Multiple potentially toxic elements in urban gardens from a Brazilian industrialized city

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Abstract

Urban agriculture should be promoted as long as the food produced is safe for consumption. Located in the metropolitan region of São Paulo-Brazil, Santo André has intense industrial activities and more recently an increasing stimulus to urban gardening. One of the potential risks associated to this activity is the presence of potentially toxic elements (PTEs). In this study, the concentration of PTEs (As, Ba, Cd, Co, Cu, Cr, Ni, Mo, Pb, Sb, Se, V and Zn) was evaluated by soil (n=85) and soil amendments (n=19) in urban gardens from this municipality. Only barium was above regulatory limits in agricultural soil, although enrichment of all elements was observed. A multivariate statistical approach was applied and indicated two groups of elements with strong influence of the petrochemical complex located in this region. However, carcinogenic, and non-carcinogenic risks were not observed. Soil amendments were identified as a possible source of contamination for Ba, Zn and Pb and for pathogenic bacteria. Besides that, the occurrence of antimicrobial resistance suggests some soil management practices are necessary.

1. Introduction

In Brazil, during the Covid-19 pandemic, food insecurity increased due to social inequalities and reinforcing national nutritional problems, which can have health impacts on individual, community, and growth levels at short and long-term. Currently, urban, and peri-urban agriculture (UA) are considered a sustainable alternative (Salomon et al., 2020). Especially, in developing countries (Nabulo et al., 2012), UA contributes to food security and strengths communities under vulnerability (Roese & Curado, 2004). On a global scale, communities have identified UA as a viable option to increase their access to health, nutrition, quality of life and low-cost fresh products (Deelstra & Girardet, 2000; Schram-Bijkerk et al., 2018). Approximately 1.1 billion people are involved in some type of UA in the world (Mougeot, 2015).

However, the introduction of toxic substances (deliberately or not) and inefficient management practices in urban environments can cause the degradation of soil quality and, consequently, affect food security, groundwater quality and human health. Potentially toxic elements (PTEs), such as As, Cd, Cu, Pb and Zn, are often detected in urban soils and in vegetables grown on them (Nabulo et al., 2012; Wiseman et al., 2013; McBride et al., 2014; Spliethoff et al., 2016; Laidlaw et al., 2018, Ćwieląg-Drabek et al., 2020). These elements can cause ecological risks and can accumulate in the human body resulting in several adverse health effects.

Another risk associated with UA is the contamination by pathogenic microorganisms. Microorganisms can come from the soil itself, from the manure and fertilization system (for example, fresh animal waste or non-composted urban waste that is in direct contact with edible parts of plants), from irrigation water, or even from incorrect handling and hygiene in the post-harvest (Urra et al., 2019). Concern about pathogens in vegetables has increased due to the increased number of disease outbreaks caused by the consumption of fresh, whole foods, cut and minimally processed vegetables (Sant'Anna et al., 2020).

Eighty five percent of the Brazilian population lives in urban areas (IBGE-PNAD, 2015) and 23.3% of the urban families are still suffering from food and nutrition insecurity (IBGE, 2013). The support for UA became part of the national policy for poverty reduction and food security guarantee from the early 2000s. In 2018, the Ministry of Social Development launched the National Program for UA (Brazil, 2018), which aims to contribute to the promotion of health and healthy habits and the food and nutritional security. It was estimated that there are more than 600 initiatives of urban and peri-urban food production in the country for both self-consumption and commercial purposes (Sant'Anna de Medeiros et al., 2020).

Santo André is a city of metropolitan region of São Paulo (MRSP), Brazil, one of the largest urban conglomerates in the world, with > 20 million inhabitants (UN, 2019). It is one of the cities of an automobile industry cluster (so-called ABC) organized around the Anchieta motorway, which connects São Paulo to the Port of Santos (Fernández-de-Sevilla and dalla Costa, 2020). The city has a fleet of more than 500,000 vehicles (DENATRAN, 2019). PTEs emissions from vehicular source can contaminate urban gardens (UG) by atmospheric deposition (Uzu et al., 2014). In the city there is also an industrial complex, the Capuava complex, with 125 hectares and 14 chemical plants that produce polyethylene, naphtha, cement and fertilizers. The industrialization process began in the 1950's, with the construction of a petrochemical refinery. This refinery produces about 30% of the fossil fuels consumed in the MRSP (Caumo et al., 2017). Petrochemical plants may be responsible for supplying PTEs to the environment (Manno et al., 2006; Marques et al., 2020).

This region has experienced an intensification of UA in the last decade (Amato-Lourenço et al., 2020). It is hypothesized that this long history of industrial activities and environmental pollution may cause the accumulation of some PTEs in soils affecting UA and may pose risk to the urban gardeners. In this scenario, the present study aimed (i) to evaluate the levels of PTEs in soils and soil amendments (SAs) in the UG of Santo André; (ii) to assess human health effects in these topsoils; (iii) to investigate the possible sources of PTEs; (iv) to assess the presence of pathogenic microorganisms in bed soils and SAs. The findings of the present study may provide baseline data needed to plan and improve UA.

2. Material And Methods

2.1. Site selection

The studied UG were selected at Santo André, Brazil. The city has about 715 thousand inhabitantsThe sites were selected into urban perimeter on the surroundings of the industrial area of Capuava (maximum 10 km of distance) (Fig. 1). Sites with the following characteristics were selected: (i) larger than 500 m^2; (ii) with commercial purpose; (iii) at least 3 years old of existence; (iv) well known on the surrounding community.

2.2. Sampling

Sampling of soil and SAs occurred in the dry season of 2019. The number of samples varied widely depending on the number of beds and number of lettuce heads cultivated per bed, as described in Table 1.

Three subsamples of topsoil samples (0–20 cm) were collected with a stainless-steel manual drill at each point; thus, a composite sample of 2 kg was produced, following a grid of one sample every three beds using a zig-zag sampling scheme, according to EMBRAPA (2011) recommendations. Samples were collected, homogenized, removing demolition waste, such as brick, tiles, steel, wood, plastic, glass, rubbers, and others. All studied gardens amended their bed soils, however the kind of amendment (Table 1) and the amount applied was very diverse among the sites. Circa of 250 mg of SAs samples were collected directly from the piles or storage containers. All samples were packed in plastic bags and immediately taken to laboratory.

Table 1
Details of sampling location, type and number of samples of the selected urban gardens of Santo André.

