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

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Abstract

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

Keywords:

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

Introduction

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

Material and Methods

Sampling strategy

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

Geochemical and mineralogical analyses

The filters (polypropylene filter (3 M™) made of several layers were split and sub-samples were taken before digestion using ultrapure HNO_3 in Digitubes (SCP Science) on a hot plate at 100°C in a class 10000 clean room at Toulouse. Samples were then centrifuged and an aliquot was analyzed using a AMETEK SPECTRO ARCOS FHX22 ICP-OES. Three different layers of the Vésinet filters were analyzed and confirms that aerosols and trace metals were homogeneously distributed over the whole filter.

For Pb isotope analysis, after evaporation, samples were diluted by passage of 1 ml of HBr 0.8M. The solution was then loaded onto 0.05ml of AG1X8 resin contained in micro-column (Bio-spin column) form Bio-Rad. The matrix is eluted using 2 ml (4*0.5 ml) of HBr 0.8M. Lead was then recovered into 2 ml HCl 6.0 M[9]. Samples were dried down and subsequently recovered in 1 ml 0.5 ml HNO_3 0.3 M. Samples were then analyzed with MC-ICP-MS (Neptune + from Thermo). After dilution to adjust the signal to the standard (NIST 981), Tl (NIST 997) was added to the samples (similarly to the standard) to a ratio 1/10 compared to Pb and used to correct for mass bias (White et al. 2000). The Tl values used to correct were those obtained by DS TIMS[10]. The total procedural blank (<500 pg) was negligible in front of the Pb introduced by the samples.

Le Vésinet filters were placed on an aluminum stub and covered by a thin (~20 nm) carbon layer. They were analyzed with a JEOL JSM 6360LV scanning electron microscope (SEM) set to an accelerating voltage of 20 kV. The Pb-bearing particles were localized and imaged using the backscattered and secondary electron detectors and the presence of Pb was determined by energy dispersive X-Ray spectroscopy (using a Bruker Quantax 10 silicon drift detector).

Hysplit modelling

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

Results and Discussion

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

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

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

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

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

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

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

Conclusions and perspectives

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

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

Supporting information

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

Acknowledgements

This research was funded by the CNRS chantier Notre Dame de Paris. We thank the 3 reviewers for their extremely constructive comments.

