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Temporal risk assessment – 20th century Pb emissions to air and exposure via inhalation in the Swedish glass district

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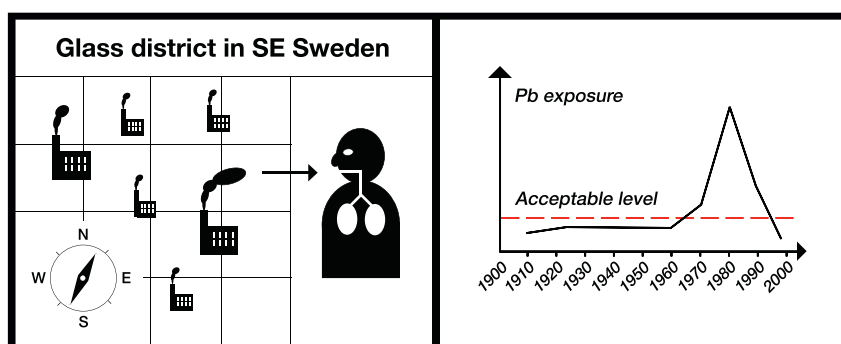
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HIGHLIGHTS

- Archives can provide data series that may be used to model historical emissions
- Pb emissions to air was modelled for the 20th century and used to assess exposure
- Around the studied glassworks, emissions and exposure peaked during 1970–1980
- By year 2000, the inhalation of Pb was comparable to that found in urban regions

GRAPHICAL ABSTRACT



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ABSTRACT

The objective of the present study was to assess historical emissions of Pb to air around a number of glassworks sites in southeastern Sweden, and the possible implications for human exposure. To do so, a four-step method was applied. First, emissions of Pb to air around 10 glassworks were modelled for the 20th century. Second, an assessment of the resulting exposure was made for a number of scenarios. Third, the number of people potentially exposed at different times was estimated, and fourth, measurements of “current” Pb concentrations in PM₁₀ material from four sites were conducted in 2019. The results show that the highest emissions, and exposures, occurred from 1970 to 1980. It coincides with the time period when the highest number of people resided in the villages. At this time, the average Pb concentration in air around the six largest factories was about 2.4 µg Pb/m³, i.e. 16 times the present US national ambient air quality standard (NAAQS) of 0.15 µg Pb/m³. By year 2000 the modelled average concentration had dropped to 0.05 µg Pb/m³, a level that is normal for urban regions today. The PM₁₀ measurements from 2019 indicate a further decline, now with a mean value of about 0.02 µg Pb/m³. Over the entire study period, inhalation hazard quotients (HQs) exceeded the dietary HQ by many orders of magnitude, indicating that inhalation has been the most prevalent exposure pathway in the past. At present, both pathways are judged to be associated with low exposures. Even if only roughly approximated, a picture of the historical exposure can increase our understanding of the connection between exposure and disease, and can be valuable when risks are to be communicated to residents near contaminated areas.

1. Introduction

An inconvenient legacy of past industrial activities is the contamination of soil and water. In the EU member states alone there are approximately 2.5 million potentially contaminated sites (Van Liedekerke et al., 2014),

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and in the US the corresponding number is 1.3 million (USEPA, 2022d). Residence near contaminated sites has repeatedly been shown to be a significant risk factor for negative health effects (Dolk et al., 1998; Hellström et al., 2007; Thomas et al., 2009), and the need to assess and manage environmental contamination and its effects are non-debatable. In this context, there are a number of risk assessment frameworks that are well established internationally (CCME, 2006; Lijzen et al., 2001; UKEA, 2006; USEPA, 1996). Within these frameworks, human health risks are assessed by comparing estimated exposure levels with toxicological reference values, e.g. tolerable exposure levels. However, when exposure levels are characterized from contemporary data, a drawback is that one then neglects the possibility that observed health effects may not derive from current exposure but rather from a historical situation that may deviate significantly from the present. In a recent article, Torén et al. (2022) highlighted the critical role of interpreting dose-response relationships and risks in a historical context. The same point of view was expressed by Coudon et al. (2021), who argued that a high incidence of diseases linked to e.g. air pollution - despite generally decreasing pollutant concentrations - may be expected for conditions where there is a long latency period between exposure and manifested illness, especially different forms of cancer. But while the understanding of past exposure can be key for a proper assessment and communication of current risks, or for the interpretation of elevated incidences of illness in certain populations, it is seldom discussed or put in relation to today's situation.

One of the most heavily contaminated regions in Sweden is the glassworks region in the south-eastern part of the country, where glass has been produced at >50 sites over the past 300 years (Nordström, 1999). Production flourished in the 20th century but has been minimal over the past two decades. The production of artistic and crystal glass has been associated with the use of several different metal oxides, in particular lead (Pb), which is a major component in crystal glass. The full-leaded crystal glass produced in the region contained as much as 24 weight-% pure Pb oxide (Council of the European Union, 1969; Magnusson, 1988), and a common feature of almost all glassworks sites today is thus exceptionally high concentrations of Pb in the soil. Mean concentrations are often found at thousands of mg per kg dry soil, and maximum concentrations in certain sub-volumes of landfilled material can even exceed 100,000 mg/kg (i.e. 10 % pure Pb). Other metal(loid)s are also frequently found in high concentrations in the soils of the glassworks villages, in particular arsenic (As), cadmium (Cd), antimony (Sb) and barium (Ba) (Hagner et al., 2018; Zhang et al., 1999). Of these inorganic As and Cd are classified as carcinogenic to humans (Group 1) by the International Agency for Research on Cancer (IARC) and Pb and inorganic Sb as probably carcinogenic (Group 2A). But the elements are also associated with a number of adverse non-cancer health effects, including kidney and bone damage, as well as neurological, cardiovascular and reproductive disturbances (EFSA, 2010; Nordberg et al., 2015; Rahman et al., 2009; Satarug and Moore, 2004). Recent studies imply that cancer incidences in the glassworks villages have been higher than the national average over the past decades (Helmfrid et al., 2019; Nyqvist et al., 2017), and while not investigated, there is a widespread concern among glassworks village residents that their various medical conditions are due to exposure to local contamination. In addition, the municipalities and county administrative boards, which regularly hold information meetings for residents in the region, express concern that people are now avoiding moving to the glassworks villages for fear of getting sick.

Substantial work has been undertaken to understand present-day exposure to the toxic metal(loid)s in the glassworks villages, both after consumption of local vegetables and drinking water from private wells. These previous studies imply a low contamination impact on local vegetables (Augustsson et al., 2018; Augustsson et al., 2015) as well as drinking water (Augustsson et al., 2016). As further described in these publications, the low solubility, leachability and phytoavailability of the soil metals can be explained by the geochemical conditions in the regional soils as well as within the glass waste dumps. Consumption of local food and drinking water is thus unlikely to explain the observed cancer incidences or other kinds of disease. Besides, the risks associated with the dietary exposure

pathways should not have been higher in the past. However, inhalation of airborne particles can also be a significant route of metal exposure (Cao et al., 2016; Glorennec et al., 2016), and this was probably significantly higher in the past than it is now. Today, the production of glass is limited to only a few sites. Where production is still carried out, emissions of toxic substances to air are now low following the substitution of the toxic raw materials. As a consequence, it is possible that any health impairments in the region are related to historical rather than contemporary exposure, and probably related to the inhalation of airborne particulate matter.

