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Potential Toxic Elements Accumulation in Several Food Species Grown in Urban and Rural Gardens Subjected to Different Conditions

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Abstract: Urban agriculture increased in Seville (South Spain) in the last 20 years and play different roles in the urban context. Edible species can be contaminated by soil and airborne contamination leading to health risks. Samples of different crop and fruit species and their soils were collected in urban and rural gardens, including urban gardens from a mining area to investigate the potential contamination in food and soils. Results show that soils from mining gardens were the most contaminated. In the city, crops were generally not more contaminated those in the rural area. Most differences were observed between species, chard and lettuce were the species that reached the highest level of most elements' accumulation and fruits always had lower metal accumulation than leafy vegetables. Arsenic, Cd, and Pb concentrations did not exceed the FAO/HWO and European legal maximum levels for vegetables studied, so their consumption would be safe for human health. The concentration of Cr, Cu, Mn, and Ni can be considered in the range cited in the bibliography. Special attention should be paid for leafy green vegetables (lettuce and chard) since high values of Ba and Zn were found, up to 42 and 123 mg kg^{-1}, respectively, and the risk to human health associated with consuming these species should be studied.

Keywords: urban agriculture; contamination; heavy metal; south of Spain

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1. Introduction

Urban agriculture (UA) is a growing activity in many cities worldwide and contributes to the achievement of several of the Sustainable Development Goals established by the UN for 2030 [1]. Among them, achieve food security and promote sustainable agriculture, guarantee sustainable consumption and production patterns and adopt measures to combat climate change. However, the list of reported benefits of the AU is more extensive and can also be cited, for example, food sovereignty and subsistence survival, improved physical and mental health in city dwellers, improved aesthetics, community building, employment opportunities, improved local land prices, shortened supply chains and, thus, reduced price differentials between producers and consumers, reduced food transportation distance, carbon sequestration, potentially reduced urban heat island effect, provision of habitat for wildlife, and waste recycling [2]. UA is practiced in both developed and developing economies, although generally serving different purposes, such as recreational in the first case or for subsistence and food security in the second [3]. Hence, research on urban agriculture is also growing from multiple perspectives that address social and economic matters [4], urban planning [5], several technical and agronomical aspects [6,7] as well as human health [8], and the environment [9].

Nevertheless, one of the UA disadvantages lies in the risks due to the possible contamination of urban products. Cities are places prone to contamination in all of the environmental compartments (air, soil, water, waste, and living beings), and so, by the mere fact of where UA is carried out, fruits and vegetables are susceptible to being contaminated by undesirable organic or inorganic substances [2,9–13]. The presence of heavy metals such as As, Cr, Cu, Fe, Mn, Ni, Pb, Sn, and Zn in urban soils is of great environmental concern due to the problems that their excessive exposure creates for health [14]. The pollutant with the greatest impact is Pb, which appears in numerous studies [13,15–18] and can affect consumers and gardeners [19,20]. A number of studies reporting pollution in soils and plant products of urban origin continue to appear [9–13,18,21–23] which means that this issue should be studied further, due to the evident risk for the food chain and human health. In view of the significant levels found of some contaminants, recent studies discuss recommendations to facilitate monitoring of edible tissues and to reduce risk [24,25].

However, it is also necessary to point out that in other studies there are no signs of contamination of soils or urban vegetables [2,26,27]. These contradictory observations are possibly due to the fact that different local conditions can significantly affect the contamination of vegetables of urban origin [28]. Among these local conditions, the geology of the place, vehicle traffic level, plant species, urban design, and punctual pollution phenomena by waste, compost, or phytosanitary products have been cited [12,29].

Generally, studies on pollution in urban orchards focus on specific cities or on comparing gardens in cities from different regions. Orchards are rarely compared on a regional scale that, however, may be subjected to different environmental impacts. The aim of this research was to compare the concentration of potentially toxic elements (PTE) in food species grown in orchards from a rural area, a mining site, and a medium–large city to detect if food security might be compromised based on the origin of the products.

2. Materials and Methods

2.1. Study Sites and Sample Collection

Seville, the capital city of Andalusia, is a medium-sized city with about 700,000 inhabitants with an extended suburban and metropolitan area with 800,000 inhabitants. Daily, about 400,000 thousand vehicles move in the city [30]. Seville has a total of 13 urban gardens that practice organic agriculture and cultivate crop and fruit species in the undisturbed soils. During 2019 and 2020 plant and soil samples were collected from the most popular food species grown in urban gardens. Three urban gardens inside the city were selected: Miraflores Norte Pino Montano, Vega de Triana, and Alamillo (Figure 1). Miraflores Norte Pino Montano is located in the northeast of the city, with a total surface of 24,574 m^2 and 226 allotments; Vega de Triana is an urban garden in the west of the city, close to Guadalquivir river, with 1777 m^2 and 37 allotments; Alamillo is located in the south of the city inside a metropolitan park and nearby a main road, it has 179 allotment in a surface of 10,366 m^2. These samples have been classified as City. Geologically, City soils corresponds to Quaternary fluvial terrace materials [31].

Another three urban gardens were selected in two villages close to the old mining area of Rio Tinto (Rio Tinto and Nerva, coordinates 376.881.540, −65.998.720; 376.885.520, −66.039.130), an area of great interest as it is one of the most contaminated fluvial–estuarine systems with heavy metals in the world [32] in the mining region of the Iberian Pyrite Belt. These samples have been classified as Mining.

A rural garden located in Aracena Mountain nearby the small village of Los Marines (coordinates 378.931.000, −66.322.870) was also chosen, and these samples were named Rural. It is a private garden about 100 km away from Seville. Geologically Mining and Rural soils are located in the southern Iberian Massif, in the South-Portuguese Zone (SPZ) the Mining sites and in the in the Ossa-Morena Zone (OMZ) the Rural ones [33]. Rural and Mining allotments also follow organic agriculture practices as well as City gardens.

