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SUITMA 8: SOILS AND SEDIMENTS IN URBAN AND MINING AREAS

An exploratory study of potential As and Pb contamination by atmospheric deposition in two urban vegetable gardens in Rome, Italy

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Abstract
Purpose A preliminary study was carried out in Rome (Italy) to assess the potential role of atmospheric deposition in trace element contamination in urban vegetable gardens relative to human health risk from crop consumption.
Materials and methods Two sites were selected on the basis of previously known contamination issues. Atmospheric deposition, parent material, soils properties affecting trace element mobility, and various anthropogenic inputs were considered. Soil samples were taken at depth from two points in each garden, within 5 cm of sampled crops. Inputs and crops were sampled and analysed for As and Pb content. A rain and dust gauge was set up in each garden for the duration of 93 days (late spring to late summer) for atmospheric deposition sampling.
Results and discussion Atmospheric deposition influx was high at both sites (2.22 and 2.32 As and 2.67 and 3.42 Pb $\mu g\ m^{-3}\ day^{-1}$). Soil pH was between 6.70 and 7.57 and texture varied from loamy sand to clay loam (3.4 to 31.9 % clay content). CEC ranged between 21.6 and 54.2 meq/100 g within rooting depth, rising almost commensurately with soil organic carbon (SOC) content (1.87–8.37 %). Somewhat high total soil Pb content (80.8–522.7 ppm) contrasted with negligible exchangeability and crop content (<0.01 ppm). Total soil As (17.0–32.0 ppm) corresponded with exchangeable and crop As for one site in one of the gardens. Leaves evinced high As accumulation levels (16.0–41.2 ppm) in all crops. High amounts of atmospheric Pb combined with negligible amounts of extractable Pb and Pb plant tissue content point to particulate inhalation and ingestion as a main health threat in the case of Pb. In contrast, food intake seems to be of greater concern relative to high As contamination. Greater soil As solubility may be explanatory, but the mostly low amounts of extractable As and the high atmospheric As suggest an airborne route being playing an important role.
Conclusions Preliminary results suggest that research on trace element contamination in urban gardens should consider atmospheric deposition as a major contributing source.

Keywords Arsenic · Atmospheric deposition · Lead · Trace element contamination · Urban gardens

1 Introduction

Over the last two decades, there has been increasing interest in and diffusion of urban community vegetable gardens in Rome (Italy), partly spurred by public sensitivity to environmental and food quality issues (Attili 2013; Pinto et al. 2010). However, trace element contamination problems in cities and associated rising concerns may ultimately stymie what could be viable urban cropping systems (Brown et al. 2015; Meuser 2010; Säumel et al. 2012). Improvements in site-specific understanding of contamination processes can help identify potential health risks and develop preventive techniques that, if explicated and diffused effectively, can contribute to raising urban food productivity while minimising deleterious exposure, at least via crop consumption.

Numerous studies have focused on identifying trace element contamination sources (e.g. Hursthouse et al. 2004; Pouyat et al. 2007; Douay et al. 2008; Bourennane et al. 2010; Meuser 2010; Jean-Soro et al. 2015) but contributions

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from atmospheric deposition have tended to be insufficiently (if at all) considered or to be studied through soil, rather than also through dust content. This is especially in the case of urban vegetable gardens, even if foliar uptake has been shown to be a contaminant pathway (Salim et al. 1992; Bondada et al. 2004; Schreck et al. 2012). Hence, the project focused on the relative role of atmospheric deposition in trace element contamination in urban vegetable gardens. Aside from atmospheric deposition, known major variables influencing trace element content and fluxes were considered, namely parent material, CEC, clay content, soil organic carbon (SOC), and pH (e.g. Impellitteri et al. 2001), as well as direct anthropogenic inputs (e.g. De Miguel et al. 1998)—in this case, irrigation water, imported soil, and straw cover.

Urban vegetable gardens were chosen on the basis of characteristics facilitating the detection of atmospheric deposition effects. As and Pb were selected so as to address local concerns, to include trace element contaminants more common to Rome, and to capture the pH-dependent range of trace element solubility.

2 Field site characteristics

The extent and major sources of soil and crop As and Pb were studied for the Forte Prenestino (FP) and Orto Insorto (OI) urban vegetable gardens of Rome (Italy). Both sites are underlain by sediment derived from Pb-enriched grey pozzolanic ash (GsF 2012; Ventriglia 2002). No agrochemicals are used and irrigation and on-site crop processing rely on local piped water.

