



Trace metal contamination and bioaccessibility in two Ulster County Urban Community Gardens, New York State (USA)

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Abstract

Purpose In this short communication, soil trace element mobility factors are compared to the bioconcentration factor (BCF; plant tissue to soil total concentration) and evaluated for their effectiveness in estimating contamination pathways between soil and cultivated vegetables. These mobility factors are bioaccessibility (BAF, the ratio of exchangeable to total soil trace metal concentrations) and translocation (TF, the ratio of leaf to root trace metal total concentrations).

Methods Kale was grown in two urban gardens in Ulster County, New York. Soil pH tests were carried out alongside analyses by ICP-OES on soil samples for total and exchangeable trace element concentrations and on kale roots and leaves for total metal concentrations (29 paired soil-kale samples), namely Al, Ba, Cd, Cr, Co, Cu, Pb, Sr, and Zn.

Results Total soil Cd, Pb, and Zn were above permissible levels in weakly alkaline soil, and kale was contaminated with Ba, Cu, and Sr. It was found that BAF explains kale contamination by Ba, Cu, Sr, and Zn more than the other indicators.

Conclusions The exchangeable fraction should be included in assessing trace metal contamination. This comparison of mobility indicators is a novel way of examining contamination problems in urban food production and improves the understanding of how evaluating soil dynamics helps identify ways to develop health-protective ecologically sustainable practices in cities.

Keywords Soil contamination · Soil trace element bioaccessibility · Vegetable contamination · Urban soil · Urban vegetable gardens

1 Introduction

Soils in cities provide many functions enhancing ecological sustainability and human health, such as in flooding attenuation, carbon sequestration, pollutant storage or neutralisation, and green space development (Cannava et al. 2014; Scharenbroch et al. 2018). Urban soils have also become increasingly important because of a burgeoning interest in food production within cities. Urban food production contributes to sustainability through many social, public health, and environmental benefits (Eigenbrod and Gruda 2015; Martin et al. 2016; Nogeire-McRae et al. 2018; WinklerPrins

2017). However, a role for urban soils as pollutant sink presents challenges to urban food production, alongside widespread airborne and waterborne pollutants (Bridges 1989; Hooda 2010; Jean-Soro et al. 2015; Malone 2022). Metals like Cd and Pb are among the most common contaminants that raise public health concerns (Lupolt et al. 2021; Paltseva et al. 2018).

Conventional soil testing services for the wider public provide total soil element concentrations, but such information addresses exposure potential by particle ingestion or inhalation rather than food consumption. Thus, it furnishes limited information on contaminant transfer to vegetables or bioaccessibility (Menefee and Hettiarachchi 2018). Soil trace element bioaccessibility is defined as the potential amount of trace elements accessible to an organism for absorption (Ehlers and Luthy 2003). Research results reported herein focus on metal concentrations in edible plant parts and exchangeable fraction of mainly soil trace metals. These are components to estimating public health repercussions of urban food production and consumption. However, the exchangeable

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fraction tends to be omitted in studies concerned with bioaccessibility assessments. The purpose of this short communication is to compare soil-vegetable trace element transfer indicators and evaluate their relative effectiveness in assessing bioaccessibility, especially in situations where *in vitro* studies are not feasible.

Plants derive nourishment mainly by means of absorbing soil water containing dissolved nutrients. This is also the main soil-borne metal contamination pathway (Cai et al. 2016). Values of total metal concentrations in soils and plant biomass are often used to estimate plant uptake, referred to as the bioconcentration factor, or BCF (Bourennane et al. 2010; Boim et al. 2016; Hooda 2010). The BCF was accordingly calculated as:

$$\text{BCF} = \frac{[\text{TE}] \text{ leaves}}{[\text{TE}] \text{ soil, total}} \quad (1)$$

This assumes that there is a direct relationship between total concentrations in plants and plant-available amounts in soil. Yet, plant-available metals are more associated with elemental analyses determined after extraction with, for example, CaCl_2 , MgCl_2 , EDTA, and DTPA, among other methods. This is because the extent to which plants are contaminated depends on combinations of element characteristics and soil conditions that facilitate solubility and thereby plant root uptake. Consequently, total concentrations in contaminated soil do not represent the amounts occurring in a form available for plant root uptake (Hooda 2010).

