ANTIMONY ACCUMULATION RISK IN LETTUCE GROWN IN BRAZILIAN URBAN GARDENS

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Abstract

More than 80% of the Brazilian population inhabits urban areas. Diffused poverty and the lack of fresh vegetables have generated malnutrition and unbalanced diets. Thus, the interest in growing food locally, in urban allotments and community gardens, has increased. However, urban agriculture may present some risks caused by the urban pollution. Road traffic is considered the biggest source of heavy metals in urban areas. Hence, the objective of the study was the assessment of the accumulation of heavy metals in an urban garden in the city of Recife, at different distances from a road with high traffic burden. The results showed that the distance from the street decreased the accumulation of many potentially toxic elements. Furthermore, the human health risk was estimated, revealing that greater danger was associated with the accumulation of antimony. Concentration of other elements in the leaf tissues were within previously reported thresholds.

Keywords: *heavy metals, pollutants, urban horticulture, road traffic*

Introduction

Urban agriculture in Brazil

In 1872, the Empire of Brazil conducted its first population census, recording 800'000 inhabitants in the city of Sao Paulo (Théry et al., 2013). Since then, the population of the city has grown up to the current 11 million people, while, at national level, about 150 million (more than 80%) out of the 190 million citizens today inhabit urban areas (IBGE, 2010). The transition toward the current urban Brazilian society has led to the emergence of diffused urban poverty, mainly as a result of exclusion, inequalities and violence (Skeldon, 2012). When people move from rural to urban environments, they are less likely to produce or even being familiar with food being grown. This reflects into changes in the diet with introduction of foods that are generally less fresh and transported in long distances. Consistently, growing concerns about the quality and cost of food, and food insecurity have increased interest in growing food locally in cities including in community or allotment gardens (Corrigan, 2011; Evers, 2011). Community gardens are growing in popularity and they provide a number of other benefits

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beyond food production, including community building, education, and promoting health (Turner, 2011). Consistently, urban agriculture is being promoted by public administrations all over the world, with specific national programmes also found in Brazilian cities (Winkler Prins, 2002; Branco and De Alcantara, 2012). Urban agriculture experiences in Brazil have been addressed in a number of recent reports (e.g. Villas-Boas, 2006; Orsini et al., 2009; Fecondini et al., 2010). In a study conducted in 2000 in the city of Belém (Madaleno, 2000), urban farmers were mainly women (70%), normally unemployed and/or retired (35%), and in most cases belonging to the less wealthy classes, immigrated from the countryside or other Brazilian states (50%). The garden had primary function in the production of food for household subsistence, with cultivation surfaces between 50 and 500 m^2, and land used often (20%) illegally occupied, although for long terms (generally more than 10 years, but often more than 20) (Madaleno, 2000).

