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## Lead from Notre-Dame fire plume caught 15 km from Paris

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1 **Lead from Notre Dame fire plume caught 15 km from Paris**

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36

37 **Abstract**

38 Thanks to the use of filters collected daily in the west of Paris, we confirm the passage of the lead-  
39 laden plume following the fire on the roof and spire of Notre Dame Cathedral in April 2019. The  
40 measured concentrations on the filter ( $Pb= 1.4 \mu g m^{-3}$ ), scanning electron microscopy and Hysplit  
41 simulation correspond to an estimate of a few hundred kilograms of lead that would have been  
42 volatilized and then oxidized in the form of micronic and submicronic aerosols. The concentrations  
43 found in the plume are, however, much lower than those found in the environment in the 1980s and  
44 1990s but are 100 times higher than those found in times prior and after the fire. The isotopic  
45 signature of the plume is almost identical to that of the fine Pb dust found inside Notre Dame  
46 building. It is different from the isotopic signature of the Parisian atmosphere before and after the  
47 fire, but it is similar to that of the atmospheric Pb legacy recorded by peat cores over the last 300  
48 years in France. The presence of very fine lead-bearing particles makes them potentially  
49 transportable over long distances after large urban fires. Our study shows the value of daily aerosol  
50 sampling to retrospectively trace the plumes of air pollutants from industrial accidents but also from  
51 historical monument fires such as Notre Dame in 2019.

52

53 **Keywords:**

54

55 Historical monument fire; Notre-Dame de Paris; lead plume; aerosols; Pb isotopes; air monitoring;  
56 urban fire

57

## 59 Introduction

60 Lead is a chemical element that is not essential and is harmful for living organisms, but which has been  
61 used by humans since the beginning of metallurgy.<sup>1</sup> Its global dispersion as a result of the industrial  
62 era, and in particular the use of leaded gasoline, was highlighted by the work of Clair C. Patterson, who  
63 showed that this chemical element is present in all compartments of the environment: from the ocean  
64 abyss to the poles and at the highest mountains on the Earth.<sup>2,3</sup> Lead is a by-product of silver mining  
65 in galena (PbS), is common in many ores as an oxide or carbonate and easy to work. It has been used  
66 in construction since Antiquity.<sup>1</sup> Many major European churches used lead roofs as tiles and lead  
67 roofing is still commonly used. Notre Dame cathedral was no exception and was built with 240 metric  
68 tons of Pb for its roof and spire.<sup>4</sup> Lead has been used in the construction of the cathedral since the  
69 12th-13th centuries, and was still largely used in the 19th century for the renovation of the roof and  
70 reconstruction of the spire by Viollet le Duc. The Notre Dame fire of 15 April 2019 destroyed a large  
71 part of the upper parts of the building, attacking the roof and the spire. The temperatures reached are  
72 still debated by the scientific community but the melting temperature of Pb (327.5°C) was undoubtedly  
73 reached. It is likely that in some places, the temperatures exceeded 600°C and Pb oxidized into an  
74 aerosol. The Parisian air monitoring network ( “AirParif”) is no longer monitoring Pb following the large  
75 decrease in Pb concentrations after the ban on leaded gasoline. Only a single weekly AirParif filter  
76 located under the plume in the west along the Seine River showed a significant increase in airborne Pb  
77 concentrations.<sup>5</sup> In addition, since the aerosols are only sampled on a weekly basis this leads to a large  
78 uncertainty in the concentration of the plume during its passage. To complete this single measurement  
79 point, we benefit from the OPERA network, a network of atmospheric air collectors developed by the  
80 IRSN ( Institut de Radioprotection et Sûreté Nucléaire) to monitor radioactivity in the environment.<sup>6</sup>  
81 More specifically, samples from these high flow atmospheric collectors allow us to follow the plume  
82 over a greater distance and coupled with data from a 24-hour aerosol sample from a location 15 km  
83 from the fire, to determine the Pb isotopic composition and chemical form of the aerosolized Pb.

## 85 Material and Methods

### 86 Sampling strategy

87 We assessed the trajectory of the Notre Dame fire plume using the online Hysplit Lagrangian  
88 atmospheric model<sup>7</sup> (Figure S1). Based on the plume trajectory, we requested permission from the  
89 French Institute of Radioprotection and Nuclear Safety (IRSN) to use weekly filters from the OPERA  
90 observatory collected before and after the fire from North West of France as well as at the Pic du Midi  
91 in the Pyrenees for background reference. Since IRSN has also a campus in Le Vésinet, the laboratory  
92 collects air filters on a daily basis at this location, which makes it exceptional because the collector was  
93 under the plume trajectory and less than 15 km from the Ile de la Cité where Notre Dame is located.  
94 Few data on lead concentrations in the air exist since their sharp decline following the ban on leaded  
95 petrol. However, the Limay station monitored by Airparif (weekly filter) also recorded the passage of  
96 the plume.<sup>8</sup>

### 97 Geochemical and mineralogical analyses

98 The filters (polypropylene filter (3 M<sup>TM</sup>) made of several layers were split and sub-samples were taken  
99 before digestion using ultrapure HNO<sub>3</sub> in Digtubes (SCP Science) on a hot plate at 100°C in a class  
100 10000 clean room at Toulouse. Samples were then centrifuged and an aliquot was analyzed using a  
101 AMETEK SPECTRO ARCOS FHX22 ICP-OES. Three different layers of the Vésinet filters were analyzed  
102 and confirms that aerosols and trace metals were homogeneously distributed over the whole filter.

103 For Pb isotope analysis, after evaporation, samples were diluted by passage of 1 ml of HBr 0.8M. The  
104 solution was then loaded onto 0.05ml of AG1X8 resin contained in micro-column (Bio-spin column)  
105 form Bio-Rad. The matrix is eluted using 2 ml (4\*0.5 ml) of HBr 0.8M. Lead was then recovered into 2  
106 ml HCl 6.0 M<sup>9</sup>. Samples were dried down and subsequently recovered in 1 ml 0.5 ml HNO<sub>3</sub> 0.3 M.  
107 Samples were then analyzed with MC-ICP-MS (Neptune + from Thermo). After dilution to adjust the  
108 signal to the standard (NIST 981), Tl (NIST 997) was added to the samples (similarly to the standard) to  
109 a ratio 1/10 compared to Pb and used to correct for mass bias (White et al. 2000). The Tl values used  
110 to correct were those obtained by DS TIMS<sup>10</sup>. The total procedural blank (<500 pg) was negligible in  
111 front of the Pb introduced by the samples.