While the contemporary literature to date includes some examples that address temporal trends in exposure to airborne contaminants, these examples mainly concern occupational exposure, where the earliest figures date back to when systematic exposure measurements began (end of the 1970s to the mid-1980s). Since then a number of comprehensive reviews have been published, based on data from multiple longitudinal studies, which show reduced exposures to harmful substances within the industrial sector (Symanski et al., 1998a, 1998b; Kromhout and Vermeul, 2000; Creely et al., 2007; Koh et al., 2014). In addition, concentrations of the most commonly monitored air contaminants in urban areas (carbon monoxide, nitrogen and sulfur dioxide, ozone, lead and particulate matter (PM₁₀ and PM_{2.5})) have decreased over the past decades in most cities in developed countries (Carugno et al., 2017; Coudon et al., 2021; Li et al., 2018; Olstrup et al., 2018; USEPA, 2022c). So not everything was better before.

However, despite a thorough literature search using keywords such as “inhalation”, “exposure”, “metal”, “PM10”, “archive”, “historical”, “temporal”, “time”, “past”, “century” and “decade”, we have not managed to find anything about temporal trends in exposure and/or health risks for residents living near contaminated sites. The study presented in this paper uses an approach with historical data to assess changes in inhalation exposure that, to the best of our knowledge, has never been used before. It gives new perspectives and highlights the need for further studies on the link between present-day illness and historical exposure.

The aim of the study is to put glassworks village residents' risks of exposure to air-born contamination, focusing on Pb, into a temporal perspective. Annual amounts of Pb released to the air over the 20th century (1901–2000) were first estimated for a selected number of glassworks sites. Second, these amounts were converted into Pb concentrations in air for four different residence scenarios. Present-day concentrations of Pb in air were measured for comparison. Third, the non-cancer exposure risks via inhalation were expressed as hazard quotients (HQs) for 10-year intervals over the 20th century, for each site and scenario, and compared to present-day risks via both inhalation and dietary exposure. We hypothesize that the historical exposure via inhalation of Pb has been associated with a higher risk than the one assessed after both inhalation and dietary exposure today. Finally, variations in historical inhalation exposure were related to the past century's changes in population size around the sites.

2. Material and methods

2.1. Study area and focus sites

Only a few of the old glassworks are still active, but no <36 sites are still found on the county administrative boards' lists of prioritized polluted areas due to the long-lasting and extensive use and release of Pb and other metal (loid) contaminants (Kalmar county administrative board, 2019; Kronoberg county administrative board, 2019). Depending on the type of glass produced, different substances were added to the batches. Lead oxide, for instance, was used as a stabilizer in the production of crystal glass (Magnusson, 1988). It has been estimated that approximately 3 % of this lead was emitted to ambient air at the temperatures applied when melting the batches (Kajiser and Fogelberg, 1922).

For this study, ten glassworks were selected for the assessment of historical Pb emissions to air, focusing on the 20th century (1901–2000) to cover the 100 years of most intense production. The glassworks were chosen to cover the variability in production type and produced amounts that applied to the glassworks of the region during the 1900s (Table 1). Bottles, cans and

Table 1

Operating period, glass types, and total volume of glass produced for the ten glass works selected for this study.

Glass work	Active years	Municipality	Main type of production	Amount of glass produced between 1900 and 2000 (tonnes)
Åfors ^a	1876–2012	Emmaboda	Household and ornamental glass	12,100
Alsterbro	1871–1969	Nybro	Bottles, household glass and pressed glass	16,600
Bergdala ^a	1889–	Hovmantorp	Household and ornamental glass	9400
Boda	1864–2003	Emmaboda	Household and ornamental glass	25,500
Emmaboda	1919–1930, 1934–1978	Emmaboda	Window glass	352,000
Gadderås	1874–1967	Nybro	Bottles, window glass, lightbulbs and household glass	19,000
Kosta	1741–	Lessebo	All types of glass	141,000
Målerås ^a	1890–1904, 1917–1922, 1924–	Nybro	Household and ornamental glass, medical glass, bottles, technical glass	41,200
Orrefors ^a	1898–2013	Nybro	Household and ornamental glass, cans, window glass and lamps	133,000
Pukeberg	1871–1995	Nybro	Household and ornamental glass, technical glass and lamps	62,700

^a Glassworks where complementary measurements of Pb in airborne PM₁₀ were made in 2019.

window glass were made of cheaper raw materials, without Pb or other heavy metals added. Most household glass and pressed glass were also relatively low in metals. Ornamental glass, lamps, crystal glass, medical and technical glass, on the other hand, could contain a variety of different metals, both as ingredients in the glass for certain characteristics and for coloration.

Among the sites examined in this study, Emmaboda is the only one without clearly elevated Pb concentrations, with an average (arithmetic mean) of 36 mg Pb per kg soil. For the remaining sites, average soil Pb concentrations increase in the following order: 260 mg/kg (Pukeberg) < 870 mg/kg (Gadderås) < 1990 mg/kg (Alsterbro) < 4030 mg/kg (Boda) < 4030 mg/kg (Åfors) < 4100 mg/kg (Åfors) < 4460 (Kosta) < 9760 (Orrefors) < 13,000 (Bergdala) < 31,000 (Målerås). These concentrations can be compared to the average regional background concentration of about 25 mg Pb per kg in natural C horizon till soil (Swedish Geological Survey, data delivery no 2015:0244). The relatively high background concentrations are explained by the occurrence of natural Pb-rich mineralizations in south-eastern Sweden.

2.2. Historical Pb emissions to air

An important prerequisite for estimating historical emissions is that the glass industry's manufacturing methods have not changed significantly over time and that the same is true for the emissions to air. The 20th century emissions of Pb to air (kg/yr) were calculated separately for each of the ten glassworks according to the method which is described in more detail by Larsson et al. (1999). Following this methodology, a model was set up where a conversion factor was calculated between measured annual Pb emissions to air and the reported volumes of used raw material (lead oxide, Pb₃O₄). Measurements of Pb emissions to air were regularly conducted at Kosta and Orrefors during the period 1971–1980, prior to the installation of dust filters at these facilities. Data from these two sites were retrieved from regional archives and used in the model parametrisation. Data on raw material (lead oxide) were collected from Swedish national archives and compiled for every second year during the evaluated century, then averaged for the missing years. The conversion factor was used to estimate historical emissions of annual Pb to air (in kg) from the 10 glassworks. For years where data on reported raw materials were missing, the production volume of different types of glass were used as a predictive value in a regression analysis to estimate the emissions. The results were validated by four measurements taken in the 1990s at three different glassworks (Boda, Åfors and Pukeberg).

2.3. Conversion of historical Pb emissions (kg/yr) to annual average concentrations in air (µg Pb/m³)

The greater part of the emitted Pb was assessed to be inhalable (<10 µm), which means that total emissions to air were not adjusted before

calculating inhalable concentrations. The rationale is that primary dust particles emitted from glass melting furnaces are very fine grained; typically <0.5 µm according to Stockham (1971). While these small particles tend to agglomerate to form secondary particles, they are still seldom larger than 10 µm in diameter (Kasper et al., 2007).