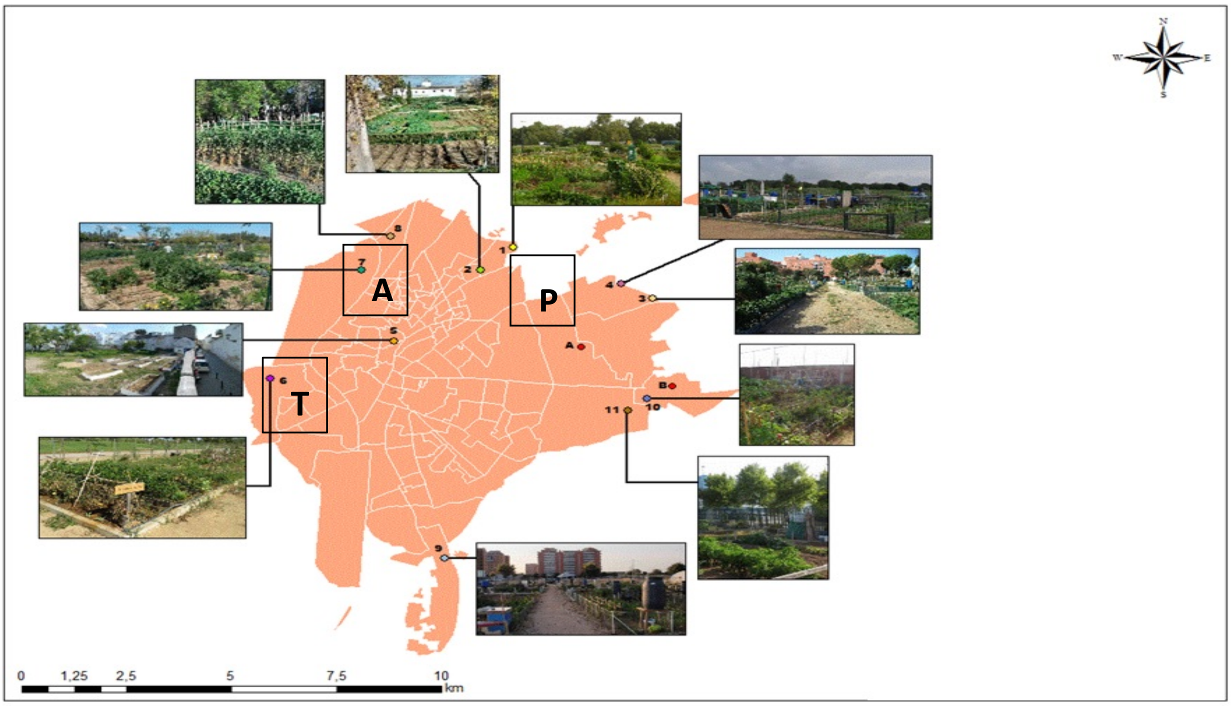

Figure 1. Urban gardens in the city of Seville selected for sampling. P, Pino Montano; A, Alamillo; T, Vega de Triana.

Samples were harvested from the most common plant species cultivated in the gardens: tomato (*Lycopersicon esculentum* Mill.), eggplant (*Solanum melongena* L.), pepper (*Capsicum annuum* L.), lettuce (*Lactuca sativa* L.), chard (*Beta vulgaris* var. *cicla* L.), and zucchini (*Cucurbita pepo* L.). In addition, samples of orange (*Citrus sinensis* (L.) Osbeck.) and tangerine (*Citrus reticulata* Blanco) were also collected in urban gardens from the mining area and strawberry in the city from Alamillo and Triana urban gardens (Figure 1). Eggplant, pepper, and chard samples were collected from the three study sites while tomato, zucchini, and lettuce samples were collected from city and rural sites since they were not cultivated in the gardens from the mining site. The number of soil/vegetable pair collected from a particular garden varied depending on sample availability. We collected a total of 63 plant samples. For each species, samples were collected from 3–5 plants depending of the species availability cutting the edible parts with scissors for leaf species.

Soil samples (0–20 cm depth) were taken in each of the allotments by combining at least 4 subsamples taken under each of the vegetables sampled.

2.2. Sample Preparation and Analysis

In the laboratory, plant samples were washed with distilled water and dried in an oven at 60 °C for several days until they were dry (fruits were peeled). After that, samples were grounded and digested with 8 mL of HNO_3 (65% Suprapur, Merk) in an open digestion system (DigiPrep, SCP Science, Montreal, QC, Canada). After cooling, plant extracts were transferred to volumetric flasks and diluted up to 25 mL with Milli-Q water, filtered, and the concentration of PTE was analyzed by inductive coupled plasma mass spectrometry (ICP-MS, Agilent 7800, Agilent Technologies, Santa Clara, CA, USA). Samples collected in the city were analyzed by inductive coupled plasma optical emission spectrometry (ICP-OES, Varian ICP 720-ES, Agilent Technologies, Santa Clara, CA, USA) due to the unavailability of the previous instrument. Quality assurance of chemical analysis in plants was performed using analytical blanks and certified reference material (NIST, Apple Leaves). All analyses performed were done in duplicate, and all results for plants and soils were calculated on a dry weight basis.

Soil samples were dried and sieved to less than 2 mm. PTE in soil samples were determined by X-ray fluorescence (XRF) following the EPA 6200 method [34]. The samples were placed in an XRF container (model SC-4331, 26-mm internal diameter, 24-mm height, Premier Lab Supply Inc., Port St. Lucie, FL, USA) capped with a 4-µm propylene film (model 240255, 63-mm diameter, Premier Lab Supply Inc., Port St. Lucie, FL, USA). The container was placed in the window of the laboratory stand of an analyzer Niton XL3t 950s GOLDD+ XRF (Thermo Scientific Inc., Billerica, MA, USA), and the soil sample was scanned in triplicate after repositioning the sample in the window using the Soil mode of the instrument. Although the usual particle size for trace element analysis is <0.25 mm, it was found that the standard deviation of the replicated measurements was very low, less than 1%, so the sieving by 2 mm was sufficient and allowed for time saving during analysis. The analysis time for each scan was 60 s and the average results of the three scans were calculated. More detailed information about the analyzer can be found in [35] and on the manufacturer's website [36]. The soil NIST 2709a [37] was used as reference material to assess the accuracy and stability of the pXRF instrument. Obtained metal concentrations for the reference material were within 82–115% in relation to certified concentrations, according to the ±20% relative difference allowed for this technique [34].