Environmental risks and heavy metal contamination in urban garden

Besides its role in emancipation and improvement of diet's healthiness, plant cultivation within cities may present environmental risks associated to both air and soil pollution (Alloway, 2004). In urban environment air pollutants generally have anthropogenic origins, due primarily to vehicular emissions and fossil fuels burning (Jean-Soro et al., 2015). With the growth of urban population and the rapid industrialization, urban air pollution has increased rapidly. Consequently, the contamination risks of urban-grown food is elevated, leading to a dietary exposure to trace elements that can cause human health risk (Massaquoi et al., 2015). In cities vehicles emissions are considered one of the main sources of contaminants (Salvagio Manta et al., 2002; Duong and Lee, 2011). The relationships between traffic-related parameters and heavy metals traces in the edible tissues of several horticultural crops cultivated in the inner city of Berlin, Germany, were investigated in Säumel et al. (2012). Results showed as high traffic burden near the planting site (<10 m) resulted in 67% of crops having lead (Pb) values which exceeded the standards of the European Union, whereas only 38% of the crops grown at the distance of more than 10 m from the nearest road exceeded these values. Low overall traffic burden corresponded with low Zn content in fruits and low Cr and Ni content in stem and root vegetables (Gherardi et al., 2009). Similar results were reported in Vittori Antisari et al. (2015), which found that in urban garden of Bologna, Italy, the amount of Cr, Ni, Sn, Zn in leaves tissues of horticultural crops cultivated nearby the road (10 m) is higher compared with the ones located far from it (60 m). Concurrently, lettuce and basil total metals accumulations did not differ between urban and rural gardens (Vittori Antisari et al., 2015). Analogous relationships between heavy metals accumulation and cultivation sites distance from the roads were described in other different studies (Nabulo et al., 2006; Sharma and Prasad, 2010; Naser et al., 2012). Urban soils can also be contaminated as they are often located on old urban sites, impacted by human activities, such as industrial activities, road traffic, waste dumps and demolition sites (Jean-Soro et al., 2015). Potentially toxic elements (PTE) can be assimilated by urban-grown products and become a danger for human health. Roadside soils are the major reservoirs of traffic-related heavy metals (Chen et al., 2010). Many studies reported that heavy metal content in roadside soil decreases exponentially with increment of roadside distance (Akbar et al., 2006; Nabulo et al., 2006).

The aim of this study is to compare heavy metals accumulation in lettuce grown in urban gardens at different distances from the roads in the city of Recife in north east of Brazil. Furthermore, in order to evaluate urban-grown products safety, Health Risk Index (HRI) for different PTE was performed.

Materials and methods

Sites descriptions and experimental design

An experiment was conducted between October 2013 and January 2014 in Recife, a Brazilian city in the state of Piauì (Fig. 1). The experiment was located in an allotment garden (coordinates 8° 04' 1'' to 8° 04' 10'' S, and 34° 56' 31'' to 34° 56' 41'' W) nearby one of the main roads of the city, BR 232 (average daily volume of 33'000 vehicles) (http://www.dnit.gov.br) and consisted of two treatments, namely:

- $Road_{0-20}$: gardens located within 20 m from the road.
- $Road_{20-50}$: gardens located between 20 to 50 m from the road.

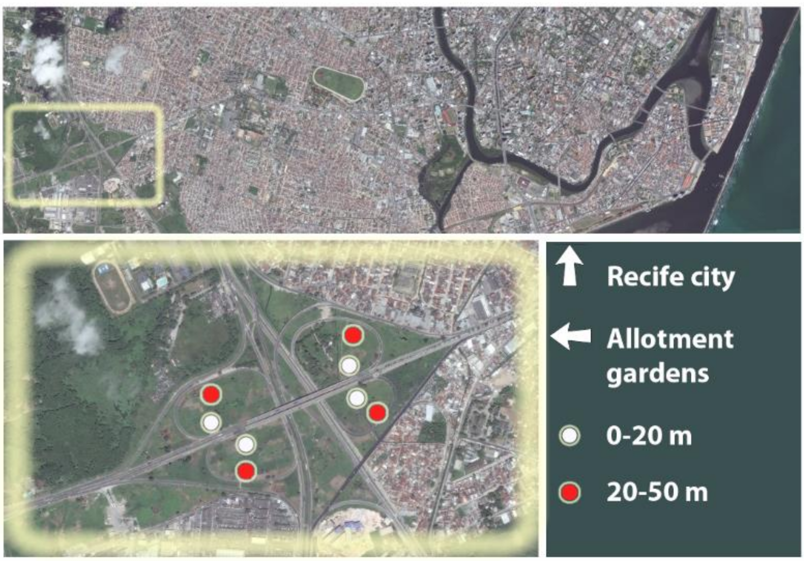

Figure1 *Recife aerial photo, with in white and red tag the location of urban garden.*

During the experimental period, six samples of lettuce (*Lactuca sativa* L.) were collected from each location at commercial maturity of the plant.

