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Urban vegetable contamination - The role of adhering particles and their significance for human exposure

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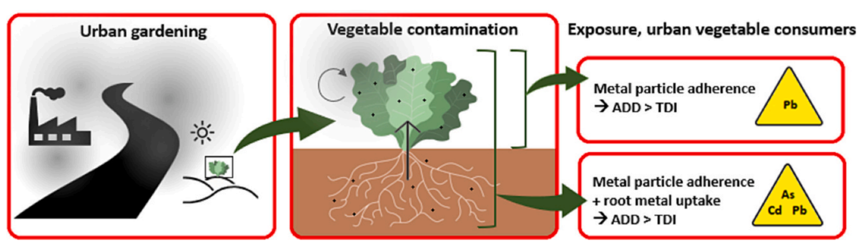
HIGHLIGHTS

- Urban vegetables contain adhering particles (APs) even after washing.
- The APs significantly contribute to the total human ingestion of particles.
- Foliar contamination by particles is crucial for the intake of several metals.
- The APs contribute most to the intake of Pb, with ADDs frequently > TDIs.

GRAPHICAL ABSTRACT



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ABSTRACT

While urban-grown vegetables could help combat future food insecurity, the elevated levels of toxic metals in urban soils need to be met with measures that minimise transfer to crops. This study firstly examines soil/dust particle inclusion in leafy vegetables and its contribution to vegetable metals (As, Ba, Cd, Co, Cr, Cu, Ni, Pb, Sb, and Zn), using vegetable, soil and dust data from an open-field urban farm in southeastern Sweden. Titanium concentrations were used to assess soil/dust adherence. Results showed that vegetables contained 0.05–1.3 wt% of adhering particles (AP) even after washing. With 0.5 % AP, an adult with an average intake of vegetables could ingest approximately 100 mg of particles per day, highlighting leafy vegetables as a major route for soil/dust ingestion. The presence of adhering particles also significantly contributed to the vegetable concentrations of As (9–20 %), Co (17–20 %), Pb (25–29 %), and Cr (33–34 %). Secondly, data from an indoor experiment was used to characterise root metal uptake from 20 urban soils from Sweden, Denmark, Spain, the UK, and the Czech Republic. Combining particle adherence and root uptake data, vegetable metal concentrations were calculated for the 20 urban soils to represent hypothetical field scenarios for these. Subsequently, average daily doses were assessed for vegetable consumers (adults and 3–6 year old children), distinguishing between doses from adhering particles and root uptake. Risks were evaluated from hazard quotients (HQs; average daily doses/tolerable intakes). Lead was found to pose the greatest risk, where particle ingestion often resulted in HQs > 1 across all assessed scenarios. In summary, since washing was shown to remove only a portion of adhering metal-laden soil/

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dust particles from leafy vegetation, farmers and urban planners need to consider that measures to limit particle deposition are equally important as cultivating in uncontaminated soil.

1. Introduction

The popularity of urban agriculture has increased in recent years with all indications being that this is a lasting trend (Kessler, 2013; Saumel et al., 2012; Warming et al., 2015; Witzling et al., 2011). This is, in part, due to its recognition as a key solution to future food insecurity, as highlighted following global crises such as the COVID-19 pandemic and the great recession in 2007–2009 (Colasanti et al., 2012; Lal, 2020; Loopstra et al., 2015; Pulighe and Lupia, 2020). Currently, about 6 % of the total cultivated land worldwide can be considered urban cropland, i.e., cultivated land in open spaces within municipality cores, in environments with high population densities and human-built structures (Thebo et al., 2014). Including also peri-urban cropland, within 20 km of cities, the fraction increases to 40 % (Thebo et al., 2014). However, with the accelerating urbanisation, a significant portion of the peri-urban cropland is projected to be lost (d'Amour et al., 2017), and this is a factor which is bound to increase the significance of initiatives for urban food production. Providing affordable and nutritious vegetables, produced with low transportation-related environmental impact while promoting green spaces and social interactions, urban gardening activities are well aligned with several of the UN Sustainable Development Goals.

Nonetheless, the physical and chemical properties of urban soils (from the Latin *word for city, 'urbs'*) often differ significantly from natural soils due to, e.g., construction activities that cause mechanical disturbances, and more frequent occurrences of unnatural components such as construction residues and contaminants from industries, domestic heating, heavy traffic and waste disposal (Lehmann and Stahr, 2007; Mamehpour et al., 2021; Puskás et al., 2008; Rezapour et al., 2022). It has repeatedly been shown that urban soils contain elevated concentrations of heavy metals (Bretzel and Calderisi, 2006; Clarke et al., 2015; Mitchell et al., 2014; Szolnoki et al., 2013), and as these elements are non-degradable they constitute a persistent challenge to produce safety. While plant-based foods are associated with many health benefits and should make up a significant proportion of our diet, many food agencies have identified vegetables, fruits and cereals as main sources of exposure to the most toxic metals (Cao et al., 2016; EFSA, 2009a; EFSA, 2009b; EFSA, 2010; Glorennec et al., 2016; Parveen et al., 2016). The metals typically considered most relevant in the context of produce safety, meaning that they can contaminate crops to levels that exceed toxicological reference values for human consumers, are As, Cd, Cr, Cu, Hg, Pb, Zn, Sb, Co and Ni (Tóth et al., 2016). Exposure to these elements can result in a spectrum of adverse effects, contingent upon the specific metal and dosage involved, encompassing renal, hepatic, and musculoskeletal impairments, along with neurological, cardiovascular and reproductive disorders (Nordberg, 2014).

The ability of cultivated crops to uptake and accumulate metal contaminants means that it is crucial to minimise health risks when increasing urban food production (Meharg, 2016). In this context, knowledge about metal contaminant transfer to urban produce and its associated risk implications is critical. Much research has focused on the soil to plant transfer of contaminants (i.e., uptake), and it has repeatedly been shown that cultivation in metal contaminated soil leads to elevated concentrations in crops and higher exposure in humans; in both urban environments (McBride et al., 2014; Saumel et al., 2012; Spliethoff et al., 2016) and near industrial point sources (Augustsson et al., 2015; Cui et al., 2004; Dziubanek et al., 2015; Hellstrom et al., 2007; Zheng et al., 2007). Correlations between total metal concentrations in cultivated soils and in the vegetables grown thereon are, however, often quite weak (McLaughlin et al., 2000; Menzies et al., 2007), and especially so for metals that are not easily taken up via the roots. One can, for example, compare Cd and Pb; two common soil contaminants that differ in that the first is among the trace elements with greatest propensity for root uptake, and the second representing an element of low phytoavailability, i.e., with only a small fraction of the element in the soil susceptible to plant absorption and potential accumulation in plant tissues. The uptake of Cd is closely related to its abundance in the soil and to soil (physico)chemical properties. Models aimed at predicting Cd in crops can therefore often be successfully developed based on soil data. For Pb, in contrast, the soil concentration is a weaker predictor for the concentration in crops (Brown et al., 2016; Dala-Paula et al., 2018; Hough et al., 2004; Legind and Trapp, 2010). This can partly be explained by the element's geochemical stability at the pH found in most soils (pH 5–7), which results in a low solubility even in soils with very high total concentrations (Moreno-Jimenez et al., 2011).

