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## Challenges in assessing the health risks of consuming vegetables in metal-contaminated environments

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### ABSTRACT

A great deal of research has been devoted to the characterization of metal exposure due to the consumption of vegetables from urban or industrialized areas. It may seem comforting that concentrations in crops, as well as estimated exposure levels, are often found to be below permissible limits. However, we show that even a moderate increase in metal accumulation in crops may result in a significant increase in exposure. We also highlight the importance of assessing exposure levels in relation to a regional baseline. We have analyzed metal (Pb, Cd, As) concentrations in nearly 700 samples from 23 different vegetables, fruits, berries and mushrooms, collected near 21 highly contaminated industrial sites and from reference sites. Metal concentrations generally complied with permissible levels in commercial food and only Pb showed overall higher concentrations around the contaminated sites. Nevertheless, probabilistic exposure assessments revealed that the exposure to all three metals was significantly higher in the population residing around the contaminated sites, for both low-, median- and high consumers. The exposure was about twice as high for Pb and Cd, and four to six times as high for As. Since vegetable consumption alone did not result in exposure above tolerable intakes, it would have been easy to conclude that there is no risk associated with consuming vegetables grown near the contaminated sites. However, when the increase in exposure is quantified, its potential significance is harder to dismiss – especially when considering that exposure via other routes may be elevated in a similar way.

### 1. Introduction

Consumption of food is a key route for the intake of many trace metals (EFSA, 2009a, 2009b, 2010; Tchounwou et al., 2012), and the safeguarding of acceptable exposure levels is thus partly linked to understanding when food safety may be at risk. In many parts of the world, including the EU and U.S., there are systems for quality control in place for commercial food, but not when it comes to homegrown items. According to Kessler (2013), approximately 35% of U.S. households grew food in 2012. Using Sweden as an example of an EU country, the corresponding figure was 43% (Swedish Board of Agriculture, 2014). Following the trend of increasing cultivation in private gardens, community gardens and school gardens (Kessler, 2013) there may be an increased risk of exposure to metals and other toxic compounds, as a significant portion of these gardens are located in urban areas and/or bordering old industrial sites.

On the one hand, it has been shown that metal concentrations in

homegrown vegetables from contaminated environments may exceed the levels permitted in commercial crops (Douay et al., 2013; Gaw et al., 2008; Khan et al., 2008a), and that metal exposure following consumption of these may result in an intake above toxicological reference values (Augustsson et al., 2015; Cui et al., 2004; Dziubanek et al., 2015; Hellstrom et al., 2007; Liu et al., 2013; Swartjes, 2009; Uddh-Soderberg et al., 2015; Xu et al., 2013; Zheng et al., 2007). On the other hand, others imply that the consumption of local vegetables alone generally does not provide an intake above what is deemed tolerable, even in areas which are known to be polluted (Beccaloni et al., 2013; Cao et al., 2010; Dudka et al., 1996; Liu et al., 2013; Pelfrene et al., 2013; Sipter et al., 2008). In summary, there are substantial gaps in our knowledge of the significance of the exposure pathway of homegrown vegetables and to what extent the risk is generally increased for individuals who reside in close proximity to contaminated sites and cultivate their own kitchen garden vegetables and/or consume wild berries or mushrooms. For this issue to be addressed, studies need to include

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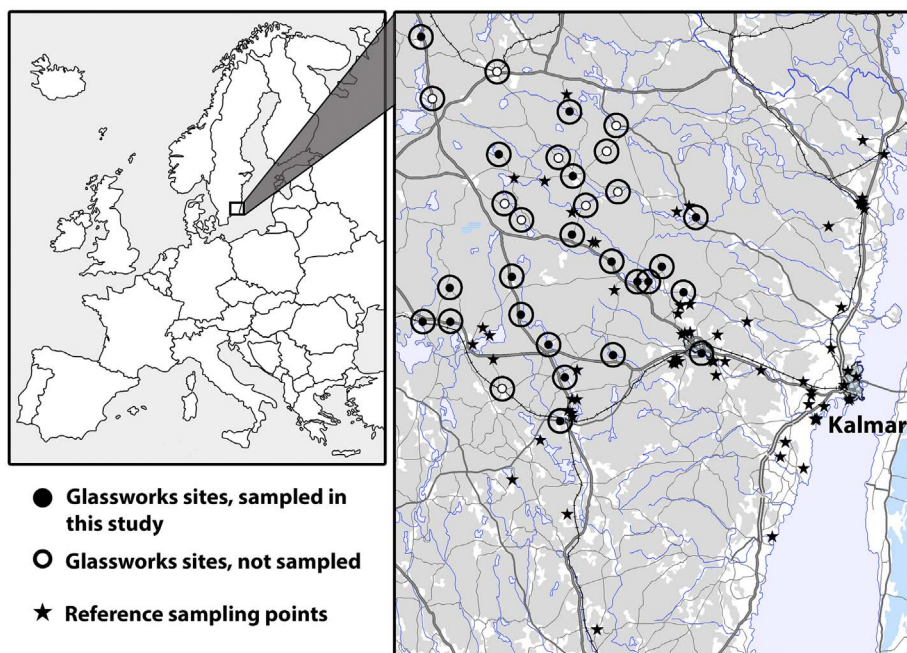