112

113 Le Vésinet filters were placed on an aluminum stub and covered by a thin (~20 nm) carbon layer. They  
114 were analyzed with a JEOL JSM 6360LV scanning electron microscope (SEM) set to an accelerating  
115 voltage of 20 kV. The Pb-bearing particles were localized and imaged using the backscattered and

116 secondary electron detectors and the presence of Pb was determined by energy dispersive X-Ray  
117 spectroscopy (using a Bruker Quantax 10 silicon drift detector).

118

## 119 Hysplit modelling

120

121 As an example, we performed different simulations using the offline Lagrangian Hysplit model<sup>11</sup> with  
122 a potential atmospheric release of Pb from the Notre Dame fire. In a first simulation, we first ran the  
123 model with an emission rate of 40kg/h of Pb for 3 hours. These conditions are relatively similar to a  
124 more extensive modelling by INERIS estimating that 63 kg and 75 kg of lead were emitted into the  
125 atmosphere<sup>12</sup> respectively from the roof and the spire. Based on our scanning electron microscope  
126 results, we considered that Pb can be aerosolized in a similar manner as as <sup>137</sup>Cs, that is to say a part  
127 of Pb was aerosolized by high temperature processes and is of micronic in size and we used thus the  
128 initial deposition parameters of Hysplit model.<sup>11</sup> We thus then simulated a higher release of Pb of the  
129 order of 125 kg/hour for 4 hours at 50 m altitude. Our simulations are not intended to be totally  
130 realistic but to show the potential synergy between our geochemical and mineralogical data and future  
131 more sophisticated atmospheric modelling.

132

## 133 Results and Discussion

134

135 Only the filters located near Paris (Le Vésinet and Limay) show an unambiguous increase in Pb  
136 concentrations following the Notre Dame fire (Figure 1 & Supplementary excel spreadsheet). Le  
137 Vésinet filter was also characterized for copper and iron, and those metals do not show any increase  
138 showing the uniqueness of the plume with mainly Pb as a constituent. From electron microscope  
139 imaging, lead is found in Pb oxide aerosols in spherical form with a size of 5-10µm as well as smaller  
140 particles (~1µm) whose shape is more difficult to distinguish due to entanglement in the filter (Figure  
141 2). Daily Le Vesinet Pb concentrations before and after Notre Dame fire are about 0.01 µg m<sup>-3</sup> whereas  
142 daily concentrations of Pb at le Vesinet in the filter following Notre Dame fire is 1.4 µg m<sup>-3</sup>. No lead  
143 particles were found in the Le Vésinet filters before and after the fire. The daily filter sampled at Le  
144 Vésinet shows a higher concentration than the weekly filter in Limay (0.108 µg m<sup>-3</sup>) due to the dilution  
145 effect from non-contaminated air over the sampling period. According to the Hysplit model, the plume  
146 passed over Le Vésinet and Limay in less than 5 hours. If we consider that Pb collected in the filters  
147 comes exclusively from the plume, it is possible to cross multiply and estimate the Pb volume  
148 concentrations during the 5 hours passage of the plume, respectively 6.72 µg m<sup>-3</sup> and 3.5 µg m<sup>-3</sup> for Le

149 Vésinet and Limay. Hysplit model shows also relative higher Pb concentrations at Le Vésinet compared  
150 to Limay, which is located 50 km from Paris downtown (Figure S2**Erreur ! Source du renvoi**  
151 **introuvable.**).

152 The lead concentration, even when adjusted for the duration of the plume, has a similar magnitude to  
153 that found in the 1990s in France when leaded petrol was used.<sup>13</sup> In 1991, the concentration in the  
154 vicinity of the site in heavy traffic was  $0.7\mu\text{g m}^{-3}$  and decreased to  $0.15\mu\text{g m}^{-3}$  in 1999.<sup>14</sup> However, the  
155 concentrations are 20,000 times higher than the lead concentration measured at our reference site at  
156 Pic du Midi and 100 times more than in Le Vésinet on the day before and the day after. Our work also  
157 shows the value of filters taken on a daily basis that can be made available to the scientific and health  
158 community following an accident. Only this filter sampling interval can truly validate the extent of  
159 particulate air pollution from such incidents, based on Pb concentrations.

160 The other weekly filters to the west and north-west do not show an increase in lead although  
161 potentially under the passage of the more dilute plume. In contrast, in the North-East, at Charleville-  
162 Mézières, Pb concentration is also high ( $0.0847\mu\text{g m}^{-3}$ ), as are other metals (Cu and Fe), which  
163 highlights the importance of monitoring of air pollution composition over over the long term  
164 complementary to PM10 and PM2.5. In general, lead concentrations in the air in Northern France  
165 appear to be higher than in Normandy (Figure S5), which is logical given the Atlantic influence on the  
166 Normandy sites and the mining and industrial heritage of Northern France. Pb concentrations  
167 measured in Western France (Penly, Paluel) are in agreement with Pb concentrations measured in  
168 Normandy in 2019 (info AirNormandie- Figure S5) and by the EMEP network in France where  
169 concentrations in 2019 from the MERA-EMEP network (6 locations-<https://ebas-data.nilu.no/>) ranged  
170 from  $4.10^{-4}\mu\text{m}^{-3}$  to  $7.10^{-3}\mu\text{g m}^{-3}$  in PM10 in France on a bi-weekly network. Little is known about fires  
171 in lead-involved buildings<sup>15,16</sup> and our study has shown the influence of a building fire on air quality at  
172 a distance of 15 km away. We also assumed that lead-laden particles could travel further than assumed  
173 as shown by atmospheric modelling and thus potentially influence atmospheric records such as ice  
174 cores.<sup>17</sup>

175 A powerful tracer for identifying the origin of lead is Pb isotope composition, which can generally  
176 discriminate between natural and anthropogenic sources of lead. Here, the isotopic signature  
177 measured at Le Vésinet ( $^{206}\text{Pb}/^{207}\text{Pb} = 1.1697\pm 1.10^{-5}$ ;  $^{208}\text{Pb}/^{206}\text{Pb} = 2.0979\pm 1.10^{-4}$ ) during the passage  
178 of the plume is clearly distinct from the signatures of the filters before and after the fire (Figure 3). It  
179 is also distinct from the signatures at other air monitoring stations (Figure 3).

