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Relating mercury occurrence in soil gases at establishments hosting children to historical mercury-using activities in Paris, France

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Abstract

Volatile pollutants from former industrial sites can degrade the buildings' indoor air quality that were built after the industrial activities. Since 2010, environmental assessments have been conducted in French establishments hosting sensitive populations identified as being on or near potentially contaminated former industrial sites. These projects are based on historical studies traditionally carried out as part of managing contaminated sites, to determine which substances should be analyzed. They pinpoint former activities likely to have stored or used pollutants. We show that the historical information collected is not effective in targeting sites with increased probability of mercury being present in soil gases. Environmental history has demonstrated the existence of large-scale artisanal contamination, both prior to and concomitant with the industrial era. Classic historical studies would not take into account artisanal activities, which are less documented than industrial activities. We carried out additional research for three schools located in three different Parisian districts. Although information on activities which could have emitted mercury was relatively imprecise (in terms of location, type and duration of activities) and uncertainties exist about the completeness of the archival documents available, our investigations identified several mercury-using activities that had not been identified during the classic historical study. However, we have shown that the number of activities identified does not provide information on how mercury has affected soil gas. Consequently, although a more extensive historical research improves knowledge about the presence of potential mercury-using activities, our study shows that a systematic analysis of mercury as part of the assessment of establishments hosting sensitive populations remains relevant. This approach should be applied to other cities around the world.

Abbreviations

- 25 BASIAS: French historical inventory of industrial and service activity sites
- 26 BASOL: database of contaminated (or potentially contaminated) sites and soils
- 27 BCE: Central Base of Establishments (Base Centrale des Etablissements)
- 28 BIT: Labor Inspection Bulletins (Bulletin de l'Inspection du travail)
- 29 BITHI: Labor Inspection and Industrial Hygiene Bulletins (Bulletin de l'Inspection du travail et de l'Hygiène
- 30 Industrielle)
- 31 CITEPA: Technical Center benchmark for atmospheric pollution
- 32 FINES: National file of health and social establishments (Fichier National des Etablissements Sanitaires
- 33 et Sociaux)
- 34 LQ: limit of quantification
- 35 MEDAD, MEEM, MTE: French national ministry for the management of contaminated sites and soils
- 36 NAF: French Nomenclature of Activities (Nomenclature d'Activités Française)
- 37 OEHHA: Office of Environmental Health Hazard Assessment (Canadian)
- 38 PNSE: second national health and environment plan (Plan National Santé Environnement)
- 39 US EPA: United States Environmental Protection Agency
- 40 TRV: chronic toxicity reference value

1 Introduction

Indoor air quality has a significant impact on human health and quality of life in general (Cincinelli and Martellini, 2017). Poor indoor air quality can be harmful to vulnerable population groups such as children or young adults. Thus, air pollution is one of the major environmental threats to human health, alongside climate change (World Health Organization, 2021a). Indoor air constitutes a complex case for human health risk assessment and management due to a wide variety of pollutants, exposure levels, different possible health outcomes, differences in sensitivity of the population... Indoor air quality measurement campaigns have been conducted in buildings frequented by children, in several countries such as the USA (Adgate et al., 2004; NICHHD, 2016), England (ALSPAC, 2015), Europe (Ballesta et al., 2006; Edwards et al., 2001) or even France (Kirchner et al., 2007) and specifically in schools (Csobod et al., 2014; Liège University, 2020; USEPA, 2009). In these studies, air degradation is associated with urban pollution (exhaust, heating), industrial air emissions or localised sources inside buildings. Other indoor air quality monitoring programmes in schools, such as in New York State (Kunz et al., 1996) and Austria (Maringer et al., 2008), have been carried out because of radon vapour intrusion. They collected soil gas samples to assess the consequences of natural radioactive vapour intrusion linked to geological formations. Last few years, methodologies for the human health risk assessment related to indoor air quality have been developed (Azuma et al., 2020; World Health Organization, 2018). None of these studies looked at outgassing from soils contaminated by volatile substances used in past industrial activities.

However, volatile pollutants from former industrial sites can degrade the buildings' indoor air quality that were built after the industrial activities (Derycke et al., 2018). Several concrete examples can illustrate this, such as a secondary school in Vincennes (France), which had to close its doors because of quantified pollutants in the indoor air (from a former metal eyelet factory) (Mandard, 2002) or housing in Romainville (France), whose air is contaminated by a former factory that used to carry out chemical metal cutting (Saget et al., 2018).

In the context of the management of contaminated sites, methodologies consist of a step-by-step process, starting with a historical phase before carrying out on-site investigations. The principle of historical studies is to investigate the presence of potentially polluting activities that may influence the quality of the environments at the establishments. This work is crucial because the decision to collect and analyze samples depends on the findings of these studies. The historical phase is mostly based on the consultation of administrative and historical data, local literature, old aerial photographs and regional and local authorities' archives. They are broadly similar in most countries (For example (Environmental Protection Agency, 1991; National Environment Protection Council (NEPC), 1999; UK Government, 2019)). However, only the French authorities carried out diagnostic campaigns on the whole territory, according to a very precise framework and piloted by the ministries, to know the influence of former activities in buildings' air indoor quality. Indeed, in 2010, France created a national approach to human health risk assessment assessment at establishments hosting sensitive populations (i.e., children and adolescents) that have been identified as being on or near potentially contaminated former industrial sites. This approach results from one of the actions of the second national health and environment plan 2009-2013 (PNSE 2: "2e Plan National Santé Environnement 2009-2013"). It includes taking and analyzing samples in the various exposure media (surface soil, soil gases, indoor air and tap water) at the establishments concerned. The establishments are then classified into one of the following three health-risk categories:

- Category A: "The establishment's soil is not a problem."
- Category B: "Current facilities and uses protect people from exposure to contamination, whether the contamination is potential or proven."
- Category C: "Assessment has shown contamination that calls for technical management measures and even the implementation of health and safety measures."

The establishments covered by the environmental assessment were identified by crossing three databases: BCE (Base Centrale des Etablissements; from the French Ministry of Education), FINESS (Fichier National des Etablissements Sanitaires et Sociaux; French Ministry of Social Affairs and Health) and BASIAS, the French historical inventory of industrial and service activity sites (French Ministry for the Ecological and Inclusive Transition). BASIAS is the recorded memory of France's industrial past from the end of the 19th century to today. It provides information on industrial activities but not on the soil quality.

Mercury is investigated as part of the assessment process. Mercury's adverse health effects are now widely known : primarily respiratory, neurobehavior effects, mood changes and tremblors, hypertension and autonomic nervous system dysfunction (Bluhm et al., 1992; Calabrese et al., 2018; Lee et al., 2009; World Health Organization, 2021b). Indeed, elemental Hg vapor is harmful for the central nervous system, while inorganic Hg compounds primarily affect the kidney (Beckers and Rinklebe, 2017; Wang et al., 2012). How it contaminates various environmental media is studied globally (United Nations Environment, 2019). It may be present in any environment (soil, water, sediment, air, wildlife) in various forms (elemental, inorganic or organic mercury) (Beckers and Rinklebe, 2017; Mason et al., 1994; Obrist et al., 2018; O'Connor et al., 2019; World Health Organization, 2021b). It can migrate from one medium to another by various processes (methylation, dissolution, volatilization, colloidal or dust transport, etc.) (Leterme et al., 2013; World Health Organization, 2010). Anthropogenic emissions include mercury that is released from fuels or raw materials, or from uses in industrial or artisanal products or processes (Ballabio et al., 2020; United Nations Environment, 2019). The mercury form will depend on the activities that produced it (Wang et al., 2012). Mercury emissions in air in gaseous form (Hg(0)) may occur from former mercury-using activities Mercury may circulate in the environment for centuries to millennia (Selin, 2009) and present a risk of exposure to populations (by inhalation from air specifically), and in particular to children (Du et al., 2020; Lee et al., 2009; World Health Organization, 2021b, 2021a).

Research in environmental history shows that , in Europe, from the beginning of the 18th century, mercury was widely used in various forms (amalgam, salts, cinnabar, etc.) and in many applications (Michel and Orru, 2005). Paris was in full expansion from the end of the 18th century, to become one of Europe's largest industrial capitals. The environmental history clearly demonstrates the existence of large-scale artisanal contamination, prior to and concomitant with industrial contamination that was regulated and thus better documented in the archives. This intense industrial and artisanal activity in the city's heart had consequences in terms of public health and contamination.

As part of diagnostic testing, in Paris, mercury was systematically collected and analyzed in subsurface soil gases at a depth of 1 meter and in sub-slab air under buildings. Everywhere else in France, mercury was only occasionally investigated in soil gases by following the sampling and analytical program as defined by the French methodology for the management of contaminated sites and soils in France (MEEM, 2017), based on a relatively simple historical study (named "classic historical study"). That is, gas samples were collected and analyzed for the measurement of mercury only when the historical study revealed the presence of any activity that potentially used mercury.

