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## Historical monuments and lead pollution: Lessons from the case of Notre-Dame de Paris and Sainte-Chapelle

Justine Briard, Sophie Ayrault, Sandrine Baron, Louise Bordier, Matthieu Roy-Barman, Delphine Syvilay, Aurélia Azéma, Maxime L'Héritier

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# 1 Historical monuments and lead pollution: lessons from the case of

## 2 Notre-Dame de Paris and Sainte-Chapelle

### 4 Abstract

5 To investigate the potential spreading of leaded dust over Paris and its inhabitants,  
6 following the fire of Notre-Dame de Paris cathedral in April 2019, we developed a dedicated  
7 protocol to identify the geochemical fingerprint of the emitted particles. This method could  
8 be transposed to a contemporary monument to Notre-Dame de Paris which had not burnt  
9 and which had to undergo a major restoration phase: Sainte-Chapelle de Paris. Thus, a new  
10 study was performed on the indoor dust collected in this monument, to which the protocol  
11 developed for the dusts of Notre-Dame was applied. The comparison of the geochemical  
12 fingerprint of both monuments highlights the complexity of establishing the chemical  
13 signatures of such buildings. The homogeneity of the isotopic and elemental results obtained  
14 on the samples from Notre-Dame de Paris compared to those from Sainte-Chapelle de Paris  
15 reveals the homogenizing role of the fire, which simplifies the geochemical signature  
16 determination. Moreover, this study does not show any significant impact of anthropogenic  
17 pollution (road traffic, industry, incinerators) on the indoor environment of monuments,  
18 which are nevertheless rich in construction lead. The notion of "polluter-polluted monument"  
19 could be problematic in the event of an accident during restoration phase. These new results  
20 obtained on dust samples from a building that has not experienced any recent major  
21 deterioration underlines the multiple sources of lead present in these monuments and the  
22 importance of identifying univocally their signature. However, the concentrations of metal  
23 and metalloid in the indoor dust do not show any major pollution inside Sainte-Chapelle de  
24 Paris that could be a threat to human health.

25  
26 **Keywords:** Sainte-Chapelle de Paris; Notre-Dame de Paris; indoor dust; environmental  
27 pollution; metals; geochemical fingerprint

## 29 1. Introduction

30 The fire of Notre-Dame de Paris (hereafter referred to as Notre-Dame) that occurred  
31 in 2019 devastated an important part of its lead roof and spire. It resulted in many  
32 controversies in the media on the potential lead pollution caused by such an incident and its  
33 impact on the health of local residents, especially children [1–5]. Citizen associations and two  
34 families accused authorities to not act adequately to prevent children exposure to the lead  
35 fallout. Lead poisoning is a major societal issue. Even if leaded gasoline phasing out in the  
36 2000s in France drastically reduced concentrations in the environment [6,7], many sources of  
37 lead are reported in urban environments such as ancient foundries, incinerators or road traffic  
38 but even more in Paris city where lead was greatly used for roofing, plumbing or painting and  
39 this, until the beginning of the 21<sup>st</sup> century [8]. Historical monuments are an important part  
40 of French cultural heritage and require special care during their maintenance and renovation  
41 phases to limit possible metal pollution in the urban environment. However, following the fire  
42 of Notre-Dame, many questions were raised about the impact of a historical monument (e.g.  
43 Eiffel Tower) [9] considered as a “potential emitter” of contaminants, in particular in metals /  
44 metalloids harmful to human health, towards its proximal environment. Indeed, does a  
45 historical monument containing lead and other toxic metals pollute its environment  
46 continuously or only during degradation events (e.g. natural disasters, fires, restoration  
47 phases, lead oxidation with ambient air and acid rain)?

48 To answer this question, a first study was performed on indoor dust samples collected  
49 in Notre-Dame after the fire to determine their geochemical signature [10]. This last was  
50 approximated from tracers commonly used in the literature to determine the sources of  
51 environmental pollution: lead isotopic ratios ( $^{206}\text{Pb}/^{207}\text{Pb}$ ;  $^{208}\text{Pb}/^{206}\text{Pb}$ ) [11–14] and elemental  
52 ones (Sn/Cu; Sb/Cu; Cd/Pb; Cu/Pb) [15,16]. Our study highlighted the heterogeneity of  
53 isotopic and elemental signatures depending on the matrix collected and the selected  
54 substrate (wood; fabric; metals; stained-glass). Therefore, a rigorous sampling strategy inside  
55 a building (non-contaminating substrate, fine and homogeneous matrix) is required to define  
56 the geochemical signature of a historical monument [10]. To understand the impact of a  
57 degradation event on a historical monument signature, we will propose a geochemical  
58 signature of Sainte-Chapelle de Paris (hereafter referred to as Sainte-Chapelle) indoor dusts,  
59 a contemporary monument to Notre-Dame (construction period between the 12<sup>th</sup> and 13<sup>th</sup>

60 centuries) subjected to similar urban pollution pressures and also covered with a lead roof.  
61 This monument is located 400 m northwest from Notre-Dame in the centre of Paris, on the  
62 “Ile de la Cité”. In addition, these two monuments underwent many restoration phases over  
63 the centuries. The Sainte-Chapelle built in the middle of the 13<sup>th</sup> century went through many  
64 modifications such as the installation of its rose window in the 15<sup>th</sup> century, the repairs  
65 following its fire in the 17<sup>th</sup> c. or another important restoration phase, especially for the roof,  
66 performed in the 19<sup>th</sup> century [17]. Since 2008, restoration of the masonry and stained-glass  
67 windows of the east, north and west (with the frontage rose window) was carried out by the  
68 architectural conservator, *i.e.* Architecte en Chef des Monuments Historiques (ACMH).  
69 Concerning Notre-Dame, construction started in the second half of the 12<sup>th</sup> century and lasted  
70 till the mid-13<sup>th</sup> century. The cathedral roof was entirely renovated in the 18<sup>th</sup> century. During  
71 the 19<sup>th</sup> century, Notre-Dame was extensively restored and its emblematic spire made of  
72 wood covered with lead sheets overhanging the once again renovated lead roof was erected.  
73 A few parts (roof top and spire ornaments) were also restored during the 20<sup>th</sup> century [18].

