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# An assessment of health risks associated with arsenic exposure via consumption of homegrown vegetables near contaminated glassworks sites

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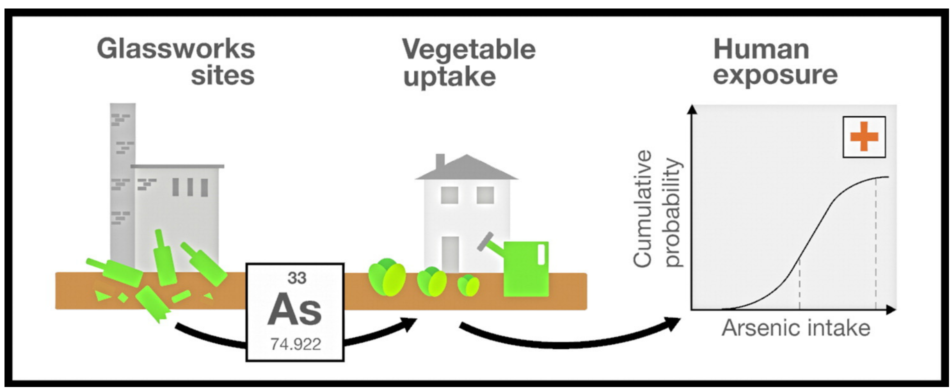
## HIGHLIGHTS

- High As concentrations were found in garden soils and homegrown vegetables near glassworks sites.
- As concentrations in soil and crops were highly correlated.
- Consumption of examined vegetables alone may lead to an unacceptable cancer risk.

## GRAPHICAL ABSTRACT

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## ABSTRACT

The health risk posed by arsenic in vegetables grown in private gardens near 22 contaminated glassworks sites was investigated in this study. Firstly, vegetable (lettuce and potato) and soil samples were collected and arsenic concentrations measured to characterize the arsenic uptake in the selected crops. Secondly, a probabilistic exposure assessment was conducted to estimate the average daily intake ($ADI_{veg}$), which was then evaluated against toxicological reference values by the calculation of hazard quotients (HQs) and cancer risks (CRs). The results show that elevated arsenic concentrations in residential garden soils are mirrored by elevated concentrations in vegetables, and that consumption of these vegetables alone may result in an unacceptable cancer risk; the calculated reasonable maximum exposure, for example, corresponded to a cancer incidence 20 times higher than the stated tolerance limit. However, the characterization of risk depends to a great extent on which toxicological reference value is used for comparison, as well as how the exposure is determined. Based on the assumptions made in the present study, the threshold levels for chronic non-carcinogenic or acute effects were not exceeded, but the cancer risks indicated highlight the need for further exposure studies, as dietary intake involves more

*Abbreviations:* ADI, Average daily intake; BCF, Bioconcentration factor; CDF, Cumulative distribution function; CR, Cancer risk; CTE, Central tendency exposure; HQ, Hazard quotient; RfD, Toxicological reference dose; RME, Reasonable maximum exposure; SF, Cancer slope factor.

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Probabilistic exposure assessment
Hazard quotient
Cancer risk

than just homegrown vegetables and total exposure is a function of more than just one exposure pathway. In addition, glassworks sites – and contaminated sites in general – contain multiple contaminants, affecting the final and total risk.



## 1. Introduction

Studies have shown that people living near arsenic-contaminated sites may be exposed to harmful quantities of the element, leading to adverse health effects (Lee et al., 2005; Liu et al., 2010; Rapant et al., 2006). One possible exposure pathway is through the consumption of homegrown vegetables (Bacigalupo and Hale, 2012; Smith et al., 2006).

Although it has been shown that crops cultivated near contaminated sites may have elevated concentrations of arsenic (Husaini et al., 2011; Lim et al., 2008; Roychowdhury et al., 2003), the solubility, and hence availability for plant uptake, in most natural soils is generally low because of a strong sorption to iron, manganese and aluminum oxyhydroxides, clays and organic matter (Bissen and Frimmel, 2003; De Vos and Tarvainen, 2006; McBride, 1994; Warren et al., 2003; Voigt et al., 1996). In surface soils, sorption is also favored as oxidized arsenic (as arsenate(V) ions; $H_2AsO_4^-/HAsO_4^{2-}$) is more efficiently sorbed than the reduced form (arsenite(III); $H_3AsO_3/H_2AsO_3^-$). Thus high total concentrations of arsenic do not necessarily equal a high bioavailability (for example uptake in plants). The toxic inorganic arsenate and arsenite species generally dominate the occurrence of arsenic in soil pore water and groundwater (Biswas et al., 2013). In plants, inorganic arsenic that has been taken up may be bio-transformed to less toxic organic arsenic forms (Peralta-Videa et al., 2009; Wei et al., 2015), but the amount of inorganic arsenic in vegetables is still significant, with the range reported in the literature being typically between 45 and 100% (Biswas et al., 2013; Chung et al., 2014; Norton et al., 2013; Signes-Pastor et al., 2008; Smith et al., 2006; Yost et al., 2004).