These elements lead to questions whether, the systematic analysis of mercury in soil gases appears relevant or whether a classic historical study, as usually carried out, would have been sufficient to distinguish between sites where mercury analysis is required, and sites where mercury analysis is not. This also leads to whether the format of historical studies, according to the national methodology and based in particular on the BASIAS inventory of mainly industrial activities, is appropriate and whether these studies provide sufficient elements to define the analytical program for mercury's soil-gas sampling. Would a more comprehensive historical study, using older sources of information and learning more about Paris's artisanal history, be useful in identifying mercury as a contaminant for investigation?

Here, we have investigated the link between the occurrence of mercury in soil gases (1-m deep soil gas and sub-slabs) in establishments hosting young children in Paris and the identification of former

mercury-using activities based on both a classic historical study and a comprehensive historical study. The aim of this study is to improve the consideration of influence of former activities in degradation of indoor air quality and to provide relevant elements to assist decision-makers faced with the presence of mercury in soil gases in cities that have had intensive artisanal and/or industrial activities.

2 Materials and methods

2.1 Sites

Elemental mercury concentrations were measured in subsurface soil gases (1-m deep soil gas and sub-slabs) in 52 early childhood establishments (33 daycare centers, 13 drop-in childcare centers, 4 preschools and 2 recreation centers) in Paris (France, Figure 1) on or near potentially contaminated soils. For confidentiality, the sites' locations are not precise, nor are the sites described in detail. These 52 establishments are located on 44 geographical sites, some of them being part of the same school campus (e.g., daycare and drop-in childcare on the same site). The sites studied are mainly located in the northeast quadrant of Paris, in the 11th, 19th and 20th districts (Figure 1).

2.2 Sampling

Sampling was carried out by a number of different consultants, between 2014 and 2018. To ensure that results were reliable and comparable from site to site, the same standardized sampling and analysis protocols were applied for all sites. The samples were collected in sub-slab soil gas probes or soil gas wells whose seals had previously been checked, by pumping air for 4 to 8 hours at a minimum controlled flow rate at the beginning and end of sampling of 0.5 to 1 L/minute depending on the site, the general guideline being 4 hours at 1 L/minute. Mercury was trapped on Hopcalite sorbent tubes (500 mg or 200 mg), with two tubes arranged in series to systematically confirm that the first tube was not saturated.

It should be noted that, as part of the national diagnostic approach, the establishments were subject to additional sampling, particularly in indoor air, where the estimated concentration in the living areas exceeded 0.03 $\mu\text{g}/\text{m}^3$. This estimate is made by weighting the concentration in soil gases at a depth of 1 meter and in sub-slab air by a dilution factor depending on building characteristics (elevator cages, piping, cracks in concrete slabs, ventilated crawl spaces, etc.) (Derycke et al., 2018). The value of 0.03 $\mu\text{g}/\text{m}^3$ corresponds to the chronic toxicity reference value (TRV) defined by the OEHA in 2008 (Bisson and Marlière, 2020; OEHA, 2015). In case of additional sampling, the soil gas sampling and analysis were repeated at the same points (Figure SI-2). Several mercury concentrations may therefore be available for one sampling point, at different dates.

The oxygen concentrations were measured in wells using a multigas sensor (Mx4, M40) to validate the structures' seals. If they were comparable to those of the ambient air ($\text{O}_2=21\%$), an air intrusion in the sampling structure was suspected and the sealing of the sampling point was redone. The temperature, pressure and humidity were also measured during sampling in the medium and in ambient air (indoor or outdoor as appropriate).

Field and transport blanks were systematically taken for analysis to check for interference during sampling procedures. Samples showing contaminations (on field and transport blanks) during collection, flow rate variations (between the start and end of the sampling) greater than 10% during sampling and sorbent tube saturation (concentration on control tube <5% of concentration on measurement tube) were excluded from the study.

A total of 137 samples were collected. To ensure independence of observations within a set of values (i.e., not giving too much importance to sampling points for which many measurements were available), multiple measurements performed on the same point taken at different dates were averaged. This affected 12 data points and results in a single mean value associated with each point. Subsequently, all the data considered correspond to different measurement points. Consequently 125 values of mercury

concentrations in sub-slab air and soil gases at a depth of 1 meter were considered unique per measurement point: 99 were taken from sub-slab air samples and 26 were taken from soil gases at a depth of 1 meter. Between one and eight measurements were performed in subsurface soil gases (1-m deep soil gas and sub-slabs) per geographical site.

2.3 Analysis

Analyzes were carried out by two different laboratories.

2.3.1 Chemicals

All the chemicals used by the laboratory 1 were :

- Nitric acid (CAS: 7697-37-2) : Baker, 65%
- Hydrochloric acid (CAS: 7647-01-0) : Merck, 37%
- Sn(II)chloride (CAS: 10025-69-1) : Merck, ACS,ISO,Reag. Ph Eur
- Potassiumbromide (CAS: 7758-02-3) : Merck, ACS,ISO,Reag. Ph Eur
- Potassiumbromate (CAS: 7758-01-2) : Merck, ACS,ISO,Reag. Ph Eur
- Ascorbic acid (CAS: 50-81-7) : VWR, 99.0 – 100.5%
- Mercury solution (CAS: 7439-97-6) : Inorganic Venture, 1 g/L

All the chemicals used by the laboratory 2 were :

- Nitric acid (CAS: 7697-37-2) : Merck, 65%
- Hydrochloric acid (CAS: 7647-01-0) : Merck, 37%
- Sn(II)chloride (CAS: 10025-69-1) : Merck, ACS,ISO,Reag. Ph Eur
- Mercury solution (CAS: 7439-97-6) : Inorganic Venture, 1 g/L

2.3.2 Method of analysis

Laboratory 1:

The extraction on hopkalite tubes was performed according to an in-house method based on EN-ISO 15587-1. The solution was made with aqua regia. Mercury concentrations were determined by atomic fluorescence spectrometry (AFS Analytik Jena Analyser) according to an in-house method based on ISO 16772. The different concentrations of the calibration standards were prepared from a 1 g/L commercial stock solution. 5 calibration levels were used: 0.1 µg/L; 0.5 µg/L; 1.0 µg/L; 5.0 µg/L; 10.0 µg/L. The recovery rate was 90 %.

Laboratory 2:

The solution from hopkalite tubes was made with aqua regia. Mercury concentrations were determined by atomic absorption spectrometry (FIMS-400 Perkin-Elmer Analyser) according to the NF EN 13211 standard. The different concentrations of the calibration standards were prepared from a 1 g/L commercial stock solution. 5 calibration levels were used: 0.1 µg/L; 0.5 µg/L; 2.5 µg/L; 10.0 µg/L. The recovery rate was 92-107 %.

The descriptive data (establishment, date of sampling, mercury concentration, average mercury concentration when multiple measurements, limit of quantification, uncertainty, laboratory) for each of the 137 samples are given in supporting information (Table SI-1). The average analytical uncertainty was about 25%. Quantification limits were 0.005 to 0.02 µg/tube, and 0.01 to 0.042 µg/m³, taking into account sampling conditions (flow rate and pumping time).

2.4 Statistical data processing

For processing the data (Kaplan-Meier analysis and Peto-Prentice test), we used the R© software (version 3.6.0) (R Foundation for Statistical Computing, 2021). The Kaplan-Meier analysis was conducted using the package “NADA” (Lee, 2020).

2.4.1 Kaplan-Meier analysis

The Kaplan-Meier method was used to interpret soil gas mercury concentration data because it is particularly suitable for data sets containing values below the quantification limits. It allows the mean, median and some quantiles to be estimated for a data population in the presence of values below the limit of quantification (LQ). It is a non-parametric method. It does not require the use of assumptions such as the mean or standard deviation that assume a particular data distribution form. Instead, it uses the relative positions (ranks) of the values in the dataset.

This approach uses an ordinal method: It uses the rank of each value without using the true numerical value. By construction, all values below the highest LQ will be censored even if they were detected.

Consider the following dataset:

Values	<1	<1	3	<5	7	8	8	8	12	15	22
Ranks	2.5	2.5	2.5	2.5	5	7	7	7	9	10	11

The rank of repeated values is equal to the median of the ranks if they were different. The three values "8", whose ranks should have been 6, 7 and 8, are assigned the rank of 7, i.e. the median of the three ranks. The sum of the ranks is thus preserved (a statistical rule used in many tests). Similarly, the four lowest values are re-censored as below the LQ of 5, and then assigned the rank of 2.5 as the median of ranks 1, 2, 3 and 4 (which would have been assigned to uncensored values). "3" is a detected value, but not knowing whether " < 5 " is equal to for instance 4 or 2.5, "3" cannot be considered as a detected value.

The Kaplan-Meier analysis is an estimate of the survival curve, which is the complement of the cumulative distribution function (cdf). This analysis comes from survival studies of a population of several individuals performed in biological tests. In these tests, individuals are placed under different

living conditions and their durability over time is reported at each stage of the test. However, the test cannot last until all the individuals reach the end of their life, some of them having a life span higher than a given value when the test stops. The dataset therefore contains several results that are censored towards the high values. Nevertheless, using the information contained in the ranks and proportions of data above and below the multiple LQ, the survival analysis can calculate the basic statistical parameters of the study population: median, mean, standard deviation, standard error, percentiles.