74 Thus, the present study focuses on indoor dust samples, collected in Sainte-Chapelle  
75 before the installation of the scaffolding necessary for its restauration planned in 2022. We  
76 applied the protocol and geochemical tracers, previously used for the study of Notre-Dame,  
77 in order to define Sainte-Chapelle geochemical fingerprint and compare it with Notre-Dame  
78 signature [10] to better understand the lead pollution originating from such environment.

## 79 **2. Research Aim**

80 This study focuses on the contribution of historical monuments, in particular Notre-  
81 Dame and Sainte-Chapelle, to the lead contamination of urban environments, with a specific  
82 application to the city of Paris. The project aims more broadly to identify the multiple sources  
83 of lead pollution in this urban environment using lead isotopic and elemental tracing. Our  
84 previous work on the fire of Notre-Dame de Paris demonstrated the ability of elemental and  
85 lead isotope ratios to discriminate between these different lead sources in the Parisian  
86 environment [10]. The project’s goal is to refine the tracing of Parisian lead by establishing the  
87 chemical fingerprint of a historical monument, having suffered no recent damage as Sainte-  
88 Chapelle, representative of the different times and uses, to tend towards a historical  
89 retrospective of the impacts of construction leads, relying on natural records of environmental  
90 contamination. Ultimately, these studies on historical monuments will help to clarify the real

91 risks of lead propagation from these edifices and will allow the addition of quantified elements  
92 to the necessary discussion to adapt the standards for carrying out restoration work. Indeed,  
93 in the presence of lead in a monument under restoration works, the “lead protocol” for safety  
94 and health at work has to protect the health of the workers and of the inhabitants of the  
95 surrounding buildings, while avoid imposing disproportionate actions that thread the welfare  
96 of the workers (such as wearing heavy personal protection equipment) and the success of the  
97 restoration project.

### 98 **3. Materials and Methods**

#### 99 *3.1. Material study strategies*

100 Defining the geochemical lead signature of a historical monument is not a  
101 straightforward question as many different lead uses coexist within each monument. Different  
102 kinds of construction leads (sealing, stained glass, roofing) were implemented during the  
103 edification or later restoration of the monument. Yet, both the isotopic and elemental  
104 signatures can differ significantly between materials depending on the type of lead  
105 construction used and its time of implementation. Indeed, the geochemical signature of pure  
106 lead varies according to the lead source (ores sources) but also depends on lead mixing which  
107 could be operated on the building site itself. This is particularly obvious during the upkeep or  
108 restoration phases of the roof, when lead is salvaged and recycled into new lead sheets. Lead  
109 supply sources are also much likely to evolve over time, throughout the Middle Ages and the  
110 modern period. Major differences in lead elemental signatures was for example highlighted  
111 in Beauvais, Bourges, Chartres and Metz cathedrals regarding lead sealing and roof sheets  
112 [19,20]. Thus, the construction of a given monument induces multiple lead signatures which  
113 correspond to different lead supplies. In addition, paintings, sculptures and organs are also  
114 potential lead sources in such monuments, each of them having its own signature.

115 Indoor dusts present monument can be analysed to define the historical lead signature  
116 knowing that this material is currently used to determine the Pb signature of potential  
117 exposure [21–24]. Surprisingly, there is no such study dedicated to historical monuments,  
118 although lead pollution is a concern in many restoration yards. This might be partly explained  
119 by the short time spent by most of the population in such indoor environments. Nevertheless,  
120 indoor dusts host multiple lead sources present inside the building (e.g. construction lead,

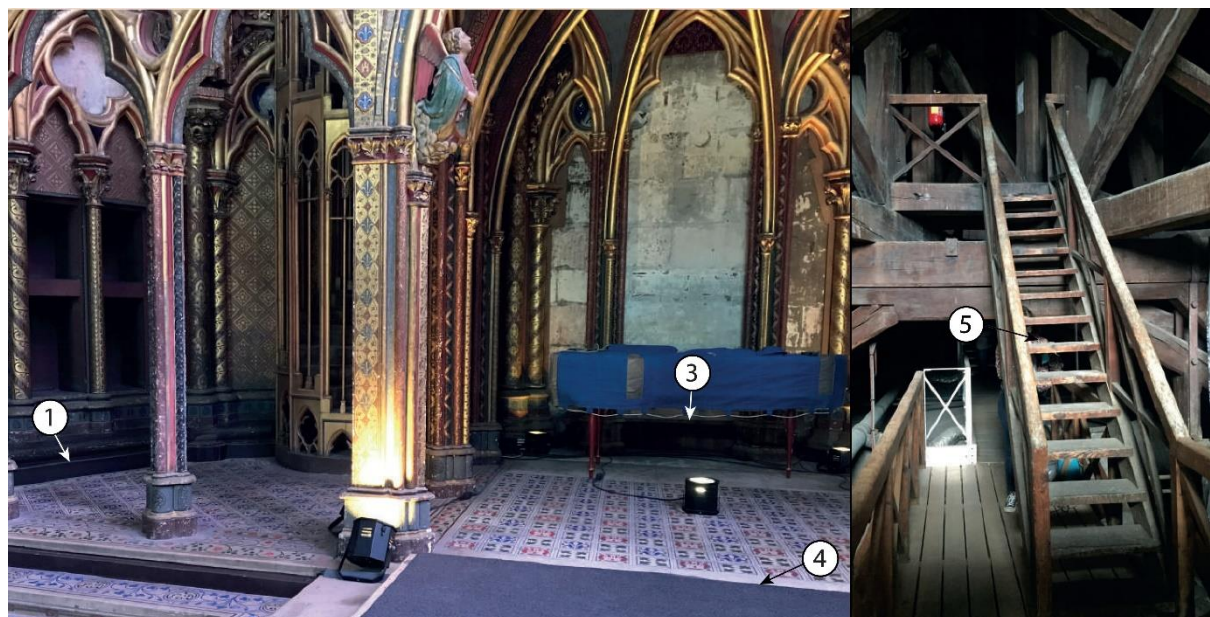
121 paints, stained glass) but they can also be mixed with the surrounding lead signature,  
122 especially in urban areas. Multiple lead sources and other metals are indeed recorded in large  
123 cities, and historical monuments are more affected by lead pollution in such environments  
124 than in rural areas where the environmental lead contamination is significantly lower [25].  
125 Thus, the dust collected inside a monument, combining the different sources of lead of the  
126 monument as well as the possible influence of the building's environment, is more  
127 representative of the monument average lead signature than every single construction lead  
128 composing the monument.