Location	Number of samples		Description of soil amendments
	Soil	Soil Amendments	
Capuava 1 (23º64'S 46º49'W)	8	3	Local compost pile and spent mushroom substrate
Capuava 2 (23º64'S 46º48'W)	9	4	Lime and quail manure
Capuava 3[a] (23º63'S 46º49'W)	-	2	Cow manure
Jd Marajoara (23º66'S 46º49'W)	15	4	Quail and chicken manure
Bairro Jardim (23º65'S 46º64'W)	22	4	Meat and bone meal
Vila Bastos (23º66'S 46º53'W)	14	-	-
Bela Vista (23º69'S 46º52'W)	17	2	Local compost pile
Total	85	19	

2.2. Sample preparation and analysis

Soil and SAs samples were dried at 40°C until constant weight and sieved to provide the < 2 mm and < 150 µm size fractions. About 400 mg of soil samples (< 2 mm) from at least 3 samples per site were analyzed for the following parameters: texture, pH, organic matter content, cation exchange capacity, potential soil acidity (H + Al), exchangeable cations (K, Ca, Mg, and P), sum of bases (SB) and base saturation (V%) determined by the methodology proposed by to Camargo et al. (2009). About 500 mg of soil and soil amendment were pre-digested for 48 h with 10 mL of sub-boiled concentrated HNO_3 (Synth, Diadema, Brazil). Then, the samples were heated in a digesting block at 175 ºC for 15 min according to USEPA 3051A method (Element, 2007), with some modifications according to Segura et al. (2016) and Suda & Makino (2016). The digested samples were diluted to 50 mL with type 1 water and then, 5-fold diluted.

The elements were determined using an inductively coupled mass spectrometer (ICP-MS Agilent 7900, Hachioji, Japan). A multi-element stock solution (10 mg L^{-1}) (Perkin Elmer, USA) was used to prepare the calibration curve according to Paniz et al. (2018) and internal standard Ge 10 mg L^{-1} were also used. Certified reference materials (CRMs) NIST SRM 1573 - tomato leaves and NIST SEM 695 - Trace Elements in Multi-Nutrient Fertilizer (NIST, Gaithersburg, MD, USA) and TILL-4 (soil) (Canadian Certified Reference Materials, Vancouver, Canada) were used throughout the analysis for method accuracy. The results of the analysis of the CRMs were statistically consistent with the certified values.

2.3. Microbiological analysis

For microbiological analyses, 40 g of soil and SAs samples from beds were collected randomly from different locations in the field. Then, they were immediately put into sterile 50-ml falcon tubes, transported in an ice chest with ice gel units to the lab and stored at 8ºC. A total of 21 samples were collected.

Bed soils and SAs samples were cultured in MacConkey agar (Oxoid, Basingstoke, UK) plates which were incubated at temperature of 37°C during 18–24 h. After bacterial growth, different colonies from each plate were selected and the identification of bacterial genera was performed by biochemical assays with EPM, MILi and Simmons citrate (Ewing, 1986; Toledo et al., 1982a, b) to confirm the presence of *Enterobacteriaceae* family. The bacterial isolates were stored in Brain Heart Infusion (BHI) (Difco, USA) plus 20% glycerol (Sigma-Aldrich, St-Louis, USA) media at -80°C.

The antimicrobial susceptibility of bacterial isolates was determined using the disk-diffusion in agar method, as recommended by the Clinical and Laboratory Standards Institute guidelines (CLSI, 2008; CLSI, 2018). The following antimicrobial agents were used: amoxicillin-clavulanic acid (AMC), aztreonam (ATM), cefepime (FEP), cefotaxime (CTX), ceftazidime (CAZ), cefoxitin (CFO), chloramphenicol (CLO), ciprofloxacin (CIP), fosfomycin (FOS), gentamicin (GEN), imipenem (IMP), nalidixic acid (NAL), nitrofurantoin (NIT), norfloxacin (NOR), tetracycline (TET) and trimethoprim sulfamethoxazole (SXT). All antimicrobial used were from Oxoid. Enrofloxacin (ENO) was also tested because this antimicrobial is commonly used in veterinary clinics. *E. coli* strain ATCC 25922 was used as a quality control for antimicrobial susceptibility testing.

2.4 Geoaccumulation index

Soil contamination by some PTEs in UG was estimated using the geoaccumulation index (*Igeo*) (Müller, 1969), according to Eq. 1.

$Igeo = \log_2(Cn/1.5Bn)$ (Eq. 1)

where Cn is the total concentration of an element in the tested soil and Bn is the respective concentration of the element in the reference environment from CETESB (Environmental Protection Agency of the State of São Paulo, Brazil) soil quality values were used (CETESB 2016). The constant 1.5 was used to compensate minor natural and anthropogenic fluctuations. The calculated results were compared to the seven classes proposed by Müller (1969): $Igeo < 0$, unpolluted; $0 < Igeo \leq 1$, unpolluted to moderately polluted; $1 < Igeo \leq 2$, moderately polluted; $2 < Igeo \leq 3$, moderately to severely polluted; $3 < Igeo \leq 4$, severely polluted; $4 < Igeo \leq 5$, severely to extremely polluted; $Igeo > 5$, extremely polluted.

2.5. Health risk assessment

For results obtained in the soil samples, the risk assessment was divided into non-carcinogenic (hazard quotient (HQ)) and carcinogenic risks (CR), considering three exposure routes: ingestion and inhalation and dermal according to the methodology proposed by Gabarrón et al. (2017) and Hu et al. (2017) based on The US Environmental Protection Agency methodology (USEPA, 2004) and using the selected parameters (Table S1). The details of the methods were provided in the supplementary materials. The carcinogenic risk was calculated only for As, Co, Cr, Ni and Pb and non-carcinogenic risk was estimated for all studied PTEs.

2.6 Data analysis method

In order to obtain information about the possible sources of the PTEs, the concentration of the elements was evaluated by principal component analysis (PCA) and Pearson's correlation. Statistical significance levels were expressed as $p < 0.05$. Principal component analysis (PCA), a multivariate statistical test, was performed to assess multivariate relationships.

PCA has been used to identify natural geochemical origins and anthropogenic sources. The dataset consisted of a matrix with 85 cases (collection points) and 13. Data were normalized and rotated (normalized varimax), grouping parameters. Only eigenvalues > 1 were considered in this study. Group loading values > 0.50 were used with high or strong correlation.

Hierarchical Cluster analysis (HCA) was performed in the data set. The values were normalized. Ward's method was used as a procedure of clustering. Euclidean distance was applied to measure similarity and the statistical significance of the clusters formed was checked by the linkage distance (dlink), expressed as the percentage of the range from the maximum to the minimum distance (dmax) in the data set, Dlink/Dmax*100. A dendogram ilustrates clustering of the similar variables (PTEs) considering the hierarchical structure. All statistical analyzes were performed using using Statistica software version 7.0 (Statistica (Tulsa, USA).