2.3. Data Analysis

For means calculation, the value of the detection limit was used to substitute the missing values in the case of values following below this limit.

Data were analyzed using the statistic program Statsoft. Element datasets were tested for normality with the Shapiro–Wilk's test ($p > 0.05$) and for homogeneity of variance with the Levene's test ($p > 0.05$). Data not respecting normality assumptions were log-transformed.

A one-way analysis of variance (ANOVA) was used to determine significant differences between areas (city, mining area, and control) and species. Differences among groups were tested by analysis of variance, using the LSD-test for post-hoc comparisons. When data were not normally distributed non-parametric Kruskal–Wallis and Mann–Whitney U test were used for mean comparison. When only two areas were compared because crop species were only available in two sites, the t-test was used.

From soil and plant metal concentrations, several indexes were derived. In the soil, the contamination factor (CF):

$$CF_i = c_i/cb_i \tag{1}$$

is the ratio obtained by dividing the concentration (c_i) of each metal (i) by its base line or background value (cb_i) [38,39]. The geochemical baselines of trace elements given by [33] for the Ossa-Morena Zone and the South-Portuguese Zone were used to calculate CF for the soils from the Rural and Mining areas, respectively. Geologically, City samples were included in the Guadalquivir river basin and its geochemical baseline of trace elements was obtained from [40]. The baseline values were determined by using HF for the acid digestion (HF, $HClO_4$, HNO_3, and HCl). Therefore, these are total metal contents equivalent to those determined by XRF.

The bioaccumulation coefficient (BC):

$$BC_i = C\ \text{edible part}_i/c_i \tag{2}$$

is the ratio between element concentrations in fruits/leaves (C edible part_i) and the concentration in soil. It was also calculated to determine the ability of the studied species to accumulate elements in the edible part. According to [41], the plants may be considered accumulators of an element if BC > 1.

3. Results

3.1. Metal Concentrations in Soil

Potential toxic element concentrations in soil from the three types of urban garden are shown in Table 1. Statistical differences among urban gardens were observed for all studied elements except for Sr. The urban garden soil from the Mining area showed the higher concentrations of most of the considered metals, i.e., As (77 mg kg^{-1}), Cr (117 mg kg^{-1}), Cu (206 mg kg^{-1}), Mn (1331 mg kg^{-1}), Pb (199 mg kg^{-1}), and Sn (22 mg kg^{-1}), than soils from the city (10, 28, 34, 458, 41, 11 mg kg^{-1}, respectively). The highest Ba (370 mg kg^{-1}) and Ni concentrations (49 mg kg^{-1}) were observed in City soil even if statistical differences were not found with Mining soil (332 and 39 mg kg^{-1}, respectively). The lowest Zn concentrations were observed in City soil (69 mg kg^{-1}) and the higher value in Rural soil (272 mg kg^{-1}).

Table 1. Average metal concentrations (mg kg^{-1}) ± standard deviation in the soil of the three types of urban gardens and certified or reference values in the reference material. Different letters indicate significant differences at $p < 0.05$ (Mann–Whitney U test except As, Ba, Cu, Mn, and Zn by LSD test).

Metal	City $n = 16$	Rural $n = 3$	Mining $n = 5$	NIST 2709a
As	10.4 a ± 3.8	21.0 a ± 3.4	77.0 b ± 44.1	10.5
Ba	370 b ± 50	264 a ± 129	332 ab ± 59	979
Cr	28.4 a ± 9.8	81.2 ab ± 13.4	117 b ± 22.0	130
Cu	34.5 a ±14.8	51.9 a ± 5.8	206 b ± 155	33.9
Mn	458 a ± 95	590 b ± 24	1331 c ± 128	529
Ni	48.8 ± 11.1	BDL [1]	39.5 ± 10.0	85
Pb	40.7 a ± 20.4	65.5 ab ± 4.7	199 b ± 84	17.3
Sn	11.5 a ± 2.8	7.1 a ± 0.0	22.5 b ± 9.4	–
Sr	168 ± 105	59.6 ± 25.8	164 ± 42	239
Zn	68.8 a ± 27.5	272 c ± 28	228 b ± 22	103

[1] The concentrations of metals Cd, Co, Hg, and Mo, were below the detection limit (BDL) (17, 285, 15, and 6 mg kg^{-1}, respectively). The concentrations of Ni in Control soils were below the limit of detection (23 mg kg^{-1}).

Figure 2 shows the mean CF values for each zone. Since the CFs are referred to the local geological background values, they allow a more adequate comparison between the three zones. It is evident that Mining soils were the most contaminated, except for Ni and Cr, with CF values between 3–6 for the metals As, Cu, Pb, and Zn.