### 129 *3.2. Substrate selection and dust collection methods*

130 The substrate choice to collect the indoor dust is an essential step to avoid the  
131 contamination of the samples by the substrate. Sampling dusts on metal, stained glass (that  
132 contains lead) or fabrics (which may also contain metals found in pesticides for cotton  
133 production and in dyes) must be avoided [26,27]. In this study, we favoured inert substrate  
134 such as wood, plastic and rubber in which metals and metalloids concentrations are  
135 considered negligible.

136 Several dust sampling techniques can be used, such as wet sampling on tissue. The  
137 "wipe protocol" of the AFNOR NF X46-032 standard is commonly used by French public health  
138 organizations to determine the surface lead concentrations in house, schools and nurseries  
139 [28,29]. This protocol is also used in many countries like UK or USA [30,31]. However, this  
140 technique of cleaning a known surface, usually 33 x 33 cm<sup>2</sup>, with a wipe is only certified for  
141 lead concentrations, and not for other metals / metalloids concentrations or lead isotopes yet.  
142 Moreover, the wipe protocol provides a surface concentration (i.e. mass of metal per m<sup>2</sup>)  
143 while geochemical studies use mass concentrations (i.e. mass of metal per kg of dust). Finally,  
144 the wipe protocol was developed for smooth surfaces, and is not adapted to rough surfaces  
145 often encountered in monuments. Thus, within this study, dry dust sampling (total dry matter)  
146 was favoured to obtain a sufficient representativeness of the metals / metalloids proportions  
147 composing them. All dust samples from Sainte-Chapelle (n = 4) were collected with a  
148 polypropylene spatula on inert (i.e. metal-free) substrates such as wood, plastic and rubber  
149 (Table 1). Three samples (SC\_070721\_PN, SC\_070721\_PA, SC\_070721\_MF) were collected on  
150 the floor of the upper chapel in the vicinity of the tribune located in the apse, at the eastern  
151 end "tribune", a space of the chapel which is not accessible to visitors. The last sample

152 (SC\_070721\_EF) was collected in the attic above the main vaults, on a wooden staircase  
153 leading to the base of the 19<sup>th</sup> century spire (Fig. 1). The collected material (between 3.0 to  
154 24.6 mg) was then stored in hermetically sealed polyethylene boxes.



155  
156 Fig. 1: **Location of the samplings.** Left, the tribune of the upper chapel. Right, the staircase in  
157 the main attic. 1: SC\_070721\_PN, 2: SC\_070721\_PA, 3: SC\_070721\_MF, 5: SC\_070721\_EF

### 158 3.3 Construction lead collection at Sainte-Chapelle

159 Sampling was performed on the lead joints located between the mullions of the upper bays  
160 of Sainte-Chapelle during their restoration in 2013. Eight lead samples of centimetre size were  
161 cut off from bays 107 & 109 (corresponding to the middle bays on the northern side).

162 Notre-dame construction leads were collected by Glorennec et al [29] mainly on lead elements  
163 from the roof and spire. .

### 164 3.4. Analytical methods

#### 165 3.4.1. Elemental composition analysis

166 Some metals or metalloids are commonly associated with lead (in varying proportions  
167 depending on the source and the recycling degree) in construction lead [19,20,32]. This is  
168 particularly the case for Cu, Ag, Cd, Sn, Sb and Bi. However, these metals are also present in  
169 urban environments linked to road traffic [16] (Cu ; Sn ; Sb) and incinerators [15] (Cd ; Cu ; Pb).  
170 To determine the pollution source of these metals and metalloids, the correlation degree

171 between the metals can be proposed to know whether they come from an only one source or  
172 from multiple sources and to determine the signature from the elemental ratio as Metal/Pb.  
173 These ratios highlight the pollution source origin. Elemental concentration analyses were also  
174 performed in the LSCE Laboratory (Gif-Sur-Yvette, France) with a Thermo Scientific™ iCAP™  
175 TQ ICP-MS following the same analytical protocol as the Notre-Dame dusts [10] where  
176 certified reference materials were used to evaluate the quality of the determined  
177 concentrations [10].

### 178 3.4.2. Lead isotope composition analysis

179 Lead owns four isotopes. Three of them i.e.  $^{208}\text{Pb}$ ,  $^{207}\text{Pb}$ ,  $^{206}\text{Pb}$  are coming from the  
180 radioactive decay of  $^{232}\text{Th}$ ,  $^{235}\text{U}$  and  $^{238}\text{U}$  respectively and one i.e.  $^{204}\text{Pb}$  is stable (its abundance  
181 has remained the same since the Earth formation). Lead isotopes allow the sources tracing in  
182 many different contexts (e.g. historical, geological, environmental...). They can also trace the  
183 different lead sources hosted by historical monuments [10][33,34]. For nearly fifty years, Pb  
184 isotopic ratios have been used to determine the pollution signatures and trace them in the  
185 environment [11,29,35,36]. In this study, we focused on the three most abundant lead  
186 isotopes in the environment:  $^{208}\text{Pb}$ ,  $^{207}\text{Pb}$ , and  $^{206}\text{Pb}$ . From 3.0 to 24.6 mg of Sainte-Chapelle  
187 dust samples were dissolved with 2 mL  $\text{HClO}_4$  (65-71%) and 4 mL HF (47-51%) in closed Teflon  
188 vessels under pressure on hot-plates for 360 min at 150 °C (Digiprep, SCP Science protocol).  
189 After cooling, solutions were evaporated to dryness. Samples were then digested at 120 °C for  
190 180 min (Digiprep SCP Science protocol) using aqua regia: 3.75 mL HCl (34-37%) and 1.25 mL  
191  $\text{HNO}_3$  (67%). Solutions were then evaporated to dryness. Finally, samples were dissolved and  
192 evaporated successively three times in 1 mL of  $\text{HNO}_3$  (67%) and diluted in 50 mL of ultrapure  
193 water (Milli-Q). Analyses were performed with a Thermo Scientific™ iCAP™ TQ ICP-MS in the  
194 Laboratory LSCE (Gif-Sur-Yvette, France) where mass bias and drift of the isotope ratios were  
195 corrected based on repeated measurements of the Pb reference material NIST SRM-981  
196 [10,12,37].