The annual amount of Pb to air via flue gas emissions (E; kg/yr) was converted into an estimated annual average concentration in outdoor air (C_{Pb}; µg Pb/m³) through Eq. (1), where RT is the estimated average residence time (yrs) of glassworks-emitted particles and V_a is the air volume (m³) in which these particles are distributed. Compared to other dispersion models available to simulate the transport of particulate metals in air from point sources (Daggupaty et al., 2008; Khaniabadi et al., 2018; USEPA, 2022a), Eq. (1) is a simplification. More advanced simulations would have been necessary if we did not have actual measurement data on deposition patterns, i.e. information on the distance within which the emissions from the glassworks factories are deposited (see further description below). Now that such information exists, we can achieve the purpose of this study without advanced atmospheric dispersion modeling.

$$C_{Pb} = (E * RT) / V_a \quad (1)$$

The average particle residence time (RT) is of course fundamental for assessing the contaminant concentrations in air around a point source, where an increase or decrease in this variable increase or decrease the particle-bound contaminant concentration proportionally. Particle residence times in outdoor air can, however, vary by many orders of magnitude and are affected by numerous factors (Esmen and Corn, 1971; Rossi et al., 1995). A site and situation specific assessment is thus in most cases more robust than the application of RT data from the literature. In the study region, results from previous moss surveys imply that the majority of the Pb deposition occurs close to the glasswork factories. Moss Pb concentrations decrease exponentially with distance and reach background levels at approximately 1000 m distance (Carlsson, 1984). This thus suggests that the vast majority of the deposition occurs within the first km. By assuming so, and further considering the average wind speed in the area, we arrive at a RT of 9,6E-6 yr. According to the Swedish Meteorological and Hydrological Institute, the average wind speed in the region was approximately 3.3 m/s during the time period 1961–1990 (Alexandersson, 2006).

The affected air volume (V_a) was calculated by a simplified assumption of combustion particles being distributed in a cone-shaped volume of air, with a base radius of 1000 m. An average chimney height of 30 m was selected after consulting various reports and representatives from the regional glass industry.

We took into account that the emitted Pb was not evenly distributed and deposited and therefore considered 4 different residence scenarios in this work, for which different aerial Pb loads applied. Since the moss surveys referred to above (Carlsson, 1984) imply that approximately 65 % of the Pb deposition occurs within 500 m distance and the remaining within the

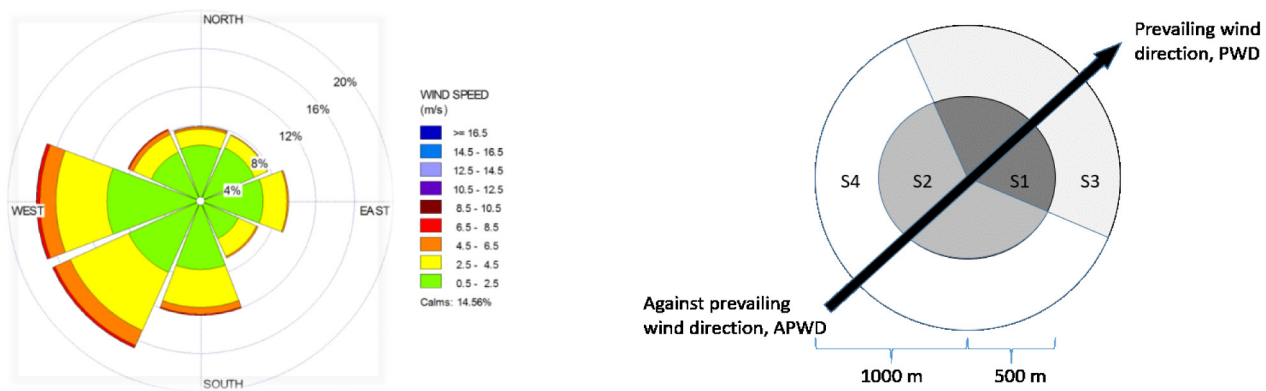


Fig. 1. a Wind rose showing prevailing wind, b. Four exposure scenarios (S1–S4) based on directions in the glass works region (SMHI, 2017) prevailing wind direction and distance to glass works.

next 500 m, a first distinction was made between an inner (0–500 m distance from factory) and an outer area (500–1000 m distance). Of the annual deposition, 65 % was assigned to the inner area and 35 % to the outer. Second, we considered that the wind in the region blows mainly from south, southwest and west (3/8 of a full circle) approximately 50 % of the time, thus resulting in the majority of deposition towards the north, northeast and east (Fig. 1a). The remaining 50 % of the time the wind comes from the remaining 5/8 directions. The four residence scenarios are illustrated in Fig. 1b, and can be summarized (with the approximate air volume (V_a) for each scenario is in the parenthesis):

Scenario 1 (S1). : In prevailing wind direction, 0–500 m ($V_a = 2.94E6 \text{ m}^3$).

Scenario 2 (S2). : Against prevailing wind direction, 0–500 m ($V_a = 4.91E6 \text{ m}^3$).

Scenario 3 (S3). : In prevailing direction, 500–1000 m ($V_a = 8.83E6 \text{ m}^3$).

Scenario 4 (S4). : Against prevailing direction, 500–1000 m ($V_a = 14.7E6 \text{ m}^3$).

The differences in V_a means that each residence scenario was assigned its own estimation of C_{Pb} when subjected to Eq. (1).

2.4. Risk assessment and calculation of hazard quotients

For metals that can both cause cancer and result in adverse non-cancer effects, common practice is to assess the cancer risks and non-cancer risks separately, usually by applying the US EPA methodology (USEPA, 1994; USEPA, 2009; USEPA, 2011) and their proposed toxicity values. For inhalation risks these toxicity values are expressed as reference concentrations for non-cancer effects (RfCs), and as inhalation unit risks for cancer effects (IURs). Several studies have assessed the cancer as well as non-cancer risks for metals (including Pb) after inhalation (Ghasemi et al., 2020; Goudarzi et al., 2018; Megido et al., 2017; Ramírez et al., 2020; Roy et al., 2019). The situation for Pb is, however, a bit unusual. While the non-cancer health effects associated with Pb exposure are well known, some of these may occur at blood Pb levels so low as to be essentially without the kind of threshold that is needed to set a reference dose or reference concentration. That is why no RfC for Pb has been proposed under e.g. the US EPA Integrated Risk Information System (IRIS) program (USEPA, 2022b), and also why both the European Food Safety Authority and World Health Organization have withdrawn their previous values of tolerable exposure levels for Pb with regards to non-cancer effects (EFSA, 2010; WHO, 2010). When it comes to cancer risks, while inorganic lead is classified as probably carcinogenic to humans (Group 2A), the International Agency for Research on Cancer (IARC) concludes in its assessment that there is limited evidence in humans for the carcinogenicity of inorganic

Pb. Consequently, there is also no IUR set for Pb under the IRIS program. An IUR value for Pb ($1.2E-5$) can be obtained from the Office of Environmental Health Hazard Assessment (OEHHA, 2022), and this is the reference value that is often used when cancer risks via inhalation is assessed (Megido et al., 2017; Ramírez et al., 2020; Roy et al., 2019). However, when comparing the IUR value of Pb to the IURs of e.g. As ($3.3E-3$ according to OEHHA) and Cd ($1.8E-3$ in IRIS), the cancer risk is still relatively low for Pb.

