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## SPECIAL SECTION: URBAN SOILS RESEARCH—SUITMA 10

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# Atmospheric sources of trace element contamination in cultivated urban areas: A review

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#### Abstract

Producing food in cities has garnered increasing attention over the past decade. Although there are ecological and social benefits, cultivated urban areas (CUAs) also bear contamination hazards, including from trace elements (TEs). Trace element contamination has been studied extensively in CUAs, but atmospheric sources remain understudied and poorly understood. A brief discussion is offered on atmospheric particulate deposition processes in cities and their implications for urban food production. Available findings are discussed and contrasted. Existing research assesses atmospheric deposition indirectly or otherwise lacks controls for other TE contaminants. There is little to no engagement with methodological guidelines from the atmospheric sciences, which reduces confidence in the findings so far attained. Suggestions are delineated to combine techniques used in the atmospheric sciences with the robust methodologies already generated by studies on TE contamination in CUAs, such as isotope and TE ratios analyses.

## **1** | INTRODUCTION

Urban food production has recently witnessed renewed institutional interest (Hou, 2017; WinklerPrins, 2017). Producing food in cities can contribute to improving ecological and social conditions (Chrisinger & Golden, 2016; Parece & Campbell, 2017), if introduced and practiced in politically empowering and egalitarian ways (Gottlieb & Fisher, 1996; Martínez, 2010; Tornaghi, 2017). However, concerns have been increasingly raised over trace element (TE) contamination undermining urban food production (Brown, Chaney, & Hettiarachchi, 2016; Cheng et al., 2015; Hursthouse & Leitão, 2016; Wortman & Lovell, 2013). Trace element contamination is a widespread problem in cities (Meuser, 2010; Webb, Rubio, & Fullen, 2019), especially in economies based on endless capital accumulation (McClintock, 2015).

Because they cannot be biologically or chemically broken down, TEs constitute a lasting health hazard (Hodges Snyder, McIvor, & Brown, 2016). High TE concentrations in human bodies can lead to impairment and death, and sources can be environmental, anthropogenic, or a combination of both (Kabata-Pendias, 2011; WHO, 2007). For example, various forms of cancer are traceable to both arsenic (As), which can originate from geothermal and bedrock sources as well as from mining, smelting, and long-term agrochemical applications, and nickel (Ni), which is also derived from bedrock and mining and related activities, as well as many electrical products involving alloying, batteries, and coating. Neural harm is associated with As, mercury (Hg), and lead (Pb), the latter two of which are often human derived. Mercury, for instance, although traceable to forest fires and volcanic emissions,

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**Abbreviations:** CUA, cultivated urban area; PM, particulate matter; TE, trace element.

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is now mostly from human sources, mainly gold mining and coal combustion. Toxic levels of cadmium (Cd), which is largely from sources like smelters, waste incineration, and batteries, is linked to kidney damage and bone fragility (Aelion, Davis, McDermott, & Lawson, 2009; Centeno et al., 2005).

Consequently, concerns are being raised about urban food production intensifying TE contamination exposure potential. Exposure can occur by inhaling or ingesting soil particles and consuming contaminated vegetables (Brevik & Burgess, 2016; Brown et al., 2016; Mitchell et al., 2014). These kinds of problems are much broader, affecting any form of food production near urban, industrial, military, and transportation areas (Boente, Matanzas, García-González, Rodríguez-Valdés, & Gallego, 2017; Graefe, Buerkert, & Schlecht, 2019; Szolnoki, Farsang, & Puskás, 2013; Tóth, Hermann, Da Silva, & Montanarella, 2016).