3. Results And Discussion

3.1. Soil results

Table S2 shows soil (0–20 cm) physical-chemical parameters of six studied gardens. Most samples consisted of neutral soils, with pH ranging from 5.3 to 7.6 and the lowest values (5.9 ± 0.6) were found at Capuava 1 garden. The samples showed content of clay between 224 and 547 g kg^{-1}, with an average of 362 g kg^{-1}, high sand content ranging from 305 to 646 g kg^{-1} and, in general, a median silt content, with an average of 200 g kg^{-1}. The soil texture varied from loam to clay soil, with a predominance of sand clay loam soil. This type of soil has a low potential for PTEs retention when compared to clay soils, present predominantly in the gardens of Bairro Jardim and Vila Marajoara. The content of organic matter (OM) varied widely from 21 to 68 g kg^{-1}, the highest values were observed at Capuava 1 (average of 61.3 g kg^{-1}).

The cation exchange capacity (CEC), which expresses the capacity of soil to retain cations, presented an average of 152 mmolc dm^{-3}. The base saturation presented was high in all gardens, varying between 88 and 96% and the soil can be classified, according to the current Brazilian soil classification system (SiBCS), as eutrophic soil ($V \geq 50\%$) (EMBRAPA, 2013). The concentrations of exchangeable phosphorus in the vegetable garden soil were all above 120 mg dm^{-3}, which is considered a minimum sufficient value for productive cultivation (Raij et al. 2011). The univariate ANOVA statistical test, with 95% confidence interval, was applied to the results presented in Table S2 and revealed that there are significant differences between the parameters in the areas, except for pH and exchangeable P.

The frequency distributions of concentrations of PTEs measured in the garden soil samples are described in Table 2. The data are not normally distributed (Kolmogorov–Smirnov test, $p < 0.010$) for most elements, except for As, Cr and Ni. Median (50th percentile) were lower than mean concentrations for Ba, Cd, Co, Cu, Mo, Pb, Sb, Se and Zn and higher for V.

Table 2
The concentrations of potentially toxic elements (mg kg^{-1}) in sampled urban garden soils in Santo André (n = 85) and the guidance values stablished by the local environmental protection agency (Environmental Protection Agency of the State of São Paulo, Brazil) (CETESB 2016).

Element	Mean	Min	Percentile				Max	State of São Paulo guidance values(a)			Melbourne, Australia(b)	Sheffield, England(c)	Belo Horizonte, Brazil(d)	Toronto, Canadá(e)
			25th	50th	75th	95th		QRV	PV	AIV	Mean (Range)	(Range)	(Range)	Mean (Range)
As	14	8.5	11	15	17	19	21	3.5	15	35	8(5–14)	(23.93–44.33)		5.0(2.8–7.1)
Ba	112	20	45	64	85	479	1001	75	120	500				
Cd	0.4	0.11	0.21	0.26	0.45	0.8	1.1	< 0.5	1.3	3.6			(0.09–0.23)	0.34(0.27–0.4)
Co	2.1	0.91	1.5	1.8	2.3	4.2	5.1	13	25	35				6.6(4.5–8.7)
Cr	59	24	46	61	74	81	89	40	75	150	17(8–32)	(86.67-160-67)		25(19–32)
Cu	51	16	28	38	62	140	237	35	60	760	40(17–102)	(46-131.33)	(10.52–64.22)	26(22–33)
Mo	1.11	0.273	0.690	1.01	1.23	2.59	4.31	< 4	5	11				
Ni	10	4.1	7.6	9.2	12	17	20	13	30	190	15(4–36)			14(11–18)
Pb	30	10	17	24	36	76	88	17	72	150	102(17–578)	(157-531.33)	(10.5–64.2)	57(19–96)
Sb	0.089	0.015	0.038	0.060	0.10	0.28	0.39	< 0.5	2	5				0.4(0.22–0.61)
Se	0.99	0.46	0.72	0.85	0.99	2.2	2.3	0.25	1.2	24				
V	106	34.5	72.1	119	143	159	173	-	-	-				
Zn	143	30	86.6	114	179	328	383	60	86	1900	218(62–950)	(177.33-727.33)		

(a)Quality reference value (QRV), Prevention value (PV) and Agricultural intervention value (AIV) established for soil in the state of São Paulo (CETESB, 2016); (b) Soils from community gardens (Laidlaw et al., 2018); (c) Urban allotment soils (Weber et al., 2019); (d) Soils from urban gardens (Dala-Paula et al., 2018); (e) Soils from community gardens (Wiseman et al., 2013).

The mean concentrations of elements detected in soil samples ranked in the following order: Zn > Ba > V > Cr > Cu > Pb > As > Ni > Co > Mo > Se > Cd > Sb. PTEs results were compared with the local environmental protection agency (Environmental Protection Agency of the State of São Paulo, Brazil) (CETESB 2016) (Table 2). The quality reference (QRV) represents the natural concentrations of chemical elements in soils without anthropic influence; the prevention value (PV) represents a sort of alert and the intervention value for agricultural (AIV) soils represents the threshold value. The PV and AIV were established based on human health risk (CETESB, 2016). The spatial variation of the PTEs in the studied UG are represented in Fig. 2.

The soils of the studied UG presented content values above the QRV for almost all elements, except for Co, Mo and Sb, whose concentration values are not shown in Fig. 3. In addition, values above the PV were observed for As, Ba, Cr, Cu, Pb, Se and Zn and above the agricultural VI for Ba in the case of the Capuava 1 garden. The areas around the Capuava Petrochemical Complex were those that presented, in general, higher levels of PTEs, especially for Ba, Cu, Pb, Se and Zn (Fig. 2).

In a similar study in soils of three UG from the metropolitan region of Belo Horizonte, Dala-Paula et al. (2018) reported values of concentration of Cu (27.9 ± 13.9 mg kg^{-1}), Pb (19.4 ± 7.7 mg kg^{-1}) and Cd (0.16 ± 0.03 mg kg^{-1}) lower than those observed in the mean values of the gardens studied in

Santo André, 51 ± 37 mg kg^{-1}; 30 ± 20 mg kg^{-1} and 0.4 ± 0.2 mg kg^{-1}, respectively. Therefore, vegetables grown in Santo André are at greater risk of contamination via soil transfer by these elements when compared to UG from Belo Horizonte, another very populated city in Southeast Brazil.