FP (41°53′29.7″N, 12°34′10.2″E; 47 m asl) is located in an elevated section of a former military fortress built and used mainly during the late 1800s. The fortress was converted into a public park by the late 1970s. The garden, a fenced area of roughly 200 m^2, was established over the last decade and is surrounded in part by trees and part of the fortress walls and hence relatively sheltered. It is divided into raised beds made from excavated material from adjacent soil. Cropping areas are permanently covered by imported straw and managed according to synergistic principles (Hazelip 2014). These entail mulching, crop residue recycling, and avoidance of soil reworking (e.g. digging, tillage) and inputs (e.g. fertiliser, insecticide) following garden establishment.

OI (41°52′19.2″N, 12°32′38.9″E; 37 m asl) is a fenced, ca. 2-ha area adjacent to a main road where gardening has been established since 2011. The area sits on a buried waste dump forming a discontinuity at 25–55 cm depth. OI is divided into open-air meeting spaces, grassy areas, fruit trees, directly cropped areas, and several plastic containers filled with imported “organic” soil (certified as devoid of contaminants or agrochemical treatments). OI is within 100 m from a construction materials plant specialising in industrial-grade paints, thermo-hydraulics, and building materials recovery. The cropping areas are located more than 50 m from the plant, but within 30 m of the adjacent road. The area was sampled and analysed for Pb in 2011, yielding values of 552–3170 ppm for total surface soil Pb (0–10 cm) in five points more than 10 m apart from each other. Crop parts per million Pb levels were found to be 0.01 for courgette, 0.03 for tomato, 0.09 for potato, and 0.47 for salad.

3 Methods

3.1 Sampling strategy and procedure

Research was conducted between 17 May and 21 August 2014. Funding and time constraints permitted only purposive sampling, which is aimed at future hypothesis refinement and testing via probabilistic sampling (Malherbe 2002). A dry foam Frisbee-type deposit dust gauge was set up in each garden prior to soil and crop sampling, following standard siting and collection protocols (Vallack 1995). Passive atmospheric deposition sampling spanned 93 days (the project’s duration). Field analyses were completed following US Natural Resource Conservations Service procedures (Schoeneberger et al. 2012). Subsequently (13–19 June), reference, fixed-interval soil samples were taken to parent material depths (at most 55–60 cm) within a 5-cm radius from crops, which were sampled on the same days for both edible and inedible parts. Two locations per garden were identified to suit above-mentioned criteria. Distances between each pair of sampling locations were no more than 5 m, and all sampling locations were within 10 m of an atmospheric deposition gauge.

Only cultivated locations were included in the study because of a focus on human health risk from crop consumption. FP01 and FP02 are raised beds that were planted respectively with *Solanum lycopersicum* (tomatoes) and *Brassica oleracea* L. var. *palmifolia* (palm cabbage). OI01 and OI02 were both planted with *Phaseolus vulgaris* (string beans). The former was directly cropped and the latter consists of imported soil in a plastic container. Crops were selected on the basis of their healthy status when sampled, widespread cultivation, trace element uptake characteristics, and their maturing stage (coincident with fieldwork). In this manner, pathogenic effects could be minimised, results could be useful towards future probabilistic sampling, and differentials between leafy and fruiting vegetables could be included. When sampled, string beans and tomatoes had grown for ca. 60 days, while palm cabbage had reached its third year of growth.

For comparative purposes as well as contaminant provenance analysis, parent material was analysed to discern background levels and geogenic influence (Pouyat et al. 2007). Previous studies were also used. The OI area selected for the present study was found to have 890 ppm total Pb according prior laboratory analysis. This is well within the 200–

1400 ppm range found for high traffic zones in Rome (Ajmone-Barsan and Biasioli 2010; Angelone et al. 1995; Calace et al. 2012; Salzano et al. 2008). To the author's knowledge, no such information on As levels is available for the sites investigated or for Rome generally. However, a report to the Istituto Superiore di Sanità and the Provincial Government of Rome (Marsili 2014) cites moderate to high deposition mean yearly values of As (0.25–0.33 μg m^{-3} day^{-1}) and Pb (5.4–11 μg m^{-3} day^{-1}) for two sites within 10 km of FP and OI.

