Next generation ice core technology reveals true minimum natural levels of lead (Pb) in the atmosphere: insights from the Black Death

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Key Points:

- Pre-industrial, atmospheric lead (Pb) levels have been grossly underestimated, with significant implications for human health and development.
- Overwhelming historical evidence shows catastrophic demographic collapse caused atmospheric Pb to plummet to natural levels only once in the last ~2000 years.
- Next-generation ice-core analysis by Laser Ablation Inductively Coupled Mass Spectrometry allows for the first time an ultra-high resolution (sub-annual) record of Pb deposition.

Abstract

Contrary to widespread assumptions, next-generation high (annual to multi-annual) and ultra-high (sub-annual) resolution analysis of an Alpine glacier reveals that true historical minimum natural levels of lead in the atmosphere occurred only once in the last ca. 2000 years. During the Black Death pandemic, demographic and economic collapse interrupted metal production and atmospheric lead dropped to undetectable levels. This finding challenges current government and industry understanding of pre-industrial lead pollution and its potential implications for human health of children and adults worldwide. Available technology and geographic location have limited previous ice core investigations. We provide new high- (discrete, inductively coupled mass spectrometry, ICP-MS) and ultra-high resolution (laser ablation inductively coupled mass spectrometry, LA-ICP-MS) records of atmospheric lead deposition extracted from the high Alpine glacier Colle Gnifetti, in the Swiss-Italian Alps. We show that, contrary to the conventional wisdom, low levels at or approaching natural background occurred only in a single four-year period in the ca. 2000 years documented in the new ice core, during the Black Death (ca. 1349-1353 C.E.), the most devastating pandemic in Eurasian history. Ultra-high chronological resolution allows for the first time detailed and decisive comparison of the new glaciochemical data with historical records. Historical evidence shows that mining activity ceased upwind of the core site from ca. 1349 to 1353, while concurrently on the glacier lead (Pb) concentrations—dated by layer counting confirmed by radiocarbon dating—dropped to levels below detection, an order of magnitude beneath figures deemed low in earlier studies. Previous assumptions about pre-industrial "natural" background lead levels in the atmosphere—and potential impacts on humans—have been misleading, with significant implications for current environmental, industrial, and public health policy, as well as for the history of human lead exposure. Trans-disciplinary application of this new technology opens the door to new approaches to the anthropogenic impact on past and present human health.

1 Introduction

Although scientists and modern historians have documented the devastating effects of lead (Pb) poisoning on humans during the past two thousand years (at the very least) [*Hernberg*, 2000; *Nriagu*, 1983], the extent of population exposure to elevated atmospheric lead levels remains unclear. Despite mitigation and public health measures aimed at reducing human exposure in occupational and residential environments, lead remains a major threat to public health worldwide [*Mushak*, 2011]. The effects of even minimal human exposure include mental deficiencies [*Hernberg*, 2000, *Lanphier*, 2005], reduced fertility [*Mushak*, 2011, *Selevan et al.*, 2011, *Chang et al.*, 2006, *De Rosa et al.*, 2003], and increased aggressive behavior [*Mielke et al.*, 2012, *Reyes*, 2015]. These symptoms have been observed even at low levels of Pb blood concentration, especially in children [*Hernberg*, 2000]. Atmospheric lead pollution is both a cause of higher levels of Pb in humans and a proxy for higher concentration of aerosol Pb. Historically, Pb has been mined and smelted (along with silver), and used widely in coinage, water pipes, roofs, and more recently as an additive in paint and fuel [*Hernberg*, 2000]. Government and industry standards continue to overestimate the proportion of natural lead (Pb) levels in the environment [*UNEP*, 2010, *Richardson et al.*, 2001]. Our high- and ultra-high-resolution continuous measurements substantiate and expand upon previously published, pioneering but lower-resolution ice core studies and those from lake sediments and peat cores that suggest a steady increase in Pb levels across western Europe from ca. 1250-900 B.C.E. to the present, with periods of only moderate decline [*Hong et al.*, 2001, *Renberg et al.*, 2001, *Shotyk et al.*, 1998, *Le Roux et al.*, 2004, *Martínez Cortizas et al.*, 2013, *Montgomery et al.*, 2010, *Gabrieli et al.*, 2014].

