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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

1
2
3 Volatile pollutants from former industrial sites can degrade the buildings' indoor air quality that were
4 built after the industrial activities. Since 2010, environmental assessments have been conducted in
5 French establishments hosting sensitive populations identified as being on or near potentially
6 contaminated former industrial sites. These projects are based on historical studies traditionally carried
7 out as part of managing contaminated sites, to determine which substances should be analyzed. They
8 pinpoint former activities likely to have stored or used pollutants. We show that the historical
9 information collected is not effective in targeting sites with increased probability of mercury being
10 present in soil gases. Environmental history has demonstrated the existence of large-scale artisanal
11 contamination, both prior to and concomitant with the industrial era. Classic historical studies would not
12 take into account artisanal activities, which are less documented than industrial activities. We carried out
13 additional research for three schools located in three different Parisian districts. Although information on
14 activities which could have emitted mercury was relatively imprecise (in terms of location, type and
15 duration of activities) and uncertainties exist about the completeness of the archival documents
16 available, our investigations identified several mercury-using activities that had not been identified
17 during the classic historical study. However, we have shown that the number of activities identified does
18 not provide information on how mercury has affected soil gas. Consequently, although a more extensive
19 historical research improves knowledge about the presence of potential mercury-using activities, our
20 study shows that a systematic analysis of mercury as part of the assessment of establishments hosting
21 sensitive populations remains relevant. This approach should be applied to other cities around the world.

22

Abbreviations

23
24

- 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

41 **1 Introduction**

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

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

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

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

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

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

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

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

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

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

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

140 **2 Materials and methods**

141 **2.1 Sites**

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

149 **2.2 Sampling**

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

157 It should be noted that, as part of the national diagnostic approach, the establishments were subject to
158 additional sampling, particularly in indoor air, where the estimated concentration in the living areas
159 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
160 meter and in sub-slab air by a dilution factor depending on building characteristics (elevator cages,
161 piping, cracks in concrete slabs, ventilated crawl spaces, etc.) (Derycke et al., 2018). The value of 0.03
162 $\mu\text{g}/\text{m}^3$ corresponds to the chronic toxicity reference value (TRV) defined by the OEHHA in 2008 (Bisson
163 and Marlière, 2020; OEHHA, 2015). In case of additional sampling, the soil gas sampling and analysis
164 were repeated at the same points (Figure SI-2). Several mercury concentrations may therefore be
165 available for one sampling point, at different dates.

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

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

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

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

185 **2.3 Analysis**

186 Analyzes were carried out by two different laboratories.

187 **2.3.1 Chemicals**

188 All the chemicals used by the laboratory 1 were :

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

196 All the chemicals used by the laboratory 2 were :

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

201 **2.3.2 Method of analysis**

202 Laboratory 1:

203 The extraction on hopkalite tubes was performed according to an in-house method based on EN-ISO
204 15587-1. The solution was made with aqua regia. Mercury concentrations were determined by atomic
205 fluorescence spectrometry (AFS Analytik Jena Analyser) according to an in-house method based on ISO
206 16772. The different concentrations of the calibration standards were prepared from a 1 g/L commercial
207 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
208 recovery rate was 90 %.

209 Laboratory 2:

210 The solution from hopkalite tubes was made with aqua regia. Mercury concentrations were determined
211 by atomic absorption spectrometry (FIMS-400 Perkin-Elmer Analyser) according to the NF EN 13211
212 standard. The different concentrations of the calibration standards were prepared from a 1 g/L
213 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
214 recovery rate was 92-107 %.

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

220 **2.4 Statistical data processing**

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

224

225 **2.4.1 Kaplan-Meier analysis**

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

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

234 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 |

235 The rank of repeated values is equal to the median of the ranks if they were different. The three values
236 "8", whose ranks should have been 6, 7 and 8, are assigned the rank of 7, i.e. the median of the three
237 ranks. The sum of the ranks is thus preserved (a statistical rule used in many tests). Similarly, the four
238 lowest values are re-censored as below the LQ of 5, and then assigned the rank of 2.5 as the median of
239 ranks 1, 2, 3 and 4 (which would have been assigned to uncensored values). "3" is a detected value, but
240 not knowing whether " < 5 " is equal to for instance 4 or 2.5, "3" cannot be considered as a detected
241 value.