The soils of the Recife sites are classified as Fluvic Gleysols (IUSS, 2014) with ACg profile. Climate is classified as humid-moist monsoon transition humidity

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regime (Papadakis, 1970, 1975). The Gleysols occur on wide range of unconsolidated fluvial and lacustrine materials in depression areas and low landscape positions with shallow groundwater (Table 1).

Table 1. *Physico-chemical values average, taken from the literature, relating to Epipedon (0-40 cm) soil of survey sites.*

Soil classification (IUSS, 2014)			**Fluvic Gleysols** [(1)(2)]
Texture class [(3)]			clay loam
pH (H_2O)			4.90±0.14
TOC		g kg^{-1}	12.1±1.3
TN		g kg^{-1}	1.00±0.42
CEC (Cation Exchange Capacity)		meq^+/100g	5.9±3.1
Exchangeable bases	Ca	meq^+/100g	0.80±0.14
	Mg	meq^+/100g	0.55±0.49
	K	meq^+/100g	0.13±0.11
	Na	meq^+/100g	0.20±0.14
Acidity	Al	meq^+/100g	1.30±0,28
	H	meq^+/100g	2.95±0.92
BS (Base saturation)		%	28.5±8.2
Total oxides	SiO_2	%	27.6±1.1
	Al_2O_3	%	20.6±2.5
	Fe_2O_3	%	2.10±0.71
	TiO_2	%	0.55±0.07
	MnO	%	0,09±0.01
	P_2O_5	%	0.09±0.04

Edited: [(1)] Serviço National des Pesquisas Agronômicas (1962); [(2)] Falesi, 1964; [(3)] Schoeneberger et al. (2012)

Ions analysis

Ions analysis were conducted as described in Vittori Antisari *et al.* (2015). Briefly, samples were dried in ventilated oven (T=60 °C) and finely ground with a ball-mill. Approximately 0.25 g of leaf dried biomass, weighted in Teflon bombs, was dissolved in 8 ml of H_3NO_3 (suprapure, Merck, Roma, Italy) + 2 ml of H_2O_2 (Carlo Erba, Milano, Italia) using a microwave oven (Milestone 2100, Sorisone, Bergamo, Italy). After cooling, solutions were made up to 20 ml with Milli-Q water and then filtered with Whatmann 42 filter paper. The accuracy of the instrumental method and analytical procedures used was checked by triplication of the samples, as well as by using reference material, which was run after every 10 samples to check for drift in the sensitivity. The analytical quality of the results was checked against the following reference materials, which certify values of the studied elements close to the measured ones: CRM 060 (aquatic plants) and CRM 062 (Olive leaves) provided by the European Commission Institute for Reference Materials and Measurements.

The macro and micro elements were determined by inductive coupled plasma optical emission spectrometry (ICP-OES, Spectro Ametek, Arcos). The ICP-OES setting followed multi-standard solutions (CPI International, Amsterdam) that reproduce the matrix effect present in samples and allow the lowering of detection limits (DLs). ICP-OES calibrations were performed by the standard solution of Bureau of Collection Recovery (BCR-909) and some internal standard were used (AMS-MO1 and AMS-ML1) (Vittori Antisari et al., 2014).

Health Risk Index

In order to better address the comprehension of the potential health risks associated with the consumption of urban-grown vegetables, the application of Daily Metal Intake and Health Risk Index was performed.

Health Risk Index is related to daily estimated consumption of contaminated food. The US Environmental Protection Agency's reference doses (US-EPA IRIS, 2006) were used as reference points. The Daily Metal Intake (DMI) was estimated using the eq. [1]:

$$DMI = \frac{C_{shoot} * 0.085 * DPC}{BW} \quad [1]$$

where C_{shoot} is the concentration of metals in the edible part of the shoot of the plant (as mg kg^{-1} dry weight), multiplied by a conversion factor of 0.085 to convert dry weight vegetable metal content to fresh weight, according to Rattan *et al*. (2005). Daily plant consumption (DPC) were estimated as 0.51 kg day^{-1} and 0.30 kg day^{-1} for adults and children (Santos *et al.*, 2004) and body weight (BW) were estimated based on Leclerq *et al.* (2009). Based on the Daily Metal Intake, it was possible to calculate the Health Risk Index, based on eq. [2]:

$$HRI = \frac{DMI}{RfD} \quad [2]$$

where RfD is the Reference Dose. RfD for the studied elements are respectively $3x10^{-4}$ mg kg^{-1} BW day^{-1} (As); $2x10^{-1}$ mg kg^{-1} BW day^{-1} (B); $2x10^{-1}$ mg kg^{-1} BW day^{-1} (Ba); 1.5 mg kg^{-1} BW day^{-1} (Cr); $4x10^{-2}$ mg kg^{-1} BW day^{-1} (Cu); $1.4x10^{-1}$ mg kg^{-1} BW day^{-1} (Mn); $5x10^{-3}$ mg kg^{-1} BW day^{-1} (Mo); $4x10^{-3}$ mg kg^{-1} BW day^{-1} (Pb); $4x10^{-4}$ mg kg^{-1} BW day^{-1} (Sb); $3x10^{-1}$ mg kg^{-1} BW day^{-1} (Zn) (US-EPA IRIS, 2006; Jan *et al.*, 2010). HRI values >1 are considered to pose health risks (Cui *et al.*, 2004; Rattan *et al.*, 2005).

Statistical analysis

Statistical analyses were executed with One-way ANOVA using SPSS v. 20 (IBM Corporation, Armonk, USA).

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Results

Elements accumulation as function of roadside distance

Plants grown within 20 m from the highway accumulated more Al (+47%), B (+52%), Ba (+41%), Cr (+229%), Cu (+11%), Mn (+10%), Sb (+14%) (Table 2) as compared to concentrations found in lettuces grown between 20 and 50 m from the road. Accumulations of Ag and As were below detection limit in samples cultivated between 20 and 50 m from the road, while in plants grown within 20 m were respectively 0.11 and 0.37 mg kg^{-1} DW. Contrariwise, accumulation levels of P, Se, Sn were higher on samples situated between 20 and 50 m from the road. No significant differences were associated with Ca, Fe, K, Li, Mg, Pb, S, Sr, Ti and Zn accumulations (Table 2).

Table 2. *Macroelements (g kg^{-1} DW) and microelements (mg kg^{-1} DW) in lettuce at different distances from road (0-20 m and 20-50 m). Significance with One-way ANOVA at $P \leq 0.05$ (*), $P \leq 0.005$ (**), $P \leq 0.01$ (***) and $P > 0.05$ (ns). <DL = values are below detection limits.*

Macroelements (g kg^{-1} DW)				**Microelements** (mg kg^{-1} DW)			
	0-20 m	**20-50 m**			**0-20 m**	**20-50 m**	
Al	0.13	0.09	*	**Ag**	0.11	< DL	*
B	0.03	0.02	*	**As**	0.37	< DL	*
Ba	0.01	0.01	***	**Cd**	< DL	< DL	
Ca	8.60	8.23	ns	**Cr**	2.27	0.69	*
Fe	0.17	0.16	ns	**Cu**	11.62	10.45	*
K	5.04	4.83	ns	**Li**	1.82	1.83	ns
Mg	3.85	4.12	ns	**Mo**	< DL	< DL	
Mn	0.02	0.02	*	**Ni**	< DL	< DL	
Na	5.64	7.60	ns	**Pb**	2.42	2.09	ns
P	3.49	3.68	*	**Sb**	0.54	0.47	*
S	2.69	2.84	ns	**Se**	< DL	0.86	*
Sr	0.07	0.06	ns	**Sn**	0.18	0.27	**
Ti	0.00	0.00	ns	**Zn**	65.01	58.78	ns