3.2 Analytical methods

Clay content was determined using a hydrometer method (ASTM 1985). Soil pH was derived through a 2:1 water to sample mixture, using a PCE 228-R pH meter. CEC was determined using $BaCl_2$ and read by Perkin Elmer ICP Plasma 400 Spectrometer (DM 1999). Total As and Pb analyses of atmospheric deposition, irrigation water, and soil samples were carried out according to US EPA method 3051A (US EPA 2007), using a Milestone MLS Mega 1200 microwave mineraliser and Perkin Elmer ICP Optima 7000. These analyses were performed at the labs of the Department of Chemical, Materials and Environmental Engineering (DCMEE) of the University of Rome "La Sapienza". Unused soil sample portions were posted from the DCMEE to the Pisa Institute of Ecosystem Study of the National Research Council (IES), where total soil carbon and SOC were analysed (the latter via the Walkley-Black method). $CaCl_2$-extractable As and Pb were also determined, using ICP AES (ICP OES Varian Liberty model) and Milestone ETHOS900 microwave mineraliser, following US EPA method 6010C (US EPA 2000). Crop samples, prepared at DCMEE (single rinse with deionised water), were taken within 5 days to IES for drying and As and Pb content determination via acid digestion and ICP-AES analysis (Campbell and Plank 1998).

4 Results

All crops exhibited high As contamination, except at OI02 (imported soil), and negligible Pb content (Table 1). At the FP garden, straw cover was highly contaminated by Pb. Irrigation water pH was 7.56 and 7.35 at FP and OI, respectively, with <0.003 ppm As and Pb for both water sources. Atmospheric deposition (μg m^{-3} day^{-1} over 93 days), amounted to 2.22 As and 2.67 Pb at FP and 2.32 As and 3.42 Pb at OI. At OI02, the imported soil (Table 2) had increasing total As and Pb with depth, but little in their extractable form. Parent material was reached at 40–55 cm in both garden areas, except at OI02. At FP, there were slightly higher values of Pb with depth, but not with As, which remained relatively constant. OI01 and OI02 exhibited lower and higher values with depth, respectively. Except for OI2 (clay loam), all soils were characterised as sandy texture (mostly loamy sand). Clay content oscillated from 3.4 to 31.9 %, but was largely low. Values for pH ranged towards or above neutral, from 6.70 to 7.57. Soil organic carbon (SOC) range was variable and highest at OI02. Soil inorganic carbon (SIC) was mostly low and highest at OI01. While total soil As and Pb values were high at all sites, extractable Pb was very low and extractable As was appreciable. Soil cation exchange capacity (CEC) values had a 21.6–54.2 meq/100 g range within rooting depth and corresponded roughly to variations in SOC content (1.87–8.37 %).

Table 1 Total plant tissue As and Pb (ppm) from selected crops at Forte Prenestino (FP) and Orto Insorto (OI) urban vegetable garden sites in Rome (Italy)

Site	Crop	Sample	As	Pb
FP1	*Solanum lycopersicum*	Fruit	13.70	<0.01
		Leaves	35.80	<0.01
	Variable	Straw	<0.04	27.00
FP2	*Brassica oleracea* L. var. *palmifolia*	Leaves	16.30	<0.01
		Stem	<0.04	<0.01
	Variable	Straw	<0.04	30.10
OI01	*Phaseolus vulgaris*	Fruit	19.6	<0.01
		Leaves	41.2	<0.01
OI02	*Phaseolus vulgaris*	Fruit	<0.04	<0.01
		Leaves	16.00	<0.01

5 Discussion

Total Pb values coincide with the current soil surface level at OI. This suggests recent anthropogenic, rather than geogenic, Pb as a main source. At FP, there may be a combination of geogenic and anthropogenic factors to consider. The lack of marked changes in soil Pb levels with depth may reflect the outcome of transferring and mixing soil from nearby sources to build the raised beds. Relatively Pb-rich pozzolanic ash, for example, may have been dug out and included in the materials used to build the raised beds when the urban garden was first established.

Finding negligible amounts of extractable Pb and Pb plant tissue content at all four sites is to be expected, given relatively high pH compared to Pb solubility thresholds. Yet the relatively high amount of atmospheric Pb also raises issues of sources other than in situ soil particle entrainment. Additionally, for the FP site, the provenance of the straw used to enhance crop growth merits further study. The straw may be trapping airborne Pb, but since the As content is very low, it may be more likely that additional Pb is being imported into the site through the straw itself.