180 The isotopic signature at the site is variable depending on the lead objects or molten or emitted lead  
181 collected at the site. The first study to look at the isotopic signature of lead was based on single

182 collector mass spectrometry measurements which did not allow for a very accurate measurement of  
183 lead-204.<sup>18</sup> A second, more recent study<sup>19</sup> succeeded in determining the isotopic signature of lead  
184 with a more precise selection of dusts inside the building and a MC-ICP-MS measurement protocol  
185 strictly identical to ours. This selection of indoor dust seems appropriate since the isotopic signature  
186 of lead in the plume is in particular similar to that measured in sample “ND140619-06 (I)”, made of  
187 fine dust collected on the wood of the Notre Dame organ. Briard et al.<sup>19</sup> showed that the isotopic  
188 signature was distinct from the legacy lead stored in different compartments in Paris. However, put in  
189 a more general spatial and historic context, the signature of Notre Dame fire plume is relatively  
190 indistinguishable from the recent isotopic signature trend of the air in France as determined by  
191 atmospheric environmental archives such as *Sphagnum* bogs (Figure 3). We used the isotope ratios of  
192 the most abundant isotopes to lead-204 to try to distinguish a potential influence of the Notre Dame  
193 plume on the more distant filters. The Maubeuge and Villeneuve d'Ascq filters collected during the  
194 plume passage are isotopically different from the filters collected after the passage. Their signature  
195 seems to tend towards that of the plume. However, the Charleville Mézières filter also has a close  
196 signature and this is characterised by an enrichment in iron and copper, not coming from the plume of  
197 the Notre Dame fire. It is therefore difficult to demonstrate the influence of the plume on the isotopy  
198 of the more distant filters, but it cannot be excluded, particularly for the Normandy (Paluel and Penly)  
199 and Villeneuve d'Ascq filters.

200 A previous study has suggested lead contamination of Parisian honeys from the Notre Dame fire.<sup>20</sup>  
201 2019 Honey collected downwind of the fire has high Pb concentrations compared to other honeys from  
202 central Paris and Paris before the fire. However, the isotopic signature of contaminated honey is not  
203 unique and sufficiently distinct. It is possible that Parisian bees were subjected to contamination from  
204 larger local particles such as those measured by Glorennec et al.<sup>18</sup> in the vicinity of the site rather than  
205 from lead present in the form of very fine aerosols that were transported over several tens of  
206 kilometers as shown in our study. Another possibility is that the lead contamination of honey in  
207 western Paris in 2019 is coming from other sources. Multi-year monitoring of honey and other  
208 bioindicators could be useful to assess their effectiveness in monitoring acute contamination. Our SEM  
209 analyses show that the lead-laden particles are at the micron scale in size (Figure 2) showing also the  
210 intensity and temperature reached to probably render lead emission in gaseous forms and then during  
211 transport in the plume integrate it as fine Pb oxide aerosols (Figure 2). The micron size confirms the  
212 potential for transport of lead-laden particles from a building fire via air masses over long distances.<sup>17</sup>

213



## 214 Conclusions and perspectives

215

216 Our results show the interest of a sampling network of daily filters to better understand the plumes of  
217 particulate pollutants in cities or industrial sites. These daily filters make it possible for a more accurate  
218 determination of the intensity of atmospheric contamination and to validate atmospheric models. It is  
219 possible to carry out this monitoring by using devices that compile several daily filters into weekly or  
220 monthly filters in order to carry out the regulatory monitoring while allowing in case of an unexpected  
221 event a return to the high-frequency samples. This strategy already exists for radionuclide monitoring  
222 and is implemented under the Comprehensive Nuclear-Test-Ban Treaty. Some metals, including lead,  
223 can also be monitored continuously by XRF detectors coupled with particle analysers. It seems  
224 essential to us to densify the network of these two types of monitoring to allow retrospective  
225 modelling of atmospheric pollutants.

226 The legacy of lead in major European cities is still a clear environmental threat. Building fires are a  
227 potential additional source of lead to be considered in urban environmental risks.

228

## 229 Supporting information

230 Details on hysplit simulation; Hysplit plume trajectory and filter locations (Figure S1); hourly Pb  
231 concentrations modelled with Hysplit at Le Vésinet and Limay (Figure S2); Filter quality control  
232 (Figure S3 and Table S1); Pb isotopes diagram with Fe and Cu concentrations (Figure S4); 2019 Pb air  
233 concentrations from french air monitoring network (Figure S5); Pb concentrations and Pb isotopes  
234 data (xls spreadsheet S1)