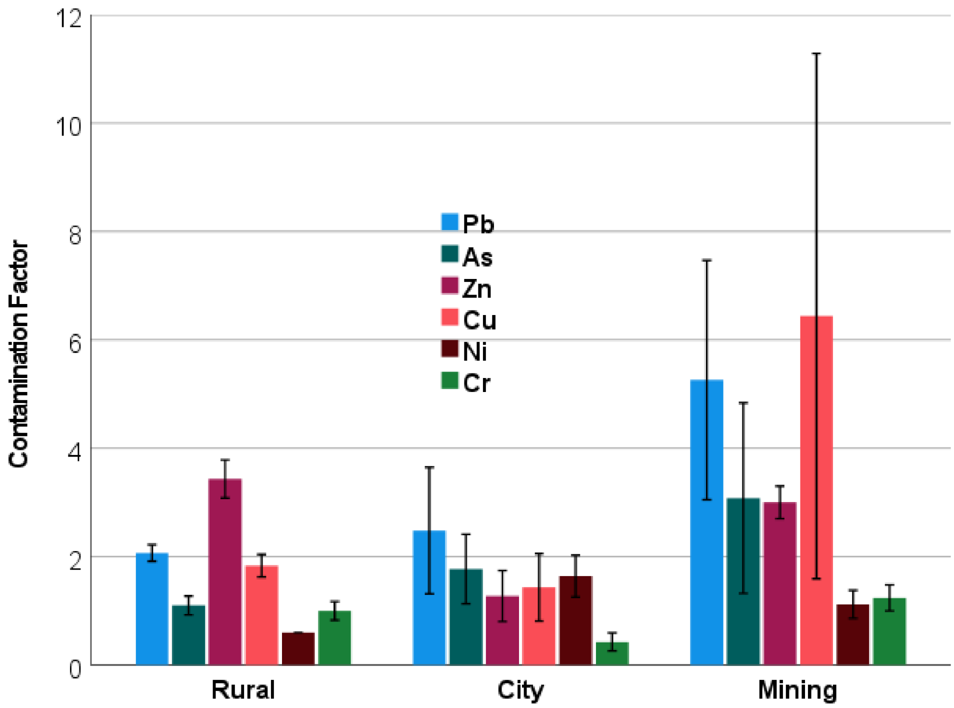

Figure 2. Mean contamination factors with standard deviation in the three types of urban gardens.

According to [38], CF values from 3 to 6 can be considered as considerable contamination. In the Rural soils, CF values for Pb, Zn, and Cu corresponded to a moderate contamination (CF = 1–3). However, it must be considered that within the geotectonic domains corresponding to Mining and Rural there is a great lithological variation that must be considered as it can give rise to large geochemical anomalies. Moderate contamination for As and especially Pb was observed in City soils, although the concentration of Pb in these soils (Table 1) was not particularly relevant.

3.2. Metal Concentrations in Plants

Metal concentrations in pepper, eggplant, and chard collected in the three sites are shown in Table 2. These species showed different element accumulation in Urban, Rural, and Mining sites, although the differences between element contents depended as well on the plant species.

In eggplant and chard, the concentration of As (0.10 and 0.13 mg kg^{-1}, respectively) and Cd (0.09 and 0.21 mg kg^{-1}, respectively) was highest in the Mining site compared to the Rural site. Although the concentration of As was not determined in plants from the City site, the As content in City soils was the lowest one (Table 1). The opposite pattern was observed for Ba and Pb, but again it depended on the species. Eggplant and chard from the Rural site showed higher Ba concentration than in the Mining site (6.41 and 42.4 mg kg^{-1}, respectively) and Pb accumulation in chard followed the same trend. Although it was not significant due to a high dispersion of data, the average concentration of Cr in chard from the Rural site was much higher (2 mg kg^{-1}) compared to the Mining values (0.29 mg kg^{-1}). Lead content also showed a big dispersion in eggplant from the City site that leads to no significant differences between sites.

The highest Mn concentrations were found in pepper and chard from the Rural site (21 and 253 mg kg^{-1}), and the highest Zn and Pb concentrations were observed in chard from the Rural site (0.83 and 102 mg kg^{-1}). Chard from the City site also showed higher Mn content than the Mining site. Eggplant showed the highest Fe concentration in the Mining site. Although it is not significant due to a high dispersion of results, the average concentration of Cr in chard from the Rural site was much higher (2 mg kg^{-1}) compared to the rest of the average values (<0.17 mg kg^{-1}).

Table 3 shows the concentrations of Cu, Fe, Mn, and Zn in samples from the City and Rural sites. In this case, only the comparison between these two sites was possible, although the results showed trends in accordance with those reported in Table 2. Tomato and lettuce plants showed the highest Mn concentration in the Rural site (17 and 55 mg kg^{-1}) and Zn was also particularly high in lettuce from Rural site (123 mg kg^{-1}). Tomato plants from Rural site also showed the highest Fe concentration (60.8 mg kg^{-1}).

The element accumulation (mean value) in strawberry from urban gardens of Seville and orange and tangerine from urban garden of Rio Tinto mining area are shown in Figure 3. Significant differences were observed only for Mn and Zn, with strawberry being the species with the highest values.

Table 2. Potential toxic element accumulation in pepper, eggplant, and chard cultivated in different urban gardens. Values are mean ± standard deviation expressed as mg kg^{-1}.

		As	Ba	Cd	Cr	Cu	Fe	Mn	Ni	Pb	Zn
Pepper	City (*n* = 7)	—	—	BDL	BDL	10.1 ± 3.18	30.6 ± 5.45	9.01 ± 2.10	0.65 ± 1.73	BDL	10.4 ± 3.29
	Mining (*n* = 6)	0.05 ± 0.02	0.94 ± 0.53	0.05 ± 0.01	0.10 ± 0.05	7.30 ± 3.95	35.5 ± 15.1	8.81 ± 0.88	0.55 ± 0.46	0.07 ± 0.03	14.9 ± 2.66
	Rural (*n* = 3)	0.02 ± 0.00	1.42 ± 0.21	0.04 ± 0.00	0.10 ± 0.00	10.8 ± 0.61	43.0 ± 0.58	21.3 ± 1.08	0.33 ± 0.02	0.07 ± 0.00	17.5 ± 0.28
	Significance	ns	ns	ns	ns	ns	ns	*	ns	ns	ns
Eggplant	City (*n* = 3)	—	—	BDL	BDL	8.41 ± 2.00	22.2 ± 4.02	12.9 ± 2.75	BDL	0.57 ± 0.98	18.3 ± 0.36
	Mining (*n* = 6)	0.10 ± 0.00	1.27 ± 0.67	0.09 ± 0.03	0.09 ± 0.01	7.69 ± 0.85	30.1 ± 3.51	14.0 ± 3.05	0.30 ± 0.12	0.06 ± 0.01	18.4 ± 1.07
	Rural (*n* = 3)	0.02 ± 0.00	6.41 ± 1.32	0.03 ± 0.00	0.09 ± 0.02	7.68 ± 0.99	25.4 ± 2.34	17.0 ± 0.65	0.06 ± 0.01	0.06 ± 0.01	17.1 ± 1.13
	Significance	*	*	*	ns	ns	*	ns	*	ns	ns
Chard	City (*n* = 5)	—	—-	BDL	BDL	7.45 ± 2.31	107.6 ± 36.8	168 ± 59.7	BDL	BDL	25.0 ± 2.63
	Mining (*n* = 3)	0.13 ± 0.00	19.2 ± 0.19	0.21 ± 0.00	0.29 ± 0.05	9.94 ± 1.41	101.8 ± 10.0	54.2 ± 4.10	0.28 ± 0.00	0.39 ± 0.22	26.3 ± 0.22
	Rural (*n* = 3)	0.09 ± 0.01	42.4 ± 1.00	0.17 ± 0.00	2.05 ± 3.19	6.16 ± 0.25	65.4 ± 4.17	253 ± 18.2	0.25 ± 0.02	0.83 ± 0.07	102 ± 4.47
	Significance	*	*	*	ns	ns	ns	*	ns	*	*