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

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

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

253 **2.4.2 Peto-Prentice Test**

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

261 **2.5 Historical studies**

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

267 2.5.1 Classic historical study

268 The classic historical study consisted in consulting:

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

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

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

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

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

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

307 **2.5.2 Comprehensive historical study**

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

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

314 *2.5.2.1 Sources consulted to make the historical study more comprehensive*

315 The literature on environmental history provides a better understanding of the artisanal and/or
316 industrial context of Paris and how it changed. We consulted several vintage printed sources on Paris
317 that gave us a general view of the capital's artisanal and industrial activity. For example, Emile de La
318 Bédollière (La Bédollière, second half of the 19th century) provides insights into the how the districts
319 looked in the second half of the 19th century. We also carried out research at archival sources, including
320 certain archives at the City of Paris useful for the study of land parcels and construction and that are not
321 used in the classic historical studies, such as the "Public Health Register" and the "Real Estate Ledger".
322 The "Public Health Register" provides information on how buildings were occupied, including the
323 identification of industrial or commercial establishments. Detailed public health records (sub-series
324 3589W - Paris. Direction chargée du logement [Housing Directorate]) are the result of a health survey
325 campaign of Parisian buildings begun at the end of the 19th century. The Paris real estate ledger (sub-
326 series DQ18 - Directorate of Registration, Stamp and Domains) lists the essential information on land
327 transfers in Paris: addresses, dates and causes of transfers, names of owners and notaries) between
328 1809 and 1945.

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

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

340 By the mid-18th century, the effects of mercury on workers' health had been proven by medicine
341 (Guillermé, 2007a). We consulted sources related to occupational health and safety, the outcome of the
342 1892 establishment of the Labor Inspectorate. Our focus was on the "Occupational Diseases Table",
343 which has included occupational mercury poisoning (hydrargyris) since 1919. In particular, the Labor
344 Inspection Bulletins ("Bulletin de l'Inspection du travail" BIT) and then the Labor Inspection and Industrial
345 Hygiene Bulletins ("Bulletin de l'Inspection du travail et de l'Hygiène Industrielle" BITHI), available on the
346 Ministry of Labor website (Ministry of Labor, 2020), were drafted for the period 1893 to 1936. The Labor
347 Inspection and Industrial Hygiene Bulletins compiled official acts relating to labor law and regulation
348 (occupational health and safety) as well as technical reports from inspectors' field investigations,
349 particularly in industrial establishments.

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

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

356 *2.5.2.2 Identification of activities identified as using mercury*

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

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

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

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

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

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

387

388 *2.5.2.3 Application of the comprehensive historical study to three establishments hosting* 389 *sensitive populations*

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

395 For these three establishments, we looked for artisanal activities within a 30-meter radius, in accordance
396 with the Guidance for Evaluating the Vapor Intrusion to Indoor Air Pathway from Groundwater and Soils
397 (US EPA, 2002). From the former land registries of the districts of Paris, the addresses (numbers and
398 names of streets) included within this perimeter were identified. The Parisian land registry has changed
399 multiple times between 1810 (The first Napoleonic land registry was ordered by the law of September
400 15, 1807. The plan of the various housing parcels is available for consultation on the Paris Archives) and
401 today. Land registries before 1860 (Archives de Paris, n.d.), after 1860 (date on which Paris annexed
402 several peripheral communes and made them districts), and the current land registry were taken into
403 account (<https://cadastre.data.gouv.fr/>) (Etat français, n.d.). Research had done on activities identified
404 as potentially mercury-using (in supporting information Table SI-3) and carried out on selected addresses
405 in commercial almanacs between 1798 and 1907. An activity is retained within a given time interval if it

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

412 **3 Results**

413 **3.1 Mercury concentrations in soil gases and sub-slab air**

414 Mercury concentrations above the limit of quantification were measured at 49 of the 125 sampling
415 points (i.e., a quantification rate of about 39%). The mean of concentrations above the quantification
416 limit is $0.08 \mu\text{g}/\text{m}^3$ (i.e., greater than the TRVs applied in closed environments) and the associated
417 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
418 44 sites studied, mercury concentrations were all above the limit of quantification. Conversely, 17 sites
419 showed mercury concentrations below the limit of quantification. The other 17 sites had at least one
420 mercury concentration lower than the limit of quantification and one higher. Per site, the number of
421 points with mercury concentrations above the LQ was between 0 and 3. Figure 2 shows the
422 concentration distribution at the various sites.