Additional guidance and details on Kaplan-Meier analysis are provided in Helsel (2012) and Huston and Juarez-Colunga (2009).

2.4.2 Peto-Prentice Test

Given that numerous concentrations are below the quantification limit and that the data does not exhibit normal distribution, a non-parametric method was used. The Peto-Prentice test compares two censored populations with values below the quantification limits (Peto and Peto, 1972; Prentice, 1978; Prentice and Marek, 1979). Scores of -1 and +1 (called U-scores) are assigned to the data using the Kaplan-Meier survival functions of the two populations to be compared. The scores are then used and weighted for comparison with production of a confidence interval (Helsel, 2005). Additional guidance and details on the Peto-Prentice test are provided in Helsel (2012).

2.5 Historical studies

The principle of historical studies is to investigate the presence of potentially polluting activities that may influence the quality of the environments at the establishments. This work is crucial because the decision to collect and analyze samples depends on the findings of these studies. In our study, the former mercury-using activities were identified using a “classic historical” study and a “comprehensive historical” study.

2.5.1 Classic historical study

The classic historical study consisted in consulting:

- The National Geographic Institute's (Institut Géographique National) historical aerial photographs, available at a regular time steps;
- The BASIAS database (database of former industrial sites and service activities, <https://www.georisques.gouv.fr/risques/basias/donnees#/>) to identify the former industrial activities at or near the establishment;
- Archives within French government departments (Local authorities/Regional Directorate for Environment/regional archives and city halls), relating to these former activities (files relating to classified facilities in particular);
- Interviewing people with good knowledge of these sites;
- And consulting the BASOL database (database of contaminated (or potentially contaminated) sites and soils; <https://www.georisques.gouv.fr/articles-risques/basol>). BASOL identifies contaminated sites that are the subject of government action (investigations, monitoring and pollution remediation).

The classic historical study is coherent with the French national methodology for the management of contaminated sites and soils (MEDAD, 2007; MEEM, 2017).

The BASIAS inventory provides information on industrial activities (e.g., dates of operation, nature of activities, nature of storage, names of operators) from which, with varying accuracy, we can trace a site's history. For Paris, BASIAS inventory work took place until 2006 and identified industrial sites from files about classified facilities, whose activities took place between 1835 and 2005 (Fauray, 2007).

From this classic historical study, we identified industrial sites that might be involved in the contamination of environmental media (soil, soil gas, water) at establishments hosting sensitive

populations. Depending on the type of industrial site identified, the contaminants selected for collection and analysis in the environmental media vary. To help in selecting a specific analytical program for each assessment, the French Ministry for the Ecological and Inclusive Transition implemented in 2007 (MEDAD, 2007) a tool called the "activity-contaminant matrix" and today the "BD Activipoll" database (BRGM and MTE, 2018), used as part of the methodology for managing contaminated sites. It specifies possible occurrences in environmental media, substance by substance, according to industrial activities (listed according to their NAF (French Nomenclature of Activities) code).

For the 52 establishments, we classified the sampling points into two distinct mercury data populations according to their positioning vis-à-vis the area of influence of at least one former mercury-using activity as identified by the classic historical study. Under the Guidance for Evaluating the Vapor Intrusion to Indoor Air Pathway from Groundwater and Soils (US EPA, 2002), a distance of 30 meters around this activity was used as the influence limit for gaseous pollution. For each concentration data point, we assessed whether the sampling point was within a radius of 30 m around at least one former mercury-using activity. To facilitate the designation of these two populations, we call the population of measurements taken within the area of influence of at least one activity identified as potentially mercury-using "Mercury activity" and the population of measurements taken in the area of influence of no activity identified as potentially mercury-using "Non-mercury activity."

2.5.2 Comprehensive historical study

A comprehensive historical study consists of a more extensive literature search with the aim of identifying potential mercury sources that may not be listed in the documentation used for the classic historical study. Specifically, comprehensive historical research looked at locating activities not identified in the BASIAS database (either because they were earlier or were not covered by legislation on classified

facilities) through sources of additional information. This method has been fully applied on three establishments hosting sensitive populations in Paris.

2.5.2.1 Sources consulted to make the historical study more comprehensive

The literature on environmental history provides a better understanding of the artisanal and/or industrial context of Paris and how it changed. We consulted several vintage printed sources on Paris that gave us a general view of the capital's artisanal and industrial activity. For example, Emile de La Bédollière (La Bédollière, second half of the 19th century) provides insights into the how the districts looked in the second half of the 19th century. We also carried out research at archival sources, including certain archives at the City of Paris useful for the study of land parcels and construction and that are not used in the classic historical studies, such as the "Public Health Register" and the "Real Estate Ledger". The "Public Health Register" provides information on how buildings were occupied, including the identification of industrial or commercial establishments. Detailed public health records (sub-series 3589W - Paris. Direction chargée du logement [Housing Directorate]) are the result of a health survey campaign of Parisian buildings begun at the end of the 19th century. The Paris real estate ledger (sub-series DQ18 - Directorate of Registration, Stamp and Domains) lists the essential information on land transfers in Paris: addresses, dates and causes of transfers, names of owners and notaries) between 1809 and 1945.

Our main resource for locating mercury-emitting activities in Paris was the collection of commercial almanacs for the period 1798 to 1914 ("Almanach du commerce de Paris" 1798- 1838 (Duverneuil and De La Tyna, 1798); "Almanach-Bottin du commerce de Paris, des départements de la France et des principales villes du monde" 1842-1856 (Bottin, 1842); "Annuaire-almanach du commerce, de l'industrie, de la magistrature et de l'administration: ou almanach des 500.000 adresses de Paris, des départements et des pays étrangers" 1857-1914 (Didot and Bottin, 1857)). These almanacs are available in a digitized version in the digital library of the National Library of France (<https://gallica.bnf.fr/>). They list the names

and addresses of merchants and manufacturers in the capital, classified by profession and sometimes by street. From them we can study the geographical distribution of professions and establish the industrial nature of a neighborhood, obtaining an overview of the commercial, artisanal and industrial activity in Paris in the 19th and 20th Centuries.

By the mid-18th century, the effects of mercury on workers' health had been proven by medicine (Guillerme, 2007a). We consulted sources related to occupational health and safety, the outcome of the 1892 establishment of the Labor Inspectorate. Our focus was on the "Occupational Diseases Table", which has included occupational mercury poisoning (hydrargyrisms) since 1919. In particular, the Labor Inspection Bulletins ("Bulletin de l'Inspection du travail" BIT) and then the Labor Inspection and Industrial Hygiene Bulletins ("Bulletin de l'Inspection du travail et de l'Hygiène Industrielle" BITHI), available on the Ministry of Labor website (Ministry of Labor, 2020), were drafted for the period 1893 to 1936. The Labor Inspection and Industrial Hygiene Bulletins compiled official acts relating to labor law and regulation (occupational health and safety) as well as technical reports from inspectors' field investigations, particularly in industrial establishments.

The 1902 Bulletin, which presents "Statistics on patients with mercury poisoning who were admitted to hospitals in Paris, from 1891 to 1901," confirms that some professions targeted in this study continued in Paris until the beginning of the 20th century: gilders, mirror makers, barbers, furriers, lapidaries, tinsmiths, paper makers, brush makers, distiller boys.

The period studied for the comprehensive historical study was based on the dates of the archival documents found. The oldest document dates back to 1782.

2.5.2.2 Identification of activities identified as using mercury

The emphasis on the importance of artisanal activities in Paris led us to update the activity-contaminant matrix, which is typically used in the French methodology for the management of contaminated sites

(MEDAD, 2007; MEEM, 2017). For the information from the environmental history, and for the period that concerns us, we mainly targeted professions related to arts, crafts and working with animal skins. The intense industrial and artisanal activity in the heart of Paris in the 19th century focuses on these activities (e.g., millinery, mirror making, goldsmiths, gilding, silversmiths). Among these sources that document the use of mercury in arts and crafts or in working skins, we can cite the “Encyclopédie méthodique” and its volume devoted to “Manufacture, arts and crafts” (Lacombe, 1782) that describes the manufacturing processes implemented at the time it was published (between 1785 and 1828). This helps to identify occupations that used mercury.

Other activities from the list of "Main professions exposed to occupational diseases caused by mercury and its compounds" established by the National Research and Safety Institute (INRS) were identified. Printed sources from the research period had been consulted to validate the use of mercury by certain (now non- existent) occupations identified in almanacs.

With regard to more contemporary activities, we consulted the report (CITEPA, 2019) on greenhouse gas and atmospheric pollution emissions in France produced by CITEPA (Technical Center benchmark for atmospheric pollution) in particular.