Major TE contamination pathways include former land use, the characteristics of belowground material, and pollution from nearby sites (Zwolek, Sarzyńska, & Stawarczyk, 2019). Land use legacies can involve building debris residues, landfills, and Pb-based paint residues, among other impacts (Alloway, 2004; Chaney, Sterrett, & Mielke, 1984; Meuser, 2010; Spittler & Feder, 1979). In situ legacies can add to or magnify preexisting, belowground anthropogenic, and/or geogenic sources (Barbieri, Sappa, & Nigro, 2018; Bourennane et al., 2010; Jean-Soro, Guern, Bechet, Lebeau, & Ringeard, 2015; Pouyat, Yesilonis, Russell-Anelli, & Neerchal, 2007). Proximity to polluting sources, such as landfills, power plants, incinerators, and vehicular traffic, often adds to existing TE levels or introduces TE contamination (Adamiec, Jarosz-Krzemińska, & Wieszała, 2016; Barbieri, Sappa, Vitale, Parisse, & Battistel, 2014; Mielke, Laidlaw, & Gonzales, 2010; Nadal, Schuhmacher, & Domingo, 2004; Sung & Park, 2018; Uzu, Sobanska, Sarret, Muñoz, & Dumat, 2010). This occurs through the deposition of variable-size particulate matter (PM) that may also be transferred from great distances (Schreck, Foucault et al., 2012; WHO, 2007).

Much research has focused on soil-to-vegetable TE transfer, which is governed by multiple interrelated variables that may include human sources and impacts (Alloway, 2013; Cheng et al., 2015; Gupta et al., 2019; McBride, 1989; Menefee & Hettiarachchi, 2018; Tack, 2010). Among the impacts are cultivators' inputs, which can introduce contaminants directly or alter soil properties in ways that favor or mitigate vegetable TE absorption (Agbenin, Welp, & Danko, 2010; Bolan & Duraisamy, 2003; Bretzel, Calderisi, Scatena, & Pini, 2016; De Miguel, Jimenez de Grado, Llamas, Martin-Dorado, & Mazadiego, 1998; Nabulo, Young, & Black, 2010). Prior human uses can

#### **Core Ideas**

- Trace element contamination by atmospheric deposition in cultivated urban areas is understudied.
- Several weaknesses undermine the confidence in findings to date.
- More engagement with atmospheric sciences is warranted.
- Combining diverse techniques used effectively but thus far in isolation is recommended.

also leave substances or debris that may augment or attenuate TE availability to plants and other organisms, depending on intrinsic and evolving, human-affected soil properties (Howard & Olszewska, 2011; Hursthouse & Leitão, 2016). As a result of these multiple sources and processes of contamination, TE levels in plants do not necessarily correspond with those in the soil on which they grow (Allen & Janssen, 2006; McBride et al., 2014; Warming et al., 2015).

Atmospheric deposition is among the major reasons for a lack of direct relationship between soil and crop contamination. Since PM is often TE enriched, airborne PM deposition can play a significant role in TE contamination in both rural and urban cultivated areas (Azimi, Cambier, Lecuyer, & Thevenot, 2004; De Temmerman, Waegeneers, Ruttens, & Vandermeiren, 2015; Xiong et al., 2014). Airborne PM is eventually deposited and accumulated on soil surfaces and plants as it is rained out (scavenged) or otherwise settles out of suspension as dry deposition (Amodio et al., 2014; McCleod et al., 2011). In plants, contamination may occur through lodging in tissues (Schreck, Bonnard, et al., 2012) or direct foliar absorption, as implied in studies on foliar fertilizer application since at least the early 1900s (Fageria, Filho Barbosa, Moreira, & Guimarães, 2009) and corroborated by radionuclide research (Fismes, Echevarria, Leclerc-Cessac, & Morel, 2005; Uzu et al., 2010). With the proliferation of nanoparticles from nanotechnologies and vehicular emissions, crop TE contamination through PM absorption has been exacerbated (Larue et al., 2014; Rajput et al., 2018). However, surprisingly little sustained research has been dedicated to processes of atmospheric deposition and its relative importance compared with other contamination processes. In this review, the general mechanisms of atmospheric deposition are described in terms of their potential effects on cultivated urban areas (CUAs). Research on atmospheric TE deposition in CUAs is then discussed, and knowledge gaps are identified.

## 2 | ATMOSPHERIC FLUXES AND TRACE ELEMENT CONTAMINATION IN CITIES

There are both environmental and human-derived sources of atmospherically dispersed TEs (Nriagu, 1989; Schroeder, Dobson, Kane, & Johnson, 1987). Environmental sources include processes like forest fires, marine water sprays, and volcanic emissions. Human-derived sources include a variety of largely industrial sources, like cement and metals manufacturing, incinerators, power plants, refineries, smelters, and vehicular exhaust and dust. Proximity to vehicular traffic and Pb from leaded petrol have been known since the 1970s to be major sources (Adamiec et al., 2016; Mielke et al., 2010).