The information available on inorganic arsenic oral toxicity is extensive, and numerous effects of both non-carcinogenic and carcinogenic character have been described. Ingestion can result in fatigue, gastrointestinal symptoms, abnormal heart rhythm, bruising and impaired nerve function (ATSDR, 2007). The most characteristic effects of long-term oral exposure are hyperkeratosis and hyperpigmentation of the skin (ATSDR, 2007; IRIS, 1993). Other non-carcinogenic effects that may occur are peripheral vascular effects including cyanosis, gangrene and a condition known as “blackfoot disease”, as well as other cardio vascular effects such as high blood pressure and circulatory problems (ATSDR, 2007; IRIS, 1993). Oral exposure to inorganic arsenic has also been reported to increase the risk of cancer in the skin, liver, bladder and lungs (IARC, 2012; IRIS, 1998).

The aim of this study was, firstly, to investigate the degree to which arsenic is taken up by two common homegrown crops (potato and lettuce) that have been cultivated in garden soils around 22 heavily contaminated glassworks sites in south-eastern Sweden. Secondly, a probabilistic exposure assessment was conducted to estimate the health risk from arsenic due to the consumption of these vegetables. Arsenic, together with lead and cadmium, constitutes the major contaminant at these sites. This study is a follow-up of a previous study that focused on the two major cation-forming metals (Augustsson et al., 2015).

## 2. Material and methods

### 2.1. Study area

The study was conducted in Kalmar and Kronoberg counties in south-eastern Sweden. Since the 1700s, over 100 glassworks and smaller glass blowing sites have produced glass in the area (Nordström, 1999). Today only a fraction of these glassworks are still in use, but many sites remain contaminated by metal(loid)s (Fanger et al., 2004; Höglund et al., 2007). Earlier investigations at glassworks properties in the region revealed mean and maximum arsenic concentrations of 1000 and 13 000 mg/kg dry weight (dw) respectively (DGE Mark och Miljö, 2010; Golder Associates, 2008; Höglund et al., 2007; Mark and Vatten Ingenjörerna AB, 2005; Pettersson et al., 2011). The level of pollution becomes obvious when these figures are compared to, for example, the Swedish EPAs generic arsenic guideline for contaminated land, which is 10 mg/kg dw for residential areas and 25 mg/kg dw for non-residential areas (Swedish EPA, 2009). The 22 glassworks sites in the region that were selected for this study are presented in Fig. S1 in the supporting information.

### 2.2. Sampling

Since many exposure models used to assess contaminant intake via consumption of homegrown vegetables separate above- and below ground vegetables (Brand et al., 2007; Environment Agency UK, 2014; Swedish EPA, 2009), one common homegrown vegetable from each category was selected for this study; lettuce (leaf/stem vegetable) and potato (root vegetable). All households within a 250 m distance of the selected glassworks sites were invited to participate in the study. The criterion was that vegetables should have been cultivated in actual garden soil and not in bought soil. Additional soil samples were collected from common areas adjacent to existing gardens, where it would be possible to start future garden plots. Cultivation of lettuce (*Lactuca sativa*) and potato (*Solanum tuberosum*) was conducted in these soils at the Linnaeus University. The sampling at private gardens resulted in 40 lettuce samples and 68 potato samples and the additional cultivation at Linnaeus University resulted in 75 lettuce samples and 77 potato samples. In total 158 soil samples were collected. A more thorough description of the sampling and cultivation methods is presented by Augustsson et al. (2015).

### 2.3. Vegetable and soil sample preparation

The procedure for sample preparation and chemical analyses is also presented in more detail by Augustsson et al. (2015). In summary, dried and ground-up vegetable samples (edible parts) were digested in a microwave oven. Prior to the digestion, 0.50 g of the prepared lettuce material was mixed with 5.0 ml concentrated nitric acid ($HNO_3$) and 0.70 g of each potato sample was mixed with 3.0 ml $HNO_3$ and 2.0 ml concentrated hydrogen peroxide ($H_2O_2$) in high-pressure quartz vessels. After digestion, the samples were diluted to 25.0 ml with 18.2 MΩ/cm$^2$ Milli-Q$^{TM}$ water. Every tenth sample was prepared and analyzed in duplicates. This digestion gives total or very near total concentrations in vegetables.

The fresh soil samples were sieved to <2 mm before digestion according to U.S. EPA Method 3051A (U.S. EPA, 1986). 10.0 ml concentrated $HNO_3$ was added to 0.50 g of soil (fresh weight). As for the vegetable samples, every tenth sample was prepared in duplicates. A metal extraction procedure without hydrochloric acid (such as aqua regia) was selected because the chloride ions can interfere with analysis of arsenic on the graphite furnace (Martin et al., 1994; U.S. EPA, 1986). The digestion provides pseudo-total concentrations in the soil.