From this bibliographic research on mercury-using activities, certain activities (e.g., millinery, gilding, treatment of venereal diseases, mirror making (supporting information, Table SI-3)) were selected as potentially mercury-using, and therefore, likely to have contaminated soil gases at a depth of 1 meter and sub-slab air at or near the establishments. We classified trades related to arts and crafts in accordance with the “Artisanat d’Art” list of arts and crafts, used for obtaining the qualification of “Artisanat d’Art” or the title of Master Artisan of Art (Trades related to creation, trades related to the restoration of heritage, traditional trades), according to the Chamber of Crafts and Crafts, Burgundy region. The trade "military equipment", a term used in almanacs without more precision, can correspond to leather work, button making, metal work, etc.

Activities using all forms of mercury were identified, not just those using elemental mercury (Hg(0), which is volatile), since elemental mercury Hg(0) can be generated from other forms of mercury. This includes the biological reduction of the oxidized form of mercury (O'Connor et al., 2019; Thomassin et al., 2003).

2.5.2.3 Application of the comprehensive historical study to three establishments hosting sensitive populations

We conducted a comprehensive historical study for three schools that were diagnosed as part of the national approach. For these three sites, no industrial activity could be identified in the classic historical study. The relevance of the choice of these three establishments stems from information derived from environmental history. The aim was to understand whether artisanal and industrial activities related to environmental history explain current mercury results in soil gases.

For these three establishments, we looked for artisanal activities within a 30-meter radius, in accordance with the Guidance for Evaluating the Vapor Intrusion to Indoor Air Pathway from Groundwater and Soils (US EPA, 2002). From the former land registries of the districts of Paris, the addresses (numbers and names of streets) included within this perimeter were identified. The Parisian land registry has changed multiple times between 1810 (The first Napoleonic land registry was ordered by the law of September 15, 1807. The plan of the various housing parcels is available for consultation on the Paris Archives) and today. Land registries before 1860 (Archives de Paris, n.d.), after 1860 (date on which Paris annexed several peripheral communes and made them districts), and the current land registry were taken into account (<https://cadastre.data.gouv.fr/>) (Etat français, n.d.). Research had done on activities identified as potentially mercury-using (in supporting information Table SI-3) and carried out on selected addresses in commercial almanacs between 1798 and 1907. An activity is retained within a given time interval if it

appears at least once in an almanac within that time period. If it appears more than once, it is counted only once. The choice was made to consult the documents published every 5 to 10 years (depending on the dates of almanacs available). Activities with a duration less than this interval could therefore be obscured. For activities identified within the 30 m radius, and to know how long they lasted, the commercial almanacs were consulted for each year. The occurrence of activities in each perimeter around the three establishments was summarized.

3 Results

3.1 Mercury concentrations in soil gases and sub-slab air

Mercury concentrations above the limit of quantification were measured at 49 of the 125 sampling points (i.e., a quantification rate of about 39%). The mean of concentrations above the quantification limit is $0.08 \mu\text{g}/\text{m}^3$ (i.e., greater than the TRVs applied in closed environments) and the associated standard deviation is $0.18 \mu\text{g}/\text{m}^3$. The highest concentrations measured were $1.03 \mu\text{g}/\text{m}^3$. For 10 of the 44 sites studied, mercury concentrations were all above the limit of quantification. Conversely, 17 sites showed mercury concentrations below the limit of quantification. The other 17 sites had at least one mercury concentration lower than the limit of quantification and one higher. Per site, the number of points with mercury concentrations above the LQ was between 0 and 3. Figure 2 shows the concentration distribution at the various sites.

Figure 1 shows the distribution of maximum concentrations measured at the 44 sites. The highest values were from sites in the 20th district. Lower concentrations (between 0.09 and $0.12 \mu\text{g}/\text{m}^3$) were distributed in the 2nd, 10th, 14th and 17th districts. Hazard Quotients (HQ) were calculated for children exposed by inhalation of indoor air in nurseries considering the toxicity reference value (TRV) produced by OEHA for mercury in 2008 ($0.03 \mu\text{g}/\text{m}^3$) (Bisson and Marlière, 2020; OEHA, 2015) and standard

protective exposure hypothesis (12h/d, 220d/y). The health risk calculation was done for each soil gas measurement that showed mercury concentrations above the limit of quantification, by estimating indoor air concentrations thanks attenuation factors (Derycke et al., 2018). HQ values are between $1.7 \cdot 10^{-3}$ and 1.03 (Table SI-1). Only 1 sample at one establishment exhibits an HQ value exceeding 1. However, these results are strictly informative considering that reliable health risk calculations should be assessed on the basis of the mercury concentrations directly measured in the indoor air of buildings, rather than estimations derived from soil gas concentrations.

3.2 Relationship between mercury concentrations measured in soil gases and former mercury activity listed by the classic historical study

The number of sampling points where mercury was quantified ("quantified mercury") and not quantified ("unquantified mercury") in soil gases and sub-slab air depending on whether the sampling site is in the area of influence of a former potentially mercury-using activity or not, according to the classic historical study is reported in Table 1.

Fifty-eight sampling points were within the area of influence (30 m) of at least one former potentially mercury-using activity (Mercury activity population). For the other 67 sampling points, the classic historical study did not reveal any previous potentially mercury-using activity (Non-mercury activity population).

The ranges (min, max, mean) of the limits of quantification for the 2 populations are similar (respectively 0.010, 0.031, 0.0199 $\mu\text{g}/\text{m}^3$ for the "Non-mercury activity" population and 0.012, 0.042, 0.0197 $\mu\text{g}/\text{m}^3$ for the "Mercury activity" population).

Contrary to what might have been expected, the number of samples where mercury was quantified was greater in the Non-mercury activity population than in the Mercury activity population (respectively, 32 and 18). Similarly, the number of samples for which mercury was not quantified ($< \text{LQ}$) is greater in the

451 Mercury activity population than in the Non-mercury activity population (40, and 35, respectively).
452 Nevertheless, the highest mercury concentrations were measured at the sampling points of the Mercury
453 activity population (respectively 72.2% and 48.4% of the concentrations are above the TRV for the
454 Mercury activity population and the Non-Mercury population).

455 Comparing the measured mercury concentration distributions in each case is made difficult by the large
456 number of samples for which the measured concentrations were below the limit of quantification (about
457 61% of all samples). To best process the data, a suitable statistical method was used. This is the Kaplan-
458 Meier analysis (Helsel, 2005). An important advantage of this approach is that unquantified values can be
459 taken into account. Table 2 shows the descriptive values obtained after data processing by Kaplan-Meier
460 analysis.

461 The mean calculated for the Mercury activity population is $0.058 \mu\text{g}/\text{m}^3$ vs. $0.030 \mu\text{g}/\text{m}^3$ for the Non-
462 mercury activity population (Figure 3). However, the rate of non-quantification is higher for the Mercury
463 activity population (about 69%) than for the Non-mercury activity population (about 54%). Because of
464 this high rate of non-quantification for the Mercury activity population, the median for this population
465 cannot be calculated. We also see greater variability in mercury concentrations within the Mercury
466 activity population than in the Non-mercury activity population.

467 The associated probability figure (known as the survival curve) shows that both Mercury activity and
468 Non-mercury activity populations are close at low and medium concentrations (Figure 4). For high
469 concentrations ($> 0.1 \mu\text{g}/\text{m}^3$), the probability of observing such values is higher for the Mercury activity
470 population.

471 The non-parametric Peto-Prentice test (Peto and Peto, 1972; Prentice, 1978; Prentice and Marek, 1979)
472 was used to compare survival curves for the two populations. The null hypothesis tested, H_0 , is that of
473 the superiority of the survival curve of the Mercury activity population over that of the Non-mercury
474 activity population. The resulting p-value (probability value) is 0.2, which is not robust evidence that the

concentrations of the Mercury activity population are higher than those of the Non-mercury activity population. The mercury concentrations obtained from soil gas samples at a depth of 1 meter and from sub-slab air in the area of influence of at least one former potentially mercury-using activity are therefore not significantly different from the concentrations obtained from samples taken in areas of influence without any former site of this type.

The fact that mercury was quantified in many samples in the Non-mercury activity population and that differences in mean mercury concentrations between the Mercury activity and Non-mercury activity populations are not significant can only be explained by a lack of specificity in the classification of sampling points in the two populations. This classification is based on the results of the classic historical study. Thus, it would appear that, in Paris, the search for historical information according to the French methodology for managing contaminated sites and soils and the exploitation of these data would not be effective in targeting sites for which the probability of mercury being present in soil gases is increased. This could be explained by the assumption that the historical research approach would be incomplete: Lack of completeness of site identification or positioning given the sources of information consulted, lack of understanding of environmental history, etc.

The means used for the historical study applied as part of the diagnostic process may therefore be insufficient to identify potentially mercury-contaminated sites in Paris.

3.3 Taking the classic historical study further

The absence of a statistically significant difference in mercury concentration in soil gases between establishments identified in a classic historical study, as being in an area of influence of at least one former mercury-using site, and those that are not, led us to take the historical research further.