BDL, below detection limit; ns, not significant difference at $p < 0.05$; *, Significant differences at $p < 0.05$.

Table 3. Potential toxic element accumulation in tomato, zucchini, and lettuce cultivated in the City and Rural urban gardens. Values are mean ± standard deviation expressed as mg kg^{-1}.

		Cu	Fe	Mn	Zn
Tomato	City (n = 6)	5.62 ± 1.95	18.5 ± 4.37	7.33 ± 1.78	14.0 ± 3.66
	Rural (n = 3)	6.40 ± 0.89	60.8 ± 12.9	17.0 ± 2.41	20.6 ± 2.07
	Significance	ns	*	*	*
Zucchini	City (n = 4)	11.5 ± 3.8	49.9 ±22.8	15.9 ± 5.4	44.9 ±22.5
	Rural (n = 3)	8.12 ± 0.06	56.2 ± 5.6	21.7 ± 1.4	57.1 ± 3.2
	Significance	ns	ns	ns	ns
Lettuce	City (n = 5)	9.03 ± 2.67	92.8 ± 41.7	33.6 ± 11.0	41.0 ± 21.4
	Rural (n = 3)	10.5 ± 0.38	46.5 ± 1.40	55.1 ± 1.92	123 ± 3.38
	Significance	ns	ns	*	*

ns, not significant difference at $p < 0.05$; *, Significant differences at $p < 0.05$.

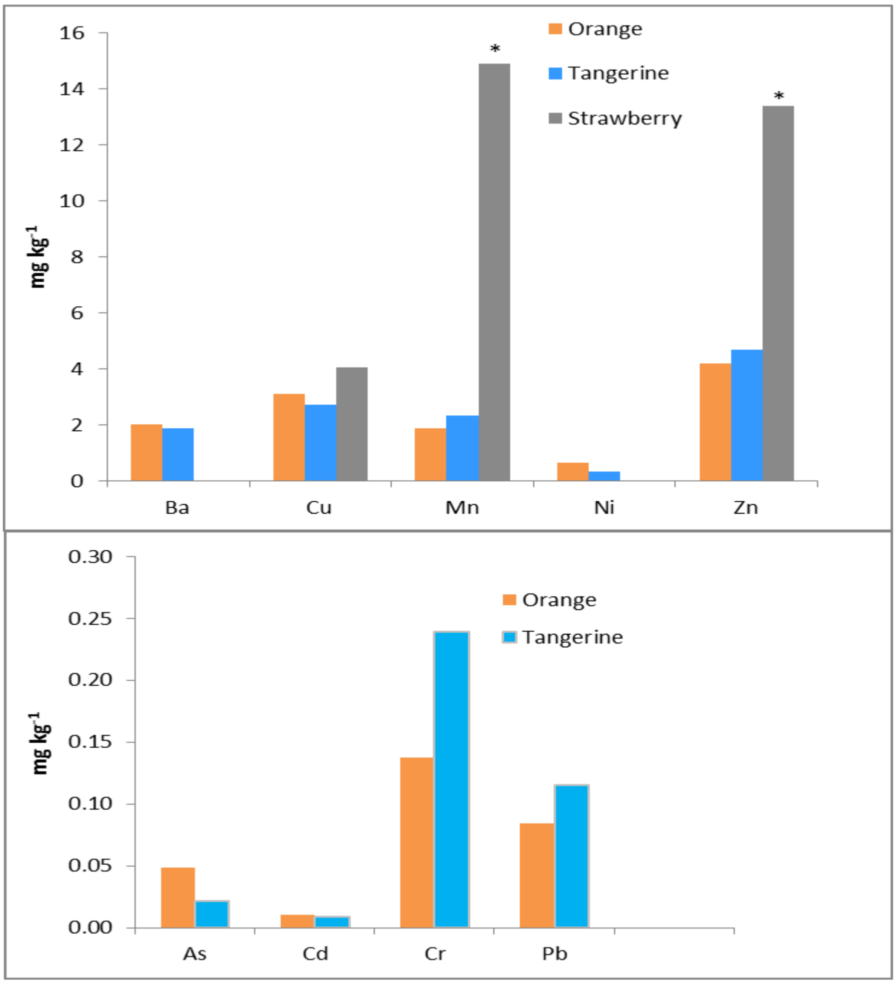

Figure 3. Element accumulation (mean value) in strawberry from urban gardens of Seville and orange and tangerine from urban garden of Rio Tinto mining area. Asterisk indicates statistical differences among species.

Remarkable differences in PTE accumulation were observed between the studied species for all elements except for Ni. Chard accumulated more As (0.13 mg kg^{-1}) compared with the other species (Table 2). In general, chard and lettuce were the species that reached the highest PTE accumulation (Ba, Cd, Mn, Pb, and Zn for chard and Mn and Zn for lettuce) with the only exception of Cu, with accumulation that was similar in almost all of the species expect for orange, tangerine, and strawberry that showed lower levels (Figure 4). Vegetables were grouped into two groups: (non-fruits) and fruits (including zucchini, tomato, pepper, and eggplant), and statistical differences were found for all PTE level except for Cu and Ni ($p > 0.005$). Non-fruits always showed the highest concentration of PTE (Figure 5).