In most cities, contaminants tend to be poorly retained and quickly exported to other areas or stored in soils, sediments, and organisms within urban and peri-urban areas. This relative transience and rapid short- to long-term storage are due to the presence of predominantly impervious surfaces that can themselves be contaminant sources (Diamond & Thibodeux, 2011; Webb et al., 2019). In such environments, PM deposition is among the main mechanisms of contaminant movement, which is affected by wind conditions (e.g., predominant direction, speed, duration, magnitude, and timing) and deposition surface properties (type, size, proximity to emissions, and geometrical aspect relative to contaminant sources). Particulate matter deposition can also be concentrated on the leeward side of buildings in areas like courtyards or adjacent urban gardens (Aristodemou et al., 2018; Chang, Kao, Wu, & Huang, 2009; Mei, Wen, Xu, Zhu, & Xing, 2018; Seinfeld & Pandis, 2006). Particle characteristics also affect whether deposition will occur and how. Fine particles at or below 1 µm are deposited through diffusion, as they are more gaslike, whereas particles of larger diameter influence airflow and tend to fall by sedimentation (Janhäll, 2015; Vawda, Colbeck, Harrison, & Nicholson, 1989).

Once deposited, PM can be incorporated into various semiporous surfaces, like soils, or trapped and in part absorbed by vegetation and, to a minor extent, organisms (Pandey & Pandey, 1994; Wuana & Okieimen, 2011). Although not a mechanism considered in this review, PM can also be precipitated into bodies of waters and travel across a city by hydrological mechanisms. These include surface runoff (e.g., flooding) and groundwater movement (e.g., throughflow). Unless buried and sealed by impervious materials or trapped in sediment at the bottom of water bodies, deposited PM can be resuspended and redeposited (or remobilized hydrologically), thereby redistributing substances that include TEs (Laidlaw & Filippelli, 2008; McKenna-Neuman, 2011).

## 3 | IMPLICATIONS OF ATMOSPHERIC DEPOSITION PROCESSES FOR CULTIVATED URBAN AREAS

As indicated in the above discussion, TEs can be temporarily stored in various media (including cultivated soils and vegetables), given off by different kinds of surfaces, and repeatedly redistributed within and away from urban areas, as well as absorbed by plants and concentrated in soils over the short or long term. Particulate matter redeposition occurs as well by way of cultivation inputs, rather than aerial processes (Clark, Hausladen, & Brabander, 2008; Paltseva et al., 2018). The introduction of imported sediment or soil amendments may simultaneously spread TE-bearing particles in cultivated areas, involving distal or proximal sources, or both. Cultivated urban areas form part of these PM and TE redistribution processes within and across urban areas and through which TEs are also diffused.

Given these mechanisms, TE foliar uptake in crops grown in CUAs should be of major concern. Trace elements can contaminate crops by entering through stomata and various permeable or semipermeable tissue openings or breaks within any aboveground plant portions. The extent and rate of specifically foliar uptake will depend on the location and relative abundance of such entry points, plant phenology, and morphology (including leaf size and architecture), as well as TE quantity and chemical species characteristics (De Temmerman et al., 2015; Hu et al., 2011; Luo, Bing, Luo, Wang, & Jin, 2019; Pandey & Pandey, 2009; Žalud, Száková, Sysalová, & Tlustoš, 2012).

With respect to atmospheric sources, the diffusion and extent of particle deposition in CUAs will depend, beyond already cited variables, on the layouts of cultivated parcels and adjacent buildings, as well as on the characteristics of concrete surfaces and vegetation, in addition to proximity to known sources of contamination like vehicle emissions (Mei et al., 2018; Sung & Park, 2018). Atmospheric deposition can occur as long-range PM influx (distal sources), local or intra-urban redeposition of PMs from urban surfaces outside the receiving cultivated areas (proximal sources), and PM fluxes within CUAs due to cultivation practices and the effects of localized meteorological phenomena (internal sources). These sources are not mutually exclusive, as internal and proximal sources can become available for long-range transport.