Studies concerning concentrations of PTEs in soils in the MRSP are scarce. Therefore, the concentration values of As, Ba, Cr and Zn obtained in the soils of this study, collected in the urban region of Santo André, were compared to the study carried out by Figueiredo et al. (2011). In this study, the authors evaluated PTEs contents in superficial soils collected in different public parks in the city of São Paulo, 20 km far from Santo André. These parks in São Paulo are located in different scenarios of urban zoning (central, residential and industrial areas). In 9 of the 12 parks studied by the authors, the levels of Ba concentration exceeded the residential VI of CETESB (2014) and the reported concentration range was 284 to 1022 mg kg^{-1}, which is higher than that observed in the soils of the gardens of Santo André (19 to 1000 mg kg^{-1}).

The highest values of Ba in our study were observed in the Capuava 1 area (Fig. 2), which is located less than 30 m from the petrochemical complex, in high-traffic vehicular routes and next to a vehicle radiator grinder. Arsenic concentrations found in the soil of the gardens ranged from 8.5 to 21 mg kg^{-1}, these values were higher than those reported in the parks of São Paulo (1.2 to 16 mg kg^{-1}) (Figueiredo et al., 2011). The highest content of As were observed in Bairro Jardim (Fig. 2), which is a neighborhood closer to the central region of the city. It is possible to observe in Fig. 2 that the pattern of distribution of this spatial element was similar to that observed for Cr and V, suggesting that these elements come from the same source.

Chromium values observed in the soils from our studied ranged from 24 to 89 mg kg^{-1} and were higher than those observed in the soils of urban parks in São Paulo (21 to 70 mg kg^{-1}). Regarding Zn, the values observed in Santo André vary from 30 ± 383 mg kg^{-1} and were higher than those observed by Figueiredo et al. (2011), which range from 15 to 179 mg kg^{-1}. Thus, it is possible to infer that despite the municipality of Santo André being less populous and with a much smaller vehicle fleet than a municipality in São Paulo, the concentration levels of As, Cr and Zn elevates in the soils of UG were higher than those reported in São Paulo soils in a region with high vehicular traffic and industrial sources of atmospheric pollution.

Comparing our results with those of other UG soils in the world it emerges that Cu, Cd values are higher than those observed in Melbourne (Laidlaw et al., 2018), Sheffield (Weber et al., 2019) and Toronto (Wiseman et al., 2013) (Table 2). However, Co, Ni, Pb, Sb and Zn values observed in soils from our study are below those reported in the international studies described in Table 2.

The levels of As (23.93–44.33 mg kg^{-1}) and Cr (86.67-160.67 mg kg^{-1}) observed in Sheffield, England, UK, were also higher than those observed in our results, probably due to this city has a long history of industrial activities and environmental pollution (Weber et al., 2019). Therefore, it is possible to conclude with these comparisons that there is a need to locally assess the levels of application of PTEs, because observations made for other regions cannot be generalized.

3.2 *Geoaccumulation index*

The geoaccumulation index of elements in the soil was calculated for those that have guiding values for soil (CETESB, 2016). However, the results presented in Fig. 3 correspond to the elements that presented minimally some points with I*geo* > 0, as following: As, Ba, Cd, Cr, Cu, Ni, Pb, Se and Zn. Considering the median obtained for each element, the soil of the studied gardens can be classified as: unpolluted to moderately polluted by Ba, Cd, Cr, Cu, Cd and Zn for almost all the studied gardens; with the exception of the vegetable garden in Capuava 1, which was moderately to severely polluted by Ba and moderately polluted by Cu and Pb. I*geo* indicated that all gardens presented moderate pollution by As and Se, except for the garden of Capuava 1, which presented moderate to severe pollution by Se.

3.3. Soil amendments results

The concentrations of PTEs in the SAs collected are presented in Table 3. Consistent with the results of collected soils, the median concentrations of elements detected in SAs ranked in the following order: Zn > Ba > Cu > V > Cr > Pb > Ni > Co > As > Mo > Se > Cd. SAs with the highest total PTEs concentration ranked in the following order: Spent mushroom substrate (SMS) > Local compost pile (Capuava 1) > Quail manure (Capuava 2) = Cow Manure = Meat bone meal > Chicken manure = Quail manure (Jd. Marajoara) > Castor cake = Lime > Local compost pile (Bela Vista).

Overall, the two SAs collected in Capuava 1 (SMS and local compost pile) were those with the highest total concentration of PTEs. SMS refers to the biomass waste generated from mushroom production (Hanafi et al., 2018) and it has been used as fertilizer (Grimm and Wösten, 2018). SMS composition varies according to geographical location and also according to mushroom species (Grimm and Wösten, 2018). Studies reporting Ba content in SMS are scarce. Kalembasa and Wiśniewska (2009) reported a lower content of Ba (52.5 mg kg^{-1}) than ours in Italian SMS. The PTEs content of SMS determined within this study is compared to levels reported in previous studies which it verifies that the Ba and Zn levels are elevated, while Cd and Cu is invariably lower that those reported in other countries (Kalembasa and Wiśniewska, 2009; Jordan et al., 2010; Medina et al., 2012).

Table 3

Mean concentration in mg kg^{-1} of the potentially toxic elements in the soil amendments collected in the urban gardens of Santo André.