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FIGURES

Figure 1

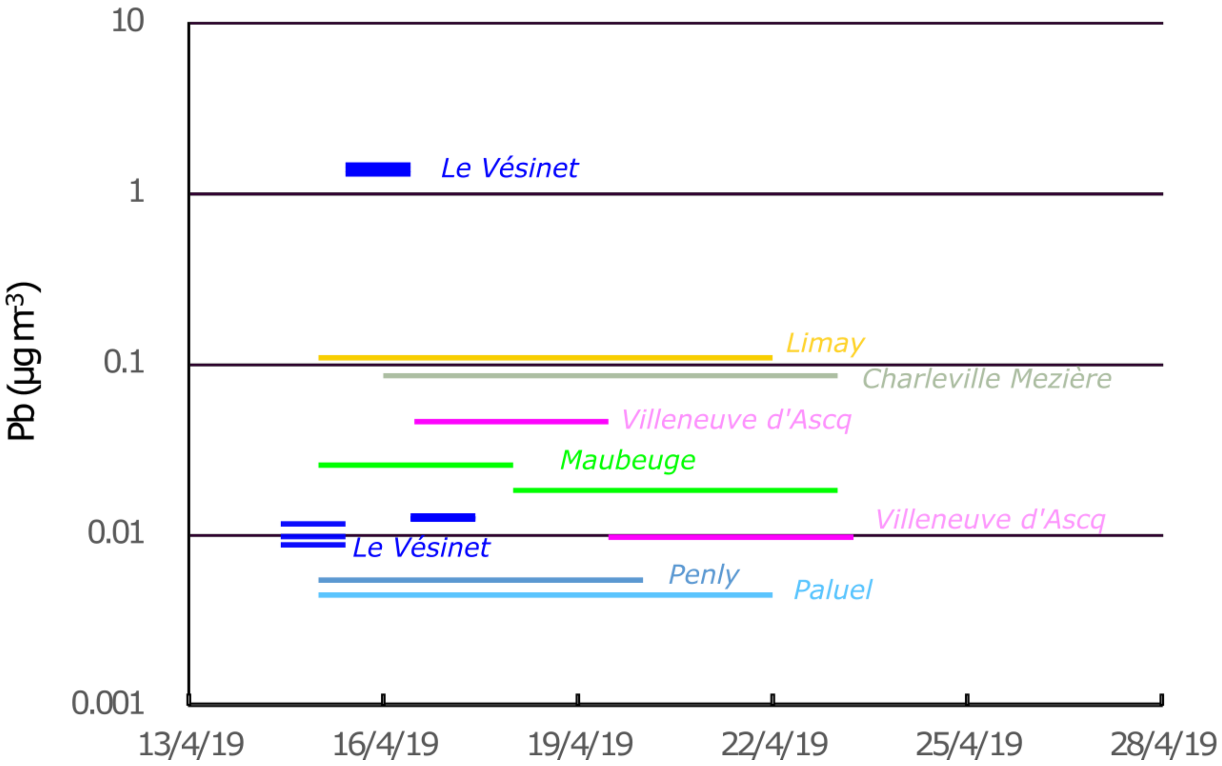

Figure 1: Pb concentrations (µg m^{-3}) measured in air filters from OPERA monitoring program. For le Vésinet, triplicates are indicated by different bars. Limay data is from AirParif

Figure 2

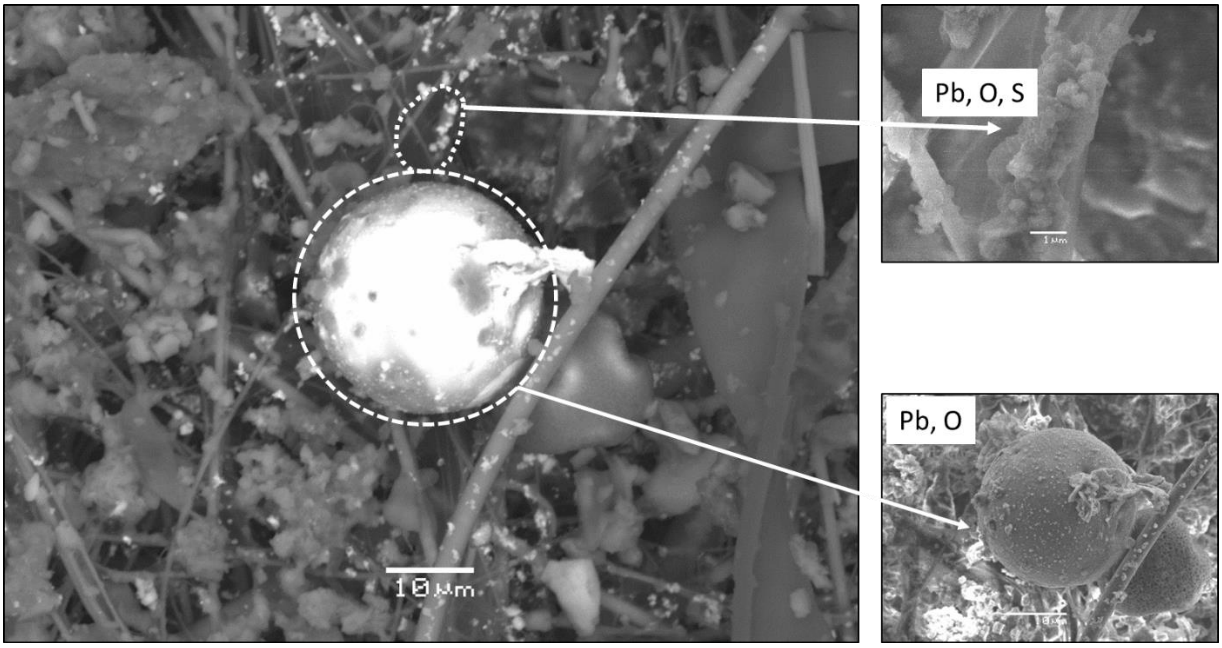

Figure 2: Examples of Pb particles found in Le Vésinet filter sampled from 15 April 2019 to 16 April 2019 identified with SEM. Backscattered electron image on the left, secondary electron images on the right.