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

428 protective exposure hypothesis (12h/d, 220d/y). The health risk calculation was done for each soil gas
429 measurement that showed mercury concentrations above the limit of quantification, by estimating
430 indoor air concentrations thanks attenuation factors (Derycke et al., 2018). HQ values are between 1.7
431 10^{-3} and 1.03 (Table SI-1). Only 1 sample at one establishment exhibits an HQ value exceeding 1.
432 However, these results are strictly informative considering that reliable health risk calculations should be
433 assessed on the basis of the mercury concentrations directly measured in the indoor air of buildings,
434 rather than estimations derived from soil gas concentrations.

435 **3.2 Relationship between mercury concentrations measured in soil gases and** 436 **former mercury activity listed by the classic historical study**

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

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

445 The ranges (min, max, mean) of the limits of quantification for the 2 populations are similar (respectively
446 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
447 the “Mercury activity” population).

448 Contrary to what might have been expected, the number of samples where mercury was quantified was
449 greater in the Non-mercury activity population than in the Mercury activity population (respectively, 32
450 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

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

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

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

492 **3.3 Taking the classic historical study further**

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

496 3.3.1 Artisanal and industrial history of Paris until the 19th century

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

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

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

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

527 **3.3.2 Comprehensive historical study on three Parisian schools**

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

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

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

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

544
545 The three selected establishments, named Case 1, Case 2 and Case 3, are in the 15th, 3rd and 10th
546 districts, respectively. The comprehensive historical study focused on the artisanal activities listed in in
547 supporting information (Table SI-3).

548 Case No. 1

549 Case No. 1 is in the 15th district. The establishment concerned was located in the "Grenelle Plain" (Huard,
550 1790), which was, until the middle of the 19th century, intended for agriculture. As Paris expanded, this
551 more out-of-town district experienced a development of more recent artisanal activities, in connection
552 with the annexation of Grenelle into Paris in 1859. Historical research in this area has identified a few
553 activities within a 30 m radius of the establishment that may have used mercury. Three horologists
554 having exercised between 1861 and 1870, with a duration of activities between 1 and 7 years and a
555 chemical factory (between 10 and 15 years of activity) were identified (Table 3). This activity is retained
556 with reservations, given the little information obtained as to the nature of the products manufactured.
557 Mercury was not quantified in soil gases at this establishment ($<0.018 \mu\text{g}/\text{m}^3$).

558

559 Case No. 2

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

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

569 Mercury is quantified in soil gases at the establishment, but concentrations are relatively low (up to
570 $0.021 \mu\text{g}/\text{m}^3$, i.e., less than the TRV of $0.03 \mu\text{g}/\text{m}^3$) in light of the school's environmental history.

571 Case No. 3

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

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

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

587 3.4 Discussion

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

590 The finding that mercury is measured in soil gases in many establishments, while the classic historical
591 study does not identify the presence of old mercury-using activities near these establishments, means it
592 will not be possible to effectively target sites for which the likelihood of mercury in soil gases is increased
593 by searching for historical information in accordance with the French methodology for the management
594 of contaminated sites and soils (MEEM, 2017). This can be explained by the number or the
595 contemporaneous period of information sources consulted (historical aerial photographs, BASIAS
596 database, local and municipal archives and BASOL interviews and database). The mere consultation of
597 these sources of information may limit the number of sites identified and the determination of their
598 location. Other countries such as the United Kingdom speak of industrial archeology and stress the
599 importance of old maps locating industries, produced by The Ordnance Survey (OS), from 1834,
600 developed for all the cities. These cards do not necessarily exist for French cities (RPS Consultants Ltd,
601 1994). As a result, some sites potentially causing mercury degradation of soil gas quality may be ignored
602 because they are unidentified or incorrectly positioned. It could be happened in the case of conversion
603 from old industrial building to residential use when insufficient information is available on past uses
604 (Gochfeld, 2003; World Health Organization, 2010). This is reinforced by the fact that the national
605 diagnostic process being carried out systematically throughout the territory, does not necessarily take
606 into account the historical specificities of each territory, in which each establishment is located. For
607 Paris, the BASIAS inventory lists industrial sites whose activities occurred between 1835 and 2005 (Faury,
608 2007). This period therefore does not cover the previous period (notably the 18th and early 19th
609 Centuries) during which mercury-using activities were numerous. In addition, choices have been made
610 regarding types of activities recorded in BASIAS. The activities identified as a priority in the inventory