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238

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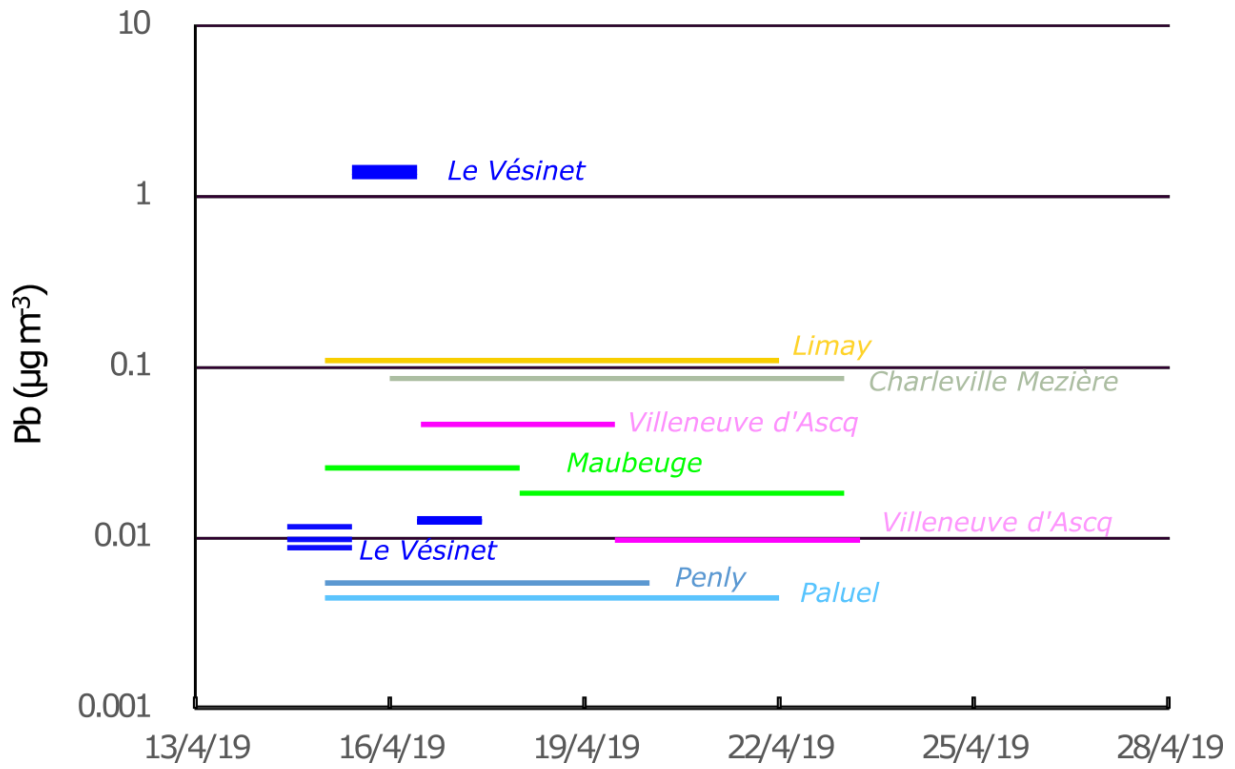
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327 FIGURES

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329 **Figure 1**



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332 *Figure 1: Pb concentrations ( $\mu\text{g m}^{-3}$ ) measured in air filters from OPERA monitoring program. For le Vésinet, triplicates are*  
333 *indicated by different bars. Limay data is from AirParif*

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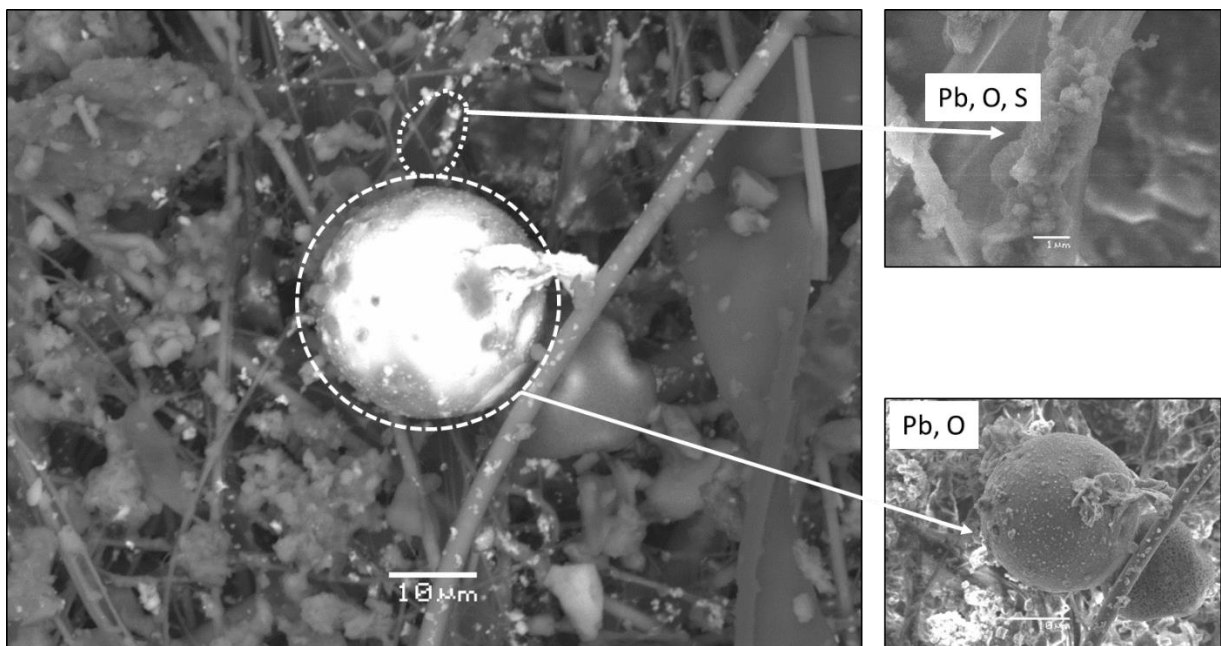
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341 **Figure 2**