Location	**Description of soil amendments**	**As**	**Ba**	**Cd**	**Co**	**Cr**	**Cu**	**Mo**	**Ni**	**Pb**	**Se**	**V**	**Zn**
		(mg kg^{-1})											
Capuava 1	Local compost pile	4.8 ± 0.1	257 ± 53	0.58 ± 0.04	2.9 ± 0.4	32.6 ± 0.9	68.7 ± 0.8	1.7 ± 0.3	9.5 ± 0.1	48.6 ± 0.3	0.97 ± 0.03	39 ± 0.1	261 ± 20
	Spent mushroom substrate	3.1 ± 0.2	4137 ± 150	0.055 ± 0.003	1.7 ± 0.2	37.3 ± 0.8	9.48 ± 0.04	0.546 ± 0.001	8.8 ± 0.2	4.4 ± 0.5	0.52 ± 0.02	16.2 ± 0.6	690 ± 11
Capuava 2	Quail manure	0.8 ± 0.1	63 ± 0.8	0.02 ± 3	1.44 ± 0.06	4.1 ± 0.5	303 ± 10	5.7 ± 0.5	2.9 ± 0.1	0.5 ± 0.03	0.7 ± 0.05	0.95 ± 0.04	297 ± 16
	Lime	2.15 ± 0.09	199 ± 0.2	0.028 ± 0.002	1.28 ± 0.03	8.5 ± 0.1	2.5 ± 0.01	0.077 ± 0.008	3.1 ± 0.1	3.93 ± 0.08	0.37 ± 0.02	5.33 ± 0.06	21 ± 0.2
Capuava 3	Cow manure	4.02 ± 0.07	101 ± 8	3.209 ± 0.004	3.34 ± 0.09	38.5 ± 0.3	102 ± 4	3.4 ± 0.03	29.5 ± 0.7	72.2 ± 0.5	0.88 ± 0.02	21.2 ± 0.9	295 ± 32
Jd Marajoara	Quail manure	1.17 ± 0.08	66 ± 2	0.05 ± 0.001	0.92 ± 0.04	4.1 ± 0.2	47 ± 2	3.74 ± 0.01	2.83 ± 0.11	3.6 ± 0.2	1.19 ± 0.07	6.0 ± 0.1	333 ± 0.1
	Chicken manure	1.04 ± 0.07	62 ± 3	0.051 ± 0.005	1.1 ± 0.2	3.45 ± 0.02	58 ± 1	4.44 ± 0.01	3.2 ± 0.2	3.16 ± 0.09	1.35 ± 0.01	4.9 ± 1.0	344 ± 0.5
B. Jardim	Castor cake	4.6 ± 0.5	46.2 ± 0.8	0.029 ± 0.002	1.26 ± 0.06	8.2 ± 1.3	29 ± 1	1.07 ± 0.11	84 ± 3	1.5 ± 0.1	0.82 ± 0.07	4.6 ± 0.5	102 ± 6
	Meat bone meal	31 ± 2	511 ± 5	0.038 ± 0.005	0.464 ± 0.002	4.2 ± 0.8	3.7 ± 0.2	0.261 ± 0.001	1.15 ± 0.01	4.5 ± 0.1	14.6 ± 0.4	1.7 ± 0.6	85 ± 2
Bela Vista	Local compost pile	1.23 ± 0.01	39 ± 2	0.099 ± 0.003	0.61 ± 0.06	4.3 ± 0.9	13 ± 1	0.43 ± 0.04	1.7 ± 0.1	6.63 ± 0.08	0.36 ± 0.001	11.6 ± 0.1	64 ± 3
	Median	1.23	68	0.05	1.25	5.2	29.9	1.2	3.05	4.0	0.85	5.4	263
	Range	0.48-32	37-4287	0.016–3.213	0.46–3.43	3.3–39	2.5–314	0.07–6.11	1.14-86	0.47-73	0.35-15	0.92-39	20–701
Cuban urban agriculture soil amendments[(a)]		nd-98.0	-	0.2–6.2	-	16.7-297.3	-	-	10.6-455.7	13.5-1,100	0.8–204	-	-
Spanish soil amendments[(b)]		0.50–7.7	24–270	< 0.5–0.51	0.50–3.5	5.5–57	60–244	0.63–3.6	3.2–53	2.7–30	-	-	160–700
Spanish urban waste composts[(c)]		-	-	nd-3.5	-	-	52–829	-	-	33–223	-	-	200–1149
Chinese soil amendments[(d)]		-	-	0.24–1.03	-	-	-	-	-	26.52-295.24	-	-	27.16-461.74

[(a)]Filter-cake compost, horse manure + crop-residue compost, horse manure + soil, rabbit-manure compost, cow-manure compost, cow manure + crop-residue compost, cow manure + soil, earthworm humus, crop-residue compost, crop residue + soil, municipal-solid-waste compost (Alfaro et al., 2017); [(b)]Sewage sludge, swine manure and organic fraction of solid municipal waste (Margenat et al., 2020); [(c)]Composts obtained from source separated organic fraction of municipal solid waste;, Compost obtained from source separated organic fraction of municipal solid waste mixed with green waste, Compost obtained from municipal garden trimmings mixed with sewage sludge, Manure vermicompost, Composted pine bark (Paradelo et al., 2020); (d)Lime, Rice straw, Pig manure, Sheep dung and peat (Han et al., 2013).

When comparing our findings with other studies of SAs (Table 3) a notable difference is seen regarding the levels of Ba and Mo in SAs samples, the levels of these elements are elevated in SAs of Santo André (Han et al., 2013; Alfaro et al., 2017; Margenat et al., 2020; Paradelo et al., 2020).

Brazilian legislation (Brazil, 2016 and 2019) maximum allowed values in SAs are 15, 10 and 200 mg kg^{-1} for As, Cd and Pb, respectively. The results in Table 3 demonstrates that in meet bone meal sample the As content (31 mg kg^{-1}) was above the Brazilian legislation.

Despite these results of PTEs levels, it is known that some organic compounds can decrease the bioavailability of PTEs in soils for plants through various mechanisms, such as precipitation, complexation, redox reactions, ion exchange and electrostatic interaction (Margenat et al., 2020;

Palansooriya et al., 2020). However, the misapplication of these compounds may be contributing to increase the supply of PTEs in the soil of UG in Santo André and also may pose risk for the gardeners' health.

We noticed a limited knowledge about the best practices for management and application of these amendments in the studied gardens and the gardeners may be unaware about the risks of misapplication. In fact, rates of application, incorporation practices into the soil, maturation status and storing procedures of these compounds were very distinguished across the gardens. In Brazil, the government allows some SAs to be used for crop nutrition, with specifications clearly defined by appropriate regulation (Brazil, 2016 and 2019). The partnership between governments, universities and representative institutions plays a strategic role in promoting risk awareness and knowledge about soil testing and fertilization.

3.4 *Source identification and relationships between PTEs*

To define clearly dominant or discrete sources of PTEs in UG soils is difficult, due to the complex temporal scale and spatial distribution of distinct inputs of PTEs. Data set was evaluated by multivariate statistics. The first factor represents 64.54% (Fig. 4.a) of the data variation and indicated a directly strong (> 0.7) proportional association of Ba, Cd, Co, Cu, Mo, Ni, Pb, Sb, Se and Zn in the 1st and 4th quadrants. Therefore, this suggests that these elements are from the same source, probably from Capuava Petrochemical complex particulate matter and from soil amendments. This assumption was reinforced by the PCA representation by cases (Fig. 4.b) where samples from Capuava UG were discriminated in the same quadrants (1st and 4th quadrants).

At same time, factor 2 discriminated samples by its contents of As, Cr and V and represents 18.53% (Fig. 4.a). Then, Marajoara, Bairro Jardim and Vila Bastos UG were placed in the 1st and 2nd quadrants due to the higher levels of these elements (Fig. 4.b). As mentioned before, Santo André is in a region of many automobiles and auto-parts industries, such as General Motors, Volkswagen, Mercedes-Benz, Scania and others. Thus, the region has a long history of foundries and metallurgical process companies which contributes to As, Cr and V (alloys elements) inputs in the environment (Lange et al., 2017; Freire et al., 2021).