Trace element deposition from distal sources is typically associated with upwind point sources of pollution, but sources can conceivably include mines, contaminated farmed soils, and military installations in the countryside or in peri-urban zones (Morman & Plumbee, 2014; Schreck, Bonnard, et al., 2012). Redeposition from proximal sources can occur through erosion and entrainment of TE-bearing PMs from nearby soils, including from nearby cultivated areas (Clark et al., 2008). Other proximal sources may introduce TEs from outside of cities, mainly in the form of industrial emissions and vehicular traffic (Mielke et al., 2010; Vittori Antisari, Carbone, Ferronato, Simoni, & Vianello, 2012), which combine imported materials and fuels to produce contaminants that will eventually be redistributed within a city (Laidlaw & Filippelli, 2008; Webb et al., 2019). Vehicular traffic burden is typically indicative of the intensity of TE contribution that can be expected generally (Adamiec et al., 2016; Al-Taani, Nazzal, & Howari, 2019), and for CUAs in particular (Säumel et al., 2012).

Internal source contamination is possible from cultivation practices that can dislodge TE-bearing soil particles from depth or the surface and lead to their translocation within and away from CUAs (Egendorf et al., 2018). Findings by Paltseva et al. (2018) also point to vegetablespecific Pb uptake relative to distance from soil surface. Carrots [Daucus carota L. ssp. sativus (Hoffm.) Arcang.] absorb Pb directly, but radishes (Raphanus sativus L.) and lettuce (Lactuca sativa L.) trap nonwashable Pb-laden particles. In contrast, tomatoes (Lycopersicon esculentum Mill.), being farther away from the soil surface, show negligible Pb amounts (see also Cai, McBride, & Li, 2016). Activities promoting TE movement and redeposition can include digging associated with bed preparation and sowing, infrastructure construction (e.g., garden tool sheds), as well as irrigation-induced splash-derived particle displacement. Weather events, aside from depositing dust, can lead to soil erosion and particulate redistribution within and away from CUAs by such processes as raindrop impacts, internal aeolian particle loosening, and entrainment with internal redeposition.

## 4 | STUDIES ON TRACE ELEMENT ATMOSPHERIC DEPOSITION IN CULTIVATED URBAN AREAS

Research on CUA contamination is relatively recent, not going much further back than the 1960s (Spittler & Feder, 1979). Monitoring of atmospheric TE fluxes is also of recent vintage. In Canada, for instance, it was not until the 1990s that precipitation chemistry monitoring began and that studies emerged to analyze atmospheric TE diffusion trends (Pilgrim & Schroeder, 1997). Suspicions of windborne TE recontamination of urban soils also emerged by the 1990s (Aschengrau, Beiser, Bellinger, Copenhafer, & Weitzman, 1994). Nevertheless, studies on atmospheric deposition in CUAs remain rare, even if long- and shortrange atmospheric sources of TE contamination have been recognized for decades as contributors of TE contamination in agriculture (Azimi et al., 2004; Hovmand, Tjell, & Mosbaek, 1983; Pandey & Pandey, 2009; Peris, Micó, Recatalá, Sánchez, & Sánchez, 2007; Voutsa, Grimanis, & Samara, 1996).

Research on atmospheric sources during the 1990s in St. John, NL, Canada, showed tendencies for higher TE content, especially Pb, in urban garden soils and vegetables that could be linked to atmospheric deposition (Pilgrim & Schroeder, 1997). Atmospheric deposition samples obtained daily were bulked to obtain weekly readings. Soils and vegetables were sampled in nine urban gardens in the city of St. John and compared with one rural garden. The authors interpreted the analytical results on snow and rain samples as showing that aboveground parts of vegetables were more likely contaminated by atmospheric deposition. Meteorologically affected TE flux variability was addressed by monthly sampling and analysis over different seasons. However, TE content differentials shown among soil, snow, and rain did not entirely support such conclusions. As the reported findings lacked spatially explicit information on soil and vegetable samples, it is not possible to discern potential associations of soil-borne contaminant to levels of contaminants in vegetables. The soil depth sampled was also unreported, bringing further data ambiguity. It was also unclear which parts of vegetables were sampled or whether entire plants were analyzed. A lack of sampling of gardening inputs also adds another confounding factor, thereby hampering the discernment of atmospheric sources.