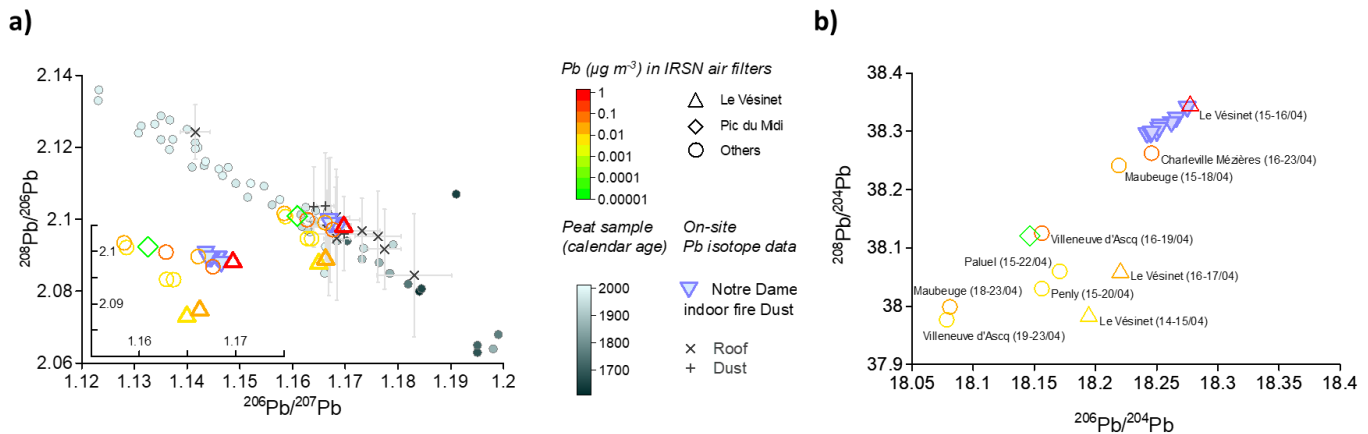
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343 *Figure 2: Examples of Pb particles found in Le Vésinet filter sampled from 15 April 2019 to 16 April 2019 identified with SEM.*  
344 *Backscattered electron image on the left, secondary electron images on the right.*

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346

347 **Figure 3**



348

349 *Figure 3 : Pb isotopes measured in air filters from OPERA network following Notre Dame fire. Uncertainties from our*  
 350 *measurements are smaller than the symbol sizes. A)  $^{206}\text{Pb}/^{207}\text{Pb}$  vs.  $^{208}\text{Pb}/^{206}\text{Pb}$  diagram: Pb isotopes measured by*  
 351 *Gloennec et al.<sup>18</sup> in Notre Dame dusts and melted Pb are shown in purple (2 “outliers” are not figured), Notre Dame indoor*  
 352 *fire dust are from Briard et al.<sup>19</sup> Background Pb isotope chronological evolution of French atmosphere is based on dated peat*  
 353 *layers from eastern and south France, Belgium and Swiss Jura (original data Hansson et al.<sup>21</sup>, Forel et al.<sup>22</sup>, Shotyk et al.<sup>23</sup>,*  
 354 *Allan et al.<sup>24</sup>). The inner panel is a zoom on the filter data and the data from Briard et al. B)  $^{206}\text{Pb}/^{204}\text{Pb}$  vs.  $^{208}\text{Pb}/^{204}\text{Pb}$*   
 355 *diagram with indoor dust data from Briard et al.<sup>19</sup>*

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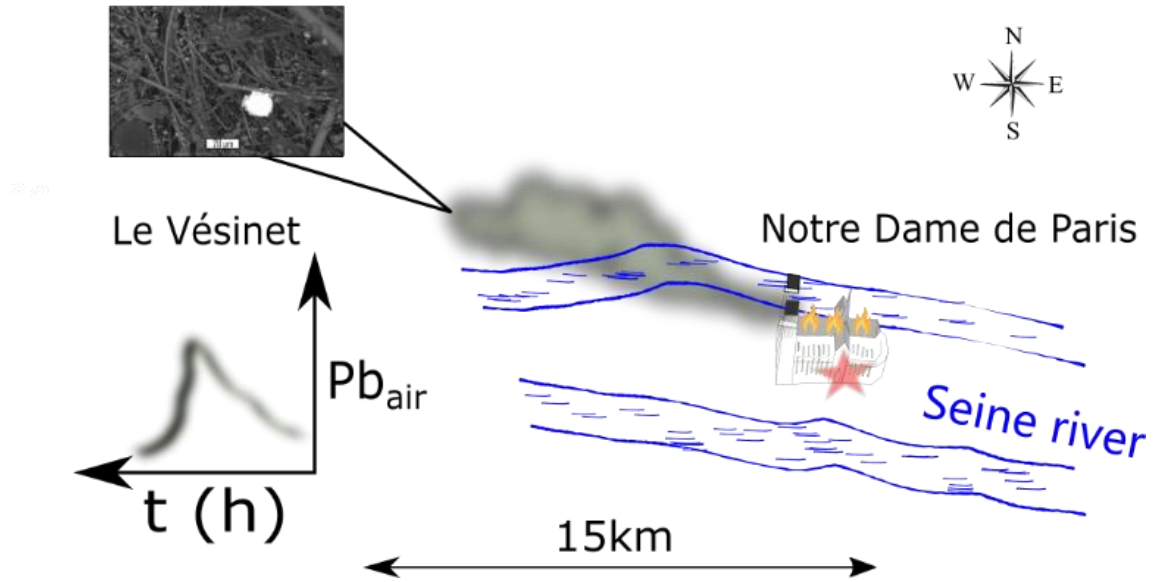
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360 For TOC Only"

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366 **Lead from Notre Dame fire plume caught 15 km from Paris**

367 **Supplementary information**

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369 Olivier Saunier<sup>5</sup>, David Baqué<sup>1</sup>, Thierry Camboulive<sup>1</sup>, Julien Berger<sup>6</sup>, Thierry Aigouy<sup>6</sup>, Françoise