Metal in elemental form can be found in soil solution and solid phases. Metals adsorbed onto charged colloid surfaces can also be released back into soil solution and exchanged with other dissolved ions. The portion that can be thus exchanged is the exchangeable fraction. The degree of exchangeability depends on a variety of conditions (e.g., extent of element saturation, clay mineralogy, microbial activity, redox reactions). Most metals become more soluble at pH values below neutral. Since soil pH is the most influential variable, the analysis of soil characteristics was limited to pH (Tack 2010).

Aside from direct vegetable tissue analysis, calculating metal bioaccessibility is a more effective estimation of soil-borne vegetable contamination compared to analyses of total TE concentration (Hooda 2010). Bioaccessibility was measured as exchangeable fractions predicted by CaCl_2 extraction relative to total concentrations. It is herein referred to as bioaccessibility factor (BAF), calculated as:

$$\text{BAF} = \frac{[\text{TE}] \text{ exchangeable}}{[\text{TE}] \text{ total}} \quad (2)$$

Moreover, root-absorbed trace metals often accumulate within root tissues without reaching the rest of the plant. Hence, another way to assess soil-derived contamination in edible portions is to calculate the translocation factor

(TF), the ratio of leaf to root TE concentrations (Coakley et al. 2019; Islam et al. 2020):

$$\text{TF} = \frac{[\text{TE}] \text{ leaves}}{[\text{TE}] \text{ roots}} \quad (3)$$

To interpret results on total soil concentrations, the maximum allowable limits recommended by the New York State Department of Environmental Conservation were applied (NYS DEC 2006). Values for unrestricted land use (corresponding to ecological resource protection criteria) were chosen because of the high potential for exposure to trace metal contamination when gardening in urban areas. There are few to no guidelines regarding vegetable-specific trace element concentration screening for the purpose of human health, even as studies have been numerous (Lupolt et al. 2021; Rai et al. 2019). Hence, the FAO/WHO's Codex Alimentarius (FAO, WHO 2019) and the European Union's regulatory framework (European Commission 2008) were used for contamination standards.

2 Materials & methods

2.1 Project locations and sampling procedures

Research, including field and laboratory portions, was carried out during April-August 2022 in two urban community gardens in Ulster County, New York; GWE (latitude: 41.926257°; longitude: -74.015778°; 145 m asl) and MF (latitude: 41.732709°; longitude: -74.089035°; 75 m asl). The regional climate is classified as Dfb (humid continental) according to the Köppen system. During the project period, daily temperatures ranged from a minimum of -1.4 °C in April 2022 to a maximum of 38.2 °C in August, with an average of 21.3 °C. Total rainfall was 328.43 mm and no precipitation occurred in 114 of 153 days of the project period. The maximum amount and most of daily rainfall was in April (70.61 mm maximum; 101.86 mm total).

GWE, established in 2016, is used by local residents and school children and was part of the school parking lot in Kingston. MF is on the southern edge of a college campus in New Paltz. It was established in 2015 on a human-made knoll comprised mainly of discarded building materials at the edge of a college campus. The area is also used as a holding site for landscaping material and campus construction debris. Prior to the piling of construction debris, the area was forested. At both sites, the vegetable growing medium is composed of transported mixed mineral and composted materials introduced in 2020 at GWE and 2015 at MF. Maximum depths do not exceed 30 cm, below which there is plastic liner underlain by unconsolidated asphalt and construction debris (GWE) or construction debris characterised by

predominantly coarse clasts of roughly 5–15 cm in diameter (MF). Field analysis did not indicate soil profile development (IUSS Working Group WRB 2015).

For the study, *Brassica oleracea* var. *acephala* (Winterbor Kale)—a known metal bioaccumulator widely grown in this area—was selected (Chollet and Brock 2008; Ngugi et al. 2022). Seeding was done in a greenhouse, according to local gardener practices, and the vegetables were transferred to beds after four weeks of growth. The kale was sampled along with adjacent soil on 26 and 28 July 2022 at MF and GWE, respectively. While wearing lab gloves, kale leaves and roots were cut with stainless-steel scissors and stainless-steel trowels were used to collect soil samples. One to five leaves were collected from each specimen, depending on availability of whole, undamaged leaves per individual plant. Soil samples were collected at 0–15 cm depth within 5 cm around each plant basal stem and just below the roots. Vegetable specimens were placed in paper bags to avoid degradation and soil samples were transferred into standard plastic soil sampling bags. To prevent cross-contamination, instruments were cleaned after each use with 10% phosphate-free detergent solution and deionised water. They were then dried with new paper towels (Jones 2017; Malone 2022; Paltseva et al. 2018). Nine paired kale-soil samples were sampled at GWE and 20 at MF for a total of 29 samples each for leaves, roots, and soil.