Health risk associated with consumption of urban-grown lettuce

Health Risk Index associated with consumption of lettuce by adults was below the safety limit for all considered elements. HRI was significantly higher in plants cultivated nearby the highway as compared to plants grown farther away, respectively for B (0.107 vs 0.070), Ba (0.038 vs 0.027), Cr (0.0009 vs 0.0003), Cu (0.180 vs 0.162), Mn (0.086 vs 0.078), Sb (0.829 vs 0.728). HRI for Pb and Zn were not affected by the distance from the road, reaching values of 0.349 and 0.128 (as a mean of the two distances), respectively (Fig. 2A). Only antimony (Sb) HRI for children exceeded the value of 1, that is considered the limit of danger for the consumption (Khan et al., 2008), reaching the value of 1.31 and 1.15 respectively on plants grown near or far from the road (Fig. 2B).

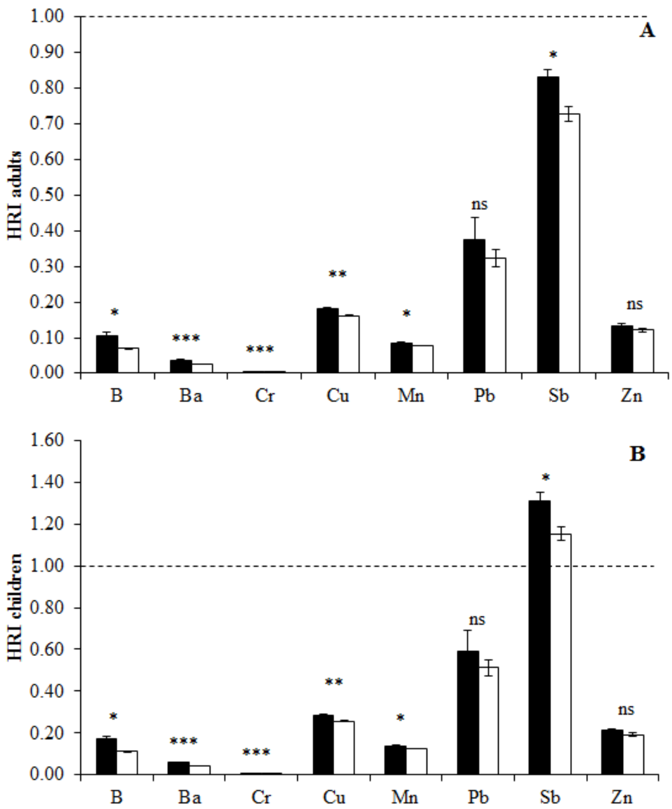

Figure 2
Health Risk Index for lettuce in Recife referred to adults (A) and children (B) at different distances from road (black bars = 0-20 m; white bars = 20-50 m).

Significance with One-way ANOVA at P≤0.05 (*), P≤0.005 (**), P≤0.01 (***) and P>0.05 (ns).

Discussion

Comparison between Recife urban allotments and worldwide distributed urban gardens

Plants collected in Recife were generally safer than other urban-grown leafy vegetables described in literature. Indeed, lettuce grown between 0 and 20 m from the highway BR-232 in urban gardens of Recife resulted in lower amount of Zn (-25%), Cu (-11%), Ni (not detected vs 0.58 mg kg^{-1} DW) and Cd (not detected vs 0.17 mg kg^{-1}) compared to leafy vegetables of urban gardens of Berlin (Säumel et al., 2012). On the other hand, Cr accumulation nearby the road was more than 2 times higher in Recife than in Berlin between 0 and 20 m from the road, while similar amount of Cr were detectable in Recife ($Road_{20-50}$) and Berlin (Säumel et al., 2012). The accumulation of Pb was also similar in the urban gardens of the two cities. Guerra et al. (2012), when analysing samples of lettuce cultivated in urban gardens of São Paulo (Brazil), found higher amount of Pb (+131%) and Cr (+245%) as compared to the results obtained in the present study. Consistently, lettuce grown in Nigerian urban gardens (Agbenin et al., 2009) was reported to present a 2-fold higher amount of Cu, 3-fold higher amount of Zn, and a 12-fold