However, the weak correlation observed for some metals, like Pb, may also partly relate to these metals originating from sources other than the cultivated soil. In these cases, efforts to understand metal uptake from the soil will fail to provide a clear understanding of crop contamination mechanisms. Correlating the plant uptake to the phytoavailable fraction of the metal in the soil, rather than to the total concentration, may refine estimates (McLaughlin et al., 2000). But in addition to root uptake, plants can also receive metals through adherence or incorporation of fine soil or dust particles (McBride et al., 2014; Schreck et al., 2012; Uzu et al., 2010; Cary et al., 1994), and gaseous species can adhere to the plant surfaces for some metals (Rodushkin et al., 2007). These too will contribute to the contaminant load detected during chemical analysis. Particles may derive from the cultivated soil as well as other sources, such as road dust, local industrial emissions and long-range aerial transfer. Particle inclusion was highlighted as a significant transfer route of contaminants to urban crops in the recent review by Engel Di-Mauro (2021), and there are publications dealing with the analytical aspects of having exogenic material contaminating vegetable samples, as summarised by Wyttenbach and Tobler (2002). Despite this, contamination via soil or dust adherence remains an overlooked pathway in risk assessments, possibly due to a common belief – sometimes even explicitly stated - that particulates are efficiently removed when the produce is washed (Brown et al., 2016). In a recent study, however, we showed that the majority of most contaminant metals remain after common household washing (Augustsson et al., 2023). This implies that the recommendation to wash vegetables is often inadequate in effectively mitigating exposure to soil and particulate matter. We therefore agree with the sentiments of the US EPA (2013) in that the lack of published estimates of soil adherence on vegetables is a knowledge gap with potential impact on the validity of metal exposure and health risk assessments for urban agriculture. The interest in adherent particles arises from the probable disparity in oral bioaccessibility between metals associated with lithogenic material and metals taken up as free ions from the soil solution, where the latter become integrated in diverse biomolecules within the plant. It has been suggested that only a minor part of the adhering particles is bioavailable (Kozlov et al., 2000), and this is to be expected for lithogenic material that contain particles that are more or less resistant to digestion.

This paper is, to the best of our knowledge, the first whose objectives are to; (i) quantify the amount of soil/dust that adheres to crops from an urban environment, (ii) characterise the fraction of different contaminant metals (As, Ba, Cd, Co, Cr, Cu, Ni, Pb, Sb, Zn) associated with the adhering material, and (iii) evaluate and discuss the implications for risk assessments and urban gardening practices.

2. Materials & methods

In the first part of this study, our focus is on examining the presence of particles in/on outdoor leafy vegetables and their contribution to the analysed concentrations of metals in the vegetable samples. Here, data from an open-field urban farm located in southern Sweden was used to conduct the analysis. In a second part, to relate the metal contamination via adhering particles to root uptake, we included data on root uptake from an indoor cultivation experiment with soils from 20 different urban gardens across several European countries. By integrating the information obtained about both particle adherence and root uptake, we could calculate total concentrations of different metals in the vegetables from the 20 urban soils. Finally, we evaluate the average daily metal doses for vegetable consumers, taking into account the respective contributions from adhering particles and root uptake. Risks are evaluated using hazard quotients (HQs).

2.1. Outdoor site, sampling & vegetable washing

The analyses of particle adherence and its significance for different metals are based on analyses of vegetables, soil and dust that were collected from an open-field urban farm in Malmö (55°34′35″ N, 13°3′5″ E), Sweden, in 2020. Established in 1876, the Botildenborg farm has a longstanding history of cultivation. The cultivation is ecological, and no fossil-fueled machinery is used. Due to urban sprawl the farm is now an integral part of urban Malmö, situated in a densely populated area and adjacent to a heavily trafficked road with ca. 70,000 vehicles passing every day. Further details about the site, including a description of the soil's basic geochemistry, information on the vegetable sampling and weather conditions leading up to the sampling is provided by Augustsson et al. (2023). The research conducted in this study relies on analyses of 63 vegetable samples with paired soil samples; the latter collected from the root zone of the harvested plants. The vegetable samples encompassed 17 samples of lettuce (*Lactuca sativa*), 16 of chard (*Beta vulgaris* var. *cicla*), 15 of kale (*Brassica oleracea* var. sabellica) and 15 of parsley (*Petroselinum crispum*). In order to obtain an adequate amount of material for the chemical analyses, each sample was generated by combining material from multiple plants that grew in close proximity to each other. In addition to the soil and vegetable samples, 6 composite samples of dust material that could be assumed to reflect airborne material reasonably well were collected by placing tarpaulins on the ground next to where the vegetables grew, whereafter the dust deposited on them was carefully collected.

All collected vegetables were divided in two equivalent subsamples. One was analysed unwashed, and the other was washed in a way intended to mimic typical domestic preparation of vegetables; under running water and stirring in a plastic colander for 10 s.

2.2. Chemical analyses

All elemental concentrations were determined by ICP- sector field mass spectrometry (ICP-SFMS) at the commercial laboratory ALS Scandinavia in Luleå, Sweden, operated as described by Rodushkin et al. (2005) and with QC/QA procedures as further outlined by Augustsson et al. (2023). This laboratory specialises in high-quality analyses of elements at very low concentrations. All analyses are conducted in a cleanroom environment and adhere to strict protocols for quality control and quality assurance. Certified reference materials, internal standards, blanks, and a number of duplicate samples were included in all analysed sample batches. The elements analysed for this study are the potential metal pollutants As, Ba, Cd, Co, Cr, Cu, Ni, Pb, Sb, Zn, and the major soil constituents Ti, K, and Si.

The treatment of the vegetable samples is detailed in the paper by Augustsson et al. (2023). In summary, 0.5 g of dry material from each sample (N = 63) was digested with concentrated HNO3 and HF; 10.0 and 0.02 ml, respectively, thereby rendering total concentrations of elements associated with the plant material.