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

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

617 By consulting complementary archives as part of the comprehensive historical study for three
618 establishments we identified activities that could have used mercury within a 30 m radius. Among the
619 documents consulted, the almanacs and the old Parisian land registry proved to be the most useful
620 sources. The presence of these activities, including a hospital, jewelers, button makers (metal in Figure 5)
621 and lamp makers could explain the quantification of mercury in soil gases in 2 of the 3 establishments
622 studied (Case 2 and Case 3). However, the number of activities identified does not provide information
623 on the level of degradation of soil gas quality by mercury. The number of activities identified that could
624 have used mercury within a 30 m radius is much higher in Case 2 (209 activities) than in Case 3 (9
625 activities and the hospital) whereas the maximum concentrations measured in soil gases are higher in
626 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
627 identified in more extensive historical research near Case 1, yet mercury is not quantified in soil gases.
628 Mercury is used in different chemical forms in processes (chloride mercury, calomel, etc.) (Guérin et al.,
629 2014; Wang et al., 2012). Some forms are not volatile (Mason et al., 1994; O'Connor et al., 2019) and, as
630 a result, if conditions are favorable (Thomassin et al., 2003), may not be found in soil gases. Once
631 deposited to the terrestrial environment, Hg preferentially adsorbs to clays and organic matter
632 (Varekamp et al., 2003). Furthermore, in Lee et al. (2009) events potentially affecting children were most
633 frequently classified as non-volatilization events. Moreover, not all identified activities have necessarily
634 caused degradation of soil gas quality, either because they have never used mercury, or have used it in

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

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

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

663 ***What is the solution?***

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

673 ***International concern***

674 The results of this work can be transposed into other French or European cities which have had intensive
675 artisanal and/or industrial activities that used mercury. First banned from use at the beginning of the
676 18th century in England and France, mercury was reintroduced in various processes (Mathews David,
677 2019) as in Lyon, "*With Paris, Lyon was then the only city in France authorized to carry out the refining of*
678 *precious metals (...) they also turned gold to antimony or to mercuric chloride*", or in Marseille "*(...)*
679 *banned in the 17th century, surreptitiously introduced in Paris in 1730, mercury acquired its legitimacy in*
680 *Marseille (...)*" (Leroux, 2014). Similarly, cities around Paris, including Senlis, Chartres, Fontainebleau,
681 Versailles and Meaux secreted it (Guillerme, 2003). Other European cities also used a lot of mercury,
682 such as London (Mathews David, 2019), and Birmingham and Bath specializing in button making

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

689 **4 Conclusion**

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

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

699 From deeper historical research around the three schools located in three different Parisian districts, we
700 can locate the presence of artisanal and industrial activities that potentially emitted mercury and were
701 not identified in the classic historical study. Our analysis also makes it possible to estimate how long
702 these activities lasted at the addresses identified. In that respect, having expanded the sources of
703 information consulted was fruitful, providing interesting complementary elements compared to a classic
704 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

912 **Figure 1.** Distribution of maximum concentrations for each of the 44 geographical sites (corresponding to
913 52 early childhood establishments) (in $\mu\text{g}/\text{m}^3$). Sites with concentrations below the limit of
914 quantification are identified by a star. The three selected Parisian establishments for a
915 comprehensive historical study are numbered.

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

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

923 **Figure 4.** Survival curve obtained following Kaplan-Meyer analysis.