It appears that several years elapsed before other similar studies were published. Remarks on the possibility of airborne sources appeared in some works, but without any direct study of atmospheric deposition effects on crop contamination. For example, Romic and Romic (2003) found high levels of TEs in surface cultivated soils in Zagreb. Using factor analysis, they concluded that multiple human sources were likely, ranging from waste disposal and upstream mining to fuel combustion from nearby transport. However, these were total soil concentrations, which, as discussed above, rarely reflect what is phytoavailable. There was also no accounting for atmospheric deposition variability, including from transfers of contaminated soil particles in the soil surfaces studied. Similar problems were obtained in a study in Shenyang, China, by Jiang, Qin, Zhang, Li, and Liang (2008). Since soil copper (Cu) levels increased with proximity to suburban zones, they attributed high soil Cu levels to urban pollution. However, farmers' sewage sludge applications and air emissions could not be distinguished as sources.

A 3-yr study concerned with Pb contamination was instead more attentive to atmospheric deposition processes

(Clark et al., 2008; Clark, Brabander, & Erdil, 2006). The study was carried out on raised beds in backyard vegetable gardens in two Boston, MA, communities. Two-tiered soil sampling (surface and rooting depth) was combined with sampling of compost and four vegetable taxa known for their TE bioaccumulation potential. Results spanning 3 yr for a nearby excavation site, two raised beds, and five backvard garden areas showed an inverse relationship between grain size and Pb levels, an enrichment in the fine soil particle fraction (and in total silicon [Si] content), and no Pb concentration gradient, which is typically found with distance from vehicular traffic. The net increase over time in Pb levels in both soil and applied compost was interpreted to be likely caused by recontamination from windtransported particles from surrounding backyard garden soils. Raised beds were found to reduce Pb phytoavailability (and thereby exposure by eating vegetables), but not exposure to Pb by particle ingestion. However, it is not possible from the study to distinguish among atmospheric fluxes (internal, proximal, and distal). Furthermore, relative traffic burden and wind-barrier characteristics were not considered.

The latter was featured in another, more recent study in Berlin, Germany. The focus was on urban garden vegetables' relative proximity to vehicular traffic emissions (Säumel et al., 2012). In contrast to the Boston study, the findings were that, irrespective of taxon, the TE concentrations in vegetables grown further away from vehicular traffic were less likely to exceed allowable limits, at least in the case of central Berlin urban vegetable gardens. A first attempt of its kind in assessing the impact of vehicular emissions, the study demonstrates the importance of giving more attention to atmospheric deposition as a major contamination pathway. As the authors have themselves underlined, further research is needed to determine the role of vehicular emissions in vegetable TE contamination. For instance, no soil samples were taken and analyzed (hence, for example, no control for pH effects on root absorption rates), whereas traffic-related emissions were the only source of atmospheric deposition considered. There was also no control for gardening inputs or potential effects of contaminants from adjacent building walls (especially relative to Pb-based paint). There is also another way the results could be interpreted, at least in the case of Pb. The percentage of contamination cases >10 m away from traffic were still relatively high (up to a third or more) for TEs such as Pb. Taken together with the fact that leaded petrol was not a factor at the time of the study, the findings suggest the possibility of Pb-containing particle resuspension and redeposition, rather than vehicular emission effects. Thus, it is unclear whether the findings reflect the impact of proximity to vehicular traffic or a combination of factors, including distance from vehicular 5

emissions (which more likely contribute to Cd, chromium [Cr], and Ni enrichment, rather than Pb) that were omitted from the study. There is hence little guarantee that reducing nearby vehicular traffic will significantly reduce TE contamination without taking into consideration other potential sources of contamination.