Quantifying risks of Pb exposure is thus problematic, but presenting an easily interpreted risk measure, such as a hazard quotient (HQ), can still be a pedagogical tool. Since non-cancer effects are especially critical for Pb, we only assessed how the historical changes in aerial Pb concentrations have affected the risks for glassworks village residents through the calculation of hazard quotients (HQs) for non-cancer risks according to Eq. (2). For inhalation, this hazard quotient relates measured or modelled exposure concentrations in ambient air (EC, in e.g. $\mu\text{g}/\text{m}^3$) to a health-based reference concentration (RfC). The RfC represents the highest tolerable concentration in inhalable air, at which no observable adverse effects are expected.

$$\text{HQ} = \text{EC}/\text{RfC} \quad (2)$$

Since there is no RfC concentration established for Pb, we use the US national ambient air quality standard (NAAQS) as an approximation (USEPA, 2008). The NAAQS for Pb is set to $0.15 \mu\text{g Pb}/\text{m}^3$. The US EPA, which has provided several guidance documents on assessing the risks of inhalable contaminants (USEPA, 1994; USEPA, 2009), proposes that the exposure concentration (EC) is set after adjusting the measured contaminant concentration in air to the fraction of the time during which the exposure occurs. In this study, where people in a worst-case scenario may be nearly constantly exposed due to their residence being close to the point source, we don't reduce the EC values by assuming exposure during only a certain number of hours per day, or days per year. Instead, we consider that only a limited fraction of the residents' time is spent outdoors, and that Pb concentrations in indoor air are lower than in outdoor air. The EC values used to calculate HQs in this study were therefore calculated according to Eq. (3):

$$\text{EC} = ((C_{Pb} * f_{\text{out}}) + (C_{Pb} * f_{\text{in}} * f_{\text{din}})) \quad (3)$$

The variable of Eq. (3) that differed between the four residence scenarios was the concentration of airborne Pb in outdoor air (C_{Pb} ; $\mu\text{g Pb}/\text{m}^3$). The remaining variables were kept constant for all the evaluated scenarios. Here, f_{out} and f_{in} are the average fractions of the time spent outdoors and indoors per day. Data on average daily outdoor time (192 min) for adults aged 16–64 years were retrieved from the US EPA Exposure Factors Handbook (USEPA, 2011). f_{din} is the fraction of the airborne, inhalable concentration found outside that can also be expected inside houses. An assumption of 50 % ($f_{\text{din}} = 0.5$) was made, according to the recommendation in the Swedish EPAs' generic risk assessment model for contaminated sites (Swedish Environmental Protection Agency, 2009).

2.5. Variations in population size around the glassworks

To assess the number of people potentially exposed to harmful levels of airborne Pb over the 20th century, historical maps and aerial photographs from the Swedish National Land Survey were reviewed. We counted the number of houses within 1000 m from the glassworks sites included in the study on the maps/photographs, and multiplied this figure by the average number of people in Swedish households during each time period (SCB, 1999; SCB, 2020a; SCB, 2020b). Since the first reliable maps from the region are from around 1940, this was the first time considered (economic maps from 1941 to 1950). Thereafter new estimates were produced with 20-year intervals; thus, again for 1960 (photos from 1960), 1980 (economic maps from 1978 to 1982) and 2000 (photographs from 2002 to 2003).

2.6. Present-day concentration of Pb in air for comparison

The historical Pb concentrations in air, estimated according to the methodology described above, were compared to present-day concentrations of Pb associated with airborne particles. Such particles were retrieved through an air sampling campaign around 4 glassworks, of which two are still operational (Målerås, Bergdala) and two are closed (Orrefors, Åfors). In the former case, airborne particles could derive both from production emissions and after resuspension of soil/dust from the ground. Electricity was needed for the sampling, and thus it was not possible to sample all ten glassworks for practical reasons. Sampling of airborne particles <10 µm (PM₁₀) was conducted with two MicroVol 1100 low volume air samplers (EcoTech) to retrieve 2 samples from each site. It was performed during April–June in 2019, with a sample duration of 2 weeks at each site. The particulates were captured on pre-weighed 47 mm mixed cellulose ester membrane filters using a flow of 3 L/min. The filters were thereafter acclimatized in a room with a controlled temperature and humidity (24 °C and 55 % ± 2 %) for 48 h before they were gravimetrically analyzed using a precision microbalance with a reading precision of 10 µg (Satorius Micro MC5 P). Dust and filters were digested using HNO₃ in a microwave before analysis of elemental composition by inductively-coupled plasma mass spectrometry (ICP-MS, iCAP™ RQ: Thermo Fisher Scientific, Waltham, MA, USA) (NIOSH, 2014).

Samples were analyzed against a multi-element standard (Spectrascan). An internal standard containing scandium, rhodium, indium, and bismuth with concentration of 1 ppm as well as a reference sample from the Norwegian National Institute of Occupational Health (STAMI) were used for quality control.

3. Results and discussion

3.1. Modelled 20th century Pb emissions to air

The modelled Pb emissions from the 10 glassworks, presented for 10-year periods in Table 2, amounted to approximately 466,000 kg over the entire 20th century. About 84 % originated from the three largest glassworks, those in Kosta, Orrefors and Boda. After normalization of the data

(Fig. 2) one can see that Pb emissions were fairly stable during the first 5 decades of the century and increased from the middle of the century until the 1980s. The increase occurred mainly at the five largest glassworks, namely Kosta, Orrefors, Boda, Pukeberg and Åfors. Comparing the time period 1941–50 and 1971–80 for these five sites, the Pb emissions increased by a factor of 6.4 - from a total of 20.5 t to 132.0 t. The emissions from the five smaller sites, on the other hand, only increased from 3.3 t altogether during 1941–50 to 4.7 t in 1971–80.

After 1980, emissions clearly decreased. This was partly due to lower production volumes, and partly due to adaptations for emission control. Gas flue filters were installed at some sites, but the most common action was to abandon the use of lead oxide as a raw material. The shut-down of several facilities also contributed to the lower emissions. At Alsterbro, Gadderås and Emmaboda, for example, production ceased already in 1969, 1967 and 1978, respectively. The factories with the highest Pb emissions, however, remained active towards the end of the century. As a consequence of these multiple contributing factors, the total estimated Pb emissions from the 7 sites that were still active in 1991–2000 were only 10.8 t for the full period (Table 2), and emissions in year 2000 were only about 10 % of those in 1991.