Figure 3

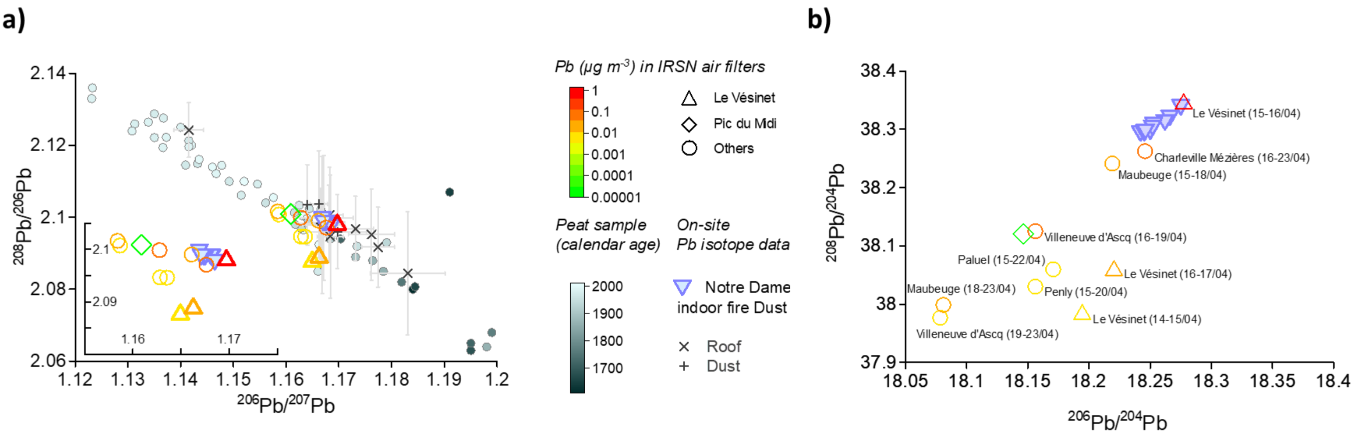

Figure 3 : Pb isotopes measured in air filters from OPERA network following Notre Dame fire. Uncertainties from our measurements are smaller than the symbol sizes. A) $^{206}Pb/^{207}Pb$ vs. $^{208}Pb/^{206}Pb$ diagram: Pb isotopes measured by Glorennec et al[18] in Notre Dame dusts and melted Pb are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

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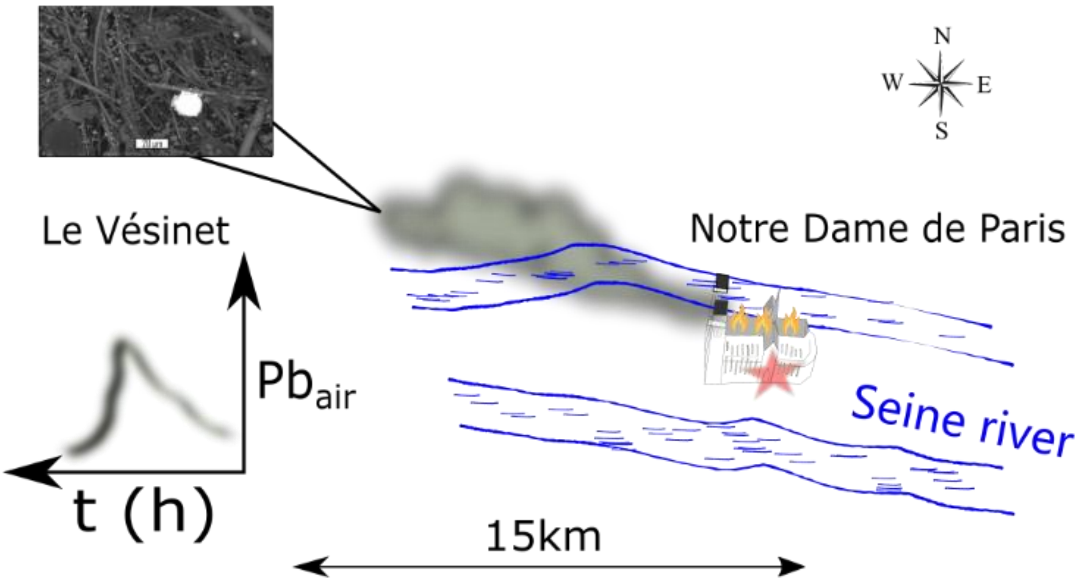
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S
Le Vésinet
Notre Dame de Paris
Pb$_{air}$
t (h)
Seine river
15km