For this paper, the vegetable dataset was complemented by analysis of 63 soil samples that were paired with the vegetable samples, and 6 composite samples of dust material, which were dried and pulverised before further treatment. First, all soil and dust samples (dried at 50 °C) were extracted by Aqua regia, where 7.5 ml 9.46 M HCl and 2.5 ml 14.4 M HNO_3 were added to 0.5 g of sample material according to standardised, accredited lab protocols (SS-EN ISO 54321). This extraction renders pseudo-total concentrations of elements and was included to facilitate comparisons with other studies, given the widespread adoption of the Aqua Regia extraction protocol. The majority of our study's calculations and interpretations are, however, based on total concentrations. These were determined according to Sutliff-Johansson et al. (2021), following a procedure which includes two extractions. The first extraction is a strong acid digestion, where 0.2 g of sample material (dried at 50 °C) is dissolved in a mixture of 6.0 ml 14.4 M HNO_3, 2.0 ml 9.46 M HCl and 2.0 ml 48 % HF. The second decomposition method is an alkali fusion, where 0.105 g of the sample (dried at 105 °C) is fused with $LiBO_2$ at 1000 °C. The total concentration is determined by selecting the extraction with the highest concentration among the two conducted for each sample and metal.

2.3. Calculation of particle adherence

The amount of exogenic particles in/on a vegetable sample can be assessed from elements with no (or a negligible) uptake via plant roots and vascular systems, and whose presence can therefore be used as an indicator of soil inclusion, for example Al or Ti, Sc or Y (Caille et al., 2005; Cary et al., 1994; Cook et al., 2009; Engström et al., 2008). Titanium is used as the indicator element in this study. Based on the assumption that all detected Ti in the vegetable sample originates from lithogenic material (such as soil), and guided by the Ti concentration found in this material, the presence of Ti in plant samples can be interpreted as a quantity or percentage of lithogenic material. For example, assume that a plant is growing in a soil that contains 2000 mg Ti/kg and is found to contain 20 mg/Ti in above ground parts. From the above rationale, we can conclude that out of 1 kg of the analysed "vegetable sample" 0.01 kg (1 %) is actually comprised of adherent particles, assuming that all the adhered particles originate from the cultivated soil. The particle adherence (PA%, in % of the vegetables' dry weight), for both unwashed and washed vegetable samples, was achieved by dividing the total concentration of Ti in the vegetable (Ti_v) by the total Ti concentration in the exogenic source material, i.e., in the soil or dust source that the particles are assumed to come from (Ti_s):

$$PA\% = \frac{Ti_v}{Ti_s} \times 100 \tag{1}$$

However, the characterisation of Ti_s is obviously complicated in an outdoor environment where fine particles ending up in/on aboveground vegetation may originate from multiple sources. While the relative contribution from local soil and airborne dust (hereafter referred to only as 'dust') cannot be assessed from concentrations of indicator elements, the PA% for each vegetable sample was first calculated using the soil Ti as the benchmark and, second, using the average concentration of Ti in dust. That way the range of possible particle adherence outcomes should be covered.

The PA% is then used to calculate the contribution of adhering particles for specific elements (E%), together with the total concentration of the element in the soil (E_s) and in vegetable (E_v):

$$E\% = \frac{E_{s\times} PA\%}{E_v} \tag{2}$$

The means of particle adherence were compared among crop types using a Welch ANOVA, followed by the Games-Howell post hoc test. P-values were adjusted using Tukey's method.

2.4. Risk characterisation

As a screening measure of potential risks, hazard quotients (HQs) can be characterised. These relate the average daily dose (ADD) to a tolerable daily intake (TDI) – the latter defining the dose that can be consumed on a long-term basis without resulting in a deleterious health outcome.

$$HQ = \frac{ADD}{TDI} \tag{3}$$

We calculated HQs to assess potential risks following consumption of urban vegetables, with a particular focus on the fraction related to ingestion of adhering particles. Four consumer scenarios were considered, based on average and high (95th percentile) vegetable consumption in adults (40–69 years old) and children (3–6 years old).

The TDIs used in Eq. (3) are given in Table 1. Of these, the Pb TDI deserves some extra attention. The current understanding is that there is no threshold level below which Pb exposure can be considered safe. The value of 0.5 μg/kg/day in Table 1 is therefore a provisional TDI, selected for the calculations of this study. It corresponds to the lowest derived BMDL01 following dietary Pb exposure, with developmental neurotoxicity in children as the endpoint (EFSA, 2010).

To define the contribution of particulates to the ADD following vegetable consumption, data from the outdoor sampling at Botildenborg was used. The contribution from root uptake was based on an indoor cultivation experiment, where lettuce and chard were grown in soil from Botildenborg plus 19 other urban gardens in 6 cities across Europe: Malmö in Sweden, Copenhagen in Denmark, Madrid in Spain, Liverpool (Widnes) in the UK and Pribram City in the Czech Republic. Here, the aim was to evaluate metal uptake from urban soil matrices under conditions with minimal impact from aerial deposition and soil splash. The cultivation conditions, as well as analyses, are described by Qvarforth et al. (2022).

Metal concentrations in/on vegetables (lettuce and chard) were used to define ADD values to Eq. (3), separately calculated for each of the 20 soils. ADDs were calculated for the fraction taken up via the roots ($ADD_{vegetable}$), and the fraction associated with adhering soil/dust ($ADD_{particle}$). The sum of the two gives the total ADD, or ADD_{total}. Table 2 shows the input data for all scenarios.

$$ADD_{vegetable} = (C_{soil}{}^{*}BCF)^{*}Ig \tag{4}$$

$$ADD_{particle} = \left(Ig^{*}PA\%^{*}C_{particle}\right) \tag{5}$$

$$ADD_{total} = ADD_{particle} + ADD_{vegetable} \tag{6}$$

For the calculation of $ADD_{vegetable}$ (Eq. (4)), C_{soil} is the metal concentration in the different soils. The BCF value corresponds to the bioconcentration factor, or the ratio of the concentration of the metal in edible vegetable tissue to the concentration in the soil, as reported by (Qvarforth et al., 2022) and summarised in the supplementary material Table S2. The BCFs were soil- and crop-specific, i.e., given separately for each of the 20 soils and for lettuce and chard. Ig is the consumption of home-produced vegetables according to the US EPA Exposure factors handbook (US EPA, 2011). This variable expresses the consumption per kilogram of body weight, which is why body weight is not explicitly included in any of Eqs. (4)–(6).