The same associations observed in PCA were observed in the hierarchical dendrogram (Fig. 4.c) Two main groups were identified. The first group by Ba, Cd, Co, Cu, Mo, Ni, Pb, Sb, Se and Zn, which can be associated to the petrochemical complex. The remarkable association between Ba and Zn in this cluster can be attributed to the inputs caused by spent mushroom substrate (Table 3). The second cluster comprises As, Cr and V.

Table S3 presents the results of Pearson's correlations between the PTEs concentrations and the soil characterization parameters. Acidity is considered a key parameter in the mobility of elements in soils, however in this study pH did not significantly correlate with PTEs and this may be related to the small variation of the pH value between the samples. The elements Ba, Cd, Cu, Mo, Ni, Se, Sb and Zn correlated with OM. The elements Ba, Co, Cu, Ni, Se, Sb and Zn correlated significantly with CEC. Therefore, organic compounds commonly used by farmers added to soil may also result in an increase of CEC. The sand fraction correlated inversely with As content, and directly with Cd and Pb content, indicating that the latter two may be associated with a natural origin or, most probably from fine particulate matter. Arsenic correlated directly with clay contents, indicating that the areas that presented higher clay contents were able to accumulate this element the most.

3.5. Risk associated with the presence of PTEs in the soil

Table 4 shows the concentration of PTEs in the soil of UG in Santo André corresponding to the 95th percentile (Csoil), the toxicity values (RfD and FC), the chronic daily doses (DDC), the hazard quotients (HQ), hazard indices (HI), carcinogenic risks (CR), total carcinogenic risk probabilities (TCR) obtained for adults, that is, the risk to which urban gardeners are exposed.

For most PTEs, the results obtained for the non-carcinogenic risk quotient indicated that the route of exposure that contributed most to the total risk was the oral route, followed by dermal absorption and finally the route by inhalation of soil particles, except for Ba and Cd, for which the inhalation route contributed more than the dermal route. The oral route of exposure to PTEs in urban soils is the most common (Gabarrón et al. 2017).

The non-carcinogenic hazard index (HI) results obtained for all PTEs, considering adults, were less than 1.0, therefore, health risks are not expected to occur (USEPA, 1989). The order of HI values for the PTEs observed was As > Cr > Ba > Pb > Cu > Ni > Zn > Sb > Co > V > Cd. The total carcinogenic risk values obtained for all PTEs were below the acceptable range for regulatory purposes of 10^{-6} to 10^{-4} (USEPA, 2009) for adults. The decreasing order of TCR for PTEs observed was Cr > As > Ni > Pb > Co.

Table 4
Non-carcinogenic and carcinogenic risk indices associated with PTEs in the topsoil of the Santo André vegetable gardens.

	As	**Ba**	**Cd**	**Co**	**Cr**	**Cu**	**Ni**	**Pb**	**Sb**	**V**	**Zn**
RfD ing	3.00E-04	2.00E-02	1.00E + 00	2.00E-02	3.00E-03	4.00E-02	2.00E-02	3.50E-03	4.00E-04	5.00E + 00	3.00E-01
RfD dermal	3.00E-04	1.40E + 01	1.00E + 02	5.40E-03	6.00E-05	1.20E-02	1.60E-02	5.25E-04	4.00E-04	9.00E-02	6.00E-02
RfD inh	1.50E-05	1.40E-01	2.90E-03	2.00E-02	2.86E-05	4.00E-02	6.00E-06	3.50E-03	4.00E-04	2.00E-03	3.00E-01
FC ing	1.50E + 00				5.00E-01			8.50E-03			
FC dermal	1.50E + 00			8.40E-01	2.00E-01		9.80E + 00	8.50E-03			
FC inh	1.50E + 00			8.40E-01	4.10E-01		9.80E + 00	4.20E-02			
C_{SOIL} (95%)	16.7	638	0.8	4.2	81	140.0	17	76	0.28	159	328
DDC_{ing}	8.0E-06	3.0E-04	3.8E-07	2.0E-06	3.9E-05	6.7E-05	8.1E-06	3.6E-05	1.3E-07	7.6E-05	1.6E-04
DDC_{dermal}	3.2E-08	1.2E-06	1.5E-09	8.0E-09	1.5E-07	2.7E-07	3.2E-08	1.4E-07	5.3E-10	3.0E-07	6.2E-07
DDC_{inh}	1.2E-09	4.6E-08	5.8E-11	3.0E-10	5.9E-09	1.0E-08	1.2E-09	5.5E-09	2.0E-11	1.2E-08	2.4E-08
HQ_{ing}	2.66E-02	1.52E-02	3.82E-07	1.00E-04	1.29E-02	1.67E-03	4.06E-04	1.04E-02	3.34E-04	1.52E-05	5.22E-04
HQ_{dermal}	1.06E-04	8.68E-08	1.52E-11	1.48E-06	2.57E-03	2.22E-05	2.02E-06	2.76E-04	1.33E-06	3.37E-06	1.04E-05
HQ_{inh}	8.05E-05	3.29E-07	2.00E-08	1.52E-08	2.05E-04	2.53E-07	2.05E-04	1.57E-06	5.06E-08	5.75E-06	7.91E-08
HI	2.68E-02	1.52E-02	4.02E-07	1.02E-04	1.57E-02	1.69E-03	6.13E-04	1.06E-02	3.36E-04	2.43E-05	5.33E-04
CR_{ing}	1.20E-05				1.93E-05			3.08E-07			
CR_{dermal}	4.77E-08			6.72E-09	3.09E-08		3.17E-07	1.23E-09			
CR_{inh}	1.81E-09			2.55E-10	2.40E-09		1.21E-08	2.31E-10			
TCR	1.20E-05			6.98E-09	1.94E-05		3.29E-07	3.10E-07			

3.6. Microorganisms in bed soils and soil amendments

The microbial results revealed the identification of 6 bacterial genera (Table 5) and 8 isolates were unidentified. Around 30% of non-animal amendments samples were contaminated either with *Serratia* or *Klebsiella* genera, however no antimicrobial resistance was observed. Animal amendments that were positive for at least one microorganism accounted for 37.5% of this type of sample and four bacterial genera were isolated. Among them, resistance to TET and NIT was identified for *Citrobacter* and *Enterobacter* genera was resistant to CFO. Antimicrobial presence in animal manures was reported previously and may be related to the resistance observed in our findings (Bloem et al., 2017).