## 197 **4. Results and discussions**

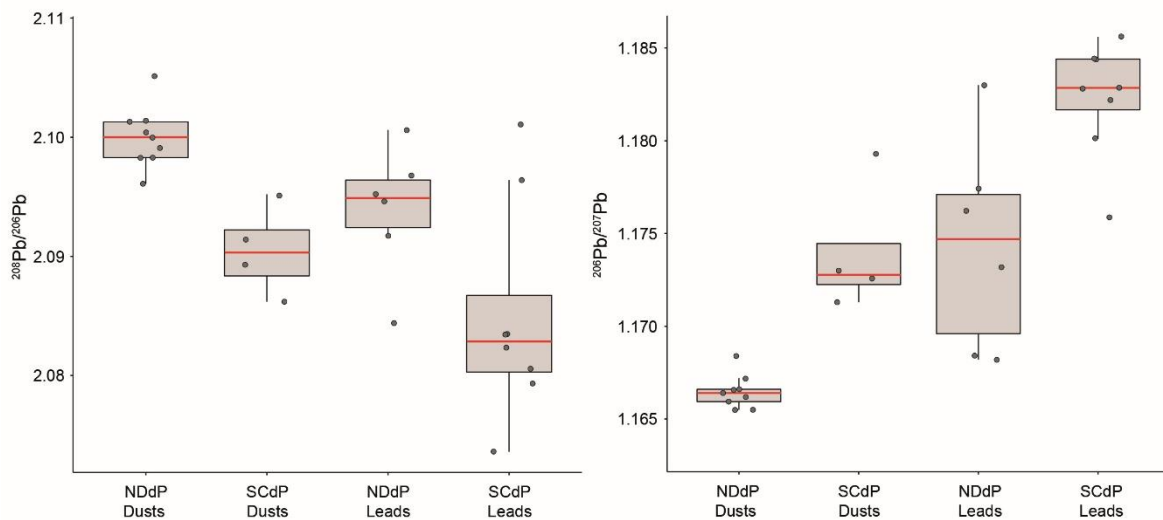
### 198 *4.1. Isotopic signature of historical monuments*

#### 199 4.1.1. Lead isotopic signature variability



200 The isotopic results of Sainte-Chapelle dusts vary between  $1.1713 \pm 0.0056$  and  $1.1793$   
 201  $\pm 0.0070$  (median = 1.1728) and between  $2.0862 \pm 0.0086$  and  $2.0951 \pm 0.0144$  (median =  
 202 2.0904) for the ratios  $^{206}\text{Pb}/^{207}\text{Pb}$  and  $^{208}\text{Pb}/^{206}\text{Pb}$  respectively (Table 1; Fig. 2). Indeed,  
 203 signatures of Sainte-Chapelle appear to be lower in  $^{206}\text{Pb}/^{207}\text{Pb}$  and higher in  $^{208}\text{Pb}/^{206}\text{Pb}$  and  
 204 with a greater values dispersion compared to the dust samples of Notre-Dame (Fig. 2). Similar  
 205 observations are made for construction leads.

206 The fact that these general features are not seen for the  $^{208}/^{206}\text{Pb}$  ratio of NDdP and  
 207 SCdP leads is attributed to outlier values in the measurement of the  $^{208}/^{206}\text{Pb}$  ratio of NDdP  
 208 [29] ) as underlined in Briard et al [10] ) and Le Roux et al [38] ).



209  
 210 **Fig. 2. Boxplot of indoor dusts and lead construction lead isotopic ratios of Sainte-Chapelle (SCdP)**  
 211 **and Notre-Dame (NDdP) [10,29].** The top of the box represents the 75<sup>th</sup> percentile, the bottom of the  
 212 box represents the 25<sup>th</sup> percentile, and the red line in the boxes represents the 50<sup>th</sup> percentile (or  
 213 median). The whiskers represent the highest and lowest values which are not outliers or extreme  
 214 values. Data are represented with grey circle. Mean values are statistically different (U Man Whitney  
 215 test at  $\alpha = 0.05$ ), except for the 208/206Pb ratio of NDdP and SCdP leads. The ratio values variability  
 216 (Kolmogorov-Smirnov test,  $\alpha=0.05$ ) is lower for NDdP than SCdP, except for the  $^{208}/^{206}\text{Pb}$  ratio of  
 217 NDdP and SCdP leads.

218  
 219 Regarding construction lead signatures, they reflect the isotopic compositions of the  
 220 different types of materials located in the monument. Indeed, in this study (Table 2),  
 221 construction lead of Notre-Dame originated from remaining pieces of the roof and spire [29]  
 222 and those of Sainte-Chapelle were sampled from lead joints of the upper bays [10]. It is worth  
 223 noticing that the Pb signature of the construction lead collected at Sainte-Chapelle are more

224 scattered than those from NDdP. They are in range of lead construction signatures of both  
225 NDdP and Chartres cathedral.

226           Regarding dust samples, given the geographical proximity of the two monuments, they  
227 experience the same anthropogenic pressure and their different isotopic signatures cannot be  
228 produced by particular urban pollutions. Thus, it must be due to the variability in lead  
229 materials used for construction and renovation phases. Indeed, the fire in Notre-Dame which  
230 led to a greater influence of the 19<sup>th</sup> century roof and spire in the chemical composition of the  
231 dusts collected inside the monument [10]. On the other hand, the dust signature of Sainte-  
232 Chapelle is significantly different and could be influenced by its numerous wall paintings  
233 (compared to Notre-Dame), which may contain lead and impact the isotopic signature of the  
234 monument [39,40].