3.3.1 Artisanal and industrial history of Paris until the 19th century

Many older activities that may have used mercury are not listed in the databases used for the classic historical study. Starting with a rather artisanal organization and relatively modest workshops for mercury-using activities, from the 1760s Paris moved to an industrial scale. But both sides coexisted. On the one hand, in the 1810s some Paris districts held a concentration of small workshops, for instance Arcies and Lombards districts [straddling the current 3rd and 4th districts] where 200-300 metal gilders worked, or the Sainte Avoie and Saint Martin districts [mainly in the current 3th district] where hundreds of milliners congregated (Guillerme, 2007b). On the other hand, the first major Parisian hat factories appeared from the 1760s onwards. They were accompanied, among other things, by mercury nitrate production. Button makers also moved to industrial production and settled on the outskirts, such as button-maker Chardin (Guillerme, 2007b), located in the current 15th Paris district.

In connection with this artisanal and industrial increase, the decree of October 15, 1810 introduced the “*Commodo et incommodo*” (suitable and unsuitable) survey, in order to check that establishments operated properly (Guillerme, 2007b; Leroux, 2014). These inspections result in reports prepared by the Public Health Council. The introduction of industrial regulations, which began in 1810, encouraged entrepreneurs in some undesirable industries to leave central Paris. An industrial suburb slowly emerged in connection with an increasingly industrial Paris. (Le Roux, 2011).

For the post-1830 years, a group of mercury-emitting “industries” was described by E. La Bédollière in one of the chapters of his book on the Temple District (La Bédollière, second half of the 19th century) [in the current 11th district] located near the Conservatoire National des Arts et Métiers: “*there are goldsmiths, silversmiths, jeweler-decorators, gold jewelers, lapidaries, jeweler-chain makers, copper and wood gilders, gold beaters, metal workers, embossers, copper finishers, chisellers, engravers, stampers, splicers, polishers, enamellers, trinket makers, etc.*,” which confirms the information identified in almanacs and the intensive use of mercury in this sector.

Environmental history thus shows how mercury-generating/using activities changed and how uneven they were, in terms of nature of activity, size, duration of activity and geographical distribution, throughout Paris. In connection with these historical elements, we cannot rule out former activities before the 19th century playing a role in the presence of mercury in soil gases at establishments for children or adolescents, whatever their location in Paris. Therefore, to illustrate the elements collected by environmental history, extensive documentary research in various archives was conducted around three establishments hosting sensitive populations.

3.3.2 Comprehensive historical study on three Parisian schools

A comprehensive historical study was done for three Parisian establishments pinpointed as part of the national approach (Figure 1) and which the classic historical study had not identified as being within the influence of a former potentially mercury-using activity. They have been chosen based on 2 criteria: 1) establishments had to be located in different zones of Paris, from the center of Paris to the outlying departments, in order to take into account the environmental history of Paris, and 2) they had to be located in arrondissements suspected of having a significant number of artisanal or industrial activities that potentially used mercury (i.e., 3rd and 15th districts).

In connection with information from the environmental history of Paris and its industrial and artisanal past, the three establishments were selected as follows:

- One establishment for which mercury has not been quantified in soil gases and environmental history indicates the presence of factories that may have generated mercury;
- One establishment for which mercury has been quantified at low concentrations in soil gases at the establishment; but elements of the neighborhood's environmental history have shown that many activities that have used mercury are concentrated there;

- One establishment for which mercury has been quantified in soil gases and where artisanal and industrial activities that have used mercury have been identified by environmental history.

The three selected establishments, named Case 1, Case 2 and Case 3, are in the 15th, 3rd and 10th districts, respectively. The comprehensive historical study focused on the artisanal activities listed in supporting information (Table SI-3).

Case No. 1

Case No. 1 is in the 15th district. The establishment concerned was located in the "Grenelle Plain" (Huard, 1790), which was, until the middle of the 19th century, intended for agriculture. As Paris expanded, this more out-of-town district experienced a development of more recent artisanal activities, in connection with the annexation of Grenelle into Paris in 1859. Historical research in this area has identified a few activities within a 30 m radius of the establishment that may have used mercury. Three horologists having exercised between 1861 and 1870, with a duration of activities between 1 and 7 years and a chemical factory (between 10 and 15 years of activity) were identified (Table 3). This activity is retained with reservations, given the little information obtained as to the nature of the products manufactured. Mercury was not quantified in soil gases at this establishment ($<0.018 \mu\text{g}/\text{m}^3$).

Case No. 2

Case No. 2 is in the 3rd district. Historical research on available almanacs has shown that the site is located in one of the most important districts for gilding, jewelry, luxury goods, etc. From Figure 5 we assess the density of activities identified within a radius of 30 m, at different time periods. A total of 236 activities were identified, including 33 activities superimposed on the establishment, with a majority of activities in the category "jewelry-silversmith-goldsmith-watchmaking" over the period 1790-1910. This

number may be reduced because not all almanacs were studied, either because not all almanacs were available, or because, given the large number of almanacs, only one selection (every 5 to 10 years) was selected. None of these activities had been identified in the classic historical study, in particular because they were not identified in the BASIAS database.

Mercury is quantified in soil gases at the establishment, but concentrations are relatively low (up to $0.021 \mu\text{g}/\text{m}^3$, i.e., less than the TRV of $0.03 \mu\text{g}/\text{m}^3$) in light of the school's environmental history.

Case No. 3

Case No. 3 is in the 10th district. This district, relatively far from the center of Paris (outside the walls of Paris until 1790), developed mainly at the beginning of the 19th century. The St. Louis Hospital, the central point of this district, was built in the 17th century to quarantine contagious patients. Its activities have been specialized in skin and syphilitic diseases since 1801 (Tilles, n.d), and used mercury-based treatments (Guillerme, 2007b, 2003).

In addition, the comprehensive historical study of the sector revealed the presence of mercury-using activities between 1837 and 1878, within a radius of 30 m, and in particular those presented in Table 4. The identification of such activities requires in particular the consultation of specific literature, so these activities were not identified in the classic historical study.

In addition to the artisanal activities identified around case No. 3, the proximity of the hospital is an additional historical element that could explain the quantification of mercury in soil gases at the school (up to $0.116 \mu\text{g}/\text{m}^3$). However, the lack of information on the quantities of mercury used or how effluents were managed means that we cannot make a more precise determination of its influence on the quality of soil gases at the establishment. The maximum concentration of quantified mercury is higher than that of case No. 2, while the number of activities identified is much lower.

3.4 Discussion

Classic historical studies fail to target sites with increased probability of mercury being present in soil gases

The finding that mercury is measured in soil gases in many establishments, while the classic historical study does not identify the presence of old mercury-using activities near these establishments, means it will not be possible to effectively target sites for which the likelihood of mercury in soil gases is increased by searching for historical information in accordance with the French methodology for the management of contaminated sites and soils (MEEM, 2017). This can be explained by the number or the contemporaneous period of information sources consulted (historical aerial photographs, BASIAS database, local and municipal archives and BASOL interviews and database). The mere consultation of these sources of information may limit the number of sites identified and the determination of their location. Other countries such as the United Kingdom speak of industrial archeology and stress the importance of old maps locating industries, produced by The Ordnance Survey (OS), from 1834, developed for all the cities. These cards do not necessarily exist for French cities (RPS Consultants Ltd, 1994). As a result, some sites potentially causing mercury degradation of soil gas quality may be ignored because they are unidentified or incorrectly positioned. It could be happened in the case of conversion from old industrial building to residential use when insufficient information is available on past uses (Gochfeld, 2003; World Health Organization, 2010). This is reinforced by the fact that the national diagnostic process being carried out systematically throughout the territory, does not necessarily take into account the historical specificities of each territory, in which each establishment is located. For Paris, the BASIAS inventory lists industrial sites whose activities occurred between 1835 and 2005 (Faury, 2007). This period therefore does not cover the previous period (notably the 18th and early 19th Centuries) during which mercury-using activities were numerous. In addition, choices have been made regarding types of activities recorded in BASIAS. The activities identified as a priority in the inventory

were defined by the April 3, 1996 bulletin (Faury, 2007). This bulletin focused primarily on the identification of former landfills, hydrocarbon deposits, chemical, petrochemical or metallurgical industries. These activities, mainly industrial or service activities, do not integrate artisanal activities potentially using mercury, so numerous and diverse in Paris.

Do comprehensive historical studies improve knowledge about the presence of potential mercury-using activities?