The Pb emissions in 1991 amounted to approximately 1.8 t according to our calculations. This figure appears realistic when compared to the one assessed by Statistics Sweden: 1.3 t of Pb to ambient air from Swedish glassworks in 1990 (SCB, 2022). With 1.8 t of Pb emitted within a 1000 m radius from 7 sites (over a total area of 22 km²), the average Pb deposition around these glassworks equalled 81 kg/km²/yr in 1991. For the country as a whole, with a total of 357 t Pb emitted to air at that time when considering all sources of Pb emissions to air and not only glassworks (SCB, 2022), the average deposition was about an order of magnitude lower (0.79 kg/km²/yr).

During 1971–1980, when glassworks-related emissions peaked (Table 2), our estimated Pb emissions corresponded to as much as 620 kg/km²/yr on average. So, while data on historical emissions from point sources are both scarce and difficult to reconstruct, these rough estimations still help point towards the high significance of the glasswork for 20th century Pb emissions to air in Sweden.

3.2. Pb concentrations in air

Pb concentrations in air (µg Pb/m³) around the 10 investigated glassworks sites, and for each of the investigated residence scenarios, are given in Table 3. Concentrations that exceed the US national ambient air quality standard (NAAQS) of 0.15 µg Pb/m³ (USEPA, 2008) are marked in red.

All Pb concentrations that were calculated for the four sites with lowest emissions (Alsterbro, Bergdala, Gadderås and Emmaboda) were below the NAAQS reference concentration, varying from 0.0000 to 0.14 µg Pb/m³ when considering the entire study period, and with an average value of 0.013 µg Pb/m³. The 20th century concentrations at Alsterbro and Bergdala are all found towards the lower end of the concentration spans recently reported for urban areas across Europe, but are clearly elevated compared to

Table 2

Total amounts of Pb (kg) emitted to air from the 10 investigated glassworks, given for 10-year periods during the 1900s.

	Kosta	Orre-fors	Boda	Puke-berg	Åfors	Målerås	Alster-bro	Berg-dala	Gadde-rås	Emma-boda	Sum
Year 2000	21	125	6	n/a	11	12	n/a	1	n/a	n/a	176
1991–2000	1361	6669	341	198	1879	244	n/a	69	n/a	n/a	10,762
1981–1990	30,556	44,123	7518	1593	5998	2603	n/a	389	n/a	9	92,790
1971–1980	70,618	40,908	10,001	4446	6040	3505	n/a	1158	n/a	10	136,686
1961–1970	20,161	27,916	11,261	3850	5036	1829	1281	693	83	10	72,120
1951–1960	9882	15,492	7161	2301	1928	4142	889	593	84	10	42,482
1941–1950	8136	7487	3110	773	1017	2598	490	133	20	10	23,774
1931–1940	7551	5370	4072	2969	185	2574	349	103	122	10	23,304
1921–1930	7538	8243	6784	4574	319	2866	370	393	15	10	31,111
1911–1920	9355	6029	3388	2422	n/a	211	362	8	16	1	21,792
1901–1910	5153	4028	1791	75	n/a	70	162	n/a	n/a	n/a	11,279
Sum	170,310	166,265	55,428	23,201	22,401	20,642	3903	3540	341	70	466,101

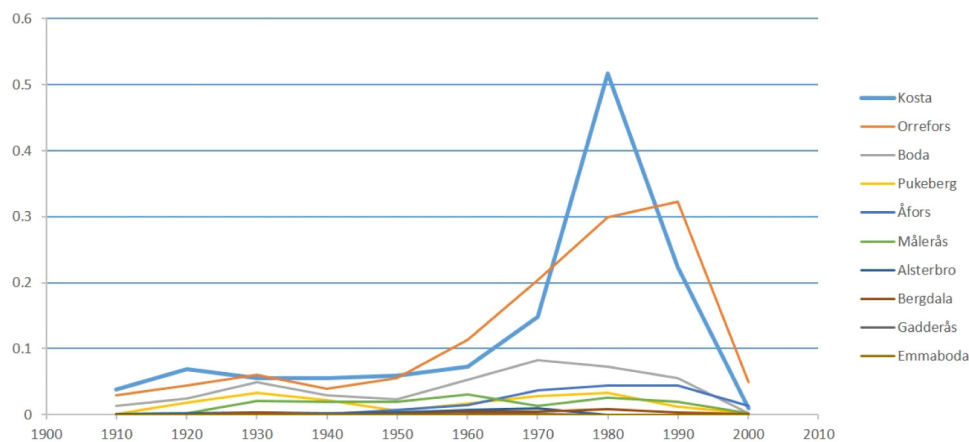


Fig. 2. Normalization of the data in Table 2 (20th century Pb emissions to air (kg/year) for the 10 study sites), i.e. rescaling the dataset into the range 0–1.

present-day background concentrations (Dimitriou and Kassomenos, 2017; Gruszecka-Kosowska, 2018; Huremović et al., 2020; Pérez et al., 2008; Rampazzo et al., 2008; Resongles et al., 2021; Terzi et al., 2010; Van Den Heuvel et al., 2016). Present background concentrations in Sweden, in rural environments without close-by point sources, are found in the range of 0.001–0.003 $\mu\text{g Pb/m}^3$ (IVL, 2019). Studies from medium-sized cities such as Thessaloniki (Terzi et al., 2010), Venice (Rampazzo et al., 2008), Kraków (Gruszecka-Kosowska, 2018), Sarajevo (Huremović et al., 2020), Dortmund and Bielefeld (Dimitriou and Kassomenos, 2017), Barcelona (Pérez et al., 2008) and Antwerp (Van Den Heuvel et al., 2016), report average concentrations of Pb in PM_{10} particles of 0.01–0.09 $\mu\text{g/m}^3$, where the higher figures of this interval derive from sites with high traffic loads. Historical Pb concentrations in air around the smaller glassworks are thus comparable to the levels found in urban areas today. High concentrations today are mainly detected in the vicinity of point sources, such as mining sites, where e.g. a study by Serbula et al. (2017) reported mean lead concentrations of 0.2 $\mu\text{g/m}^3$, and in megacities of developing countries, where an often-highlighted example is the Delhi metropolitan area with Pb concentrations in air frequently found to be above 0.5 $\mu\text{g/m}^3$ (Shen et al., 2016). But even though there are some examples where high Pb concentrations in air can still be found, the occurrence of Pb in urban air was obviously significantly higher prior to the ban on leaded gasoline, where the phase-out was completed in most countries at the turn of the millennium (Tong et al., 2000). Taking London as a European example, Resongles et al. (2021) sum up the development by describing how average Pb concentrations at central London background sites were around 0.5–0.6 $\mu\text{g/m}^3$ prior to the ban on leaded gasoline in the early 1980s, but that Pb concentrations thereafter have decreased to about 0.02 $\mu\text{g/m}^3$ in year 2000 and now, about two decades later, typically are found around 0.01 $\mu\text{g/m}^3$. In Helsinki too similar concentrations of Pb were reported before the Pb gasoline ban; 0.3 $\mu\text{g/m}^3$ for the years 1964–1977 (Mattsson and Jaakkola, 1979). So, the past exposure to airborne Pb for people living in the glassworks villages of Alsterbro, Bergdala, Gadderås and Emmaboda should have been moderate relative to what urban populations have experienced, even when glass production was flourishing there.