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

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929

Tables

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

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

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

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

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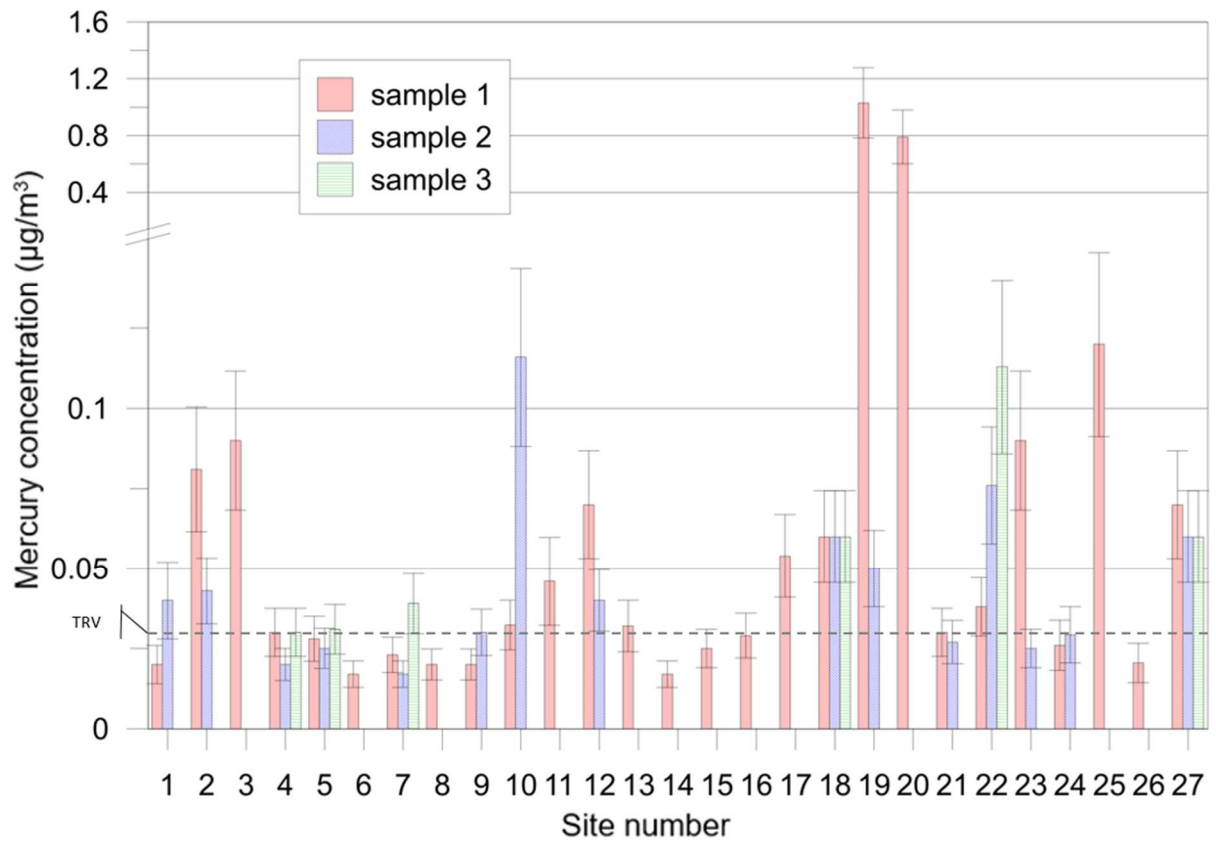


Figure 2: Concentrations ($\mu\text{g}/\text{m}^3$) of mercury at the 27 sites where one concentration at least is higher than the quantification limit (Logarithmic scale. The error bars represent the analytical uncertainty). The toxicity reference value (TRV) is represented by the dotted line.