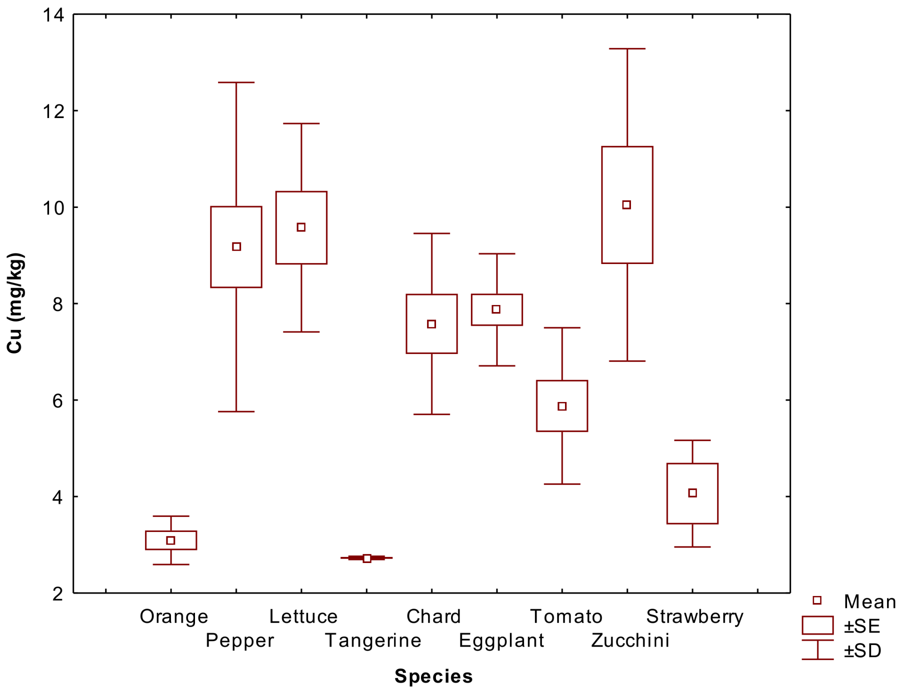

Figure 4. Copper accumulation in the different studied species growing in urban gardens.

Bioaccumulation in the studied vegetable species collected in the different types of urban gardens are shown in Table 4. The bioaccumulation coefficient was always <1 for all elements and species in all urban gardens. The highest coefficients corresponded to Cu and Zn with average values for all plants and sites of 0.16 and 0.23. The higher values of the coefficient corresponded to Zn in lettuce and zucchini of the City sites. Bioaccumulation coefficients for the rest of the elements were very low.

Table 4. Bioaccumulation coefficient ((BC) = C edible part/C soil) in the studied vegetable species collected in different urban gardens.

		As	Ba	Cr	Cu	Mn	Ni	Pb	Zn
Tomato	City	–	–	–	0.13	0.01	–	–	0.19
	Rural	–	–	–	0.10	0.02	–	0.001	0.07
Zucchini	City	–	–	–	0.27	0.03	–	–	0.57
	Control	–	–	–	0.15	0.03	–	–	0.21
Pepper	City	–	–	–	0.28	0.01	0.01	–	0.13
	Mining	0.0008	0.004	0.0008	0.05	0.006	0.01	0.0003	0.06
	Rural	0.0008	0.003	0.001	0.23	0.03	–	0.0009	0.08
Lettuce	City	–	–	–	0.26	0.07	–	–	0.63
	Rural	–	–	–	0.18	0.08	–	0.007	0.49
Eggplant	City	–	–	–	0.21	0.02	–	–	0.11
	Mining	0.001	0.003	0.0009	0.05	0.01	0.007	0.0003	0.08
	Rural	0.0009	0.02	0.001	0.09	0.02	–	0.0009	0.06
Chard	City	–	–	–	0.18	0.30	–	–	0.33
	Mining	0.002	0.06	0.002	0.07	0.04	0.006	0.002	0.11
	Rural	0.004	0.16	0.02	0.09	0.42	–	0.01	0.37