For the calculation of $ADD_{particle}$ (Eq. (5)), PA% refers to the fraction of the vegetable dry weight that is composed of soil/dust, as obtained in this study. $C_{particle}$ is the concentration of the target metal in the exogenic source material. Here, only soil data (i.e., not dust) are used, to enable the inclusion of the soils from the Qvarfort et al. study (Qvarforth et al., 2022).

Table 1
Tolerable daily intake (TDI) values via oral exposure for non-cancer effects. The values for As, Cd, Ni and Pb are retrieved from the European Food Safety Authority (EFSA, 2009a; EFSA, 2009b; EFSA, 2010; EFSA, 2015). For Ba, Cr and Sb, the TDIs are from the US EPA IRIS database, and for Co, Cu and Zn they come from the Dutch National Institute of Public Health and the Environment, RIVM (Baars et al., 2001; Tiesjema and Baars, 2009). When there is a difference between the toxicological point of departure and the stated TDI this is typically due to the application of different uncertainty factors.

Metal	Toxicological point of departure	TDI (μg/kg/day)
As	$BMDL_{01}$ for ***hyperpigmentation, keratosis*** and possible ***vascular complications*** = 0.3 μg/kg/day	0.3
Ba	$BMDL_{05}$ for ***kidney toxicity*** = 63 mg/kg/day	200
Cd	***Kidney toxicity***, where an intake of 0.36 μg/kg/day renders >1 μg Cd/g creatinine in urine for a maximum of 5 % of the population.	0.36
Co	LOAEL for ***cardiomyopathy*** = 0.04 mg/kg/day	1.4
Cr (VI)	NOAEL for ***tissue accumulation*** = 2.5 mg/kg/day	3.0
Cu	NOAEL = 8 mg/kg/day (critical effect not specified)	140
Ni	$BMDL_{10}$ ***post-implantation loss*** = 1.3 mg/kg/day.	13
Pb	$BMDL_{01}$ for ***developmental neurotoxicity*** = 0.5 μg/kg/day	0.5
Sb	LOAEL for effects **longevity, blood glucose, and cholesterol** = 0.35 mg/kg/day	0.4
Zn	LOAEL for ***decrease in female erythrocyte superoxide dismutase (ESOD) activity*** = 1 mg/kg/day	500

3. Results & discussion

3.1. Particle adherence

The calculated fractions of adhering particles (PA%), as summarised in Table 3, are clearly influenced by the material that is used to

Table 2
Input data for the derivation of ADD. The Ig data from the US EPA exposure factors handbook is one of few sources that provide information on consumption of home-grown vegetables. The consumption of only lettuce or chard is, however, not reported. This means that the ADD calculations assume that the collective vegetable consumption contains the same amount of particles as the analysed leafy vegetables (lettuce and chard) do. This method of calculation is likely to result in an overestimation of particle intake.

Parameter	Unit	Value	Ref
Ig = consumption home-grown vegetables (fresh weight) per kilo bodyweight. These values were converted to dry weight in the ADD calculations.	g/kg/day	Average adult = 2.1 95%ile adult = 6.9 Average 3–6 yr old child = 2.5 95%ile child = 7.7	(US EPA, 2011)
PA% = fraction of the vegetable dry weight that comes from adhering particles.	–	0.5 % 1.0 %	This study
$\mathbf{C_{particle}} = \mathbf{C_{soil}}$ = average concentration of metal in the exogenic source material (soil) from Botildenborg and the 19 other urban allotment sites included in the risk characterisation.	mg/kg	See supplementary material, Table S1	(Qvarforth et al., 2022)
BCF = bioconcentration factors for lettuce and chard that were grown in soil from Botildenborg and the 19 other allotments included in the risk characterisation.	–	See supplementary material, Table S2	(Qvarforth et al., 2022)

Table 3
Fraction of adhering particles (PA%) in dry plant material, assessed from the total Ti concentration in soil and dust.

		PA% (from soil Ti)		PA% (from dust Ti)		Removed by washing
		Min - Max	Mean	Min - Max	Mean	
All crops	Unwashed	0.10–4.2 %	1.1 %	0.051–2.7 %	0.68 %	
	Washed	0.099–1.3 %	0.38 %	0.053–0.65 %	0.23 %	66 %
Lettuce	Unwashed	0.80–4.2 %	2.1 %	0.39–2.7 %	1.2 %	
	Washed	0.17–1.3 %	0.50 %	0.086–0.65 %	0.28 %	76 %
Chard	Unwashed	0.30–3.2 %	1.2 %	0.18–2.0 %	0.77 %	
	Washed	0.12–0.90 %	0.46 %	0.082–0.56 %	0.30 %	61 %
Kale	Unwashed	0.25–1.5 %	0.86 %	0.14–1.06 %	0.55 %	
	Washed	0.17–0.77 %	0.42 %	0.10–0.52 %	0.27 %	51 %
Parsley	Unwashed	0.10–0.60 %	0.23 %	0.051–0.31 %	0.13 %	
	Washed	0.10–0.30 %	0.14 %	0.053–0.16 %	0.079 %	39 %

characterise the exogenic source. The figures are nearly twice as high when assessed from soil Ti as compared to dust. Using soil Ti as the benchmark, the amount of adhering material on unwashed vegetables is assessed to be 1.1 % on average, with a min-max span of 0.10–4.2 % when considering all four crop types. Using Ti in dust lowers the average fraction to 0.68 % and narrows the min-max span to 0.051–2.7 %. These differences are explained by different abundances of Ti in soil compared to dust, where the former contained on average 1.80 g/kg and the latter 2.98 g/kg. Table 4 summarises the concentrations of Ti in soil and dust at the study site, together with concentrations of the other studied elements.

Table 3 further shows that washing the produce under running water reduces the adhering material by on average 66 %, which was indicated already by Augustsson et al. (2023). After washing, the vegetables contained on average 0.38 or 0.23 weight-% of fine soil/dust particles, depending again on whether calculations are based on Ti in soil or dust.

