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Concentrations of lead, cadmium and barium in urban garden-grown vegetables: The impact of soil variables

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ABSTRACT

Paired vegetable/soil samples from New York City and Buffalo, NY, gardens were analyzed for lead (Pb), cadmium (Cd) and barium (Ba). Vegetable aluminum (Al) was measured to assess soil adherence. Soil and vegetable metal concentrations did not correlate; vegetable concentrations varied by crop type. Pb was below health-based guidance values (EU standards) in virtually all fruits. 47% of root crops and 9% of leafy greens exceeded guidance values; over half the vegetables exceeded the 95th percentile of market-basket concentrations for Pb. Vegetable Pb correlated with Al; soil particle adherence/incorporation was more important than Pb uptake via roots. Cd was similar to market-basket concentrations and below guidance values in nearly all samples. Vegetable Ba was much higher than Pb or Cd, although soil Ba was lower than soil Pb. The poor relationship between vegetable and soil metal concentrations is attributable to particulate contamination of vegetables and soil characteristics that influence phytoavailability.

1. Introduction

Urban environments are variably contaminated with substances such as metals and persistent organic pollutants as a result of human activities including transportation, construction, manufacturing, fossil fuel combustion, and incinerator emissions (Alloway, 2004; Biasioli et al., 2007; Bradley et al., 1994; Norm et al., 2001; Peltola and Aström, 2003). Consequently, urban garden soils can be moderately to severely contaminated by one or more metals, with lead (Pb), cadmium (Cd), and mercury (Hg) reported to be most likely to pose some hazard for human health (Alloway, 2004; Chaney et al., 1984; Preer et al., 1980; Stilwell et al., 2008). Elevated barium (Ba) has also been found in urban environments, including soils and airborne particulate matter adjacent to roads (Monaci et al., 2000; Paterson et al., 1996). Because of the widespread use of Ba in manufactured materials such as tiles, automobile clutch and brake linings (ATSDR, 2007), rubber, brick, paint, glass, and other materials, unusually high concentrations of this metal in soils may be a marker for anthropogenic activity, including traffic (Monaci et al., 2000).

Numerous studies have investigated the relationship between metal contamination of urban garden soils and garden-raised foods, particularly for Pb and Cd (Alloway, 2004; Moir and Thornton, 1989; Sánchez-Camazano et al., 1994; Spliethoff et al., 2013). For Pb, the highest levels in vegetables generally occur where soil Pb levels are the highest (Bielinska, 2009; Huang et al., 2012; Jorhem et al., 2000; Moir and Thornton, 1989; Samsøe-Petersen et al., 2002). Some studies using sensitive methods for analyzing vegetable metals concentrations have indicated a near-linear relationship between soil Pb and vegetable Pb concentrations, so that bioconcentration factors (BCFs) could be estimated (e.g., 0.001, 0.002 and 0.05 for lettuce, potato and carrot [with peel], respectively) (Samsøe-Petersen et al., 2002). However, despite some success in linking concentrations of metals in vegetable crops to soil contamination levels, the results overall have been inconsistent, particularly for Pb (Hough et al., 2004; Jorhem et al., 2000; Peris et al., 2007; Samsøe-Petersen et al., 2002; Säumel et al., 2012).

The difficulty in establishing a quantitative relationship between vegetable Pb content and soil Pb content that is frequently (but not always) observed (Hough et al., 2004; Jorhem et al., 2000;

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Murray et al., 2011) may be in part because the typically low levels of soluble and bioavailable Pb are not simply dependent on the total soil Pb burden, but are subject to control by important soil properties including pH, organic matter content, and dissolved organic matter (Sauvé et al., 2000, 1998). For example, uptake reported for Pb is typically quite low except where soils are strongly acidic because of the strong tendency for Pb to be immobilized in neutral- and higher-pH soils by adsorption and precipitation reactions (Sauvé et al., 2000, 1998; Mosbaek et al., 1989). However, even after accounting for pH and other important soil properties, prediction of Pb uptake by vegetable crops from urban gardens has been generally unsatisfactory (Hough et al., 2004). Jorhem et al. (2000) found a relationship between vegetable Pb content and soil total Pb and pH, but their study sites represented a wider range of soil pH than seen in many studies of garden soils. For most garden soils, crop type has proven to be a stronger determinant of the edible crop metal concentration than soil contamination level (Alexander et al., 2006; Douay et al., 2013; Moir and Thornton, 1989; Samsøe-Petersen et al., 2002).

In order to make some sense of the apparently contradictory nature of Pb uptake results taken as a whole, which Jorhem et al. (2000) referred to as “information with a lack of unanimity,” it is necessary to consider some critical factors that have compromised data and obscured trends in many (particularly older) studies of Pb in food crops:

1. Insufficient analytical sensitivity to measure low Pb concentrations in vegetables. Market vegetables grown in uncontaminated rural regions have quite low Pb levels (e.g., median Pb concentrations < 0.006 mg/kg (US FDA, 2010, 2007)) that require very sensitive methods such as inductively coupled plasma mass spectrometry (ICP-MS) to measure correctly (McBride, 1998).
2. External sources of Pb (e.g., deposition from air). This was a major problem decades ago when leaded gasoline was in widespread use and could still be a concern in some urban environments. Many earlier plant uptake studies may have been compromised by aerial contamination of vegetables that obscured impacts of soil Pb on vegetable concentrations (Chamberlain, 1983; Dalenberg and Driel, 1990; Prasad and Nazareth, 2000; Sheppard and Evenden, 1992).
3. Highly variable physical contamination of vegetables (e.g., from dust, soil splash, traffic-related aerial contamination) related to local conditions, and the type of vegetable (with differing surface area and roughness of plants) (McBride et al., 2012; Nali et al., 2009; Säumel et al., 2012; Uzu et al., 2010).
4. Highly variable spatial distribution of Pb in soils of urban environments, from the scale of several hundred meters (Säumel et al., 2012; Shinn et al., 2000) to the minute scale of the root zone, soil aggregates and microscopic particles (Tai et al., 2013; Wharton et al., 2012).