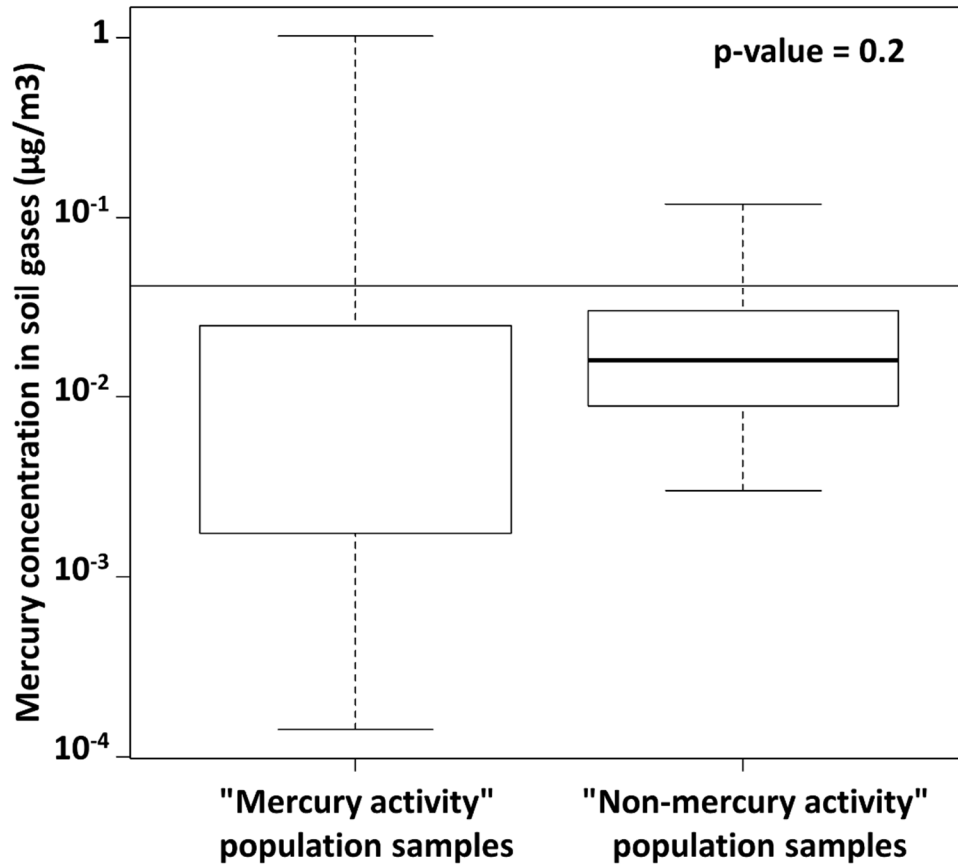


Figure 3: Box plot of mercury concentrations measured in soil gases at a depth of 1 meter and sub-slab air for samples collected in the area of influence of at least one former potentially mercury-using activity (Mercury activity population) and in the area of influence of no potentially mercury-using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

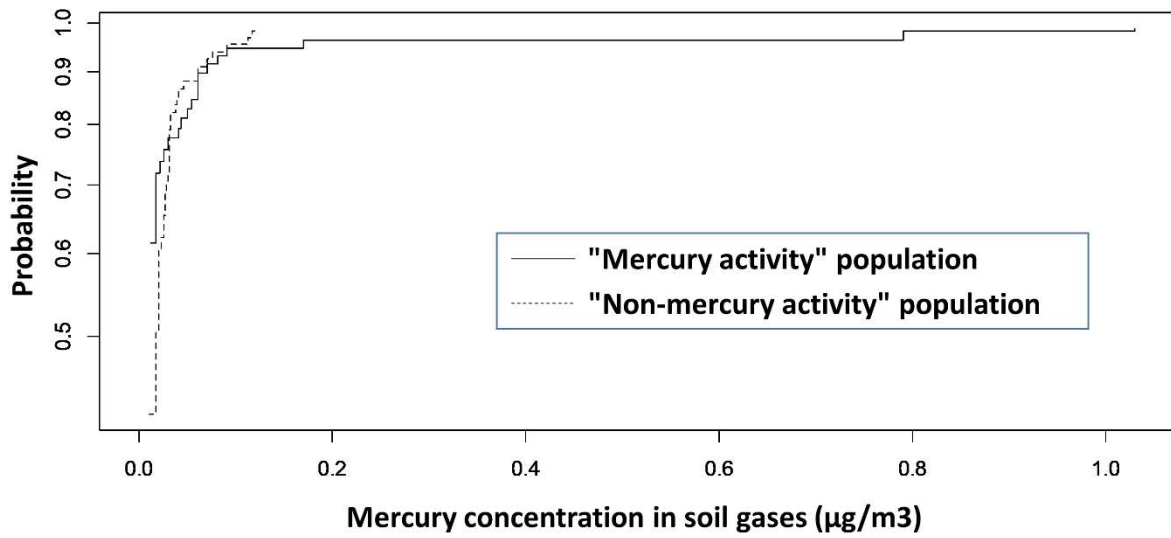


Figure 4: Survival curve obtained following Kaplan-Meier analysis.