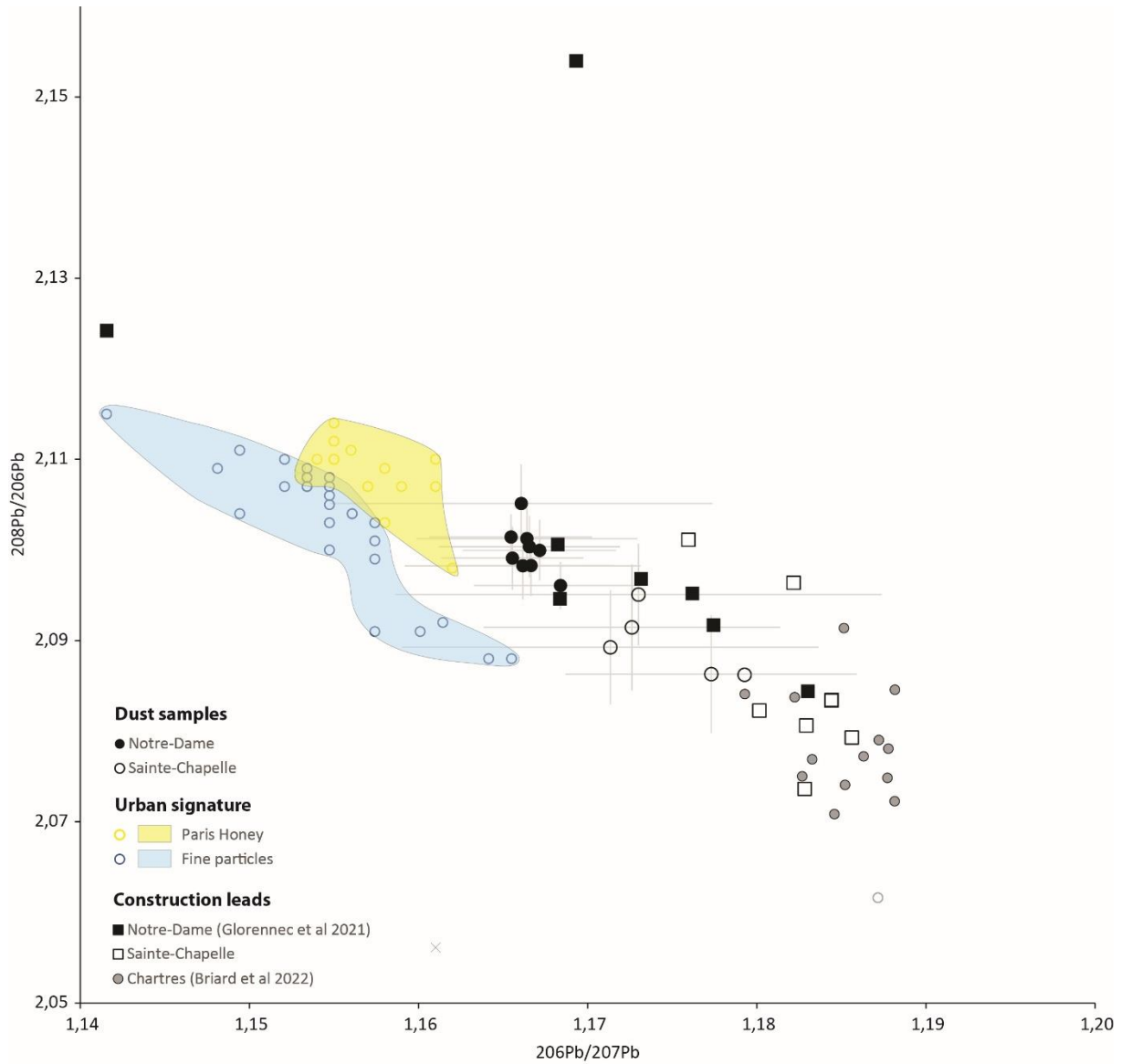
#### 235 4.1.2. Urban lead impact on lead isotopic signature of historical monuments

236 For both Sainte-Chapelle and Notre-Dame, an enrichment in  $^{206}\text{Pb}/^{207}\text{Pb}$  (from 1.1748 to  
237 1.1829 for Sainte-Chapelle and from 1.1664 to 1.1747 for Notre-Dame, calculated from  
238 medians) and an impoverishment in  $^{208}\text{Pb}/^{206}\text{Pb}$  (from 2.0904 to 2.0828 for Sainte-Chapelle  
239 and from 2.1000 to 2.0949 for Notre-Dame calculate from medians) are observed in dust  
240 samples compared to construction lead samples (Fig. 2). The U-Mann Whitney test ( $\alpha=0.05$ )  
241 confirms a significant difference between dusts and metallic construction leads in  $^{206}\text{Pb}/^{207}\text{Pb}$   
242 with a p-value of 0.029 and 0.007, for Notre-Dame and Sainte Chapelle, respectively.  
243 Regarding  $^{208}\text{Pb}/^{206}\text{Pb}$ , the test is significant and insignificant with p-values calculated at 0.029  
244 and 0.081 for Sainte-Chapelle and Notre-Dame, respectively. Overall, the isotopic signatures  
245 of both monuments are closer than their construction leads to the urban signature (Fig. 3),  
246 characterized by low  $^{206}\text{Pb}/^{207}\text{Pb}$  values and high  $^{208}\text{Pb}/^{206}\text{Pb}$  values and illustrated by isotopic  
247 ratios from Parisian honey [14] and fine atmospheric particles [11] (Fig. 3).

248

249           Thus, we can assume that the indoor dust signature of a historical monument, that did  
250 not burn, is a mixture between its construction lead isotopic signature and the urban lead  
251 isotopic signature.