Lead from Notre Dame fire plume caught 15 km from Paris

Supplementary information

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9 pages

5 supplementary figures

1 supplementary table included in this text file

1 supplementary XLS spreadsheet

Online and offline hysplit simulations:

An online simulation was run on hysplit the day after the fire to assess the potential of OPERA IRSN filters to track the particulate plume from the Notre Dame fire.

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

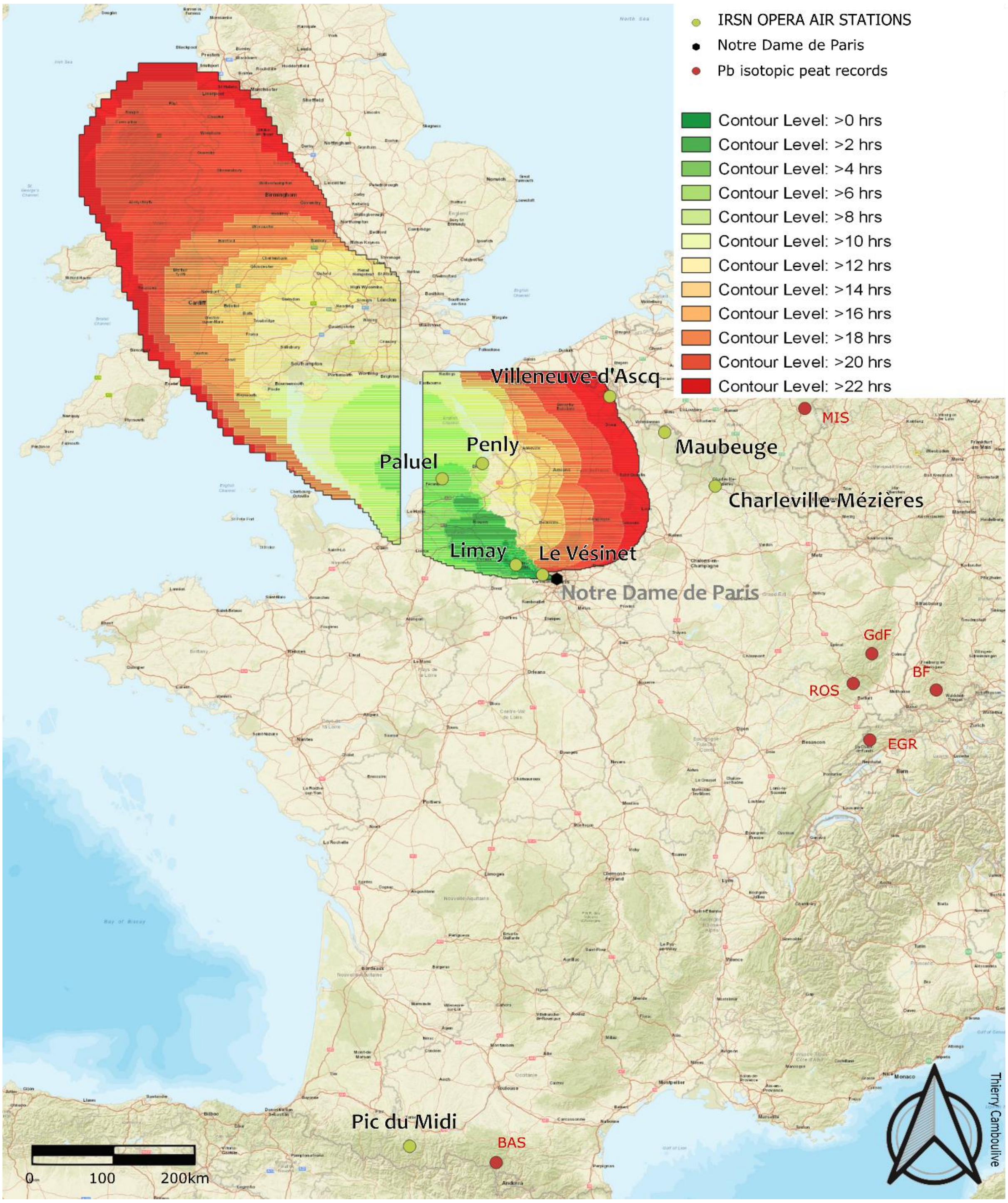

Figure S1 : Hysplit plume trajectory with OPERA AIR STATIONS and peatland locations used as Pb isotopic records – contour levels indicate time of arrival of the plume

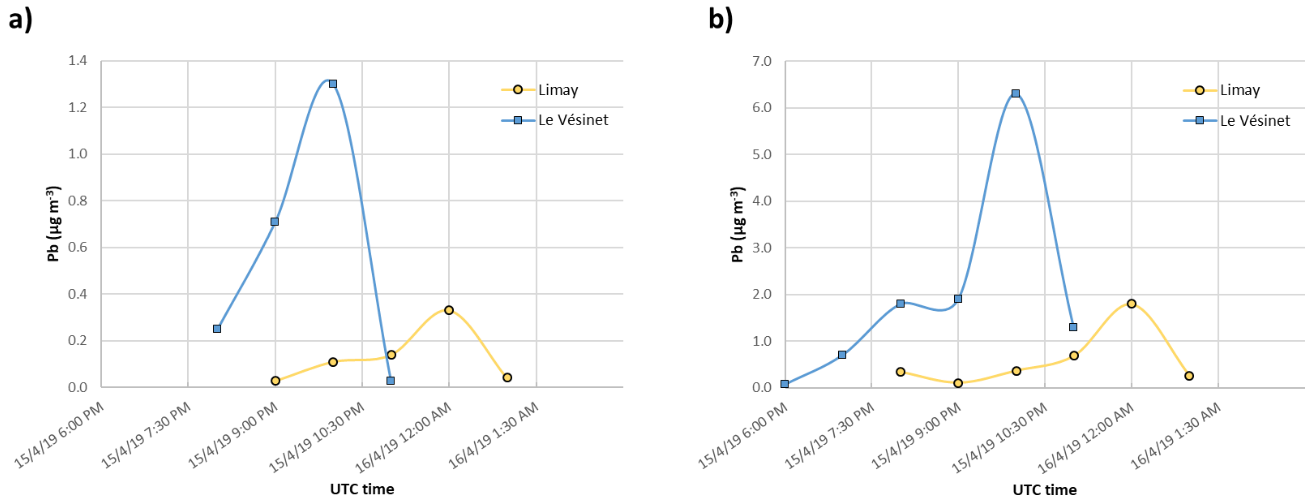

Figure S2 : a) offline hysplit Pb simulation at Le Vésinet (blue squares) and Limay (orange circles) locations using an emission of 40 kg of Pb emitted during 3 hours; b) offline hysplit Pb simulation using 125 kg/h rate of Pb emission during 4 hours at 50m agl and with a similar behaviour as ^{137}Cs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

Filter quality control :

OPERA filters are made of several filter layers ((polypropylene filter (3 M^{TM}). To evaluate potential variability between the different layers, we measured triplicates made of subsamples of three layers. A known surface of each filter was subsampled 3 times, digested using 3 ml of ultrapure 69% HNO_3 (Optima™, for Ultra Trace Elemental Analysis, Fisher Chemical™) at 105°C overnight in 50ml PP DigiTUBEs (SCP Science, Villebon sur Yvette-France), then adequately diluted with ultrapure water (Veolia Purelab, Toulouse-France) and measured with a ICP-OES (AMETEK SPECTRO ARCOS FHX22). For Pb isotope measurements, we selected an intermediate filter.