2.2 Sample preparation and analyses

Laboratory procedures followed Jones (2017) for soil and vegetable specimen preparation and US EPA (2000) for analysis. Roots were separated from stems and leaves and washed in water followed by 20 mM EDTA in 200 mL polyethylene bottles to remove trace metals attached to leaf and root surfaces (Jean-Soro et al. 2015). Kale leaves and roots were then daubed dry with lab-grade absorbent wipes and oven dried (24 h at 80 °C). The specimens were placed in a muffle furnace and ashed for 36 h at 500 °C to ignite the organic carbon.

Soil samples were oven-dried at 50 °C and ground by mortar and pestle. pH measurements were recorded for each soil sample in a 2:1 by mass deionised water to soil slurry. Soil samples were each homogenised and subsampled. Subsamples were oven-dried for 24 h at 50 °C. To determine total and exchangeable fractions, two extraction procedures were performed on each sample: a strong acid digestion and a CaCl_2 extraction. For the former, 10 mL of 50% HNO_3 was added to each sample, which was then agitated at room temperature for 10 min before heating at 70 °C for 15 min. Next 5 mL of concentrated HNO_3 was added and samples were returned to the 70 °C water bath for 30 min. Three mL of 30% H_2O_2 was then added, followed by heating at 70 °C for 20 min. Finally, 10 mL of HCl was added and the samples were shaken at room temperature for at least one hour before centrifuging (2000×g) for 10 min.

To determine exchangeable trace metals in soils, CaCl_2 was used as extraction reagent because it has been shown to be indicative of the root-available fraction (Jones 2017). A CaCl_2 concentration of 0.01 M was chosen for its similar ionic strength to many soils' average salt concentrations. Such a solution also extracts trace metals at the pH of the soil since the solution is not buffered. A 0.01 M CaCl_2 solution was added to soil subsamples in a 10:1 ratio by mass and mixed by an orbital vortexer followed by overnight mechanical shaking. The mixture was centrifuged to clear the supernatant for analysis to determine the trace metal fraction extractable by 0.01 M CaCl_2 .

Ashed kale roots and leaves were solvated in 3:1:6 concentrated hydrochloric acid, concentrated nitric acid, to deionized water (v/v/v). The solution was centrifuged to clear the supernatant for analysis. Two method blanks were prepared for each extraction technique (Jones 2017).

Nine elements were examined for soil total, CaCl_2 extractable, and kale leaf and root concentrations by ICP-OES analysis. They include Al, Ba, Cd, Cr, Co, Cu, Pb, Sr, and Zn. Total soil extractions were subsampled and diluted 1:10 to determine total Al concentrations which exceeded the extracts' 25-ppm standard. Wavelengths for each element were selected from EPA 6010c method (US EPA 2000). Nine external standards, including a blank, were prepared from a multielement standard including Al, Ba, Cd, Cu, Sr, and Zn and from oven dried primary standard salts including Cr, Co, and Pb, ranging from 0.1 to 25 ppm. Standard regression curves were recorded three times during analysis of the three extractions with R^2 values > 0.985 for each regression. The 7.5 ppm standard was analysed every 16 samples to detect signal drift.

3 Results and discussion

3.1 Soil pH and trace metals in soil and kale

Average soil pH was 7.56, ranging between 7.43 and 7.84 (standard deviation: 0.10). The pH 7.60 average at GWE was slightly higher than at MF (pH 7.54). Weakly alkaline soil pH values suggest low trace metal solubility and thereby a tendency for low bioaccessibility.