252



253

254

255 **Fig. 3. Comparison between the lead isotopic signature of Sainte-Chapelle and Notre-Dame indoor**  
 256 **dusts and the construction lead of these two historic monuments [10].** The urban signatures  
 257 correspond to data acquired on Parisian honeys (yellow envelope) [14] and on atmospheric particles  
 258 (brown envelope) [11]. The construction lead signatures correspond to the data acquired on the lead  
 259 from Chartres cathedral and Versailles Palace (grey envelope) [10,41]. Uncertainties are shown in gray  
 260 lines

261

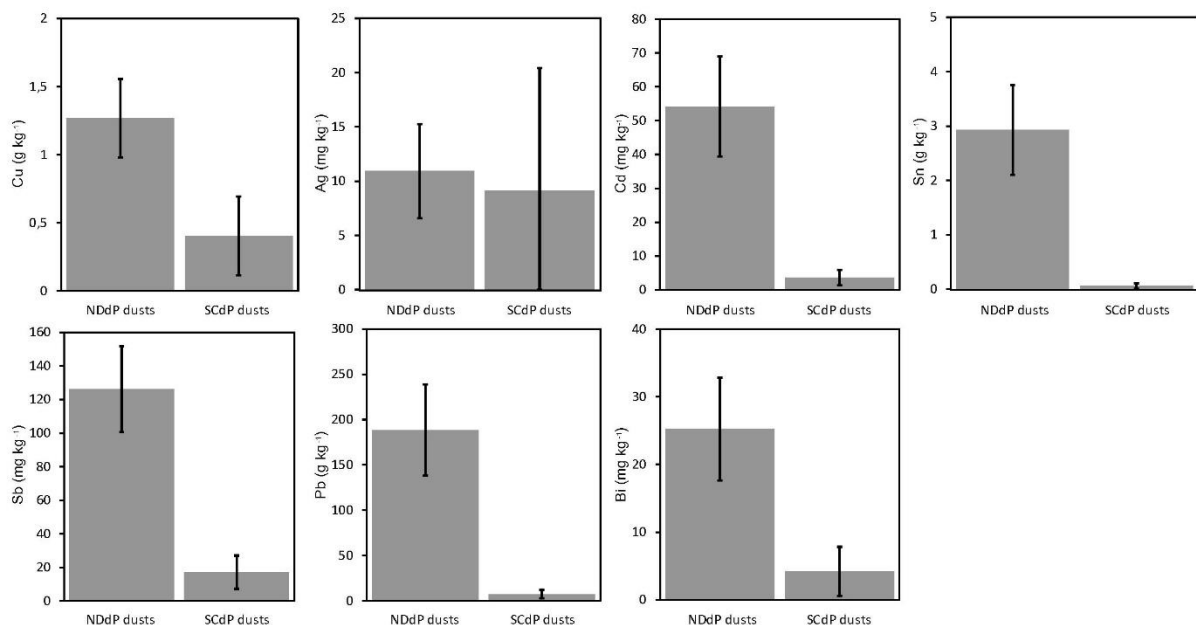
262 *4.2. Elemental signature of historical monuments*

263 *4.2.1. Monument fire impact on metals / metalloids concentrations in indoor dusts*

264 There is a significant variability in metal content among Sainte-Chapelle dust samples  
 265 (Table 1). Indeed, the concentrations range between 0.05 and 0.67 g kg<sup>-1</sup> (average: 4.0 g kg<sup>-1</sup>)

266 for Cu, between 2.2 and 26.0 mg kg<sup>-1</sup> (average: 9.1 mg kg<sup>-1</sup>) for Ag, between 0.5 and 5.4 mg  
 267 kg<sup>-1</sup> (average: 3.6 mg kg<sup>-1</sup>) for Cd, between 0.01 and 0.12 g kg<sup>-1</sup> (average: 0.06 g kg<sup>-1</sup>) for Sn,  
 268 between 2.5 and 23.5 mg kg<sup>-1</sup> (average: 17.26 mg kg<sup>-1</sup>) for Sb, between 2.3 and 12.5 g kg<sup>-1</sup>  
 269 (average: 7.4 g kg<sup>-1</sup>) for Pb, and between 0.3 and 8.0 mg kg<sup>-1</sup> (average: 4.2 mg kg<sup>-1</sup>) for Bi (Table  
 270 1). Only Ag shows an important variability within samples notably with dust sample  
 271 SC\_070721-PN collected from plastic substrate which is much richer in Ag (Fig. 4).

272 To determine if a monument blaze has an impact on the elemental concentrations of  
 273 metals and metalloids found in indoor dust, we compare the elemental concentrations of the  
 274 dusts of Sainte-Chapelle and Notre-Dame [10] (Table 1; Fig. 4). For most elements, the  
 275 concentrations are much higher in Notre-Dame than in Sainte-Chapelle. They range from 3  
 276 times higher for Sn to 200 times higher for Pb (Fig. 4). This seems to correlate with the amount  
 277 of lead that was scattered during the fire, with an estimated 150 to 1000 kg [42,43]. The fire  
 278 therefore seems to have had a major impact on the metal / metalloid composition of the  
 279 dusts.



280

281 **Fig. 4. Comparison between elemental concentrations in indoor dust of Sainte-Chapelle (SCdP) and**  
 282 **Notre-Dame (NDdP) collected on inert substrates [10]. Grey rectangle corresponds to the**  
 283 **concentrations average in g kg<sup>-1</sup> for Cu, Sn, and Pb and in mg kg<sup>-1</sup> for Ag, Cd, Sb and Bi. The black bar is**  
 284 **the standard deviation (SD) of the mean value.**

285 We also note that the Pb concentrations between 2.3 and 12.5 mg kg<sup>-1</sup> measured in  
 286 Sainte-Chapelle dust samples are significantly lower than the indoor dust Pb concentration

287 measured worldwide (mean  $\approx 120 \text{ mg kg}^{-1}$ ; range: 14.2 – 7800  $\text{mg kg}^{-1}$ ) [44] [45]. The Pb  
288 concentrations recorded in the Sainte-Chapelle dusts are also well below French health  
289 recommendations for Pb content in soil which limit the concentration to 100  $\text{mg kg}^{-1}$  for  
290 agricultural soils and 300  $\text{mg kg}^{-1}$  for recreative areas such as playgrounds [46]. Thus, the lead  
291 pollution generated by the construction and decorative lead in the Sainte-Chapelle remains  
292 very limited inside the building. However, it would be interesting to estimate the lead roof  
293 impact on the outdoor dust that could be collected in the gutter, at the gutter exit and on the  
294 sidewalk.

