is necessary to adopt healthy agricultural practices to limit unnecessary exposure in patients or susceptible population to tolerable levels of toxic metals, especially those with liver, kidney or hematological comorbidity (Wani et al., 2016).

Prevention strategies with Public health impact should be implemented in areas or countries that use contaminated or residual waters to irrigate crops. Such actions should include application of biocarbonized products in soils, implementation of precise, less expensive and low toxicity potential methods to detect heavy metal levels in fruits and vegetables, and increase special washing techniques that have demonstrated to be highly efficient to remove As, Pb and Cd from fruits, vegetables and cereals (El-Naggar et al., 2019, 2020; Habibollahi et al., 2019; Sharafi et al., 2019).

4. Conclusion

In general, this study evaluated heavy metal concentrations in vegetables of frequent consumption in the Colombian diet. It was identified the mean concentration of three (As, Pb and Cr) out of eight heavy metals exceeded the allowed limits according to international standards. Accumulation of heavy metals in different parts of the vegetables studied was directly proportional to the transfer of metals from the soil to the plant. This transfer was significantly higher in parsley, due to greater foliage density parsley has in comparison to the other two plants. Based on heavy metal physiochemical properties correlation analyses demonstrated heavy metal transfer was associated with the presence of other heavy metals, as a case in point Pd and Cu. In the context of this study the geographical location where samples were collected was influenced by hydrographic sources contaminated by effluents from leather industries, which are used to irrigate crops. Therefore, heavy metal contamination observed in this investigation could be accounted in part by this fact.

Last, limitations of this research include the need of more samples at different sampling points. In addition, samples from different plants to establish comparisons by type of material would have been desirable. In the near future it is essential to search for plant species that have the capacity to accumulate heavy metals and that can be used in phyto-remediation processes (Franco et al., 2018) to provide solutions for Colombian agricultural practices.

Declarations

Author contribution statement

María F. Lizarazo, César D. Herrera, Crispín A. Celis, Sandra P. Forero, Javier R. Velandia, Fabio E. Díaz, William A. Andrade: Performed the experiments.

Oscar E. Rodríguez: Conceived and designed the experiments; Analyzed and interpreted the data.

Aníbal A. Teherán, Luis G. Piñeros: Analyzed and interpreted the data.

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Competing interest statement

The authors declare no conflict of interest.

Additional information

No additional information is available for this paper.

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Table S1. Marginal Means - Vegetable species × Metal

| Vegetable species | Metal | Marginal Mean | SE  | 95% CI          |
|-------------------|-------|---------------|-----|----------------|
|                   |       |               |     | Lower | Upper |
| Cynara scolymus   | As    | 1.025         | 0.003 | 1.019 | 1.030 |
|                   | Cd    | 0.947         | 0.003 | 0.942 | 0.953 |
|                   | Co    | 0.972         | 0.003 | 0.967 | 0.978 |
|                   | Cr    | 1.072         | 0.003 | 1.067 | 1.078 |
|                   | Cu    | 1.064         | 0.003 | 1.058 | 1.069 |
|                   | Ni    | 0.757         | 0.003 | 0.752 | 0.763 |
|                   | Pb    | 1.047         | 0.003 | 1.041 | 1.052 |
|                   | Zn    | 1.031         | 0.003 | 1.026 | 1.037 |