The high degree of variability and apparent randomness inherent in data for Pb measured in vegetables is disconcerting, and indicates that even more intensive research will be needed to identify factors that contribute to vegetable contamination by metals such as Pb. Though available data are limited, when the number of environmental and soil variables is reduced, as is done with single-site field studies and greenhouse research with one particular soil containing a range of Pb concentrations, a clearer relationship between vegetable Pb and soil Pb emerges (McBride, 2013; and unpublished results).

The present investigation was undertaken as a follow-up to a study designed to measure trace metals of potential concern in urban community gardens of New York City (NYC). New York State (NYS) Department of Environmental Conservation Soil Cleanup Objectives for residential land use (NYSDEC, 2006) developed for the NYS Environmental Remediation Programs were considered as guidance values for comparison with garden soil metal concentrations in a study of more than 500 garden soil samples (Mitchell et al., 2014), which identified Pb and Ba as metals that commonly exceeded guidance values. Concentrations of arsenic (As), nickel (Ni), copper (Cu), chromium (Cr), and zinc (Zn) rarely exceeded guidance values in the results reported by Mitchell et al. (2014) and so were not included in the present study. Cd is included in the present study because of its potential for uptake into vegetables even at relatively low soil concentrations, and because analytical limitations encountered with inductively coupled plasma optical emission spectrometry (ICP-OES) in the pilot study led to uncertainty about the range of Cd concentrations present in the soils.

The immediate objectives of the present study were to measure: 1. Total Pb, Cd and Ba in the washed edible portion of a range of vegetable types grown in community gardens and farms of urban areas in NYS, 2. These same metals in soil samples collected from the exact location of vegetable collection, and 3. Aluminum (Al) in the vegetables. Al is quite insoluble and unavailable for plant uptake at the near-neutral pH levels generally found in urban garden soils, and its presence in vegetable samples can be taken as an indicator of the physical presence of soil particles either in or on the washed vegetable samples (McBride et al., 2012). For example, washed vegetables were found to contain from 0.07 to 4.88% soil on a dry-weight basis (using immobile soil elements such as Al as indicators of vegetable contamination) (Cary et al., 1994).

The overall goals of the study were: 1. To determine the degree to which the concentrations of these metals in vegetables could be linked to metals concentrations in soils, 2. To compare metals concentrations in urban garden produce with concentrations found in market-basket produce, and 3. To determine whether consumption of the urban garden produce could represent a significant health hazard based on comparison with available health-based guidance values.

2. Materials and methods

Vegetable samples (80 fruiting vegetables, 67 leafy, 16 herb and 32 roots) were collected over the growing seasons in 2011 and 2012 from seven community gardens in NYC and ten gardens and urban farms (hereafter referred to as “gardens”) in Buffalo, NY. Each vegetable sample was collected simultaneously with a paired surface soil sample (0–15 cm) from the same location in the garden plot. A total of 195 pairs of soil and vegetable samples were processed (160 from NYC and 35 from Buffalo). The number of soil/vegetable pairs collected from a particular garden varied widely depending on sample availability. In some cases, samples of multiple crop types were harvested from the same garden bed. Sampling locations were not pre-screened for soil contaminant level, as the intent was to collect samples from a range of soils. Upon arrival at Cornell University, vegetable samples were washed thoroughly under tap water, scrubbed with a vegetable brush when necessary to remove visible soil (e.g., for all root crops), and blotted dry with paper towels. Root crops were not peeled. The vegetables were cut into small pieces, placed into labeled brown paper bags (most vegetables) or open-topped glass jars (for juicy vegetables such as tomatoes), and dried in an oven at 70 °C for several days to a week until the samples appeared dry based on visual inspection. Once dry, samples were ground into a coarse powder using a coffee grinder, and stored in sealed labeled Whirl-Pak™ bags for later digestion and analysis. The grinder was cleaned of all plant residue between samples using a pressurized air stream to prevent cross-contamination.

Soil samples were prepared by air-drying in a laboratory hood for several days and passing through a 2 mm plastic sieve, and were subsequently stored in closed cardboard containers. Soil pH was determined by weighing out 10 g of each soil sample from the cardboard containers into small glass jars, adding 20 mL of distilled water, mixing the soil-water slurry, allowing it to stand for 30 min, and determining the pH of the supernatant using a glass electrode.

All samples underwent microwave digestion with HNO_3 US EPA SW-846 Method 3051, (US EPA, 2012) prior to metals analysis. Soil samples were analyzed for Pb, Cd, and Ba either by a commercial laboratory (H2M Labs, Inc.) certified by the NYS Environmental Laboratory Approval Program using ICP-MS (US EPA SW-846 Method 6020) or by the Cornell Nutrient Analysis Laboratory (CNAL) using ICP-OES (US EPA SW-846 Method 6010) (US EPA, 2012). 39 soils were analyzed by