Fifty percent of the bed soil samples showed at least one bacterial genera. *Shigella* isolate demonstrated the broadest spectrum of antimicrobial resistance, being resistant to three antimicrobials (SXT, FOS and NAL). *Citrobacter* isolate was resistant to CFO, while *Klebsiella* isolate did not present any resistance.

Table 5
Identification of bacterial genera and antimicrobial susceptibility

Sample type	Bacterial isolates	Resistance
Non-animal amendment	*Serratia sp.*	None
	Klebsiella sp.	None
Animal amendment	*Citrobacter sp.*	TET, NIT
	Escherichia sp.	None
	Enterobacter sp.	CFO
	Shigella sp.	None
Bed soil	*Shigella sp.*	SXT, FOS, NAL
	Citrobacter sp.	CFO
	Klebsiella sp.	None
TET: tetracycline; NIT: nitrofurantoin; CFO: chloramphenicol; SXT: trimethoprim sulfamethoxazole; FOS: Fosfomycin; NAL: nalidixic acid; CFO: cefoxitin.		

None means sensibility for all antimicrobials tested in this study.

4. Conclusion

Our findings showed an enrichment of some PTEs in the soils of some gardens, but at concentration levels below the agricultural intervention values established by the local environmental protection agency, except for Ba. The multivariate analysis of the data revealed that some PTEs are important discriminators of urban pollution inputs and may provide valuable information for mitigation strategies. Results also revealed that the organic matter directly influences the geochemistry of PTEs in the soils of these gardens. Some organic compounds used in these gardens showed high levels of As, Ba, Pb and Zn. The risk assessment revealed that gardeners are not subject to health damage from exposure to PTEs. However, pathogenic, and resistant microorganisms were identified in soil beds and amendments. Despite of the hostile environmental situation of the studied region, the results herein showed an optimistic scenario in the soils of urban gardens of Santo André. Future studies to investigate PTEs in the food produced in these areas are strongly recommended. The studies about the atmospheric deposition of these elements can also contribute to the mass balance and source identification.

Declarations

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Conflicts of interests

The authors have no conflicts of interest to declare that are relevant to the content of this article.

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Figures

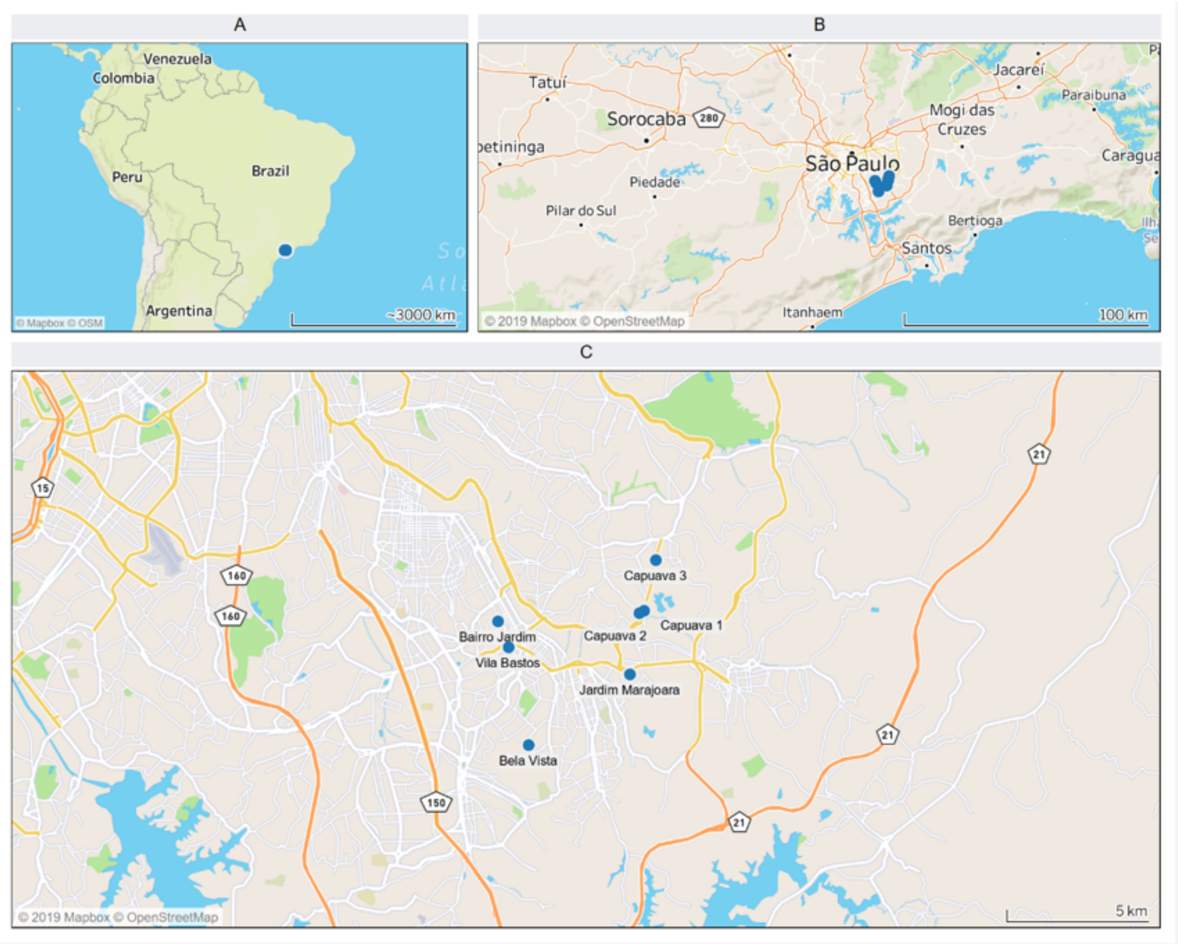

Figure 1

Locations of the selected urban gardens in the city of Santo André.