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and 14-fold higher amount of Pb and Cr, respectively. Leafy vegetables grown in urban gardens of New York (McBride et al., 2014) resulted in lower content of Al (-73%) and 4-fold higher content of Ba as compared to lettuce grown in Recife. According to the Brazilian National Agency for Sanitary Vigilance (ANVISA, 2013) the accumulation limits in tissues of leafy vegetables are 0.30 mg kg^{-1} FW for Pb and As (about 3.5 mg kg^{-1} DW) and 2.5 mg kg^{-1} FW for Sn (about 29.4 mg kg^{-1} DW). All samples of lettuce did not exceed these limits for Pb, As, Sn. Even if there are not Brazilian limits for Cr content, the accumulation of Cr in the presented study (respectively 2.27 mg kg^{-1} DW in samples grown close to the road and 0.69 mg kg^{-1} DW in samples grown far from the road) exceeded the safety limit (0.5 mg kg^{-1} DW) reported by Chinese National Food Sanitation Standards (Li et al., 2006).

Influence of distance from road on PTE accumulation

Lead (Pb) found in the samples from the present study did not show differences between the two distances, even if many studies reported the correlation between distance from road and accumulation of Pb in vegetable tissues (Nabulo et al., 2006; Naser et al., 2012; Säumel et al., 2012). However, Rodriguez et al. (1982) fixed the limit for the risk of Pb accumulation at 33 m from the road. Contrarily, Nabulo et al. (2006) found higher content of Pb in lettuce cultivated at 1 m from the road but no differences between 5 and 10 m, explaining the absence of significant differences hereby described.

Chromium (Cr) accumulation in plants grown more than 20 m from the road was significantly lower than the plants grown nearby the highway, while Zn accumulation was not affected by the distance. These results confirm the findings of Säumel et al. (2012) which described a significant correlation between distances and Cr content in leafy vegetables and a non-significant correlation between Zn and distance from road. Barium (Ba) accumulation significantly decreased with distance from road. Consistently, Ba content in urban vegetable crops is traffic related, deriving from lubricating oil additives and fuel, as reported in Monaci and Bargagli (1997).

Antimony (Sb) is a rarely studied element, even if the associated health risk is elevated due to its carcinogenic effects (Gebel et al., 1997). The accumulation levels founded in this study for lettuce were significantly affected by the distance from the road, reaching 0.54 mg kg^{-1} DW ($Road_{0-20}$) and 0.47 mg kg^{-1} DW ($Road_{20-50}$). This value is 9-fold and 8-fold higher than the mean content of terrestrial plants, equal to 0.06 mg kg^{-1} DW (Kabata-Pendias, 2010). On the other hand, Sb content in historical mining areas in leafy vegetables could be higher, reaching up to 2.2 mg kg^{-1} DW in endive and between 0.31 and 1 mg kg^{-1} DW in corn salad (Wolfram et al., 2000). Other sources in urban environment may be light bulb production, pigment industries, burning of fire retardants in fuel (He et al., 2012). Furthermore, vehicular traffic is one of the main sources of antimony in the highly populated urban environments (Gomez et al., 2005), causing by the abrasion dust of the vehicles brakes (von Uexküll et al., 2005; Iijima et al., 2008).

Health risk associated with urban grown vegetables in Recife

Health risk index is highly affected by the Daily Plant Consumption (DPC). In this study it was decided to use the total consumption of vegetables and derived products (respectively 0.51 kg day^{-1} for adults and 0.30 kg day^{-1} for children) assuming that all the vegetables derived from the urban garden, describing the worst scenario. Higher values are evaluated for Sb and Pb (Fig. 2), which resulted below the safety limit of 1 (Khan et al., 2008).