By consulting complementary archives as part of the comprehensive historical study for three establishments we identified activities that could have used mercury within a 30 m radius. Among the documents consulted, the almanacs and the old Parisian land registry proved to be the most useful sources. The presence of these activities, including a hospital, jewelers, button makers (metal in Figure 5) and lamp makers could explain the quantification of mercury in soil gases in 2 of the 3 establishments studied (Case 2 and Case 3). However, the number of activities identified does not provide information on the level of degradation of soil gas quality by mercury. The number of activities identified that could have used mercury within a 30 m radius is much higher in Case 2 (209 activities) than in Case 3 (9 activities and the hospital) whereas the maximum concentrations measured in soil gases are higher in Case 3 ($0.116 \mu\text{g}/\text{m}^3$) than in Case 2 ($0.021 \mu\text{g}/\text{m}^3$). Also, several mercury-using activities have been identified in more extensive historical research near Case 1, yet mercury is not quantified in soil gases. Mercury is used in different chemical forms in processes (chloride mercury, calomel, etc.) (Guérin et al., 2014; Wang et al., 2012). Some forms are not volatile (Mason et al., 1994; O'Connor et al., 2019) and, as a result, if conditions are favorable (Thomassin et al., 2003), may not be found in soil gases. Once deposited to the terrestrial environment, Hg preferentially adsorbs to clays and organic matter (Varekamp et al., 2003). Furthermore, in Lee et al. (2009) events potentially affecting children were most frequently classified as non-volatilization events. Moreover, not all identified activities have necessarily caused degradation of soil gas quality, either because they have never used mercury, or have used it in

small quantities, or have used it, but only for a period of time. Indeed, one of the limitations of using historical research to determine whether we should look for mercury during investigations is that the processes for certain activities have changed over time, particularly over the time interval 1790-1910. For example, electrolytic metal plating developed from the 1850s, gradually replacing the use of mercury (Hunt, 1973; Protection des Métaux, 2008). In the millinery field, the filing of a patent in 1818 by Malard and Desfossés, with a view to replacing mercury nitrate with a mixture of caustic soda and slaked lime (Guillerme, 2007a), also contributed to reflections on the need to abandon mercury in this field, in view of the health risks for workers. At the same time, in the 1929 Labor Inspection Bulletin, a "Note sur l'hydrargyrisme professionnel dans les ateliers de dorure au mercure sur métaux [Note on Professional Mercury Poisoning in Mercury Gilding Workshops]" confirms the use of mercury for gilding purposes despite advances in electrolysis. Since we cannot know precisely how craftsmen or factories stopped using mercury over time, it is possible that mercury was not used for each of the activities identified in the more extensive historical study. Finally, improvements at the establishment have changed the soils' structure (provision of backfill, reworking, etc.) and therefore also their quality.

Although the more comprehensive historical study improves knowledge about the presence of potential mercury-using activities near establishments hosting sensitive populations, Cases 1 and 2 illustrate that our results do not clearly identify sites for which the assessment will reveal the presence of mercury. The more extensive historical research conducted was not sufficient to identify other activity explaining the origin of the quantified mercury. More broadly, therefore, more extensive historical research is not exhaustive. In the absence of identification of mercury-using activities around an establishment as a result of such research, the absence of mercury in soil gases near the establishment cannot be guaranteed with certainty. In addition, others interfering sources of mercury emissions could constrain and complicate the landscape. For instance, combustion of coal is a major source of mercury emissions (Charvát et al., 2020; Streets et al., 2019, 2011). Together with the wind and the atmospheric lifetime of

Hg species (Selin, 2009), it could explain a regional contamination footprint (Li et al., 2017; Rodríguez Martín and Nanos, 2016; Sherman et al., 2012). These "interfering" pollution sources (identified in the supporting information; Table SI-3) could be responsible for significant dust production (potentially containing mercury) and may be additional to the artisanal pollution sources targeted in our study.

What is the solution?

Systematic analysis of mercury is therefore necessary to identify problematic situations. In terms of the cost of investigation, systematic consideration of mercury does not represent a very large additional budget for an assessment that already includes other substances to be investigated. Thus, the main cost of taking mercury into account is associated with its laboratory analysis. This cost remains low (approximately 50 euros/sample) in relation to the cost corresponding to the time spent carrying out a historical study. Indeed, on the basis of the time spent by the consultants involved in the diagnostic process, we estimate this time at about 2 days per establishment for the completion of a classic historical study. We estimate it at about 5 days for establishing a more complete history, based on the time spent for the three Parisian establishments we studied more specifically for this article.

International concern

The results of this work can be transposed into other French or European cities which have had intensive artisanal and/or industrial activities that used mercury. First banned from use at the beginning of the 18th century in England and France, mercury was reintroduced in various processes (Mathews David, 2019) as in Lyon, "*With Paris, Lyon was then the only city in France authorized to carry out the refining of precious metals (...) they also turned gold to antimony or to mercuric chloride*", or in Marseille "*(...) banned in the 17th century, surreptitiously introduced in Paris in 1730, mercury acquired its legitimacy in Marseille (...)*" (Leroux, 2014). Similarly, cities around Paris, including Senlis, Chartres, Fontainebleau, Versailles and Meaux secreted it (Guillerme, 2003). Other European cities also used a lot of mercury, such as London (Mathews David, 2019), and Birmingham and Bath specializing in button making

(Guillerme, 2007a, 2003). The Flemish made many hats in the 18th century and were the first importers of mercury from Ydria (Greece) (Guillerme, 2003). On the other side of the Atlantic, studies shows that the hat-making industry was present there too during the late 19th-century to 20th century, for example in Connecticut (USA) (Anatone et al., 2020; Janick, 1990). Mercury is also found in the sediments of Long Island Bay (Varekamp et al., 2003). These results raise the question of the potential presence of mercury in other environmental media in Paris, including the sediments of the Seine River.

4 Conclusion

The research and exploitation of historical information in Paris, in accordance with the French methodology for the management of contaminated sites and soils, does not lead to the effective targeting of industrial sites for which the probability of mercury in soil gases is increased. In this context, the usual historical studies conducted for Paris are insufficient to determine whether to retain mercury in the soil-gas investigation program.

If the consideration of mercury in the investigations had been based solely on the results of the classic historical study, mercury would not have been looked for at certain establishments and this would not have uncovered situations where, based on the results of the assessment, management measures related to the presence of mercury in soil gases were necessary.

From deeper historical research around the three schools located in three different Parisian districts, we can locate the presence of artisanal and industrial activities that potentially emitted mercury and were not identified in the classic historical study. Our analysis also makes it possible to estimate how long these activities lasted at the addresses identified. In that respect, having expanded the sources of information consulted was fruitful, providing interesting complementary elements compared to a classic study.

705 Yet the sources of information we consulted have not resulted in quantification of activities or
706 estimations of their environmental impact in terms of the quantities of mercury potentially emitted.
707 Moreover, a thorough historical study, however deep, cannot be considered completely exhaustive, if
708 only because of uncertainty about the completeness of the archival documents available. Thus, if such a
709 study results in the absence of identification of activity that used mercury, doubt remains, and it would
710 be risky to say that no activity had ever degraded soil gas quality. It is therefore difficult, even after a
711 more comprehensive historical study, to distinguish between cases for which mercury analysis is justified
712 and those for which the analysis is not. Therefore, a systematic analysis of mercury in Paris in the context
713 of the assessment of establishments hosting sensitive populations remains relevant. When combined
714 with the detection of other contaminants, systematic mercury review does not represent a substantial
715 extra cost. This approach also appears valid for a large number of cities around the world that have had
716 intense artisanal and/or industrial activity that used mercury. Furthermore, this paper shows the
717 influence of former activities which could degrade buildings air indoor quality. The indoor air
718 degradation source by former industrial/artisanal activities could be better taken in indoor air quality
719 monitoring programmes.

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Figures

Figure 1. Distribution of maximum concentrations for each of the 44 geographical sites (corresponding to 52 early childhood establishments) (in $\mu\text{g}/\text{m}^3$). Sites with concentrations below the limit of quantification are identified by a star. The three selected Parisian establishments for a comprehensive historical study are numbered.

Figure 2. Concentrations ($\mu\text{g}/\text{m}^3$) of mercury at the 27 sites where one concentration at least is higher than the quantification limit (Logarithmic scale. The error bars represent the analytical uncertainty. The toxicity reference value (TRV) is represented by the dotted line).

Figure 3. Box plot of mercury concentrations measured in soil gases at a depth of 1 meter and sub-slab air for samples collected in the area of influence of at least one former potentially mercury-using activity (Mercury activity population) and in the area of influence of no potentially mercury-using activity (Non-mercury activity population).

Figure 4. Survival curve obtained following Kaplan-Meier analysis.

Figure 5. Density of artisanal mercury-using activities (a total of 236 activities) within a 30 m radius around the establishment "Case No. 2" (categories are detailed in supporting information, Table SI-3).

Tables

Table 1. Contingency table showing the number of quantifications/non-quantifications of mercury in soil gases and sub-slab air depending on whether or not measurements are made in the area of influence of a former potentially mercury-using activity.

Table 2. Descriptive statistics of measured mercury concentrations ($\mu\text{g}/\text{m}^3$), depending on whether the sampling points are located in the area of influence of a former potentially mercury-using activity (LQ = limit of quantification). These statistics were obtained after data processing by Kaplan-Meier analysis.

Table 3. List of artisanal activities that may have used mercury around Case No. 1 (within a 30 m radius around the establishment).

Table 4. List of artisanal activities that can use mercury around Case No. 3 (within a 30 m radius around the establishment).