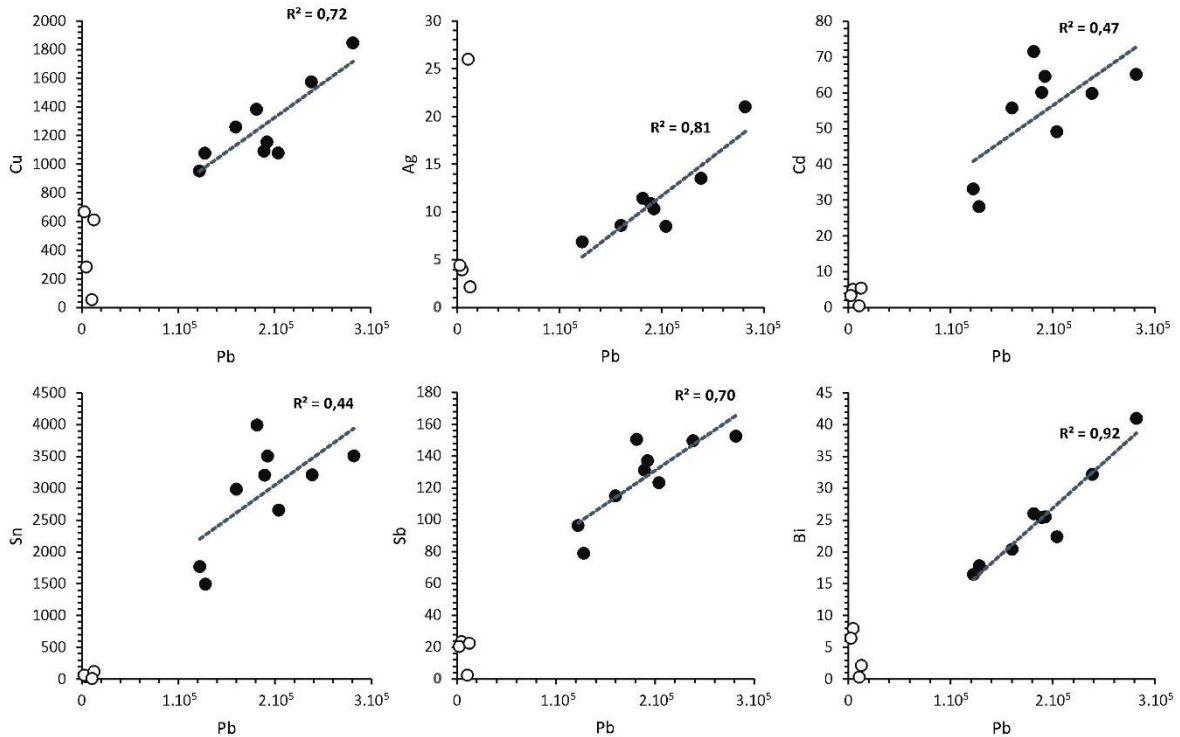
295           Concerning Ag, the averages seem similar between both historical monuments, yet  
296 noting a greater variability of Ag concentrations in the dust samples of Sainte-Chapelle (Fig.  
297 4). This similarity excludes the lead roof fire as a source of Ag in these buildings. A local ~~urban~~  
298 source of Ag can therefore be considered. As the most concentrated sample (SC\_070721\_MF)  
299 was collected next to the tribune where the great silver Chasse which held the relics used to  
300 be, this local increase in Ag concentration could be the result of its melting during the French  
301 Revolution in 1791.

#### 302 4.2.2. Historical monuments: multiple sources of metals and metalloids

303           The lack of correlation (Pearson test at  $\alpha = 0.05$ ) of Cu, Ag, Cd, Sn, Sb and Bi with Pb  
304 concentrations for Sainte-Chapelle indoor dusts (Fig. 5) and their variable Pb isotopic  
305 signatures highlight the presence of multiple lead sources emitting metals (e.g. stained  
306 glasses, paintings, roof) [20]. Conversely, Cu, Ag, Cd, Sb and Bi concentrations are correlated  
307 with lead concentrations observed in Notre-Dame indoor dusts, with  $R^2$  between 0.62 and  
308 0.90 for Sb and Bi, respectively (Fig. 5). It indicates a common source with variable degree of  
309 dilution by metal-free material, or similar mechanisms of mobility [13,47].

310

311



312

313 **Fig. 5. Bivariate concentration plots of selected trace element concentrations versus Pb**  
 314 **concentration for Sainte-Chapelle dust samples and Notre-Dame dust samples [10] [10] (unit : mg**  
 315 **kg<sup>-1</sup>).** Each colour corresponds to localities where the dust samples were collected (white = Sainte-  
 316 Chapelle; black = Notre-Dame). The coefficients of determination ( $R^2$ ) are obtained from the elemental  
 317 concentrations calculated in the Notre-Dame indoor dusts.  $R^2$  values in bold means that the Pearson  
 318 correlation p values is lower than 0.05, proving the correlation between element X and Pb  
 319 concentration values.

320

321 However, Notre-Dame contains lead from as many different sources as Sainte-  
 322 Chapelle, but the Pb content of the Notre-Dame fire dust originated mainly from the roof and  
 323 the spire. Indeed, the fire and the associated air motions could have homogenized the dusts  
 324 inside the cathedral. This hypothesis also seems to be supported by the more homogeneous  
 325 isotopic data for Notre-Dame dusts than t Sainte-Chapelle ones. In view of the correlation  
 326 between metals and metalloids, elemental ratios can be proposed to trace the signature of  
 327 Notre-Dame fire [10]. Conversely, it is impossible to compare these ratios with those of Sainte-  
 328 Chapelle where the variability is more important. Definitely, intensive sampling of indoor and  
 329 outdoor dusts at Sainte-Chapelle is needed to infer the full picture of the Pb sources in this  
 emblematic monument of the 13<sup>th</sup> century.