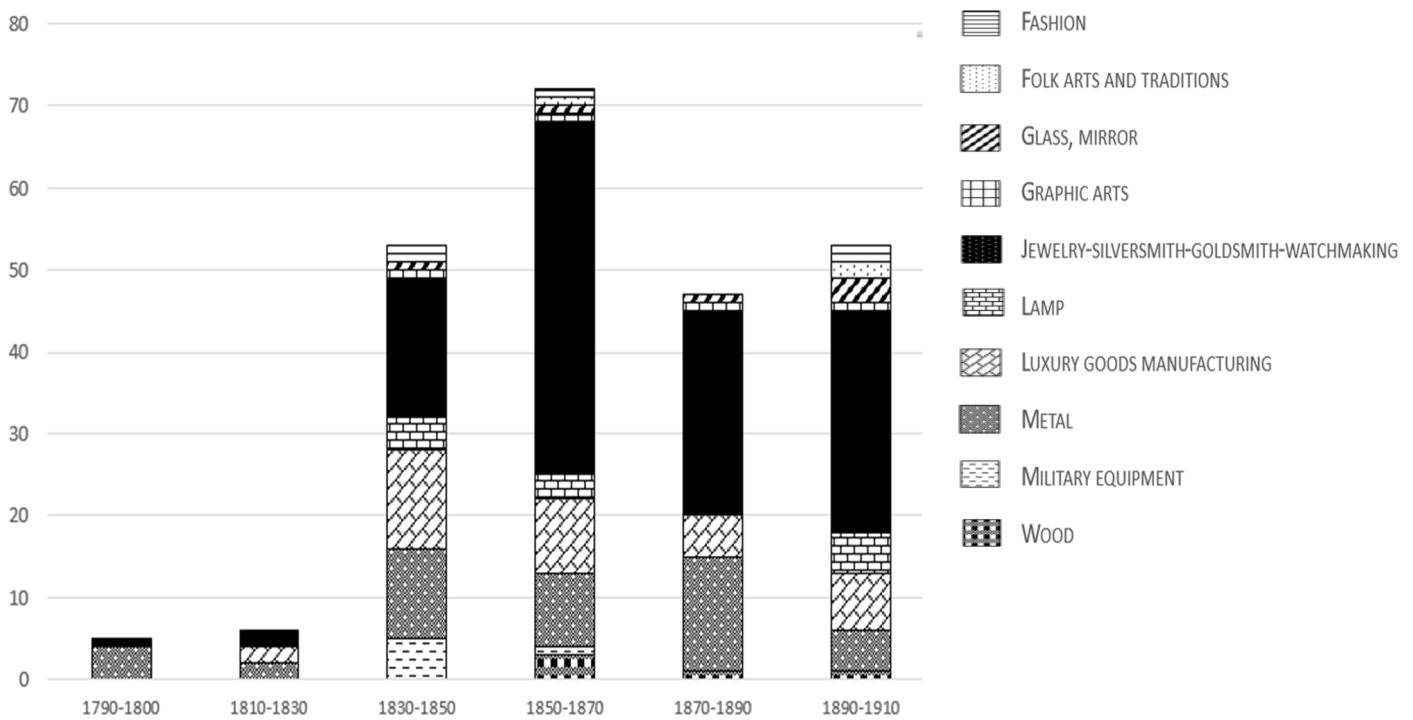


Figure 5: Density of artisanal mercury-using activities (a total of 236 activities) within a 30 m radius around the establishment "Case No. 2" (categories are detailed in supporting information, Table SI-3).

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 Ainé | 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