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403 9 pages

404 5 supplementary figures

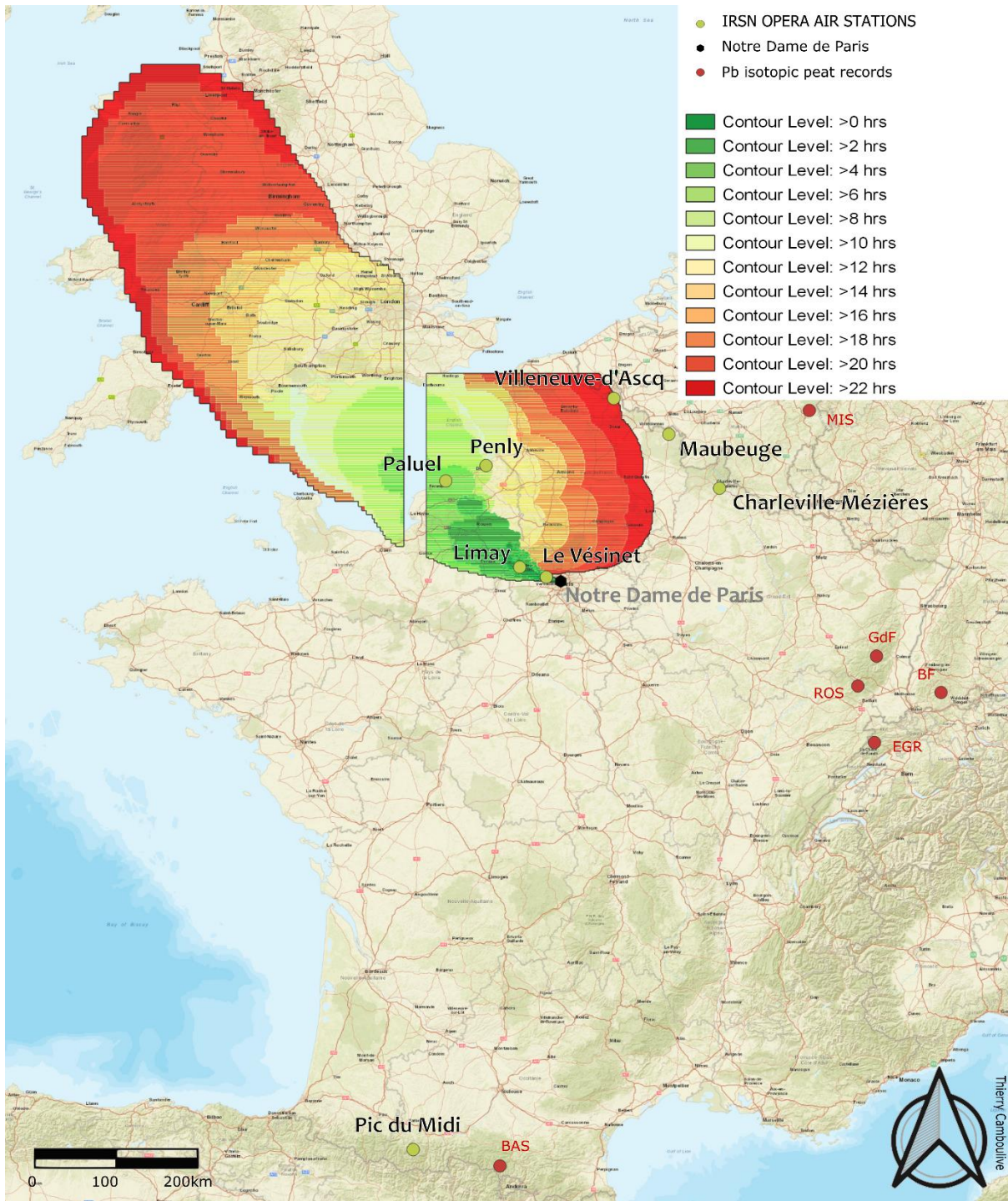
405 1 supplementary table included in this text file

406 1 supplementary XLS spreadsheet

407 Online and offline hysplit simulations:  
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409 An online simulation was run on hysplit the day after the fire to assess the potential of OPERA IRSN  
410 filters to track the particulate plume from the Notre Dame fire.

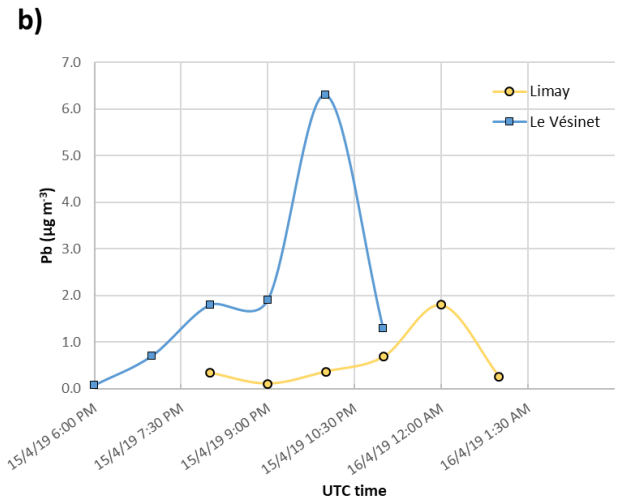
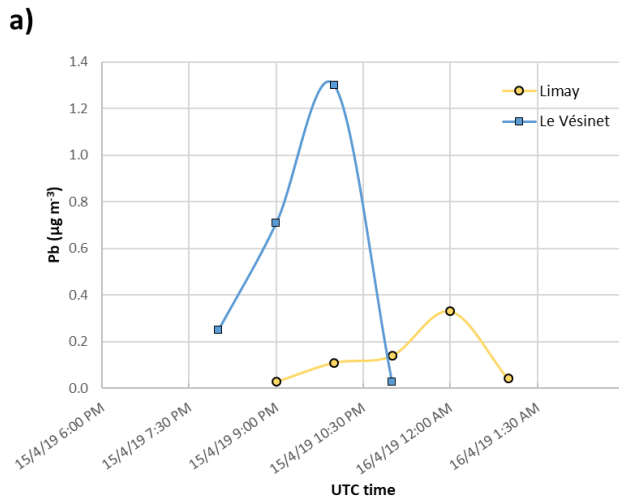
411 In addition, 2 offline rough simulations have been produced as examples. The first is a simulation of a  
412 lead emission of 40kgs/h of Pb from 50m to 100m height one hour after the start of the fire. The  
413 second simulation considers an emission of 125kgs/h of Pb for 4 hours one hour after the start of the  
414 fire. To obtain the potential hourly air concentrations in Limay and Le Vésinet, we used the offline  
415 version of Hysplit (particle mode concentration model). For the 1<sup>st</sup> simulation, no wet deposition  
416 forcing was included. For the 2nd simulation, we used the default wet deposition parameters similar  
417 to <sup>137</sup>Cs considering a micron size for the aerosols.