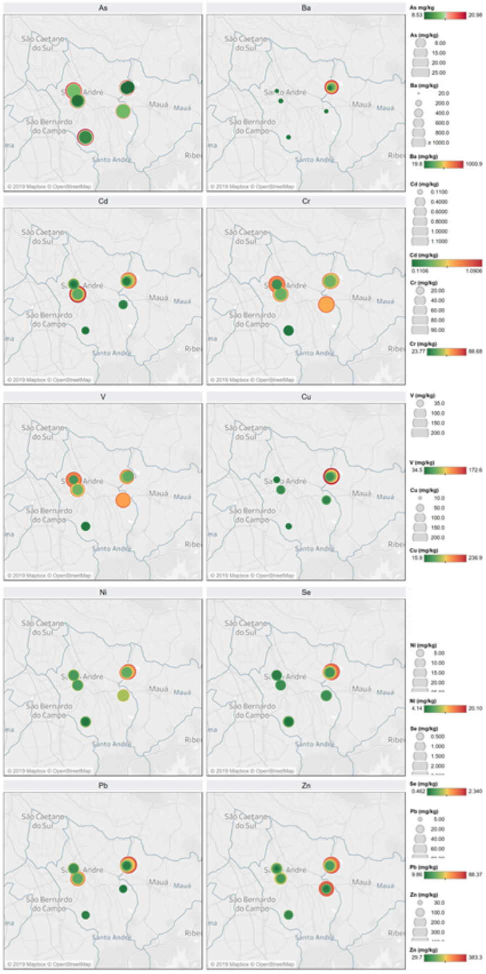

Figure 2

Spatial distribution of potentially toxic elements in soils of urban gardens from the city of Santo André.

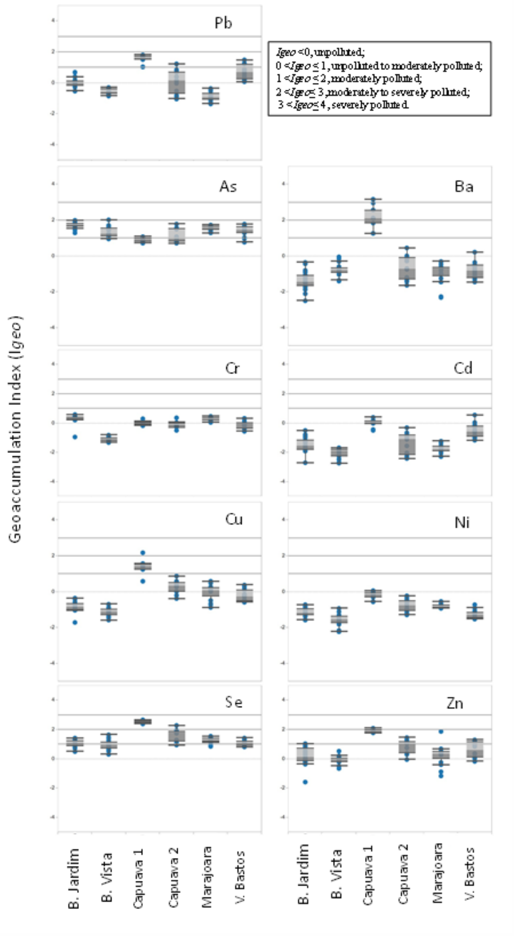

Figure 3

Boxplot of the results of geoaccumulation index of Pb, As, Ba, Cr, Cd, Cu, Ni, Se and Zn in soil samples (0-20 cm) from UG, in relation to the Quality Reference values of the State of São Paulo (QRV) (CETESB, 2016).

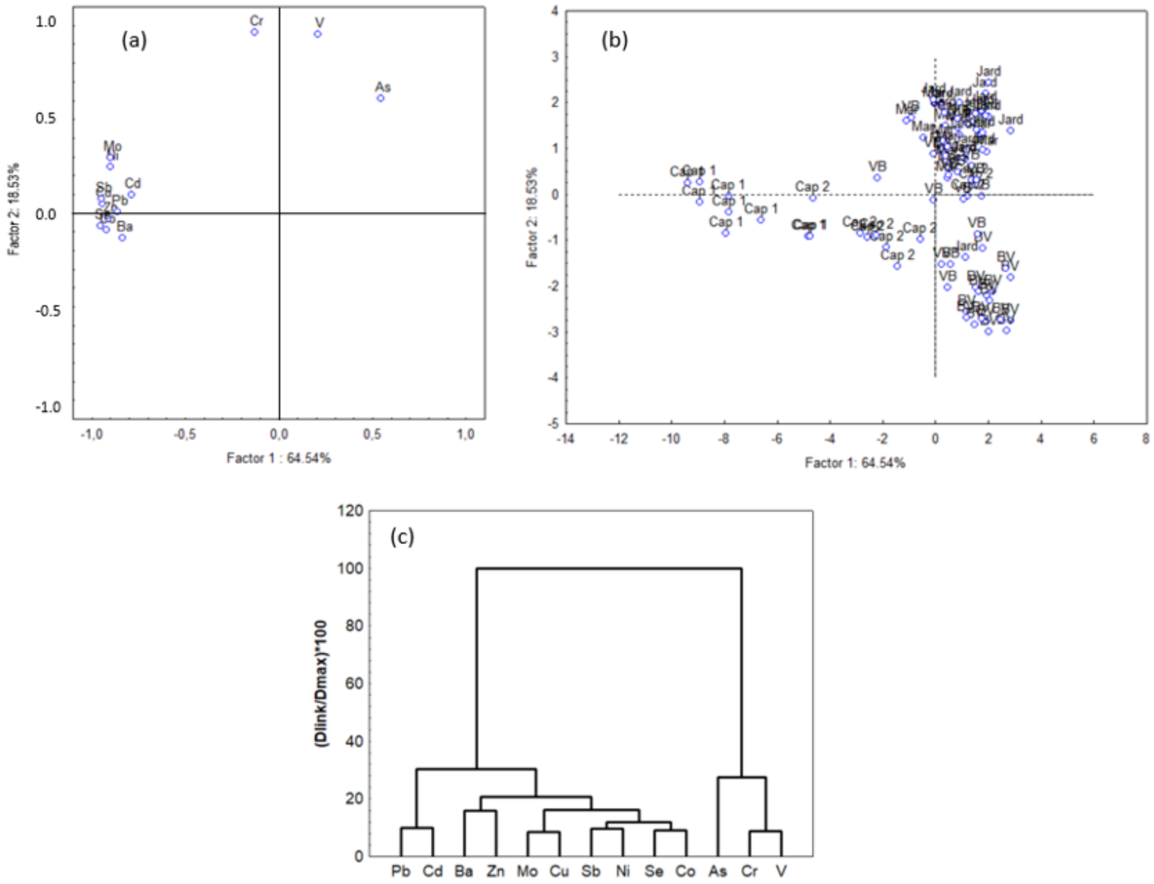

Figure 4

Principal component analysis with (a) Factor 1 versus Factor 2 and (b) Factor 1 versus Factor 2 loading plots by gardens. (c) Dendrogram obtained by cluster analysis of the concentrations of elements determined in the soil of urban gardens in Santo André.

Supplementary Files

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