330

## 5. Conclusions

331 This new study performed on the indoor dust of Sainte-Chapelle to determine its  
332 geochemical fingerprint highlights a significant disparity of lead isotopic and elemental  
333 signatures. As the sampling strategy excludes dust contamination by the substrate, these  
334 disparities reflect the multiple sources of lead in the building. Unlike Sainte-Chapelle, dusts  
335 collected in Notre-Dame are homogeneous for lead isotopes and elemental ratios despite the  
336 presence of different sources in the construction and roof lead. This homogeneity is most  
337 likely caused by a signature homogenization by air motions during the fire of 2019.

338 Comparison of indoor dusts and construction leads for Sainte-Chapelle and Notre-  
339 Dame highlights a shift between the two lead isotopic signatures. Indoor dusts always have  
340 lower values in  $^{206}\text{Pb}/^{207}\text{Pb}$  and higher in  $^{208}\text{Pb}/^{206}\text{Pb}$  compared to construction leads. It could  
341 suggest a potential impact of urban lead pollution on indoor dust under typical monument  
342 operating situations. To conclude, a potentially polluting monument may itself be polluted by  
343 its surrounding environment.

344 The metals and metalloids concentrations in Sainte-Chapelle dusts are very low (except  
345 for Ag) compared to Notre-Dame, which indicates that without the fire triggering the lead roof  
346 destruction and the lead dust release, the lead level is low, with no risk for the population.  
347 This result also suggests that the Pb background level in Notre-Dame before the fire was much  
348 lower and also had a different signature compared to the Pb released by the fire. However, it  
349 would be interesting to continue the study of Sainte-Chapelle dust during and after the work  
350 phase planned for 2022-2023 to determine whether a renovation phase can change the  
351 geochemical signature of a historical monument by remobilizing urban lead from facades or  
352 internal lead sources coming from the monument. It would also be interesting to complete  
353 this new study with the collection of outdoor dust to determine the impact of roof lead runoff  
354 on Sainte-Chapelle close environment and to highlight if their signatures are significantly  
355 different from indoor dust and closer to urban lead.

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**Table 1. Elemental and lead isotope ratios of Sainte-Chapelle dust samples and Notre-Dame dust samples [10].**

Historical monuments	Dust samples ID	Inert substrate	Cu	Ag	Cd	Sn	Sb	Pb	Bi	$^{206}\text{Pb}/^{207}\text{Pb}$	2SE	$^{208}\text{Pb}/^{206}\text{Pb}$	2SE
			<i>g kg<sup>-1</sup></i>	<i>mg kg<sup>-1</sup></i>	<i>mg kg<sup>-1</sup></i>	<i>g kg<sup>-1</sup></i>	<i>mg kg<sup>-1</sup></i>	<i>g kg<sup>-1</sup></i>	<i>mg kg<sup>-1</sup></i>				
Sainte-Chapelle (This study)	SC_070721-PN	Plastic	0.28	3.9	5.1	0.04	23.5	4.5	8.0	1.1730	0.0056	2.0951	0.0144
	SC_070721-PA	Rubber	0.67	4.4	3.4	0.06	20.5	2.3	6.5	1,1726	0,0070	2,0914	0,0088
	SC_070721-EF	Wood	0.61	2.2	5.4	0.12	22.5	12.5	2.1	1.1713	0.0063	2.0893	0.0123
	SC_070721-MF	Plastic	0.05	26.0	0.5	0.01	2.5	10.4	0.3	1.1793	0.0065	2.0862	0.0086
Notre-Dame [10]]	ND_110220-02_PO	Wood	1.08	7.3	28.2	1.49	78.9	127.8	17.8	1.1664	0.0034	2.1013	0.0065
	ND_110220-01_PO	Wood	0.95	6.9	33.1	1.77	96.5	122.1	16.5	1.1655	0.0035	2.0991	0.0042
	ND_311019-02	Wood	1.16	10.3	64.7	3.51	137.1	192.3	25.5	1.1662	0.0037	2.0983	0.0070
	ND_140619-05	Wood	1.26	8.6	55.8	2.99	115.0	160.0	20.4	1.1666	0.0034	2.1004	0.0054
	ND_311019-01	Wood	1.38	11.4	71.6	3.99	150.5	181.3	26.0	1.1672	0.0033	2.1000	0.0045
	ND_311019-04	Wood	1.09	10.9	60.1	3.21	131.3	189.2	25.4	1.1684	0.0026	2.0961	0.0051
	ND_140619-06	Wood	1.85	21.0	65.2	3.51	152.5	281.6	41.0	1.1666	0.0033	2.0983	0.0049
	ND_060220-01_B	Wood	1.08	8.5	49.1	2.66	123.3	203.9	22.4	1.1655	0.0025	2.1014	0.0048
ND_110220-03_B	Wood	1.58	13.5	59.9	3.21	149.7	238.4	32.2	1.1661	0.0043	2.1052	0.0113	

**Table 2. Lead isotope ratios of Sainte-Chapelle lead samples (this work) and Notre-Dame lead samples [29].**

Historical monuments	$^{206}\text{Pb}/^{207}\text{Pb}$	2SD	$^{208}\text{Pb}/^{206}\text{Pb}$	2SD
Sainte-Chapelle (This study)	1,1759	0,0089	2,1011	0,0259
	1,1844	0,0087	2,0835	0,0207
	1,1822	0,0077	2,0964	0,0185
	1,1828	0,0138	2,0736	0,0086
	1,1856	0,0138	2,0793	0,0116
	1,1844	0,0093	2,0834	0,0257
	1,1829	0,0118	2,0806	0,0102
	1,1801	0,0098	2,0823	0,0132
Notre-Dame [29]	2,1540	0,0155	1,1693	0,0027
	2,1242	0,0076	1,1416	0,0028
	2,0844	0,0171	1,1830	0,0071
	2,0968	0,009	1,1732	0,0034
	2,0952	0,0127	1,1762	0,0044
	2,1006	0,0132	1,1682	0,0026
	2,0917	0,0111	1,1774	0,0031
	2,0946	0,0171	1,1684	0,0025

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