In a later study of five gardens in Bologna, Italy, airborne TE deposition (and soil phytoavailability) was similarly deduced based on distance from presumed pollution sources (railway and road) compared with soil-less rooftop and rural garden samples (Vittori Antisari, Orsini, Marchetti, Vianello, & Gianquinto, 2015). Only crop leaves and soils (single-tiered) were sampled and analyzed. A coeval 3-yr study in Toronto, ON, Canada, was carried over three vegetable garden sites (one being on a rooftop), representing different traffic burden intensities (Wiseman, Zereini, & Püttmann, 2013, 2014, 2015). Soil sampling occurred incrementally to rooting depth, and curbside dust and above- and belowground vegetable tissues were also sampled. All samples were taken at multiple times during each project year, to address seasonal and longer term changes. Results were interpreted as showing that traffic burden sources are the predominant source of TE contamination in vegetables, largely through the rhizosphere. Neither the Bologna nor the Toronto study included sampling of atmospheric deposition or geogenic sources and anthropogenically contaminated soils and sediments, which can manifest themselves as redeposited particles associated with nearby building sites (Clark et al., 2008). There was also no accounting for source distance effects (internal, proximal, or distal) or atmospheric deposition duration, as Amato-Lourenco et al. (2016) pointed out for the Berlin findings. Although traffic-related TE contamination seems likely in the Berlin, Bologna, and Toronto studies, other sources of contamination may have been prematurely ruled out, especially if one considers time scales >3 yr.

Building explicitly on Säumel et al. (2012), Amato-Lourenco et al. (2016) tested for atmospheric deposition in collards (Brassica oleracea L. var. viridis L.) and spinach (Spinacia oleracea L.) over 30-90 d in 10 urban community vegetable gardens in São Paulo, Brazil. Unlike prior studies, the researchers added weather variables (temperature, humidity, rainfall, and wind speed) in addition to vertical obstacles (height, width, and distance) to vehicular traffic burden level. Vegetables were grown on uncontaminated soil in coir-covered polyethylene containers 1 m above the cultivation surface to avoid splash-related particle detachment and redeposition. The containers were placed centrally within the cultivation area. Tillandsia usneoides (L.) L. specimens (Bromeliaceae) were used as controls and for biomonitoring purposes (see also Gupta et al., 2019). Significant differences in vegetable TE content were found, related to differences in vehicular traffic exposure levels,

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vertical obstacles, and meteorological conditions. Higher traffic burden contributed to greater vegetable contamination. Conversely, vertical barriers correlated with lower vegetable TE contamination levels. The major influence of traffic burden was corroborated by the biomonitoring portion of the study (Amato-Lourenco et al., 2017). Impressively, concentrations of TEs like Cd and Pb surpassed permissible levels within a couple of months in the more exposed and traffic-burdened vegetable gardens. However, unlike several of the above-discussed studies, the brevity of the research project did not allow for seasonal differences in TE levels to be ascertained. Moreover, the placement of the specimens at 1 m above the cultivated surface is known to be insufficient in avoiding the effects of within-garden particle resuspension and contamination (see Amodio et al., 2014; Dämmgen, Erisman, Cape, Grünhage, & Fowler, 2005). Thus, internal sources may still interfere with those from proximal vehicular emissions relative to airborne particle deposition and TE contamination.

To overcome some of these ambiguities, the present author carried out preliminary research in two urban community gardens in Rome, Italy, to parse out atmospheric from other sources of contamination (Engel-Di Mauro, 2018). A three-tiered soil sampling program aimed to include geogenic effects, while soil samples, taken within a 5-cm radius of vegetables, were analyzed for the main known soil properties governing TE mobility. Soil analyses included tests for extractable As and Pb. Gardeners inputs, including irrigation water, were also sampled. Whole vegetables were harvested, with edible portions tested for As and Pb content. A bulk sampler was sited according to standard procedures (Dämmgen et al., 2005; Grynkiewicz, Polkowska, Zygmunt, & Namieśnik, 2003) within each cultivated area and used to sample atmospheric deposition at monthly intervals for 93 d. Atmospheric As and Pb influx was above permissible levels. Controlling for cultivation, soil-borne, and geogenic sources, it was postulated that atmospheric deposition may be a significant source of As contamination, mainly through foliar uptake, as As was found in amounts exceeding safe levels on vegetable leaves and on some fruiting bodies. However, replicates were insufficient, distal atmospheric sources could not be satisfactorily distinguished from internal and proximal sources, and the study duration was too short relative to seasonality.

The few studies that have included sampling for atmospheric deposition have used bulk deposition samplers. These can establish total TE influx over a 5-km radius, which may be too broad relative to the typically much smaller areas dedicated to urban food production. Nevertheless, they can theoretically account for distal sources, if coupled with sampling and analyses relevant to internal and proximal sources. It must be borne in mind that bulk deposition samplers do not separate size categories, so they cannot aid in specifying likely contaminant provenance or in modeling fluxes or processes involving the relationship between crop and atmospheric deposition. This may become especially important in identifying the effects of nanoparticle fluxes.