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

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

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

WQB-1 lake sediment	Measured	Certified
Pb (mg kg^{-1})	85	83.7
Fe (mg kg^{-1})	40020	47358
Cu (mg kg^{-1})	82	79.6

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

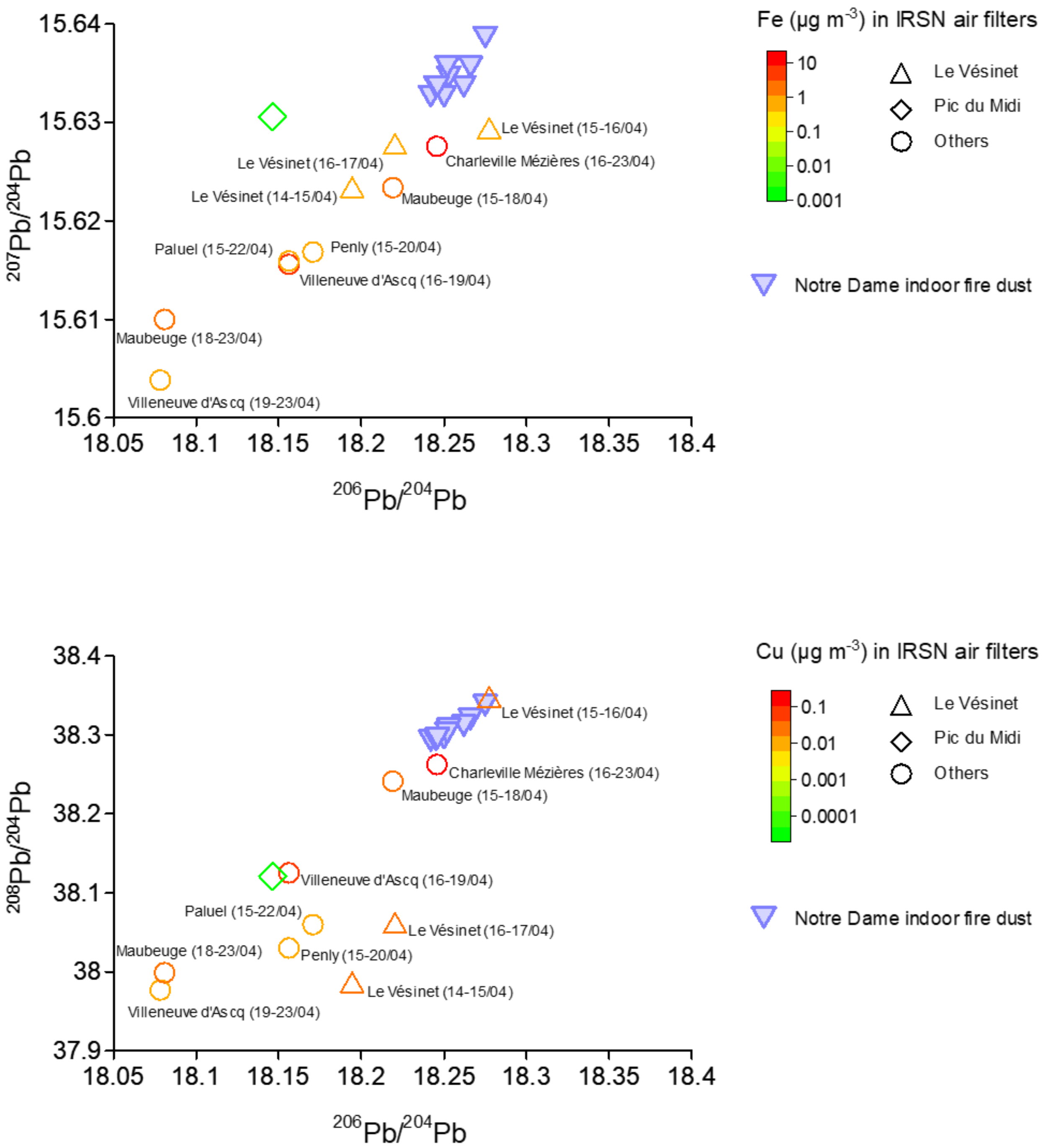

Figure S4 : Pb isotope ratio diagrams and Fe and Cu concentrations for IRSN OPERA filters