(continued on next column)
| Vegetable species | Metal | Marginal Mean | SE  | 95% CI Lower | 95% CI Upper |
|-------------------|-------|---------------|-----|--------------|--------------|
| Daucus carota     | As    | 1.009         | 0.004 | 1.002        | 1.016        |
|                   | Cd    | 0.941         | 0.004 | 0.934        | 0.948        |
|                   | Co    | 0.969         | 0.004 | 0.962        | 0.976        |
|                   | Cr    | 1.062         | 0.004 | 1.055        | 1.069        |
|                   | Cu    | 1.042         | 0.004 | 1.035        | 1.049        |
|                   | Ni    | 0.758         | 0.004 | 0.751        | 0.765        |
|                   | Pb    | 1.052         | 0.004 | 1.045        | 1.059        |
|                   | Zn    | 1.026         | 0.004 | 1.019        | 1.033        |
| Petroselinum crispum | As    | 1.024         | 0.004 | 1.017        | 1.031        |
|                   | Cd    | 0.944         | 0.004 | 0.937        | 0.951        |
|                   | Co    | 0.978         | 0.004 | 0.971        | 0.985        |
|                   | Cr    | 1.078         | 0.004 | 1.071        | 1.085        |
|                   | Cu    | 1.069         | 0.004 | 1.062        | 1.077        |
|                   | Ni    | 0.752         | 0.004 | 0.746        | 0.759        |
|                   | Pb    | 1.055         | 0.004 | 1.048        | 1.062        |
|                   | Zn    | 1.050         | 0.004 | 1.043        | 1.057        |

Table S2. Marginal Means - Organ × Metal

| Organ    | Metal | Marginal Mean | SE  | 95% CI Lower | 95% CI Upper |
|----------|-------|---------------|-----|--------------|--------------|
| Flowers  | As    | 1.029         | 0.006 | 1.017        | 1.041        |
|          | Cd    | 0.922         | 0.006 | 0.910        | 0.934        |
|          | Co    | 0.960         | 0.006 | 0.948        | 0.972        |
|          | Cr    | 1.073         | 0.006 | 1.061        | 1.085        |
|          | Cu    | 1.030         | 0.006 | 1.018        | 1.042        |
|          | Ni    | 0.753         | 0.006 | 0.740        | 0.765        |
|          | Pb    | 1.045         | 0.006 | 1.033        | 1.057        |
|          | Zn    | 1.035         | 0.006 | 1.023        | 1.047        |
| Leaves   | As    | 1.026         | 0.003 | 1.019        | 1.032        |
|          | Cd    | 0.955         | 0.003 | 0.949        | 0.961        |
|          | Co    | 0.984         | 0.003 | 0.977        | 0.990        |
|          | Cr    | 1.071         | 0.003 | 1.065        | 1.087        |
|          | Cu    | 1.075         | 0.003 | 1.069        | 1.081        |
|          | Ni    | 0.752         | 0.003 | 0.745        | 0.758        |
|          | Pb    | 1.053         | 0.003 | 1.047        | 1.059        |
|          | Zn    | 1.049         | 0.003 | 1.042        | 1.055        |
| Root     | As    | 1.001         | 0.003 | 0.995        | 1.008        |
|          | Cd    | 0.949         | 0.003 | 0.942        | 0.955        |
|          | Co    | 0.974         | 0.003 | 0.968        | 0.981        |
|          | Cr    | 1.072         | 0.003 | 1.066        | 1.078        |
|          | Cu    | 1.080         | 0.003 | 1.073        | 1.086        |
|          | Ni    | 0.753         | 0.003 | 0.747        | 0.760        |
|          | Pb    | 1.061         | 0.003 | 1.055        | 1.068        |
|          | Zn    | 1.026         | 0.003 | 1.020        | 1.032        |
| Stem     | As    | 1.021         | 0.003 | 1.015        | 1.028        |
|          | Cd    | 0.951         | 0.003 | 0.944        | 0.957        |
|          | Co    | 0.975         | 0.003 | 0.968        | 0.981        |
|          | Cr    | 1.067         | 0.003 | 1.061        | 1.073        |
|          | Cu    | 1.049         | 0.003 | 1.043        | 1.055        |
|          | Ni    | 0.785         | 0.003 | 0.759        | 0.772        |
|          | Pb    | 1.046         | 0.003 | 1.040        | 1.053        |
|          | Zn    | 1.033         | 0.003 | 1.027        | 1.039        |
Figure S1. Correlation between metal concentration and vegetable.

Figure S2. Correlation between metal concentration and soil.
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