In this, isotopic ratios can be additional aids in determining TE sources, such as discerning paint from petrol-based Pb (Clark et al., 2006) or soil- from airborne Pb (see Hu et al., 2011). Trace element ratios, such as titanium (Ti)/Pb (Clark et al., 2006), or correlations between aluminum (Al) and TEs (Paltseva et al., 2018) can help distinguish among internal sources, including from soil particle attachment to vegetable tissues (McBride et al., 2014). To raise certainty in differentiating between internal redeposition and proximal and distal atmospheric sources, specimens could be grown at multiple heights (surface, 1 m, and 2 m) and at different distances from presumed proximal sources. This could be carried out using containers as recommended by Amato-Lourenco et al. (2016) or row covers and portable greenhouses with ascertainable permeability characteristics.

There are a variety of instruments and techniques used to sample atmospheric deposition. Many overviews are available regarding atmospheric deposition sampling techniques, instrumentation, and analytical methods that can help build research designs appropriate to CUAs (Amodio et al., 2014; Dämmgen et al., 2005; Elmes & Gasparon, 2017; Grynkiewicz et al., 2003; Popoola, Adebanjo, & Adeoye, 2018). These do not yet appear to have been fully integrated into TE contamination research for CUAs.

A salient example worth revisiting is the abovediscussed study carried out in St. John, Canada, in the 1990s (Pilgrim & Schroeder, 1997). Following a 50-km urban-rural transect starting from the city center, the researchers used two kinds of methods that are meteorologically specific (late autumn 1992 in New Brunswick Province, Canada). For snow, samples were taken with plastic tubes 7 cm in diameter. For rain, an automated MIC precipitation sampler was used, consisting of a bucket with a polyethylene insert bag. For other climate regions, wetdry or dry deposition gauges will not need to be specific to snow or ice, but they may have to be much more capacious in high-rainfall areas, more sensitive to dry deposition in arid lands, and more attuned to capturing droplets from mist or fog in coastal or similar situations.

Finally, it is worth considering that Pb contamination in CUAs may not be as much of a concern than is usually assumed, and cultivation practices can minimize the hazard (Brown et al., 2016). This, however, pertains to Pb and to contaminated food ingestion and particulate inhalation pathways from sources internal to cultivated areas. It is unclear whether similar cultivation techniques are as effective with other TEs. There has also not been enough studies to determine the health impacts of atmospherically deposited TEs from proximal and distal sources, not only through ingestion, but also through inhalation of what could be polluted air, with little difference in contamination level from the rest of the city. The difference in air quality within CUAs and the rest of a city also requires greater research attention.

## 5 | CONCLUSIONS

Despite copious research on contamination in CUAs, surprisingly little attention (<10 studies) has been dedicated to TE contamination by atmospheric deposition. Most existing studies that consider airborne sources do not include sampling of PM deposition. Effects are largely inferred, rather than measured or estimated, and without studying the effects of wind conditions, physical layouts, and rainfall variability. Use of standard instrumentation in the atmospheric sciences could be combined with existing approaches to provide clarity regarding contaminant sources. Longitudinal studies spanning at least a decade are especially needed in assessing the extent and effects of TE accumulation in soils from atmospheric sources.

It is also important to consider TE contamination from a wider perspective. Atmospheric particulate emissions, which include TEs, are associated with millions of premature deaths, in addition to the development of often life-long ailments in millions more people worldwide (Campbell-Lendrum & Prüss-Ustün, 2019; Di et al., 2017; Liu et al., 2019; Morman & Plumlee, 2014). Hence, in evaluating the net health effects and potentials of urban food production, the impact of TE contamination in cultivated soils and in food produced in cities should be compared with the well-known health consequences of air pollution. Doing so will likely cast urban food production in a much more positive light and refocus attention to matters of greater urgency, such as promoting urban food production while phasing out all sources of air pollution as rapidly as possible.

#### AUTHOR CONTRIBUTIONS

The author has researched and written the manuscript in its entirety.

#### CONFLICT OF INTEREST

The author declares that the research was conducted in the absence of any commercial or financial relationships that could be construed as a potential conflict of interest.

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