Already the study by Mitchell (1960) concluded that foliar particle contamination could hamper the interpretation of vegetable analyses, but few studies since have sought to quantify the amount of adhering soil/dust in a systematic way. The current references that we have found are based on two sources, of which both present their results for rinsed vegetables and on a dry weight basis. In the first, Cary et al. (1994) assessed the soil inclusion in leafy vegetables (spinach, lettuce, cabbage, turnip leaves and beet tops) to be on average 1.43 %, ranging from 0.10 to 4.88 %. Their calculations are based on Ti in paired soil samples, and both soil and vegetable samples were analysed after a digestion that included HF to obtain total concentrations. In the second, McBride et al. (2014) used Al in soil as the indicator element, and they report a range that is both narrower and lower: 0.019–0.027 %. They, however, applied weaker extractions; concentrated HNO_3 to the vegetable samples and Aqua Regia to the soil samples. Due to Al's association with highly weathering-resistant silicate minerals, only a fraction of the total pool is brought to solution by these extractions, especially in the one using only HNO_3. This may have resulted in an underestimation of soil adherence and could explain the lower percentages in the McBride et al. study. Taking all the above-presented data together, a reasonable yet conservative assumption (for risk assessors, etc.) is that leafy vegetables contain on average about 0.5 % of adhering particles after washing, and about 1 % as a maximum.

Looking at the different crops (Fig. S1 of the supplementary material), lettuce was found to contain the greatest quantities of adhering material prior to washing, followed by chard, kale and parsley. Since the particle removal was most efficient for lettuce, and decreased in the order chard>kale>parsley (Table 3), the differences in particle content between the different crops were smaller after washing. There were statistically significant differences between crop types, both for the unwashed (Welch ANOVA $F_{3,\,27.5} = 33.6$, $p < 0.0001$) and washed ($F_{3,\,27.5} = 21.6$, $p < 0.0001$) material. Before washing, significant differences in mean PA% were found between all vegetable types, except between chard and kale (Fig. S1a). After washing, significant differences were only seen between parsley and the other three crops (Fig. S1b). We prefer, however, to focus the results on “leafy vegetables” as a wider group rather than separately for different crops, since a vegetable-specific differentiation would suggest that there is more robust knowledge than is actually the case. For example, while the proportion of particles in our study was highest for lettuce and lowest for parsley (Fig. S1), the study by Schreck et al. (2012) found that the deposition of airborne lead was about twice as high on parsley as it was on lettuce (per kg), i.e. quite the opposite. So, while the particle entrapment per unit area will depend on differences in foliar morphology and physiology (Shahid et al., 2017), and for a given deposition the concentration will also depend on the ratio between foliar surface area and foliar volume, there are multiple varieties of most vegetables. Knowing what governs foliar particle contamination is therefore not sufficient to determine between crops (e.g., lettuce or parsley) which will catch the most airborne particles.

Table 4
Total concentrations of the analysed elements in soil and dust from Botildenborg. The right-hand column reports the average % of the total concentrations that was extracted with Aqua Regia (AR) to aid comparison with a wider range of studies. These values are rounded to the nearest five digits (except for Si).

	Total conc soil (N = 63)		Total conc dust (N = 6)		% of tot conc extracted with AR
	Min-Max	Mean	Min-Max	Mean	
Ti (g/kg)	1.5–2.3	1.80	1.67–4.72	2.98	20
K (g/kg)	16–19	17.2	5.59–17.9	11.8	20
Si (g/kg)	270–330	304	251–519	333	0.5
As (mg/kg)	<LOD–18	5.58	3.57–7.32	4.78	95
Ba (g/kg)	0.33–0.43	0.40	0.269–0.613	0.382	20
Cd (mg/kg)	0.23–0.49	0.344	0.177–0.406	0.274	100
Co (mg/kg)	4.2–6.2	5.25	4.34–12.0	7.23	90
Cr (mg/kg)	18–55	29	30–120	64	60
Cu (mg/kg)	18–33	23	22–150	74	100
Ni (mg/kg)	9.8–23	14	11–31	20	90
Pb (mg/kg)	20–52	24	20–65	44	95
Sb (mg/kg)	0.31–0.80	0.46	0.70–7.0	3.4	100
Zn (mg/kg)	68–110	88	90–320	210	100

3.2. Fraction of different elements associated with soil/dust particles

Fig. 1 shows how the exogenic material translates into fractions of different metals associated with adhering particles for washed vegetables, considering all crops. The figure is framed by the results for K and Si, two major soil constituents that help illustrate that the particulate contribution to an element's total concentration in plants is highly controlled by the element's availability for root uptake. Potassium shows how an efficient uptake and metabolism of an element by plants can make the contribution from adhering soil negligible, even for elements with a high abundance in the soil. Silicon, on the other hand, is the second most abundant element in natural soils, but also one of the most geochemically inert ones, with the vast majority associated with highly resistant silicate minerals. Although dissolved monosilicic acid is taken up by plants to some degree (Haynes, 2014; Kabata-Pendias, 2011), dicots (including all crops in this study) are low accumulators of Si (Kaur and Greger, 2019). Therefore, it is likely that the vast majority of the Si analysed in plant materials grown on open land originates from exogenic particles.

The average contribution of adhering particles to the vegetable content of individual metals (in % of the total concentration) increases, when assessed using soil Ti data, from the least affected contaminant metal element Cd (on average 0.33 %) via Zn (0.91 %), Cu (1.24 %), Ba (6.08 %), Sb (6.65 %), Ni (11.7 %), As (20.2 %), Co (20.2 %) and Pb (25.3 %) to Cr (33.4 %). The fraction of many contaminant metals is, however, higher when the assessment departs from the dust composition. This is explained by the fact that element-to-titanium ratios are higher in dust. This applies to Zn, Cu, Pb, Cr and – most distinctly – Sb. As a result, the average contribution of adhering particles to these elements' concentrations in vegetables increases when dust is used to define the exogenic source material; to 1.36 % (Zn), 2.48 % (Cu), 28.8 % (Pb), 44.4 % (Cr), and to as much as 30.0 % for Sb. The metals with a lower contribution from adhering particles when assessed from dust Ti were Cd (0.16 %), Ba (3.71 %), As (9.39 %) and Co (17.1 %). The result for Ni (10.7 %) was essentially the same. Boxplots for each separate crop are found in the supplementary material, Fig. S2a–d.