In samples of lettuce cultivated in the reclaimed soil of the Pearl River Estuary (China) Li et al. (2012) found dramatically lower values of HRI for Pb (40-fold), for Zn (26-fold) and for Cu (20-fold). These differences could be explained considering that Li et al. (2012) used a Daily Plant Consumption of 32.4 g day^{-1} referred only at lettuce consumption. Contrarily, Cui et al. (2004) reported HRI values in vegetables grown in an area near a smelter in Nanning (China) ranging between 1.45 and 13.5 for Pb and between 0.25 and 0.32 for Zn, dramatically higher compared to the values found in this study. Considering the HRI associated to children only antimony exceeded the limit.

Conclusion

Pernambuco, Brazilian and world population is highly concentrated in urban areas, thus, diffusion of urban agriculture is increasing in most cities as a way to promote food security, income generation and a number of ecosystem services. The risk of cultivation in urban area deriving from vehicular traffic was here addressed. Potential toxic elements content in lettuce grown nearby the road in Recife resulted always lower than in other urban areas, with the exception of Chromium. In this study accumulation in plant tissue of Chromium, Barium, Antimony and Copper turned out to be strongly correlated with traffic. Highest Health Risk Index was associated with Antimony. The emission of this element should be more studied and solutions for the reduction of antimony pollution should be investigated.

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RISQUE DE L'ACCUMULATION D'ANTIMOINE DANS LA LAITUE CULTIVÉE DANS LES JARDINS URBAINS BRÉSILIENS

Resumé

Plus de 80% de la population brésilienne habite dans les zones urbaines. La pauvreté diffuse et le manque de légumes frais ont généré la malnutrition et une alimentation déséquilibrée. Ainsi, l'intérêt pour la culture alimentaire locale, dans des affectations urbaines et les jardins communautaires, est augmentée. Cependant, l'agriculture urbaine peut présenter certains risques liés à la pollution de la ville. Le trafic routier est considéré comme la principale source de métaux lourds dans les zones urbaines. Par conséquent, l'objectif de l'étude a été d'évaluer l'accumulation de métaux lourds dans un jardin urbain dans la ville de Recife, à différentes distances d'une route très fréquentée. Les résultats ont montré que la distance de la route affecte l'accumulation de nombreux éléments potentiellement toxiques; ceux-ci diminuent avec la distance. De plus, le risque pour la santé a été estimé et relevé que le principal problème est lié à l'accumulation d'antimoine. La concentration des autres éléments dans les tissus foliaires classe les légumes comme etant plus sains que ceux des autres jardins urbains, dans la littérature examinée.

Mots-clés: *métaux lourds, polluants, horticulture urbaine, trafic routier.*

RISCHIO DA ACCUMULO DI ANTIMONIO IN LATTUGA COLTIVATA IN ORTI URBANI NEL NORD-EST DEL BRASILE

Riassunto

Piú dell'80% della popolazione brasiliana vive in aree urbane. La povertà diffusa e la scarsa disponibilità di ortaggi freschi nelle grandi città sono tra le cause dell'aumento della malnutrizione e della diffusione di diete sbilanciate. Negli ultimi decenni si pertanto visto un aumentato interesse verso la coltivazione locale di ortaggi e verdura in orti urbani e comunitari. Tuttavia, l'agricoltura urbana può presentare alcuni rischi legati all'inquinamento di suoli e atmosfera, spesso associati all'intenso traffico veicolare. L'obiettivo del presente studio è stato, quindi, la valutazione dell'accumulo di metalli pesanti in un orto urbano nella cittá di Recife a diverse distanze da una strada altamente trafficata. I risultati hanno indicato che la distanza dalla strada diminuiva l'accumulo di molti elementi potenzialmente tossici. Inoltre, è stato stimato il rischio per la salute umana, rivelando quale elemento di maggiore rischio l'accumulo di antimonio. La concentrazione nei tessuti fogliari degli altri elementi classifica gli ortaggi come sicuri e sani, anche in comparazione ai risultati di precedenti studi in letteratura.

Parole chiave: *metalli pesanti, inquinanti, orticoltura urbana, traffico veicolare*

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