The calculated values for the six glassworks with the highest Pb emissions, on the other hand (Kosta, Orrefors, Boda, Pukeberg, Åfors, Målerås), frequently exceeded the US EPA standard of 0.15 $\mu\text{g Pb/m}^3$ (USEPA, 2008) over the past century (Table 3). From the beginning of the century and until ~1990, all the concentrations calculated for the two 0–500 m scenarios (S1 + S2) at the three largest factories (Kosta, Orrefors, Boda) exceeded this air quality standard, ranging from 0.16 to 7.5 $\mu\text{g Pb/m}^3$, with an average of 1.2 $\mu\text{g/m}^3$. Concentrations were lower over the same time period and scenarios at Pukeberg, Åfors and Målerås, although 59 % of the figures exceeded the air quality standard here too (Table 3). Values ranged from 0.0064 to 0.64 $\mu\text{g Pb/m}^3$, with an average value of 0.22 $\mu\text{g/m}^3$. During the 1970s, where

the emissions peaked, the average concentration for scenario 1 (residence in the main wind direction and within 500 m from the factory) around Kosta, Orrefors, Boda was 4.1 $\mu\text{g Pb/m}^3$ and around Pukeberg, Åfors and Målerås 0.49 $\mu\text{g/m}^3$. In the former case, the concentration was about 400 times the present-day concentrations in urban European regions, and e.g. 10 times the concentrations in London before the phasing-out of leaded gasoline. Thus, the historical Pb concentrations in air surrounding the larger crystal-producing glassworks were exceptionally high. The problem is, however, mainly a historical one. At none of the sites did the assessed Pb concentrations in air exceed the 0.15 $\mu\text{g Pb/m}^3$ standard in year 2000 (Table 3), although concentrations at all open factories except Bergdala were still higher than in remote areas without any industrial activity in Sweden (0.001–0.003 $\mu\text{g Pb/m}^3$; (IVL, 2019)). The complementary measurements of PM_{10} -Pb in air around Målerås, Orrefors, Bergdala and Åfors in 2019 (upper results row of Table 3), implied that concentrations had decreased further by year 2019. Recent collection and analyses of airborne particles were also performed at a still active glassworks site in Venice, Italy (Rossini et al., 2010). This study found considerably higher concentrations, around 0.009–2.420 $\mu\text{g Pb/m}^3$, than those found in our study.

3.3. Pb inhalation risks

The hazard quotients (HQs) related to non-cancer risks after inhalation of Pb particles are shown for all glassworks and residence scenarios in Table 4. The lowest inhalation HQs of the century, consistently below 1 for all sites and scenarios and ranging from 0.00 to 0.50, were found for the last year evaluated (year 2000). Table 4 further indicates that the risks following inhalation is – and has been – low in comparison to the risks via dietary exposure. In Table 4, inhalation HQs that exceed the average dietary HQ are marked in red. This dietary HQ, estimated to 0.16 on average, was calculated as the ratio of the average dietary exposure in the glassworks villages to a reference dose for oral exposure (RfD), i.e. according to the same principle as outlined in Eq. (2). The median dietary Pb intake via vegetables, fruits, berries and mushrooms has been estimated to 0.046 $\mu\text{g/kg/day}$ (Augustsson et al., 2018), and to 0.035 $\mu\text{g/kg/day}$ via consumption of drinking water from private wells (Augustsson et al., 2016); thus, about 0.08 $\mu\text{g/kg/day}$ in total. This dietary exposure value was then divided with a toxicological reference value via oral exposure of 0.50 $\mu\text{g/kg/day}$ to reach the average HQ of 0.16. While the previous tolerable daily intake of 3.6 $\mu\text{g Pb/kg/day}$ from WHO (1993) has been withdrawn (EFSA, 2010; WHO, 2010), and a new tolerable intake has not been established due to the abovementioned difficulties in defining a threshold dose, the lowest derived benchmark dose, BMDL₀₁, following dietary Pb exposure found in the review by EFSA (EFSA, 2010) was used as an approximation of a RfD.

Table 3

Concentrations of PM₁₀-Pb in air (µg/m³) around the 10 investigated glassworks, for the 4 assessed residence scenarios (S1–S4). S1 = residence in the prevailing wind direction and at a distance of 0–500 m from the glass works. S2 = against prevailing wind direction at 0–500 m distance. S3 = in prevailing wind direction at 500–1000 m distance, and S4 = against prevailing wind direction at 500–1000 m distance. Average Pb concentrations are based on data only from the glassworks that were running during each specific time period. Periods without production are marked as not applicable (n/a) in the table. The red figures show concentrations that exceed the US national ambient air quality standard (NAAQS) of 0.15 µg Pb/m³ in total suspended air particles (USEPA, 2008).