2 Data: A new high- and ultra-high resolution record of lead (Pb) deposition from the heart of Europe

In this study we provide a new atmospheric Pb deposition record from a ~72m ice core extracted from the Colle Gnifetti (CG) glacier (4450 m.a.s.l.) in the Swiss-Italian Alps. A discrete, high-resolution ICP-MS Pb record (Fig. 1) covers the last ca. 2000 years; an additional ultra-high resolution LA-ICP-MS record provides more detailed evidence of sub-annual Pb deposition for the years ca. 1330-1360 C.E.

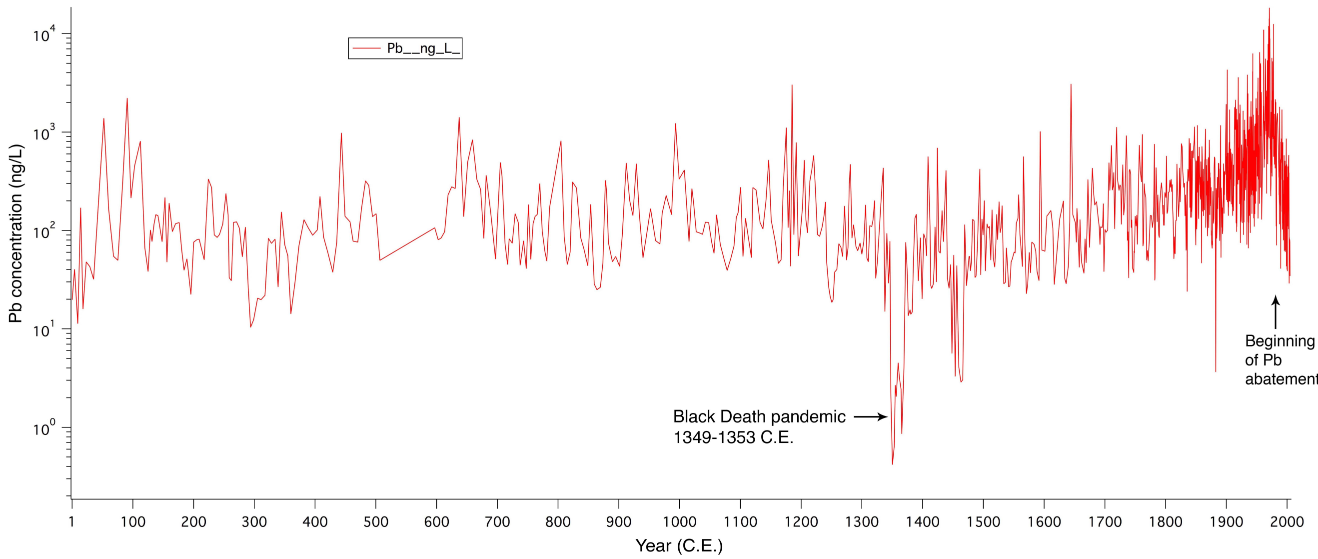

Fig. 1. Lead concentration in Colle Gnifetti ice core, from high-resolution discrete ICP-MS. The graph covers the period ca. 1- 2007 C.E. The Black Death drop marks the years 1349-1353 C.E. Values below 1ng/L here are calculated using semi-quantitative calibration data. A gap in data of 90 years around ca. 500 C.E. is shown here linearly interpolated.

Ultra-high resolution sampling of this ice core (~120 micron, allowing ~550 measurements within the year dated ~1300 C.E.) was produced using the Climate Change Institute's (CCI at the University of Maine) W. M. Keck Laser Ice Facility laser ablation inductively coupled plasma mass spectrometer (LA-ICP-MS) [*Sneed et al.*, 2015]. This new method allowed us to count highly thinned annual layers previously not detectable by conventional cm-resolution analyses. The ultra-high resolution time-series allowed us to apply the layer-counting procedure down to the beginning of the first millennium of the Common Era. Back to 1900 C.E., known time markers such as documented Saharan dust events were used to constrain the chronology of the ice core, as already demonstrated in similar alpine cores [*Gabrieli et al.*, 2014, *Bohleber et al.*, 2013, *Jenk et al.*, 2009, *Schwikowski et al.*, 2004, *Eisen et al.*, 2003, *Wagenbach and Geis*, 1989]. For the most recent ca. 800 years the resulting time scale was further corroborated by direct time series comparisons with a neighboring CG ice core dated with conventional cm-resolution analysis [*Bohleber et al.*, 2013]. The time scale for the layers dated for years before this period is currently under development using our ultra-high resolution technique. Fig. 2