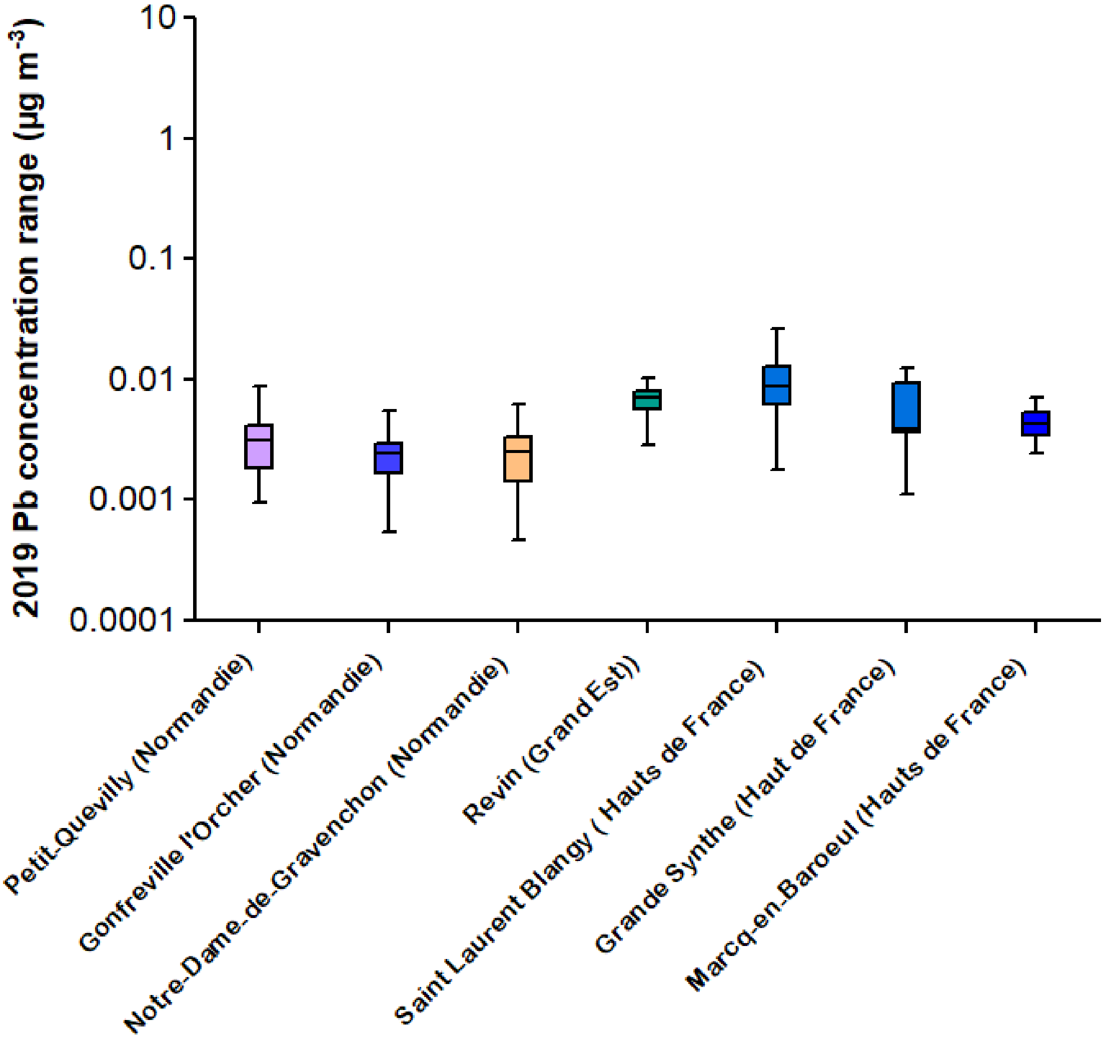

Figure S5: Lead concentration ranges in filters collected a) every other week by Atmo Normandie in 2019 on three locations. The week of the fire, no filter was measured by Air Normandie. (data courtesy from Atmo Normandie) ; b) by Atmo Grand'Est in Revin from jan. 2019 to April. 2019 every ~2weeks ;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

XLS spreadsheet S1 : Pb concentrations and Pb isotopes data (xls)