Figures

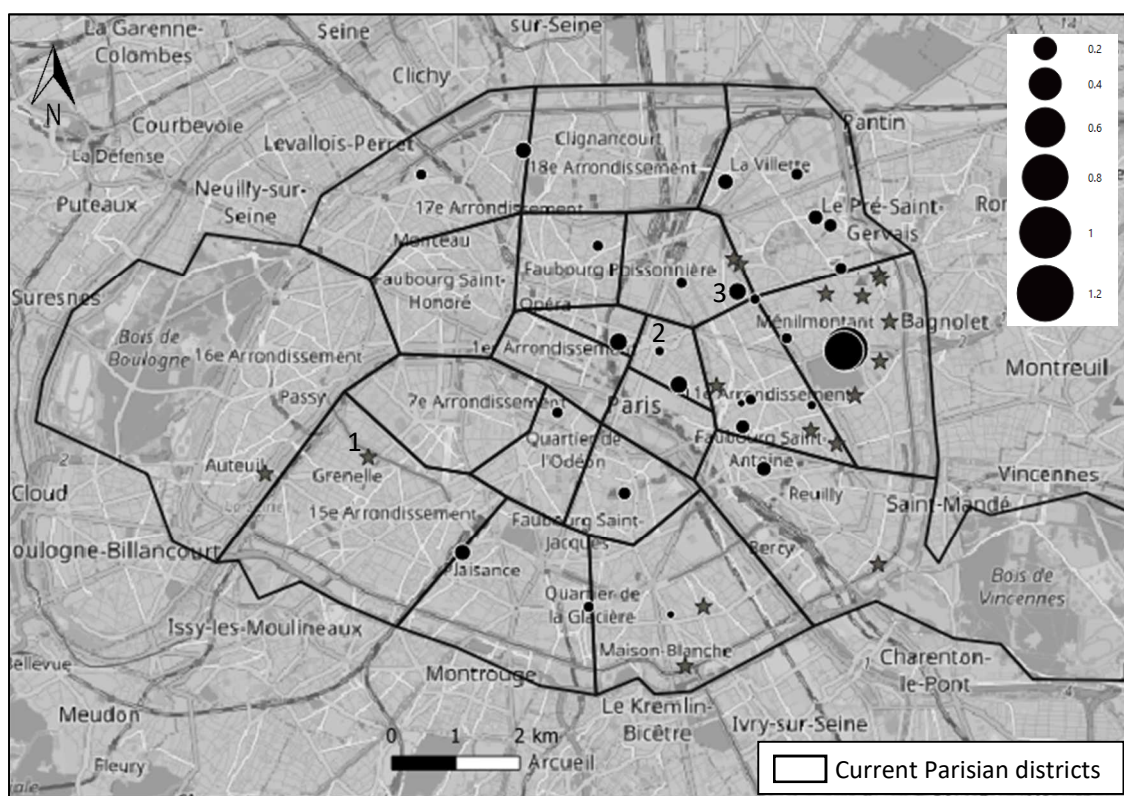


Figure 1: Distribution of maximum concentrations for each of the 44 geographical sites (corresponding to 52 early childhood establishments) (in $\mu\text{g}/\text{m}^3$). Sites with concentrations below the limit of quantification are identified by a star. The three selected Parisian establishments for a comprehensive historical study are numbered.

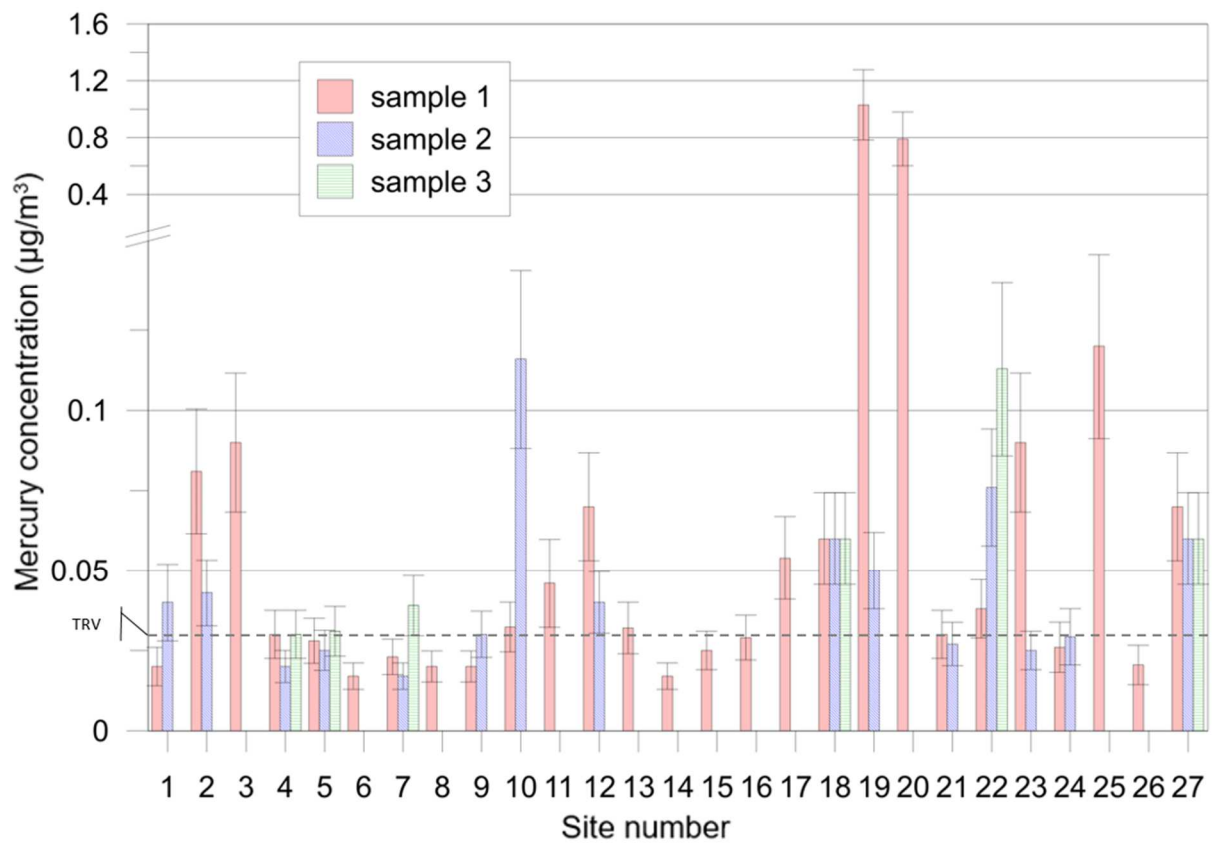


Figure 2: Concentrations ($\mu\text{g}/\text{m}^3$) of mercury at the 27 sites where one concentration at least is higher than the quantification limit (Logarithmic scale. The error bars represent the analytical uncertainty). The toxicity reference value (TRV) is represented by the dotted line.

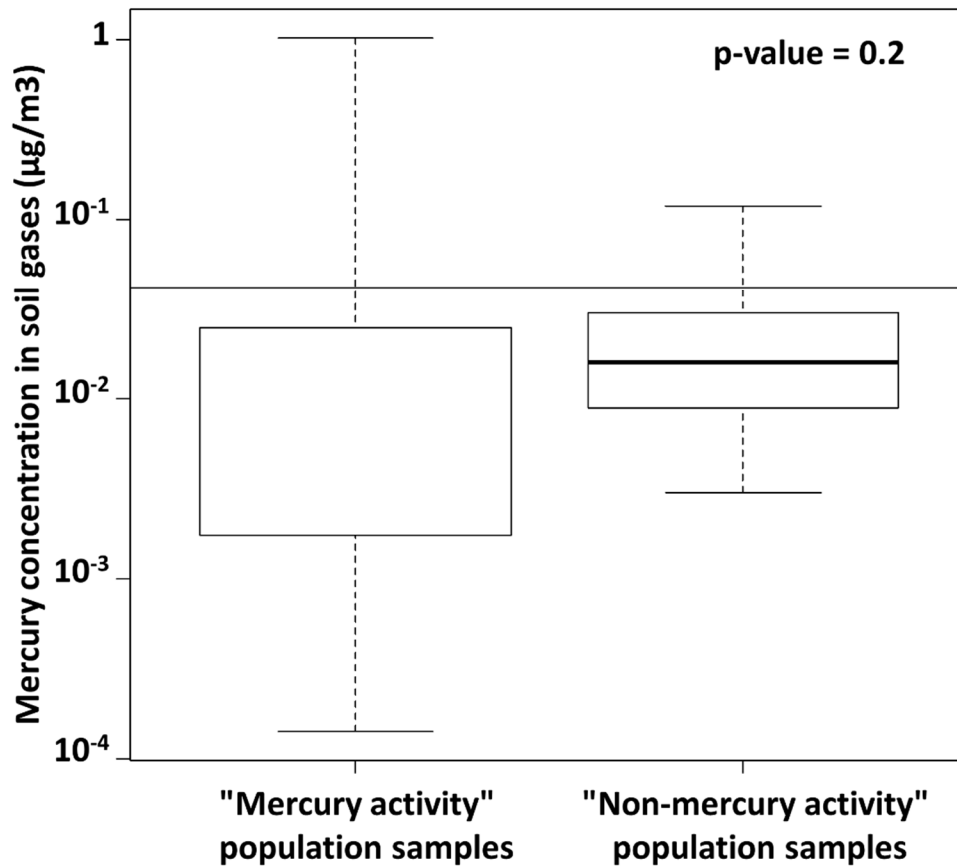


Figure 3: Box plot of mercury concentrations measured in soil gases at a depth of 1 meter and sub-slab air for samples collected in the area of influence of at least one former potentially mercury-using activity (Mercury activity population) and in the area of influence of no potentially mercury-using activity (Non-mercury activity population). These statistics were obtained after data processing by Kaplan-Meier analysis. The horizontal line is the highest limit of quantification for all samples. Because of the high rate of non-quantification for the Mercury activity population, the median for this population cannot be calculated.