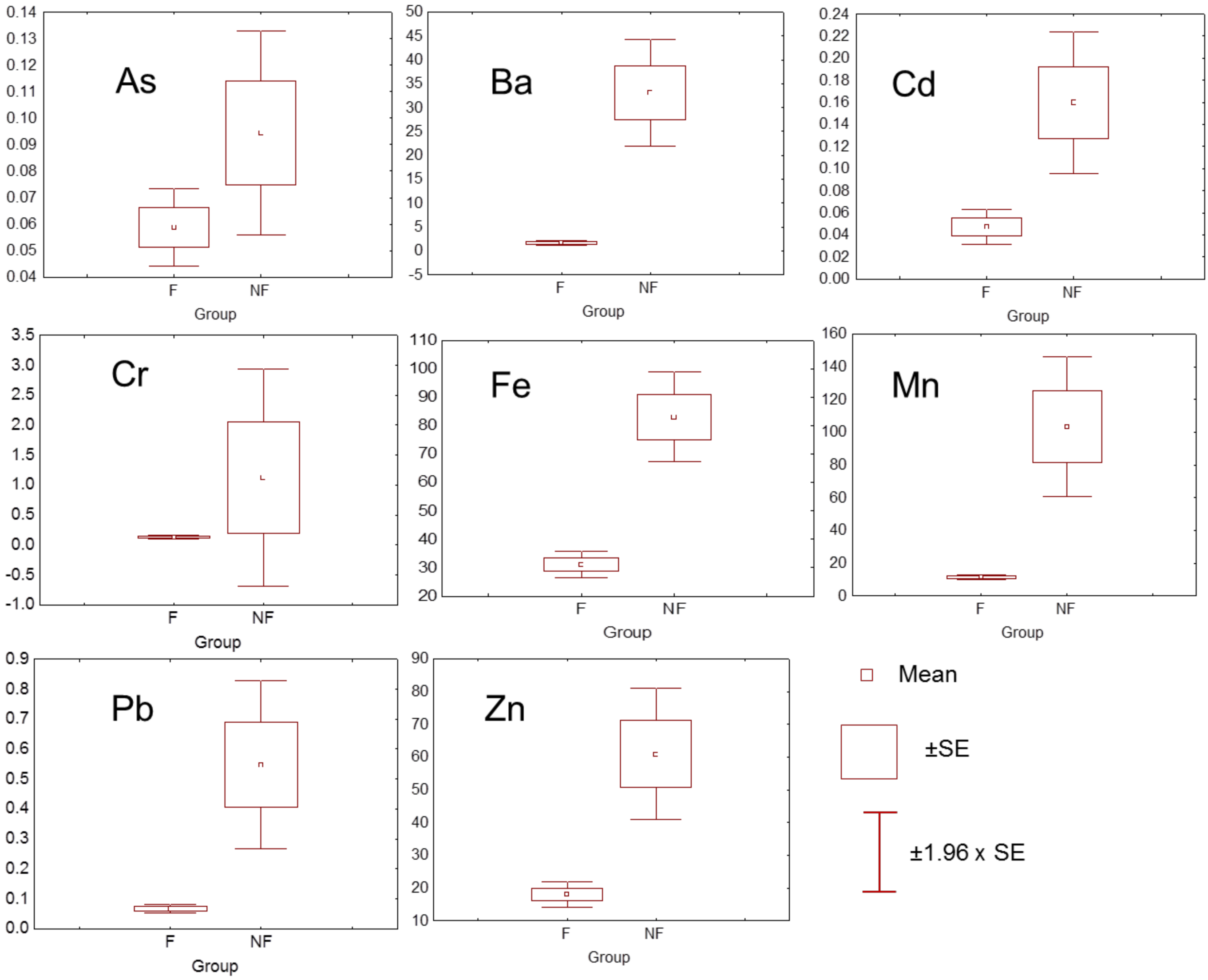

Figure 5. Potential toxic element concentration (in mg kg^{-1}) in urban garden grown species by fruit (F) and non-fruit (NF) type.

4. Discussion

4.1. Considerations on the Analytical Methodology

Elemental concentrations in soil determined with portable XRF are known to be affected by various factors, especially by soil moisture content, and it has also recently been indicated that by the content of organic matter in the soil [42]. With respect to moisture, it has no effect in this study when measurements are made on dry samples. In relation to the soil organic matter content, it should be noted that Mediterranean climate soils tend to be poor in organic matter. Although orchard soils receive important additions that could increase their content, organic matter contents generally lower than 50 g kg^{-1} have been found in urban soils of the same geographical area [15]. For these levels of organic matter, Ravansari et al. [43] calculated correction factors that showed little deviation (<2%) with respect to the same mineral soil, so the results of this study could be affected by an error of this magnitude due to the interference of the soil organic matter.

Furthermore, the certified elemental concentrations of the reference material are generally within the ranges measured in this study (Table 1). The loss on ignition (LOI) for the SRM 2709a soil was 66 g kg^{-1}, concordant with the organic matter contents that we can expect in the Mediterranean soils of this study [15]. Therefore, despite the biases inherent in this method, the utmost care has been taken to ensure the quality of the results.

For several elements, the concentrations measured in the City soils were close to the detection limits of the technique. For example, for As, Sn, and Cu, the guideline detection limits indicated by the manufacturer [36], (7, 20, and 15 mg kg^{-1}, respectively) were similar to the mean values indicated in Table 1. (10, 11 and 34 mg kg^{-1}, respectively). This means that the concentrations given for the City soils could be affected by a relatively large error, although in any case, they would always be lower values than those of the other sites.

4.2. Differences among Soil–Plant Relations

Urban gardens are often located in contaminated areas, so exposure of humans to contaminants via consumption of home-grown vegetables and fruit may constitute a potential health risk. Comparisons between element level in crop species from the City and Rural sites were possible only for Cu, Mn, and Zn, since the other element concentrations were below detection limit as analyzed by ICP-OES. Despite the results of other studies [28,44], none of these elements had a higher concentration in vegetables from the City (range for Cu: 2.8–10.1 mg kg^{-1}; range for Mn: 5.0–168 mg kg^{-1}; range for Zn: 9.0–45mg kg^{-1}) than from the Rural site (range for Cu: 6.4–10.8 mg kg^{-1}; range for Mn: 17.0–253 mg kg^{-1}; range for Zn: 17.1–123 mg kg^{-1}). These elements have a similar value in soils from the City and Rural urban gardens (Table 1), except for Zn level, which was higher in soil from the Rural site (272 mg kg^{-1}). This can explain the high values of Zn found in chard (102 mg kg^{-1}) and lettuce (123 mg kg^{-1}) in the Rural urban garden (Tables 2 and 3). Mining soils can be classified as considerably contaminated by Cu according to the CF value (Figure 2). Such a high Cu concentration in the soil is not at all surprising considering the ancient mining tradition of the area. About 60 mines were operative during the last century, mainly for S and Cu [33]. However higher accumulation of As, Cd, and Ni were observed in some crops (eggplant and chard) from the Mining site than those from Rural. Differences for As in soils from these two sites were not significant, whilst Ni concentration in the Rural site was below the detection limit (Table 1). This suggests that PTE accumulation in the studied species is mainly related to plant species and correlation between some PTE concentration in soils and PTE concentration in crops is often poor or inconsistent. Actually, soil from the Mining urban gardens had the highest Mn value (1331 mg kg^{-1}), but none of the studied species, reflected this tendency and soil from Mining and Rural sites had a similar Pb value (Table 1) but chard growing in the Rural site had a significantly higher Pb content than the Mining site (Table 2).

For non-vegetable fruits, strawberry grown in the City urban gardens showed the highest Mn accumulation (Figure 3), but the City soil presented less Mn concentration than the Mining one.