Most studies that point to the significance of soil adherence for the transfer of specific metals to urban vegetables base this conclusion on observed correlations between certain contaminant metals and elements such as Al or Ti in the analysed vegetables. A strong correlation to plant Al concentrations, leading to the conclusion of an exogenic source, have repeatedly been found for Pb in urban produce (Egendorf et al., 2021; McBride et al., 2014; Mosbaek et al., 1989; Paltseva et al., 2018) and on some occasions for As as well (Paltseva et al., 2018). While this kind of co-variation is a good indication of an exogenic source, it lacks the ability to make the kind of quantifications that we provide here. While our approach provides precise estimates, however, one must note that the calculated percentages are site-specific and should increase with increasing soil contamination levels, especially for the elements that are largely coming from adhering particles. The soil at Botildenborg contains, for example, only 23.6 mg Pb per kilo, compared to the several hundred mg/kg that have been presented for other urban gardens (Crispo et al., 2021; McBride et al., 2014; Mitchell et al., 2014; Moir and Thornton, 1989). Our percentage values for Pb thus probably represent low estimates. For comparison, Moir and Thornton (1989) estimated that 60–80 % of the Pb analysed in vegetable samples from contaminated sites in the UK was due to deposition of airborne particles. And in a study focusing on New York City community gardens, 55 % of the Pb in leafy vegetables was assessed to come from adhered soil (Spliethoff et al., 2016). The essential message here is that contamination via

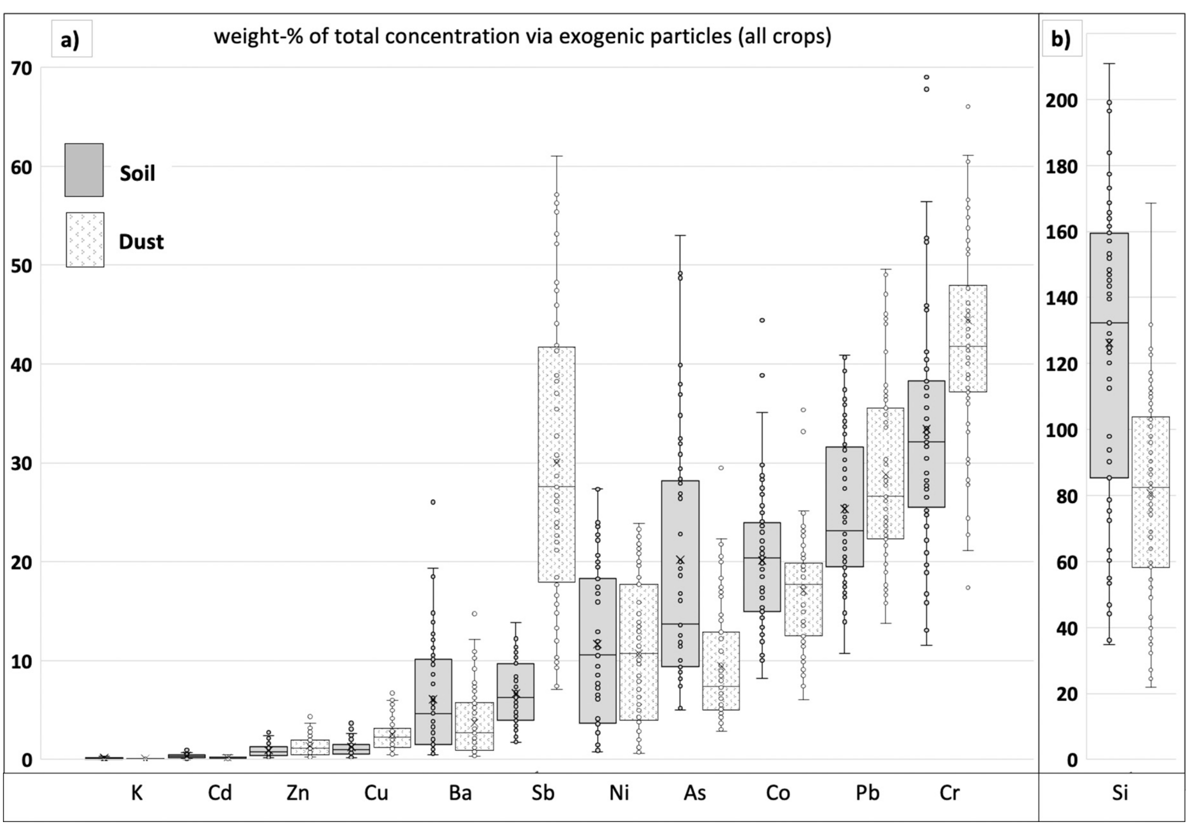

Fig. 1. The contribution (% of dry weight) of adhering particles for the total concentrations of a) potassium and the 10 contaminant metals, and b) silica, with an extended y axis. Results are presented for washed vegetables. The grey and dotted boxes show results based on the elemental composition of soil and dust, respectively. Each box is delimited by the 1st and 3rd quartile of the entire data, i.e. including results from all four studied crops, with the median value marked by the horisontal line and the mean value by the cross. The data points below or above the whiskers (outliers) are either >1.5 times the IQR below the lower or above the upper quartile. The corresponding figure for each of the studied crops separately can be found in the supplementary material, Fig. S1a–d.

particles may be as important as the root uptake for Pb, and probably also for metals such as As, Cr and Co if concentrations in surrounding topsoil are high enough.

What is not acknowledged in the previous literature about metal transfer to crops via foliar contamination, however, is that there are several metals at the other end of the scale as well, with a far lower association to adhering material, like Cd, Zn, Cu and Ba. For these elements, the exogenic fraction is typically well below 5 %. As the examples with K and Si in Fig. 1 show, and as indicated also by Augustsson et al. (2023), it is clear that metals which are readily taken up as free ions from the soil solution (like K, Cd and Zn) will accumulate within plant tissues to a degree that makes the contribution from any potential dust on the plant's surface of minor importance. Contamination via deposition of fine particles thus becomes an issue only for metals of low phytoaccessibility (like Si, Pb and Cr), for which the amount internalised in plant tissues will be lower than the amount associated with adhering particulates.

3.3. Source of particulate matter

It was recently suggested that soil splash is the main transfer route for particulate matter in/on urban vegetables, at least for Pb (Egendorf et al., 2021). If so, risks could be efficiently reduced if gardeners take action to make sure that the cultivation is carried out in clean soil. However, a clean soil will not protect crops from receiving air-borne particulates from adjacent and potentially more contaminated spaces. In this context, the Si results in Fig. 1b indicates that estimates of particle adherences departing from the cultivation soil alone seems to result in overestimations, i.e., the many data points > 100 % is not realistic. The results based only on dust material will, on the other hand, result in an underestimation. Therefore, both soil particles and airborne particles from adjacent areas must be relevant contributors to the total amount of particles in/on urban produce.