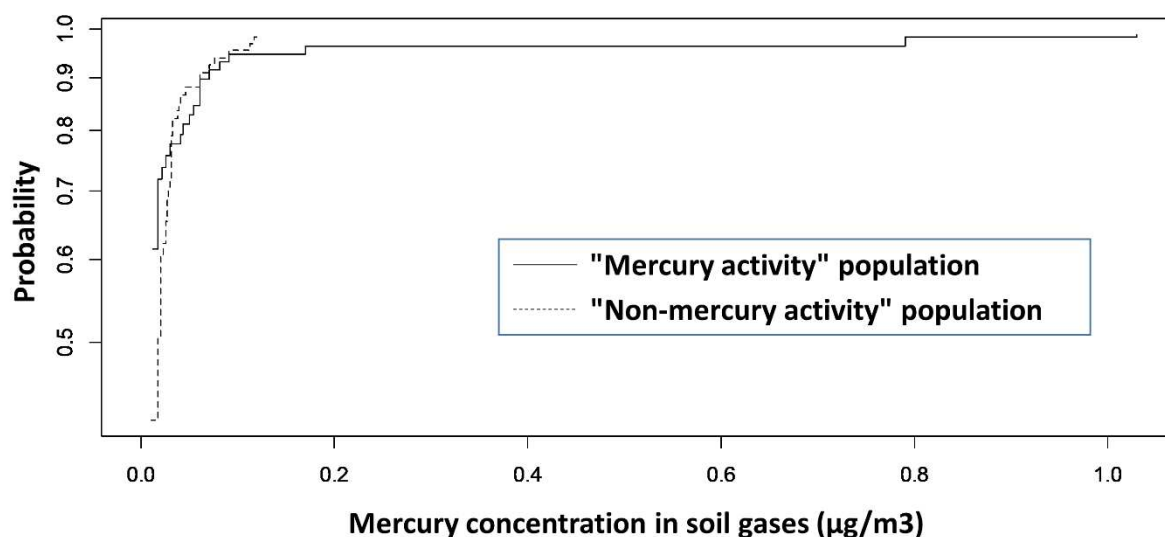


Figure 4: Survival curve obtained following Kaplan-Meier analysis.

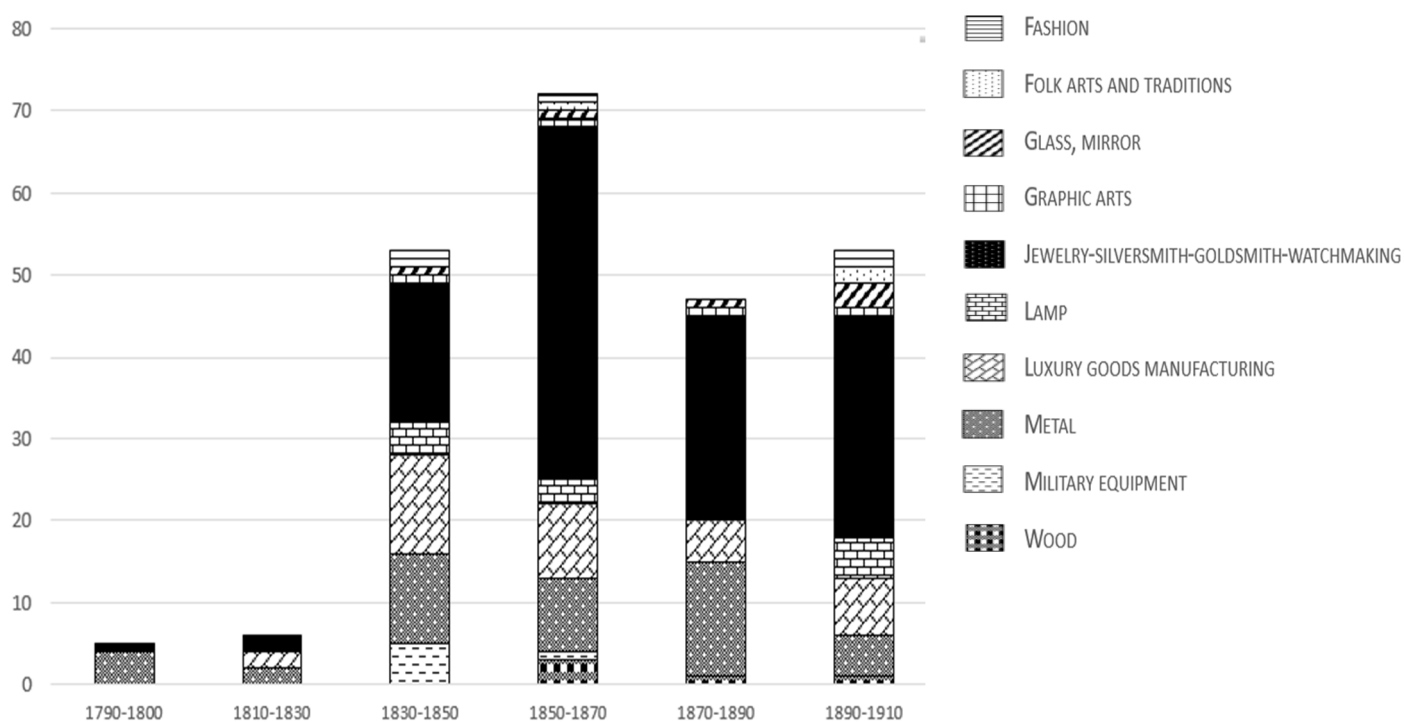


Figure 5: Density of artisanal mercury-using activities (a total of 236 activities) within a 30 m radius around the establishment "Case No. 2" (categories are detailed in supporting information, Table SI-3).

Tables

	Mercury Activity Population	Non-mercury activity Population	Total
Quantified mercury	18	31	49
Unquantified mercury	40	35	76
Total	58	67	125

Table 1: Contingency table showing the number of quantifications/non-quantifications of mercury in soil gases and sub-slab air depending on whether or not measurements are made in the area of influence of a former potentially mercury-using activity.

	Mercury activity population	Non-mercury activity population
Number of samples:	58	67
Number of samples for which the concentration is < LQ	40	36
Median ($\mu\text{g}/\text{m}^3$)	Not calculated	0.017
Mean ($\mu\text{g}/\text{m}^3$)	0.058	0.030
Standard deviation ($\mu\text{g}/\text{m}^3$)	0.17	0.024
Minimum ($\mu\text{g}/\text{m}^3$)	$1.3 \cdot 10^{-4}$	$2.9 \cdot 10^{-3}$
Maximum ($\mu\text{g}/\text{m}^3$)	1.03	0.12

Table 2: Descriptive statistics of measured mercury concentrations ($\mu\text{g}/\text{m}^3$), depending on whether the sampling points are located in the area of influence of a former potentially mercury-using activity (LQ = limit of quantification). These statistics were obtained after data processing by Kaplan-Meier analysis.

Name of principal	Business	Years for which activity was recorded on almanacs	Estimated duration of activity
Claverie	Horologist	1861-1862	2 years
Barbier	Horologist	1864-1870	7 years
Delfour	Horologist	1870	Between 1 and 6 years
Joffre	Chemicals	1899-1908	Between 10 and 15 years

Table 3: List of artisanal activities that may have used mercury around Case No. 1 (within a 30 m radius around the establishment).

Name of owner of industrial site	Activity	Years of activity recorded in available almanacs	Estimated duration of activity
Lecuyer	Ironmonger and lamp maker	1837-1864	Between 28 and 36 years
Jouvin	Jeweler	1870	Between 1 and 6 years
Husson	Mirror maker	1870-1875	Between 6 and 11 years
Roy	Painter on porcelain	1864-1876	Between 13 and 15 years
Meny-Mouton	Button maker	1857-1876	19 years
Gravereau	Painter/glazier	1875-1876	Between 2 and 5 years
Richard	Cabinetmaker	1873-1878	Between 6 and 8 years
Lignière	Cabinetmaker	1864	Between 1 and 3 years
Hiart Aîné	Button maker	1864	Between 1 and 3 years

Table 4: List of artisanal activities that can use mercury around Case No. 3 (within a 30 m radius around the establishment).

« Classic » or
« complete » historical
researches



Presence of former
mercury-using activities ?

Artisanal or
industrial activities



Presence of Hg
in soil gases



Absence of Hg in
soil gases

Systematic
mercury analysis
in soil gases