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Figure S1 : Hysplit plume trajectory with OPERA AIR STATIONS and peatland locations used as Pb isotopic records – contour levels indicate time of arrival of the plume



421

422 *Figure S2 : a) offline hysplit Pb simulation at Le Vésinet (blue squares) and Limay (orange circles) locations using an emission*  
 423 *of 40 kg of Pb emitted during 3 hours; b) offline hysplit Pb simulation using 125 kg/h rate of Pb emission during 4 hours at*  
 424 *50m agl and with a similar behaviour as <sup>137</sup>Cs bearing particles*

425

426 **Filter quality control :**

427 OPERA filters are made of several filter layers ((polypropylene filter (3 M™). To evaluate potential  
428 variability between the different layers, we measured triplicates made of subsamples of three layers.  
429 A known surface of each filter was subsampled 3 times, digested using 3 ml of ultrapure 69% HNO<sub>3</sub>  
430 (Optima™, for Ultra Trace Elemental Analysis, Fisher Chemical™) at 105°C overnight in 50ml PP  
431 DigiTUBEs (SCP Science, Villebon sur Yvette-France), then adequately diluted with ultrapure water  
432 (Veolia Purelab, Toulouse-France) and measured with a ICP-OES (AMETEK SPECTRO ARCOS FHX22).  
433 For Pb isotope measurements, we selected an intermediate filter.



434

435 *Figure S3: : Example of triplicates le Vésinet filter (16/04/2019 to 17/04/2019)*

436 The iron blank was negligible that is to say 10 times lower than the lowest concentration filter apart  
437 the Pic du Midi filter. The lead blank was under the detection limit of the ICP-OES instrument that is  
438 20 times lower than the lowest Pb concentration measured in OPERA filters. The blank for the  
439 isotope separation chemistry is typically in the sub-nanogram range and usually around 300 pg of  
440 lead.

441

442 To test the validity of our protocol, we put approximately 50mg of WQB-1 reference material (NWRI  
443 CRM-Lake Ontario Sediment) on a blank filter and digest it similarly overnight as with the other  
444 filters.

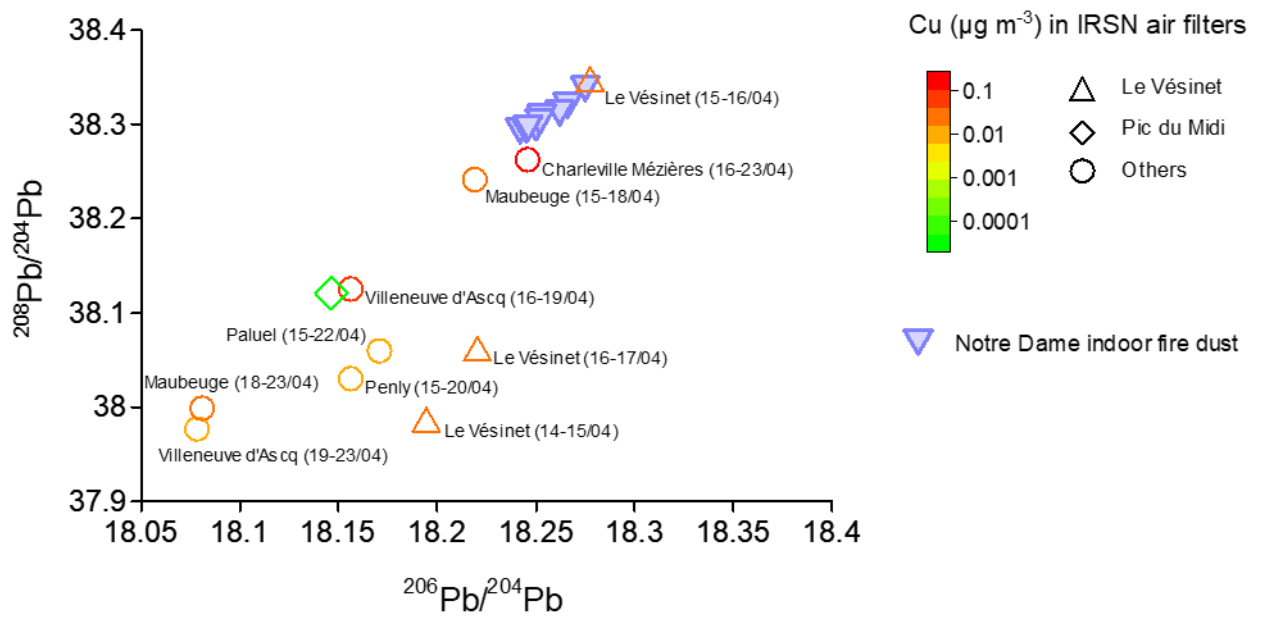
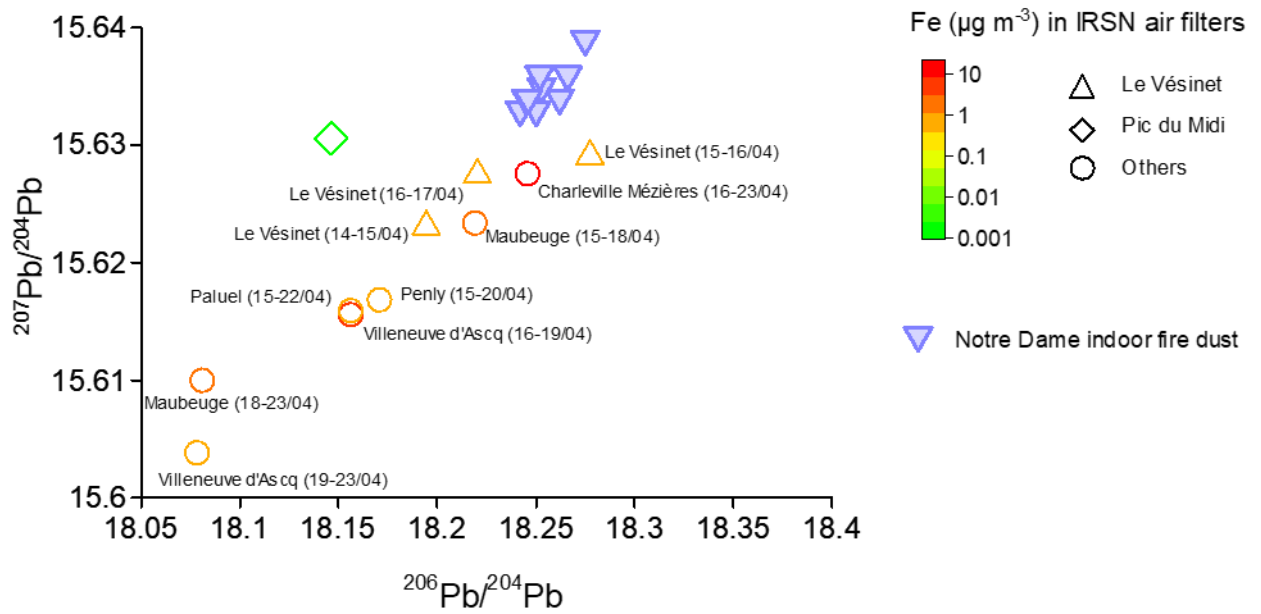
WQB-1 lake sediment	Measured	Certified
Pb (mg kg <sup>-1</sup> )	85	83.7
Fe (mg kg <sup>-1</sup> )	40020	47358
Cu (mg kg <sup>-1</sup> )	82	79.6