One study that points to the significance of air deposition for the contamination of urban produce is that of Saumel et al. (2012), which presented how the concentration of several metals in urban vegetables from Berlin, Germany, is higher near densely trafficked roads. Since paired soil samples were not analysed uncertainty exists as to whether the correlation to traffic density was caused by higher soil concentrations near trafficked road, which is to be expected (Clarke et al., 2015; Fakayode and Olu-Owolabi, 2003; Szwalec et al., 2020), or by airborne dust contamination. The authors themselves, however, propose atmospheric deposition as the main explanation. And this seems plausible because (i) some crops were grown in clean, store-bought potting compost, and (ii) Pb and Cr, which are less accessible to root uptake, were higher in vegetables grown near busy roads, whereas Cd and Cu, which are more easily absorbed from the soil solution, did not show the same correlation. The lack of a link to traffic proximity for Cd and Cu in vegetables was confirmed via personal communication with the authors. The significance of resuspended soil or dust for contaminant transfer to urban gardening plots is also highlighted by Clark et al. (2008) and Laidlaw et al. (2012), who showed that clean substrates may become contaminated over only a few years due to deposition of air-borne soil/dust from contaminated surroundings. So, despite emission controls, resuspension of soil and road dust keeps feeding significant amounts of Pb and other soil contaminants into urban air (Laidlaw and Filippelli, 2008), and the measures advocated within urban farming today do not adequately address the pollution impact that this material can have on crops.

3.4. Risk characterisation

With 1 % of the vegetable dry weight consisting of adhering particles, the average and high intake rates of vegetables for adults (Table 2) resulted in the consumption of 199 mg and 435 mg of particles per day, respectively. For 3–6 year old children the corresponding figures were 56 and 125 mg. With 0.5 % particle adherence, all these values are halved. Oral exposure to soil and dust particles is already a recognised exposure route in the US EPA's methodology as well as in most other generic risk models for contaminated land, even though its relationship to vegetable consumption is not considered. In this context, our results imply that consumption of home-grown vegetables is an important contributor to the intake of soil or dust particles. According to the US EPA exposure factors handbook (US EPA, 2011), the average total ingestion of soil particles and dust for adults and 3–6 year old children is 20 and 50 mg/day, respectively. Thus, significantly less than implied by our calculations. One plausible explanation could be that consumers of home-grown vegetables don't achieve the same particle removal when they wash their produce as is obtained in commercial cleaning processes. It is also likely that previously available estimates on soil ingestion are not representative for those who grow and eat their own produce, since the underlying studies are unlikely to include meaningful numbers of high-risk individuals, or smaller sub-populations such as consumers of self-grown vegetables.

The key studies available on soil ingestion (Calabrese and Stanek, 1995; Davis and Mirick, 2006; Davis et al., 1990; Hogan et al., 1998; Van Wïjnen et al., 1990), which are also the ones behind the US EPA recommendations, use concentrations of tracer elements in feces to estimate the oral intake of soil or dust, and they cover only short-duration periods and include only few individuals.

The output of the risk characterisation, the hazard quotients (HQs), are summarised for the 95%ile child in Fig. 2, with the total exposure via urban vegetables (ADD_{total}) distinguished from the exposure related only to adhering particles ($ADD_{particle}$). Since the 95%ile children were defined by the highest vegetable consumption per kilo body weight (Table 2), the highest HQs were calculated for this group. The HQs for the 95%ile adult, average child and average adult are consistently 89.6 %, 32.5 % and 27.3 % of the values for 95%ile children. Exact figures for all scenarios are specified in the supplementary material, Table S3a–d.

From the HQs based on ADD_{total} (Fig. 2a+c), it is clear that the metals that urban vegetable consumers run the greatest risk of ingesting in harmful amounts are Pb, Cd and As, in that order. This is consistent with the previous literature, which shows that vegetable consumption often results in ADDs above safe levels for these three metals (EFSA, 2009a; EFSA, 2009b; EFSA, 2010). However, as shown in the "% via PA" row at the bottom of Fig. 2, the adhering material constitutes only 0.5–0.9 % of the total ADD for Cd, while 39–52 % for Pb. When only exposure from adhering particulates is considered Fig. 2b+d, the maximum Cd HQ is only 0.09. The ingestion of Cd is thus problematic when the total intake is assessed, but neglectable when the contribution from adhering particles is isolated. The contribution of adhered particles to the total intake of As via urban vegetables is on average 28–43 %, and while not negligible is not sufficient to be associated with an appreciable risk (i.e., HQ < 1 when ingestion of adhered particles is assessed on its own). Lead is the only metal for which the contribution via adhering particles is frequently associated with a high probability of an intake above the TDI. In the worst calculated scenario, for 95%ile children and 1 % particle adherence, 13 out of 20 evaluated soils give Pb HQs > 1. For the group with the lowest assessed ADDs, i.e., adults with an average vegetable consumption, calculated HQs were >1 for 3 out of the 20 soils (Table S3d of the supplementary material). If one considers that Pb exposure also occurs through other pathways, even this seemingly modest proportion is concerning.

Lead is often the primary focus in discussions of urban metal contamination. It is often stated that the uptake of Pb from soils to plants is low, even from contaminated environments. By coincidence, we even penned those words in the introduction of this paper. It is true that the bioconcentration factors reported for Pb are lower than for most other metals, i.e., its concentration in plants is relatively low compared to its concentration in soil (Khan et al., 2015; Qvarforth et al., 2022; Swartjes et al., 2007). However, it is equally true that urban produce generally contains more Pb than commercial foodstuffs. According to the