Of the elements analysed, Al, Ba, Cu, Sr, and Zn occurred in both soil and kale (Table 1). The rest were detected only as part of total soil concentrations. Total soil concentrations for Cd, Cr, Pb, and Zn were above maximum allowable levels. Al concentrations are within safety levels and were also expected to be relatively high, due to the element's typical abundance in soil (e.g., a part of clay minerals). The wide range in Al values could not be explored in this study. Exchangeable fractions were detected only for Al, Sr, and Ba. Sr had the highest

Table 1 Tracematerial in Soil and Kale Roots and Leaves (29 Paired Samples, mg/kg dry Samples)

	Soil Total		Soil Exchangeable		SSL ^a	Roots		Leaves		VSL ^b
	mean	SD	mean	SD		mean	SD	mean	SD	
Al	7289	860	7.1	0.5	10,000	90.1	89.9	9.8	7.0	N.A
Ba	80.6	8.1	0.6	0.6	350	5.6	4.6	4.3	4.6	N.A
Cd	3.7	1.0	0.0	0.0	2.5	0.0	0.0	0.0	0.0	0.2
Cr	9.6	2.3	0.0	0.0	1–30 ^c	0.0	0.0	0.0	0.0	N.A
Co	1.3	0.8	0.0	0.0	20	0.0	0.0	0.0	0.0	N.A
Cu	38.4	5.0	0.0	0.0	50	3.5	3.9	3.0	1.1	0.2
Pb	96.8	12.3	0.0	0.0	63	0.0	0.0	0.0	0.0	0.3
Sr	69.4	33.2	12.0	1.5	N.A	36.8	6.3	48.2	19.6	N.A
Zn	126.4	12.9	0.0	0.0	109	30.9	9.8	41.3	9.0	N.A

^a NYS DEC Soil Screening Levels

^b EU and FAO/WHO Vegetable Screening Levels

^c For toxic hexavalent Cr, the standard is 1, whereas it is 30 for trivalent Cr. In this study, it was not possible to differentiate the two species

exchangeability, followed by Al and Ba. This suggests root absorption potential for those elements.

Results for Zn concentrations in leaves diverged from the 12–22 ppm Zn range found in a recent field experiment with *Brassica oleracea* L. cv GM Dari (Zafar et al. 2020). Cu values were within the lower range of that same study but at levels considered unsafe for daily consumption. Since no Cu or Zn were detected in the exchangeable fraction, root and leaf contamination for those elements is traceable to root-and leaf-tissue particle lodging and foliar absorption (including through local particle redeposition) respectively. In the roots' case, high surface area may have reduced the effectiveness of scrubbing and washing off particles. Another factor is root trace metal sequestration contributing to long-term accumulation; if this is occurring, we would expect divergence from shorter-term values implied in analysing the exchangeable fraction (Tack 2010; Paltseva et al. 2018).

Cd, Cr, Co, and Pb were not detected in roots, indicating that these elements were not bioaccessible, as expected relative to soil pH values (McBride 2013). This is consonant with the low root absorption of Cd, Cr, and Pb reported by Zafar et al. (2020) and from other findings of Cd absorption (0.1–0.7 ppm in kale leaves) under acid conditions (Ngugi et al. 2022). For Ba and Sr, contamination involves multiple pathways, namely root uptake, particle adherence to or lodging in roots and leaves, soluble element transfer from roots to leaves, and foliar absorption. In the case of Al, which is insoluble at the pH range found, the processes involved would be root- and leaf-tissue lodging and foliar absorption.

There is little correspondence between the unsafe levels found for total soil metal concentrations (Cd, Cr, Pb, and Zn) and metal content detected in kale leaves (Al, Ba, Cu, Sr, Zn). Hence, with respect to growing kale, even restrictive

soil screening standards such as those of the NYS DEC are only valid for Zn and cannot account for contamination by Al, Ba, Cu, and Sr.

3.2 Contamination indicators

The BCF values showed accumulation potential in kale leaves for Al, Ba, Cu, Sr, and Zn, while for the BAF this was limited to Al, Ba, and Sr (Table 2). For Cu and Zn, the BAF is inapplicable owing to a lack of detected labile forms, but the BCF values suggest accumulation in leaves through local airborne sources (e.g., splash, particles scattered by cultivation, etc.). Very low BCF and BAF values for Al correspond to the typically low mobility in alkaline conditions for that element. Due to the high concentrations of Al in soil and the tendency for some soil particles to remain adhered to produce even after washing (Paltseva et al. 2018), even these low BCF and BAF values may be overestimated.

