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Metal uptake by homegrown vegetables – The relative importance in human health risk assessments at contaminated sites

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ABSTRACT

Risk assessments of contaminated land often involve the use of generic bioconcentration factors (BCFs), which express contaminant concentrations in edible plant parts as a function of the concentration in soil, in order to assess the risks associated with consumption of homegrown vegetables. This study aimed to quantify variability in BCFs and evaluate the implications of this variability for human exposure assessments, focusing on cadmium (Cd) and lead (Pb) in lettuce and potatoes sampled around 22 contaminated glassworks sites. In addition, risks associated with measured Cd and Pb concentrations in soil and vegetable samples were characterized and a probabilistic exposure assessment was conducted to estimate the likelihood of local residents exceeding tolerable daily intakes. The results show that concentrations in vegetables were only moderately elevated despite high concentrations in soil, and most samples complied with applicable foodstuff legislation. Still, the daily intake of Cd (but not Pb) was assessed to exceed toxicological thresholds for about a fifth of the study population. Bioconcentration factors were found to vary more than indicated by previous studies, but decreasing BCFs with increasing metal concentrations in the soil can explain why the calculated exposure is only moderately affected by the choice of BCF value when generic soil guideline values are exceeded and the risk may be unacceptable.

1. Introduction

A variety of human activities have resulted in metal contamination of land, and for people who live near contaminated sites and grow their own vegetables, consumption of these vegetables may provide a significant addition to their metal intake from other sources (Hellström et al., 2007; Swartjes et al., 2007; Liu et al., 2013). Metal exposure following consumption of homegrown vegetables may also be of concern for human health following the current growth in urban gardening, as urban soils often have accumulated contaminants from a range of anthopogenic activities (Kessler, 2013).

It has been shown repeatedly that metal concentrations in cultivated crops increase with contamination level, and that crops grown in contaminated soils may fail to comply with foodstuff regulations (Khan et al., 2008a; Gaw et al., 2008; Douay et al., 2013). While several studies at contaminated sites/regions have concluded that exposure through vegetable consumption alone can lead to an intake above toxicological threshold levels (Cui et al., 2004; Zheng et al., 2007; Xu et al., 2013), others indicate that this exposure pathway alone generally does not provide an intake above what is deemed tolerable, even in areas with significant pollution (Dudka et al., 1996; Sipter et al., 2008; Cao et al., 2010; Wang et al., 2012; Beccaloni et al., 2013; Liu et al., 2013; Pelfrêne et al., 2013). Aside from inconsistent results, the ability to draw general conclusions about the risks associated with the consumption of homegrown vegetables around contaminated sites is limited by the fact that most previous exposure studies focusing on this exposure pathway are based on deterministic calculations, and often use mean or median values to characterize exposure factors (input variables), meaning that those individuals that are subjected to the highest exposure are often overlooked.

Routine risk assessments of contaminated land, for practical and economic reasons, rarely include site-specific analyses of contaminant concentrations in vegetables. Instead, a common procedure is to use generic bioconcentration factors (BCFs), which are multiplied with soil concentrations to render estimates of corresponding concentrations in edible vegetable parts (e.g. in Swedish EPA, 2009). The use of generic BCFs in site-specific risk assessments is, however, uncertain for several reasons.

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Firstly, there is high natural variability in metal uptake in plants. Uptake varies between crops, and depends largely on the geochemical properties and hydrological conditions of the soil (Dudka and Miller, 1999). There are, indeed, a number of empirical regression models that are relatively widely acknowledged where the plant trace element uptake is calculated after taking into account different physicochemical characteristics of the soil (e.g. Swartjes et al., 2007; Legind and Trapp, 2010; McLaughlin et al., 2011). Although these models can (possibly) reduce the uncertainty in the resulting BCF values, natural variability remains. Furthermore, the fact that such models require a relatively detailed site-specific characterization of input soil parameters, limits their practical usability. As many risk assessments in practice (by consultants etc.) only use BCF values to directly transform a soil concentration into a concentration in vegetables, without taking into account the influence of geochemical factors, this paper focuses on the use of BCF values in this way.

Secondly, there is considerable uncertainty associated with the adoption of many BCF values from the literature because the metal concentration in soil, which is a crucial variable when BCFs are calculated, is often determined after digestion of the soil with strong acids. It has, however, been established that the total concentration in soil often correlates poorly with the concentration in edible parts of plants (McLaughlin et al., 2000; Menzies et al., 2007; Liu et al., 2013). While the use of total or pseudo-total metal concentration in the soil is reasonable from a conservative precautionary perspective, which is what many parts of the risk assessment require, it does not lend itself as well when information about mobility or bioavailability is crucial, as is the case with plant uptake. This is particularly the case when uptake is described as a function only of the concentration in soil, without any correction for pH, content of major sorbent components etc.