	Kosta	Orrefors	Boda	Pukeberg	Åfors	Målerås	Alsterbro	Bergdala	Gadderås	Emmaboda	Average
Measured in		0.049*			0.001*	0.015*		0.003*			
2019		(closed)			(closed)	(running)		(running)			
Year 2000 S1	0.022	0.13	0.0061	n/a	0.012	0.013	n/a	0.0011	n/a	n/a	0.031
S2	0.013	0.079	0.0036	n/a	0.0072	0.0076	n/a	0.0006	n/a	n/a	0.019
S3	0.0040	0.024	0.0011	n/a	0.0022	0.0023	n/a	0.0002	n/a	n/a	0.0056
S4	0.0024	0.014	0.0007	n/a	0.0013	0.0014	n/a	0.0001	n/a	n/a	0.0034
1991–2000 S1	0.14	0.71	0.036	0.021	0.20	0.026	n/a	0.0073	n/a	n/a	0.16
S2	0.087	0.42	0.022	0.013	0.12	0.016	n/a	0.0044	n/a	n/a	0.098
S3	0.026	0.13	0.0065	0.0038	0.036	0.0046	n/a	0.0013	n/a	n/a	0.029
S4	0.016	0.076	0.0039	0.0023	0.022	0.0028	n/a	0.0008	n/a	n/a	0.018
1981–1990 S1	3.2	4.7	0.80	0.17	0.64	0.28	n/a	0.041	n/a	0.0010	1.2
S2	1.9	2.8	0.48	0.10	0.38	0.17	n/a	0.025	n/a	0.0006	0.74
S3	0.58	0.84	0.14	0.030	0.11	0.050	n/a	0.0074	n/a	0.0002	0.22
S4	0.35	0.50	0.086	0.018	0.069	0.030	n/a	0.0044	n/a	0.0001	0.13
1971–1980 S1	7.5	4.3	1.1	0.47	0.64	0.37	n/a	0.12	n/a	0.0011	1.8
S2	4.5	2.6	0.64	0.28	0.38	0.22	n/a	0.074	n/a	0.0006	1.1
S3	1.3	0.78	0.19	0.085	0.12	0.067	n/a	0.022	n/a	0.0002	0.33
S4	0.81	0.47	0.11	0.051	0.069	0.040	n/a	0.013	n/a	0.0001	0.20
1961–1970 S1	2.1	3.0	1.2	0.41	0.53	0.19	0.14	0.074	0.0088	0.0011	0.77
S2	1.3	1.8	0.72	0.25	0.32	0.12	0.082	0.044	0.0053	0.0006	0.46
S3	0.38	0.53	0.21	0.073	0.096	0.035	0.024	0.013	0.0016	0.0002	0.14
S4	0.23	0.32	0.13	0.044	0.058	0.021	0.015	0.0079	0.0009	0.0001	0.082
1951–1960 S1	1.0	1.6	0.76	0.24	0.20	0.44	0.094	0.063	0.0089	0.0011	0.45
S2	0.63	0.99	0.46	0.15	0.12	0.26	0.057	0.038	0.0054	0.0006	0.27
S3	0.19	0.30	0.14	0.044	0.037	0.079	0.017	0.011	0.0016	0.0002	0.081
S4	0.11	0.18	0.082	0.026	0.022	0.047	0.010	0.0068	0.0010	0.0001	0.049
1941–1950 S1	0.86	0.79	0.33	0.082	0.11	0.28	0.052	0.014	0.0022	0.0011	0.25
S2	0.52	0.48	0.20	0.049	0.065	0.17	0.031	0.0084	0.0013	0.0006	0.15
S3	0.15	0.14	0.059	0.015	0.019	0.050	0.0093	0.0025	0.0004	0.0002	0.045
S4	0.093	0.086	0.036	0.0088	0.012	0.030	0.0056	0.0015	0.0002	0.0001	0.027
1931–1940 S1	0.80	0.57	0.43	0.31	0.020	0.27	0.037	0.011	0.013	0.0011	0.25
S2	0.48	0.34	0.26	0.19	0.012	0.16	0.022	0.0066	0.0078	0.0006	0.15
S3	0.14	0.10	0.078	0.057	0.0035	0.049	0.0066	0.0020	0.0023	0.0002	0.044
S4	0.086	0.061	0.047	0.034	0.0021	0.029	0.0040	0.0012	0.0014	0.0001	0.027
1921–1930 S1	0.80	0.87	0.72	0.49	0.034	0.30	0.039	0.042	0.0016	0.0011	0.33
S2	0.48	0.52	0.43	0.29	0.020	0.18	0.024	0.025	0.0009	0.0006	0.20
S3	0.14	0.16	0.13	0.087	0.0061	0.055	0.0071	0.0075	0.0003	0.0002	0.059
S4	0.086	0.092	0.078	0.052	0.0036	0.033	0.0042	0.0045	0.0002	0.0001	0.036
1911–1920 S1	0.99	0.64	0.36	0.26	n/a	0.022	0.038	0.0008	0.0017	0.0001	0.26
S2	0.60	0.38	0.22	0.15	n/a	0.013	0.023	0.0005	0.0010	0.0001	0.15
S3	0.18	0.11	0.065	0.046	n/a	0.0040	0.0069	0.0001	0.0003	0.0000	0.046
S4	0.11	0.069	0.039	0.028	n/a	0.024	0.0041	0.0001	0.0002	0.0000	0.028
1901–1910 S1	0.78	0.61	0.27	0.011	n/a	0.011	0.025	n/a	n/a	n/a	0.28
S2	0.47	0.37	0.16	0.0068	n/a	0.0064	0.015	n/a	n/a	n/a	0.17
S3	0.14	0.11	0.049	0.0020	n/a	0.0019	0.0044	n/a	n/a	n/a	0.051
S4	0.084	0.066	0.029	0.0012	n/a	0.0011	0.0027	n/a	n/a	n/a	0.031

*PM₁₀-Pb measured in 2019.

Table 4

HQs via inhalation. Inhalation HQs > HQs via intake of local vegetables and drinking water from private wells, where the average HQ is 0.16, are marked in red. Values <0.01, i.e. where HQs are <1 % of the RfC, are rounded to 0.00. Periods without production are marked as not applicable (n/a).

		Kosta	Örrefors	Boda	Pukeberg	Åfors	Målerås	Alsterbro	Bergdala	Gadderås	Emmaboda
Year 2000	S1	0.08	0.50	0.02	n/a	0.05	0.05	n/a	0.00	n/a	n/a
	S2	0.05	0.30	0.01	n/a	0.03	0.03	n/a	0.00	n/a	n/a
	S3	0.02	0.09	0.00	n/a	0.01	0.01	n/a	0.00	n/a	n/a
	S4	0.01	0.05	0.00	n/a	0.00	0.01	n/a	0.00	n/a	n/a
1991-2000	S1	0.55	2.7	0.14	0.08	0.75	0.10	n/a	0.03	n/a	n/a
	S2	0.33	1.6	0.08	0.05	0.45	0.06	n/a	0.02	n/a	n/a
	S3	0.10	0.48	0.05	0.01	0.14	0.02	n/a	0.00	n/a	n/a
	S4	0.06	0.29	0.01	0.01	0.08	0.01	n/a	0.00	n/a	n/a
1981-1990	S1	12	18	3.0	0.64	2.4	1.0	n/a	0.16	n/a	0.00
	S2	7.4	11	1.8	0.38	1.4	0.63	n/a	0.09	n/a	0.00
	S3	2.2	3.2	0.54	0.11	0.43	0.19	n/a	0.03	n/a	0.00
	S4	1.3	1.9	0.32	0.07	0.26	0.11	n/a	0.02	n/a	0.00
1971-1980	S1	28	16	4.0	1.8	2.4	1.4	n/a	0.46	n/a	0.00
	S2	17	10	2.4	1.1	1.5	0.84	n/a	0.28	n/a	0.00
	S3	5.1	2.9	0.72	0.32	0.43	0.25	n/a	0.08	n/a	0.00
	S4	3.1	1.8	0.43	0.19	0.26	0.15	n/a	0.05	n/a	0.00
1961-1970	S1	8.1	11	4.5	1.5	2.0	0.73	0.51	0.28	0.03	0.00
	S2	4.9	6.7	2.7	0.93	1.2	0.44	0.31	0.17	0.02	0.00
	S3	1.5	2.0	0.81	0.28	0.36	0.13	0.09	0.05	0.01	0.00
	S4	0.87	1.2	0.49	0.17	0.22	0.08	0.06	0.03	0.00	0.00
1951-1960	S1	4.0	6.2	2.9	0.92	0.77	1.7	0.36	0.24	0.03	0.00
	S2	2.4	3.7	1.7	0.55	0.46	1.0	0.21	0.14	0.02	0.00
	S3	0.71	1.1	0.52	0.17	0.14	0.30	0.06	0.04	0.01	0.00
	S4	0.43	0.67	0.31	0.10	0.08	0.18	0.04	0.03	0.00	0.00
1941-1950	S1	3.3	3.0	1.3	0.31	0.41	1.0	0.20	0.05	0.01	0.00
	S2	2.0	1.8	0.75	0.19	0.24	0.62	0.12	0.03	0.00	0.00
	S3	0.59	0.54	0.22	0.06	0.07	0.19	0.04	0.01	0.00	0.00
	S4	0.35	0.32	0.13	0.03	0.04	0.11	0.02	0.01	0.00	0.00
1931-1940	S1	3.0	2.2	1.6	1.2	0.07	1.0	0.14	0.04	0.05	0.00
	S2	1.8	1.3	0.98	0.71	0.04	0.62	0.08	0.02	0.03	0.00
	S3	0.54	0.39	0.29	0.21	0.01	0.19	0.03	0.01	0.01	0.00
	S4	0.33	0.23	0.18	0.13	0.01	0.11	0.02	0.00	0.01	0.00
1921-1930	S1	3.0	3.3	2.7	1.8	0.13	1.2	0.15	0.16	0.01	0.00
	S2	1.8	2.0	1.6	1.1	0.08	0.69	0.09	0.09	0.00	0.00
	S3	0.54	0.59	0.49	0.33	0.02	0.21	0.03	0.03	0.00	0.00
	S4	0.33	0.36	0.29	0.20	0.01	0.12	0.02	0.02	0.00	0.00
1911-1920	S1	3.8	2.4	1.4	0.97	n/a	0.08	0.15	0.00	0.01	0.00
	S2	2.3	1.5	0.81	0.58	n/a	0.05	0.09	0.00	0.00	0.00
	S3	0.67	0.43	0.24	0.17	n/a	0.02	0.03	0.00	0.00	0.00
	S4	0.40	0.26	0.15	0.10	n/a	0.01	0.02	0.00	0.00	0.00
1901-1910	S1	3.0	2.3	1.0	0.04	n/a	0.04	0.09	n/a	n/a	n/a
	S2	1.8	1.4	0.62	0.03	n/a	0.02	0.06	n/a	n/a	n/a
	S3	0.53	0.41	0.18	0.01	n/a	0.01	0.02	n/a	n/a	n/a
	S4	0.32	0.25	0.11	0.00	n/a	0.00	0.01	n/a	n/a	n/a