Table 2 Soil and parent material analytical results from Forte Prenestino (FP) and Orto Insorto (OI) urban vegetable garden sites in Rome (Italy)

Site	Depth cm	Texture (USDA)	pH	Clay %	SOC	SIC	CEC meq	Total As ppm	Total Pb	Extractable As	Extractable Pb
FP01	0–10	Loamy sand	6.70	13.70	3.75	0.08	39.34	27.38	136.38	3.64	<0.01
	20–30	Loamy sand	7.38	12.30	1.12	0.01	24.59	26.12	128.50	<0.04	<0.01
	45–55	Loamy sand	7.48	9.60	0.76	0.01	21.01	29.84	144.98	3.52	<0.01
FP02	0–15	Sandy loam	7.17	15.80	1.42	0.01	27.08	28.88	127.86	<0.04	<0.01
	20–30	Loamy sand	7.09	10.60	1.24	0.01	23.18	29.52	131.77	<0.04	<0.01
	40–50	Loamy sand	7.49	14.90	0.90	0.08	23.55	28.61	131.49	<0.04	<0.01
OI01	0–15	Loamy sand	7.43	4.30	1.87	0.65	21.59	23.2	523.00	7.79	<0.01
	20–30	Loamy sand	7.57	3.40	1.74	0.55	21.21	22.6	243.00	<0.04	<0.01
	40–50	Loamy sand	7.22	6.50	0.57	0.05	12.27	17.0	85.10	4.17	<0.01
OI02	0–10	Clay loam	6.91	29.60	8.37	0.11	54.23	26.90	80.80	<0.04	<0.01
	20–30	Clay loam	7.05	31.90	3.66	0.07	43.49	32.00	94.00	<0.04	<0.01

SOC soil organic carbon, *SIC* soil inorganic carbon, *CEC* cation exchange capacity

Total As values had a pattern similar to that of total Pb in that higher levels occurred towards the current or original soil surface. This indicates an unlikely geogenic effect at either garden sites and, in the case of OI, a relatively low influence of the buried landfill. As mobility, in contrast, is of greater concern, especially relative to food intake (see also, e.g. Nathanail et al. 2004; Jean-Soro et al. 2015). In part, the pH may explain some of the greater solubility encountered, but there may be other processes involved, including the effects of root exudates that cannot be estimated through the lab techniques used. Additionally, as McBride (2013) reports, pH may not be a determining factor as much as sheer soil trace element load. However, the *P. vulgaris* grown on imported soil was less contaminated by As than the one cultivated directly on contaminated soil. There may therefore also be plant tissue As enrichment resulting from atmospheric sources, which were much higher than expected at both sites compared to recent monitoring data (Marsili 2014). The importance of atmospheric deposition is reinforced by the finding that (1) at OI As levels rise as one reaches the original current soil surface and (2) at FP there were marked contrasts in extractability and crop content in spite of similar clay and SOC %, CEC, and pH values. Furthermore, the *B. oleracea* L. var. *palmifolia* site is sheltered by tree canopy, unlike the other locations, and has grown continuously for 3 years. This implies a dilution effect for what is otherwise a known bio-accumulator (Gall and Rajakaruna 2013). Differential exposure to atmospheric deposition may therefore be more explanatory with respect to As contamination.

6 Conclusions

Given the constraints of this study, no inferences can be made beyond the immediate context investigated. However, some broader implications can be gleaned that form the basis for future work based on probabilistic sampling. It appears that, in the case of Pb, the main health threat is from the insoluble fraction, via particulate inhalation and ingestion. Crop contamination appears to be of greater concern with respect to As and this seems likely due to a combination of pH effects and atmospheric deposition (see also Brown et al. 2015; Jean-Soro et al. 2015). Irrespective of the above findings, the high atmospheric and total soil As and Pb noted at both gardens should be of concern.

These are among the issues that can form the basis for a future statistically viable sampling strategy that, on the basis of results so far, may shed more light on urban vegetable garden contamination processes. However, other variables should also be included in such analyses (e.g. Fe oxy-hydroxides, dissolved organic carbon) to account for other possible influential factors of trace element mobility. Finally, atmospheric deposition sampling over the course of an entire year (or over multiple years) and multiple areas will also be necessary so as to encompass seasonal and spatial variability, alongside analyses of dust particles lodged within crop tissues, downslope runoff and through-flow from nearby contaminant sources, as applicable.

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