Concentration of trace metals in plant species growing in contaminated soils were often at non-toxic levels [45,46].

4.3. Differences among Plant Species

Plants can uptake PTE from soil but atmospheric deposition could also be another important pathway that should explain these results.

Plants are able to absorb PTE but the uptake varies with plant species and organs [28,47]. The level of PTE in fruits was in general lower compared to those in vegetable as showed by other authors [44,48]. This is due to the different transport system. Fruits receive nutrients from the phloem and the delivery of some PTE is strictly controlled. The level of Pb and Cd observed in fruit species were lower than values reported in fruit from urban garden in NYC and Buffalo [49].

Chard showed high value of Ba (42.4 mg kg^{-1}) growing in the Rural site. Barium is not an essential element for plants and in tomato and fruits the mean concentration reported by Kabata and Mukherjee [50] is 2.1 mg kg^{-1}, whilst in lettuce, it had a range of 9–11 mg kg^{-1}. However, no health-based standards or guidance value exist for Ba in food crops, so we cannot interpret the value as a health risk. Urban garden soil from Rural site has 264 mg kg^{-1} Ba, a similar value than in the Mining site, but lower compared to soil from the City. Barium was considered as a marker of anthropogenic activity [51] and also in Seville city soils were enriched by Ba [52]. In the urban garden of New York City, lower Ba mean concentration was reported and no correlation between total soil Ba and crop Ba was observed [49]. Barium concentration found in soils of the studied urban garden were lower than the European topsoil's baseline [53]. Regional guideline values for this element are not available. Chard accumulated the highest concentration of As; the mean value in this species grown in urban garden from Mining site (0.13 mg kg^{-1}) is lower than values reported by Kabata-Pendias [54] for leaf species. In the same Mining area, Monaci et al. [55] reported higher As concentration in plant leaves. Paltisava et al. [17] reported higher As level in root species than in leaf species. FAO/WHO [56] established a value of 0.20 mg kg^{-1} in rice, so the value found in chard cannot be considered hazardous for human health. However, As in the Mining area should be monitored especially considering that one of the plots of the area (155 mg kg^{-1}) quadruples the guidance value (36 mg kg^{-1}) established by the regional government as a contamination criteria for soils [57].

In general, leafy vegetables (chard and lettuce) accumulated more PTE than fruits species. Similar results were reported in other studies [13,17,48,58] and metabolic differences among species have been proposed to explain this pattern [59]. The PTE concentration found in chard and lettuce are below the normal value in plant foodstuffs [54] except for Zn. The mean value of Zn observed in lettuce and chard growing in the Rural area is higher than values reported for lettuce (44–73 mg kg^{-1}, [54]), where soil has also the highest Zn concentration (Table 1), but there are no EU health-based standards or guidance value for Zn in food crops, so it is not possible to interpret the value as a health risk and a risk estimation by a different exposure scenario would be needed. If we consider the USA standard in vegetable (0.7–8 mg kg^{-1} FW) values for lettuce and chard are high. Assuming a dry matter content in the samples of around 10%, the range of Cd and Pb in the edible part of the studied species was always below the values established by European union regulation [60]: 0.05–0.2 mg Cd kg^{-1} FW and 0.1–0.3 mg Pb kg^{-1} FW. According to Egendorf et al. [25], most of the Pb accumulated in plants came from airborne contamination and in root and leaf food species the main Pb and As sources originated from adhered soil particles [17]. Even if the Mining site urban garden soils had high Pb value (Table 1), the washing of species considerably reduced the Pb level. Lead concentration of soils is not the main factor that influences the Pb concentration of vegetables, since anthropogenic and environmental factors are known factors [61]. In vegetables, the washing was effective to reduce As and Pb concentration, but when soil contamination is high the effect of washing should be strongly investigated. It should be taken into account that in soil from one of the plots of the Mining area Pb content (345 mg kg^{-1}) was higher than the limit value considered by the local government for potentially contaminated soils (275 mg kg^{-1}) [57], so the research on soils of this area should also be extended. Pepper grown in Alamillo had a high value of Ni (4.60 mg kg^{-1}). This value overcome the Ni concentration in plant foodstuffs (0.06 to 3.3 mg kg^{-1}, [50]. López et al. [15] also reported lower Ni level in lettuce grown in Miraflores urban garden of Seville. The Ni concentrations in the soil of the corresponding plots, 68.1 mg kg^{-1}, do not justify the high concentration of pepper. Near the plots where this value is recorded, there is a heating boiler that could be the origin, but this point must be investigated in greater depth to determine the cause.

The bioaccumulation coefficient (BC) was always <1 for all studied elements and species in all urban gardens (Table 3), indicating that both leaf and fruit species act as excluder and translocation to the edible part is very low. Similar results for Pb was observed by other authors [28,44]). This plant characteristic is very important for edible species and suggests that an important contamination input should come from atmospheric deposition.

5. Conclusions

Although the orchards included in this study were based on very different soils by their PTE concentrations, due to their different geology, the variations found in vegetables have been more related to the plant species than to the soil contamination. Many of the studies carried out on trace elements in urban gardens show that vegetables are contaminated by transfer from the soil or deposition of soil particles. However, this study indicates that urban agriculture can develop in soils with a certain level of contamination (up to 77 mg As kg^{-1}, 206 mg Cu kg^{-1}, 1331 mg Mn kg^{-1}, and 272 mg Zn kg^{-1}) if an adequate selection of the species to be cultivated is made. Therefore, together with soil-based remediation strategies, it would be necessary to study other strategies that are based on an adequate selection of species and varieties with limited capacity for trace element accumulations. In any case, this study, in general, did not find concentrations of PTE in food species at a toxic level for human health, even though the orchards were located in potentially contaminated sites, due to intense mining activity or urbanization, except for some specific allotments. Attention should be paid for the leaf species (lettuce and chard) since elevated concentration have been observed in some allotments for Ba (42 mg kg^{-1}), and Zn (123 mg kg^{-1}) and a more extensive studied is recommended.

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