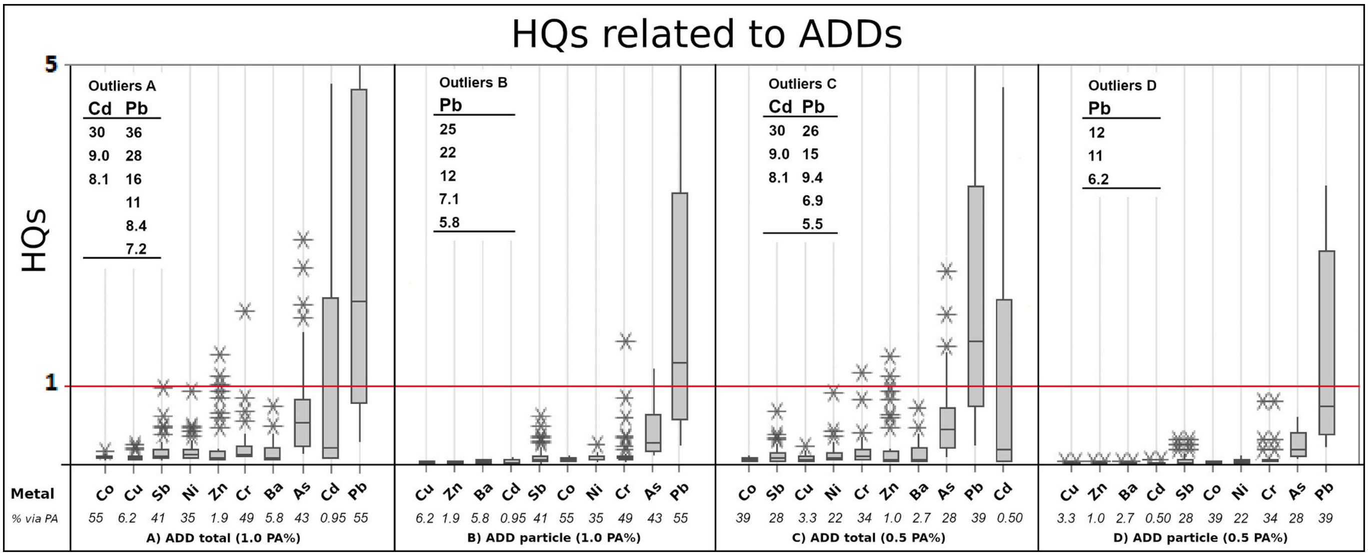

Fig. 2. Hazard quotients (HQs) according to Eq. (3), for 95%ile children. The four subfigures are based on TDIs from Table 1 and: a) ADD_{total} at 1 % particle adherence; b) $ADD_{particle}$ at 1 % particle adherence; c) ADD_{total} at 0.5 % particle adherence; d) $ADD_{particle}$ at 0.5 % particle adherence. The "% via PA" shows the fraction of the ADD_{total} that relates to the intake of particles.

European Food Safety Authority, for example, the median and 95th percentile concentration of Pb in commercially available leafy vegetables are 0.02 and 0.19 mg/kg (fresh weight), respectively. For comparison, the contemporary literature on urban vegetables reveals median and maximum values from about 0.06 to 0.48 mg/kg, and 0.59 to 2.77 mg/kg, respectively (Folens et al., 2017; McBride et al., 2014; Saumel et al., 2012; Warming et al., 2015). To enable this comparison, dry weight concentrations were converted to fresh weight equivalents. There are also several studies from urban environments, where the main issue has been about how different soil amendments can reduce levels of Pb in crops. Some of these are reviewed by Brown et al. (2016), and these studies also indicate that leafy vegetables from urban environments tend to contain relatively high concentrations of Pb. It is therefore not reasonable to claim that urban vegetables are equivalent to commercial ones in terms of their Pb content.

The question then is how the risks associated with this Pb should be assessed, and especially in the light of the high proportion that is related to the deposition of soil/dust particles. Glorennec et al. (2016), who studied Pb exposure via different exposure pathways, concluded that "*interpreting lead dietary intakes cannot be complete without perspective to soil and dust exposures*". For example, it is very probable that the oral bioaccessibility is lower for Pb in adhering exogenic material than it is for Pb that has been taken up as free ions and incorporated into plant biomolecules. The available scientific literature on the subject suggests that the bioaccessible fraction of Pb in urban soils and dust, referring to the fraction of the ingested amount that can be released in the gastrointestinal tract and subsequently absorbed into systemic circulation, ranges from 20 to 60 % (Attanayake et al., 2015; Defoe et al., 2014; Dehghani et al., 2018; Hu et al., 2011; Kelepertzis et al., 2021; Li et al., 2017; Ma et al., 2021; Najmeddin et al., 2018; Wang et al., 2021). Therefore it is clear that despite the conventional focus on Pb in soils as a potentially toxic element, there is much more to be studied and understood about this metal from the perspective of surface vegetation deposition as a pathway to risk.

4. Conclusions

This study demonstrates that particle adhesion to leafy vegetables forms a substantial contribution to human ingestion of soil/dust particles, and to the mass of several contaminant metals in/on vegetation. Even after regular washing, adhering particles can contribute significantly to human exposure of several toxic metals. Our finding that contamination via particles may be as important as the root uptake for several metals have implications for the development of more accurate models for risk assessments in urban areas as well as near superfund sites.

The conclusion regarding the likely significant importance of the adhesive material also emphasises the need for further studies regarding sources and pathways of urban vegetable contamination. These studies should include further quantification of the deposition of particulate matter on above-ground vegetables, investigation of the contribution from different sources (cultivation soil versus airborne dust), and examination of the significance of the adhering material for various metals. All of these aspects should be site-specific, and the results obtained from any one single urban farm, as the one in this study, should be cautiously extrapolated to other locations. Additionally, it is worth emphasising that the proportion of different metals linked to the adhesive material is most likely relatively lower at the studied site compared to many other urban areas, as the investigated location was not highly contaminated. Furthermore, a methodological uncertainty arises from the fact that the applied methodology cannot differentiate between particles from the cultivated soil and airborne dust, which differ in their composition. Further work is now needed to refine this method. Additionally, we did not examine geochemical fractionation of metals within adhered particles or their bioaccessibility; to fully assess risks to human health this information will be crucial.

Meanwhile, previously assumed best practices to mitigate risks, such as utilizing raised beds of uncontaminated soils in urban agriculture schemes and washing produce before consumption, may prove insufficient in fully mitigating risks associated with the consumption of vegetables grown in urban areas. To further minimise vegetable contamination, it is advisable to reduce foliar exposure to soil dust by implementing measures like fencing or other physical barriers. Green walls, for instance, could potentially serve as efficient filters. These measures can be combined with rooftop or elevated growing techniques to further prevent the contamination of edible crops. When coupled with thorough vegetation washing, the risks of metal transfer through airborne particulates can be significantly reduced.

CRediT authorship contribution statement

A. Augustsson: Conceptualization, Methodology, Formal analysis, Investigation, Writing – original draft, Writing – review & editing, Visualization, Supervision, Project administration, Funding acquisition. **M. Lundgren:** Methodology, Formal analysis, Investigation, Writing – review & editing, Visualization. **A. Qvarforth:** Formal analysis, Investigation, Visualization, Writing – review & editing. **E. Engström:** Conceptualization, Methodology, Writing – review & editing. **C. Paulukat:** Conceptualization, Methodology, Writing – review & editing. **I. Rodushkin:** Conceptualization, Methodology, Writing – review & editing. **E. Moreno-Jiménez:** Conceptualization, Writing – review & editing. **L. Beesley:** Conceptualization, Writing – review & editing. **L. Trakal:** Conceptualization, Writing – review & editing. **R.L. Hough:** Conceptualization, Methodology, Writing – review & editing.

Declaration of competing interest

The authors declare that they have no known competing financial interests or personal relationships that could have appeared to influence the work reported in this paper.

Data availability

Data will be made available on request.

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Appendix A. Supplementary data

Supplementary data to this article can be found online at https://doi.org/10.1016/j.scitotenv.2023.165633.

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