445 *Table S1 : Comparison of measured and certified values for WQB-1 reference material*

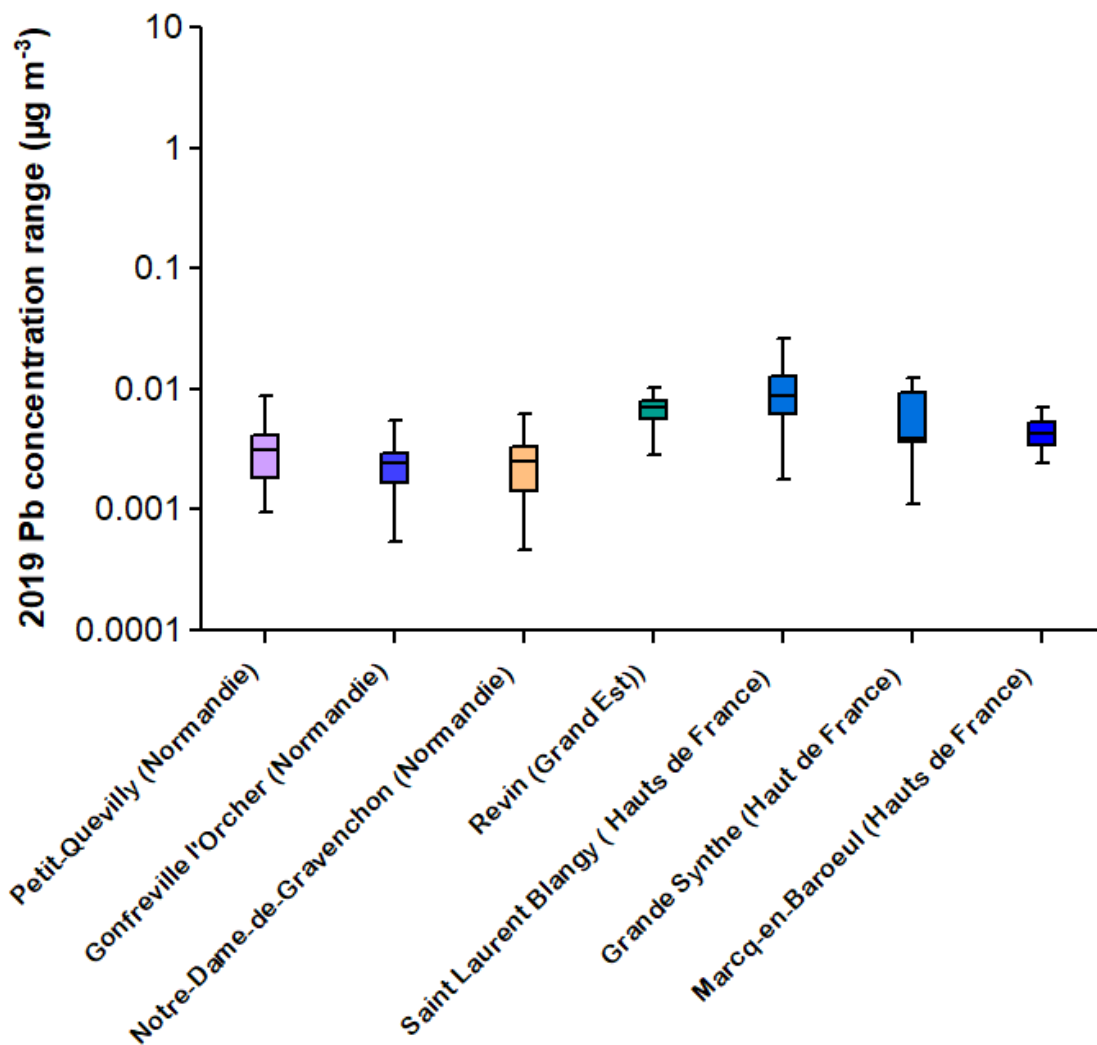
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451 *Figure S4 : Pb isotope ratio diagrams and Fe and Cu concentrations for IRSN OPERA filters*



452

453 *Figure S5: Lead concentration ranges in filters collected a) every other week by Atmo Normandie in 2019 on three locations.*  
 454 *The week of the fire, no filter was measured by Air Normandie. (data courtesy from Atmo Normandie) ; b) by Atmo*  
 455 *Grand'Est in Revin from jan. 2019 to April. 2019 every ~2weeks ; c) by Atmo Hauts de France in 3 sites in winter and summer*  
 456 *2019*

457



458 [XLS spreadsheet S1 : Pb concentrations and Pb isotopes data \(xls\)](#)

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