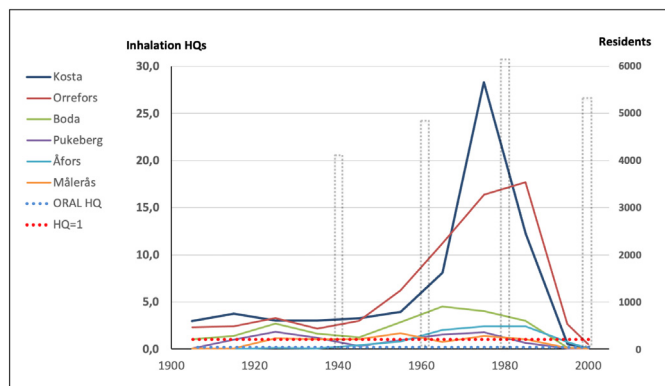


Fig. 3. Historical variation in hazard quotients (HQs) around the 6 largest glassworks factories, together with the total number of persons residing within 1000 m from these factories in the years 1940, 1960, 1980 and 2000.

Considering the entire study period, inhalation HQs exceeded the dietary HQ by many orders of magnitude for the major part of the 20th century. Fig. 3 shows the worst-case scenario (S1) for the six glassworks with the highest Pb emissions.

3.4. Exposure and population size

Fig. 3 also shows how the historical variations in HQ (left y axis) covary with the total population residing within 1 km of the 10 glassworks investigated in this study, assessed for the years 1940, 1960, 1980 and 2000 (right y axis). By 1940, when the total production and HQs were still low, there were about 6800 persons living in the glass works villages. In 1980, soon after the peak of production, the number of persons had increased to approximately 9800, and in the year 2000 there were only about 5500 left.

That the regional population decreased along with the glass production has also been described by Borgegård et al. (1995). This was, however, probably not only due to reduced employment numbers at the glass factories. Both the public and service sectors were centralized in Sweden during the 1980s, and this led to people moving from rural to urban areas (Hedlund and Lundholm, 2015). The same trend in decreased rural populations at this time was also seen in, for example, mining villages in northern Sweden (Carson et al., 2020), and in many other countries too, for example in Ireland (Cawley, 1994), Australia (Carson et al., 2016), France (Laménie, 2016), and the USA (Johnson and Lichter, 2019).

4. Conclusion

Since the risks associated with environmental pollutants depend on the dose presented to the exposed individuals, knowledge of the historical exposure can provide input that is valuable from several perspectives. There is, however, still a lack of studies that examine how exposure has changed for people that reside near pollution point sources. This study is unique in that it does just that. It uses the annual volumes of used raw material (Pb oxide) at 10 glassworks sites in south-eastern Sweden, extracted from national archives, as input in a model to simulate Pb emissions to air around these glassworks for the entire 20th century, and thereafter assesses changes in inhalation exposure and hazard quotients (HQs). What the study shows is that:

- Lead emissions to air were moderate for the first part of the century but started to increase in the 1960s. They peaked during the 1970s, with emissions from the six largest factories amounting to between 3500 and 70,700 kg Pb. These emissions correspond to average concentrations in air of between 0.37 and 7.5 $\mu\text{g Pb}/\text{m}^3$, which is between 2.5 and 50 times the present US national ambient air quality standard (0.15 $\mu\text{g Pb}/\text{m}^3$).
- By the year 2000 the modelled concentrations around the

abovementioned six factories had dropped to between 0.012 and 0.13 $\mu\text{g Pb}/\text{m}^3$. Thus, over approximately 20 years the Pb concentrations in air went from exceptionally high to a level that is normal for urban regions today. The PM_{10} measurements from 2019 indicate a further decline, with a mean value of 0.02 $\mu\text{g Pb}/\text{m}^3$.

- The highest emissions, and highest exposure levels, coincided with the time period with the highest number of residents in the glassworks villages. Thus, not only was the risk higher in the past, more people were also subjected to this risk.
- Over the entire 20th century the non-cancer risks were associated with inhalation, expressed as hazard quotients (HQs), to a much greater extent than the dietary HQs. While inhalation was the more prevalent exposure pathway in the past, both pathways are nowadays judged to be associated with low exposures and risks.

Using archives as a data source has its limitations, both in terms of data availability and in terms of reliability. As a result, it is inevitable that the emissions will be overestimated for some years and underestimated for others. Over the whole century though, the estimated emissions should be roughly correct, validated with a few measurements also found in the local archives. Even if the applied methodology can only provide a rough estimate of the historical Pb emissions to air and subsequent exposure, its strength is that it presents a fairly straightforward approach to illustrate past conditions. Information about how past exposures compare to the present-day situation can be useful, for example, when risks are to be communicated to worried residents or to people thinking of moving to a contaminated area. A picture of the historical exposure can also increase our understanding of the connection between exposure and disease.

CRediT authorship contribution statement

Anna Augustsson: Conceptualization; Methodology; Formal analysis; Investigation; Writing (original draft); Writing (review and editing); Visualization; Supervision; Project administration; Funding acquisition.

Stina Alriksson: Conceptualization; Methodology; Formal analysis; Investigation; Writing (original draft); Writing (review and editing); Visualization; Supervision.

Elin Voxberg: Formal analysis; Investigation; Writing (original draft).

Helen Karlsson: Investigation; Writing (review and editing).

Stefan Ljunggren: Investigation; Writing (review and editing).

Data availability

Data will be made available on request.

Declaration of competing interest

The authors declare the following financial interests/personal relationships which may be considered as potential competing interests: Anna Augustsson reports financial support was provided by Crafoord Foundation.

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