TF values point to translocation from roots to leaves for Al, Cu, Sr, and Zn. In the case of Cu, there was detection in one specimen's leaves but not its roots and there was no Cu detected in another specimen. Generally, there is a consistent pattern of TF values aligning with those of BCF and BAF, save for Cu and Zn. However, aerial deposition and foliar absorption effects may present confounding factors such that TF figures may exaggerate the amount of trace metal translocation.

On the other hand, BAF and BCF values for Al, Ba, and Sr suggest that a portion of leaf contamination is explainable by the soil exchangeable fraction (i.e., the root-absorbable dissolved forms of those trace metals). Due to non-normal data distributions and homoscedasticity in both factors, Spearman rank correlation and Wilcoxon signed-rank tests were carried out to assess, for each element, the relationship between the

Table 2 Contamination Indicators for Elements Showing Nonzero Results (29 Paired Soil-Kale Samples)

	BCF ^a		BAF ^b		TF ^c	
	mean	SD	mean	SD	mean	SD
Al	0.001	0.001	0.001	0.000	0.169	0.173
Ba	0.053	0.058	0.007	0.007	1.887	3.604
Cu	0.079	0.030	N.A	N.A	2.636 ^d	3.494
Sr	0.877	0.531	0.217	0.101	1.394	0.739
Zn	0.330	0.079	N.A	N.A	1.463	0.559

^a Bioconcentration Factor (ratio of leaf to total soil trace element concentrations)^b Bioaccessibility Factor (ratio of exchangeable to total soil trace element concentrations)^c Translocation Factor (ratio of leaf to root trace element concentrations)^d Due to reversed TF ratios in two specimens, the effective number of samples is 27

BAF, as independent variable, and the BCF, as dependent variable. A significant correlation was found for Sr ($r_s=0.762$; $n=29$; $p<0.001$). Wilcoxon signed rank tests showed that the BAF affects the BCF for Ba ($Z=-4.487$; $p<0.001$) as well as for Sr ($Z=-4.703$; $p<0.001$). These results suggest that, for mildly basic soils, CaCl_2 extraction can be an effective indicator of Sr and, perhaps to a lesser degree (owing to poor BAF-BCF correlation), of Ba bioaccessibility.

4 Conclusions

Findings showed that total soil Cd, Pb, and Zn were above NYS DEC soil screening levels. Typically, produce is not tested for trace metals and when such tests are done, a bioconcentration factor (BCF) is used to assess contamination (the ratio of plant or plant part to total soil trace metal concentrations). However, BCF does not consider the exchangeable fraction, which rarely corresponds to total concentrations. Yet the exchangeable form of trace metals is the form through which they can be absorbed by roots, although there are multiple variables that govern the bioaccessibility of trace metals, pH and plant characteristics being the most influential. This explains the frequent contrast between total soil and vegetable trace metal concentrations. The research results reaffirmed this well-known contrast in that some trace metals exceeding recommended levels were not detected in kale roots and leaves.

Additionally, findings point to kale contamination by Al, Ba, Cu, Sr, Zn being explained more by BAF (the ratio of exchangeable to total soil trace metal concentrations) than the other conventional indicators. This was corroborated by a congruence of BCF and BAF with TF (the ratio of leaf to root trace metal total concentrations). This raises confidence in the overall findings that the exchangeable fraction is an important variable to consider when assessing trace metal contamination and to overcome the limitations of BCF. The exercise was found to be especially useful in assessing Ba and Sr contamination.

The results support recommendations to urban vegetable growers for the sort of soil management practices that help

reduce trace metal exchangeability, such as maintaining near-neutral pH and adding reactive colloidal substances (biochar, most organic matter, zeolites, etc.) (Menefee and Hettiarachchi 2018). In this way, keeping track of the exchangeable fraction of trace metals contributes to a bioaccessibility-based evaluation process. At the same time, several inconsistencies were also found among variables. Therefore, other possible sources of contamination, such as airborne sources and additional soil characteristics (e.g., form and amounts of soil organic matter), should be prioritised in future research.

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Availability of data and material The datasets used and/or analysed during the current study are available from the corresponding author on reasonable request.

Declarations

Ethics approval and consent to participate Not applicable.

Competing interests There are no competing interests to report.

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