Prior to, or parallel with, research on how metal uptake in vegetables can be estimated with greater precision, it is thus relevant to examine the extent to which metal BCF values calculated only from total concentrations in soil and vegtables can vary, and what impact this variability has on estimated human exposure. McKone and Ryan (1989) found that variability obtained in exposure calculations after food intake largely depends on the transfer of a soil concentration to a corresponding concentration in vegetables. Subsequently, scientists at the Dutch National Institute for Public Health and the Environment (RIVM) demonstrated the importance of BCF values for calculating human exposure at contaminated sites (Swartjes et al., 2007, 2013; Swartjes, 2009). However, to the best of our knowledge, a closer examination of the variability in BCF values and their importance for exposure assessments has not been conducted.

The overall aim of this study was to quantify the variability in BCFs and to evaluate the implications of this variability in human exposure assessments. This was accomplished by focusing on concentrations of cadmium (Cd) and lead (Pb) in two common homegrown crops (lettuce and potatoes) sampled around 22 contaminated glassworks sites in southeastern Sweden. The calculated BCF values were then used in a probabilistic exposure assessment to estimate the likelihood that consumption of homegrown vegetables would generate an exposure above the tolerable daily intake (TDI). The goal is to provide advice on how to deal with BCF values in risk assessments at contaminated sites.

2. Material and methods

2.1. Study area

Glass has been produced in Kalmar and Kronoberg Counties, southeastern Sweden, at more than 50 locations over a period spanning more than 300 years, and consequently there are currently many heavily polluted sites, with Cd and Pb being the two most common cationic metals (there is also significant arsenic pollution). Previous studies have estimated that there are ca. 3100 t of Pb and 30 t of Cd in the landfills and soils of the glassworks properties (Höglund et al., 2007). Most residents in the surrounding villages live in houses that were built after the establishment of the local glasswork, and excavated soil contaminated with glass waste was often used for land preparation. Glass manufacturing has historically also been associated with substantial emissions into the air, which contributed further to the contamination of nearby land (Larsson et al., 1999). A survey carried out in four glassworks villages in the region in 2012 revealed that many residents cultivate vegetables in their own gardens (Filipsson et al., 2013).

2.2. Collection of garden soil and vegetables

In the spring of 2013, letters were sent to all households (1130) within a 250 m range of 30 of the most heavily contaminated glassworks sites in the region. The maximum Cd concentration in soil measured at these sites, according to previous site investigations, is 550 mg/kd dry weight (dw), and the corresponding figure for Pb is 63,000 mg/kg (Höglund et al., 2007). For those householders who replied that they grew potatoes and/or lettuce we offered to come and take a sample from the vegetables and soil for metal analyses. Potato and lettuce were selected to represent a root vegetable and a stem/leaf vegetable that many people cultivate, since many exposure models distinguish between root- and leaf/stem vegetables. By doing so samples of different types of potato and lettuce were obtained. Moreover, the time between planting and harvesting varied. To collect vegetables that people have chosen themselves, rather than letting everyone cultivate under the same conditions, was considered appropriate for this study since the focus was on the variability in metal uptake and on concentrations in the two crops selected. In total 70 households, located around 22 glassworks sites, responded to the offer. Some households cultivated both potatoes and lettuce, while some cultivated only one or the other.

Sampling was carried out during July and August 2013. Lettuce and potato samples were taken from several plants ($N=3$–5) to provide composite samples, and soil from the upper 20 cm was collected close to the sampled plants and also pooled to composite samples. All soil samples can be considered representative of garden plot soil. Both soil and vegetable samples were collected in polyethylene bags and kept cool between sampling and analysis. In total 70 soil samples, 59 potato samples and 31 lettuce samples were gathered.

2.3. Additional vegetable cultivation in the laboratory

Eighty-eight additional composite soil samples were collected from 18 of the glassworks sites. Some samples were taken from garden plots (generally from households that had wanted to participate but were not cultivating any of the selected crops), and some were taken from common land adjacent to residential gardens. These soils were used for the cultivation of complementary potato and lettuce samples at Linnaeus University.

In preparation for planting the potato samples, ca. 10 L of well-blended soil from each composite sample was transferred to a bucket with drainage holes in the bottom. Two pre-germinated potatoes (Bintje, *Solanum tuberosum*) were added to each bucket, which were then placed in an open area outdoors and watered with sprinklers for 15 min every day. The mean temperature during the growing period was 14.4 °C, while maximum and minimum temperatures were 24.7 °C (day) and −1.0 °C (night),