Fig. 1. The encircled black and white dots show the location of the major glassworks in the study region. The circles correspond to a 2000 m radius around each glassworks site. All reference sites (stars) were located outside the encircled areas.

all, or at least the majority, of the relevant crops frequently consumed in the study area. The intake of contaminants via consumption of local food around contaminated sites also has to be evaluated against representative reference levels, which reflect the intake for people in the same region who do not live in close proximity to contaminated sites. A local or regional reference level is particularly crucial when the suspected contaminants that are being studied are metals, as these natural elements can occur in high concentrations due to geological enrichments as well as anthropogenic activities.

One of the industrial activities that is known to lead to contamination of the surrounding environment is the manufacturing of glass, which is associated with the extensive use of various metal oxides. The potential exposure areas in focus in this study are centered around glassworks sites in southeastern Sweden. By analyzing metal concentrations in a large number of different crops, from both the glassworks villages and reference areas, the aim was to examine whether residents around the contaminated sites who consume homegrown vegetables and local berries or mushrooms are subjected to a higher exposure than the general population. The paper is limited to the three most toxic metals found at the glassworks sites: lead (Pb), cadmium (Cd) and arsenic (As).

## 2. Methods

### 2.1. Study region

All sampling areas (described below) are found in the area of Småland in southeastern Sweden, in the boreo-nemoral zone. Mean January and July temperatures are  $-2.0\text{ }^{\circ}\text{C}$  and  $17.0\text{ }^{\circ}\text{C}$ , respectively, and the mean annual precipitation is 700 mm, based on data from 2006 to 2015 (SMHI, 2016). The forests are dominated by spruce and pine plantations with podzolized soils. The soil overburden consists mainly of tills with a mineralogy that reflects the underlying granitoid bedrock. One important characteristic of the local geology with respect to this study is the occurrence of Pb-rich mineralizations (Andersson et al., 2014). Blueberries and lingonberries are common in the coniferous forests found in the study area, as are edible mushrooms such as chanterelles and various ceps. In more open terrain, e.g. clear-cut and pasture areas, wild raspberries and blackberries are often found. The Swedish Right of Public Access gives members of the public the right to

move freely in woods and fields, and to gather berries and mushrooms for personal consumption without restrictions. A survey among residents in the Swedish glassworks district shows that consumption of berries and mushrooms picked by residents is common, as is home gardening (Filipsson et al., 2017).

### 2.2. Potential exposure areas

Glass has been produced in the study area since the 18th century, in four small municipalities that together cover an area of  $3600\text{ km}^2$ . The full leaded crystal glass contains a minimum of 26 wt% lead oxide (Magnusson, 1971), but also other metals - such as As, Sb, Ba, Cd, Co, Cr, Cu and Zn - can be found in concentrations of several thousand ppm in the glass wares. That the health of glass factory workers has been impacted negatively has been documented in both the Swedish glass district (Wingren and Axelson, 1987; Wingren and Englander, 1990) and elsewhere (IARC, 1993). Although the significance of non-occupational exposure among local residents is not as well understood, recent investigations imply a general increase in the incidence of cancer in the Swedish glass region (Nyqvist et al., 2017). Effects among local residents may be attributed to the widespread contamination of both the glassworks properties and their surroundings, including private garden soils (Augustsson et al., 2015 + references in the Supplementary Information). In the most productive years, and before emission controls and waste regulations were introduced, hundreds of tonnes of waste materials were generated every year at the larger glass factories and deposited on site (Ewert and Höglund, 2012). In addition, emissions of dust and flue gases to the air were extensive (Larsson et al., 1999).

Currently, thirty-one glassworks sites in the four glassworks municipalities are found on the priority lists of the County Administrative Boards (white and black dots of Fig. 1). Twenty-one of these (black dots) were included in this study. These are essentially the sites that people live around. Since a key issue was to establish that metals at these sites occur in elevated concentrations as a result of the glassworks industries and not because of natural enrichments, available concentration data from the glassworks sites were compared to the background concentrations of natural till soil of the region. A summary is provided in Table 1, and a more thorough description of each site is given in the Supplementary Information.

Once contamination of the glassworks properties had been

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