shows an example of annual layer counting for the period ca. 1310 C.E. to 1317 C.E., illustrating seasonal variability in dust-source Ca. Fig. 2 (as well as Fig. 5) presents the raw data (red) and a smoothed line as a visual aid (black). In the time range between ca. 500 C.E. and 1500 C.E. the ultra-high resolution annual layer counting data is also backed up by ^{14}C ages, retrieved from analysis of the particulate organic carbon fraction [*Hoffmann et al.*, 2017]. This ^{14}C data was developed completely independently from the layer counting. Comparison reveals very good agreement with the annual layer counting within a 1σ error range (Fig. S1).

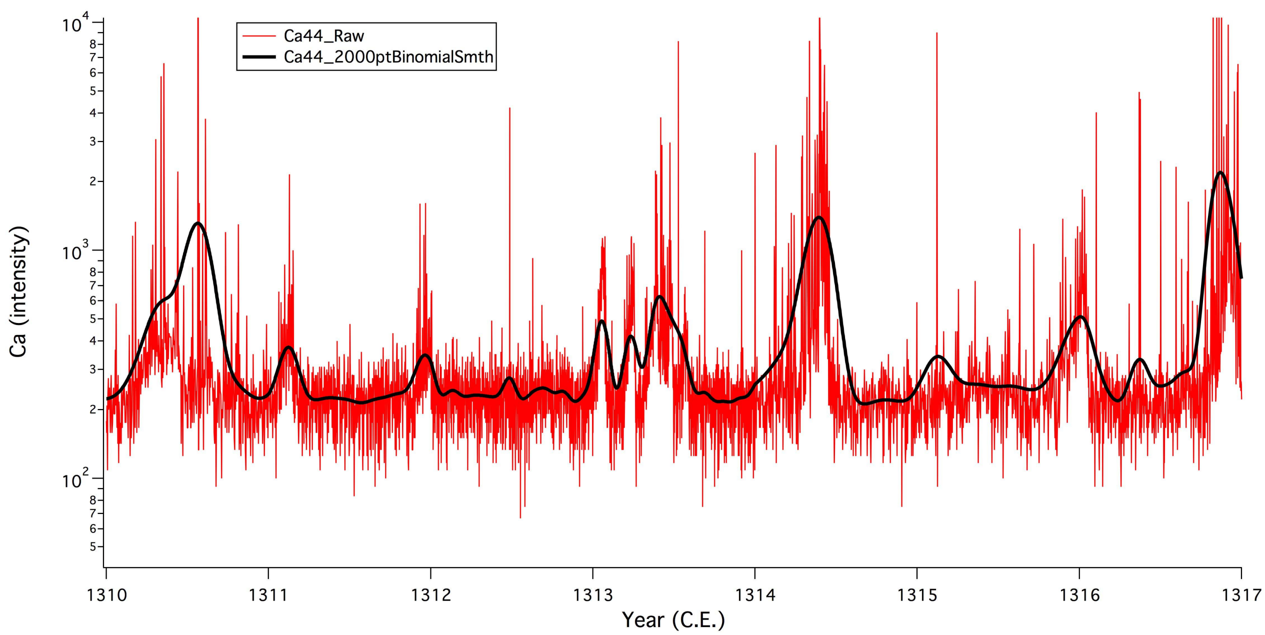

Fig. 2. Example of annual layer counting using ultra-high resolution LA-ICP-MS. Annual layers were identified as local maxima in the Ca-profile corresponding to snow deposited during high summer season. Relative uncertainty in annual layer counting within the time period represented in this figure is around one-two years. Smoothing in this figure (black line) is displayed only as a visual aid.

It is important to note that all annual layer counting for the CG ice core was completed prior to comparison with written historical evidence collected and analyzed by the Initiative for the Science of the Human Past at Harvard. The independently developed sub-annual resolution record derived from the CG ice core thus allowed testing against sub-annually resolved historical records. Sources in Latin, Middle English, English, French, German, and Italian provided sub-annual dates (months, year) for the arrival and spread of the Black Death throughout Europe, as well as decadal to sub-annual trends in mining and smelting activity from the last two millennia. Extensive archaeological and historiographical evidence corroborated our conclusions and timescale (Tables S2-5).

Prior to ultra-high-resolution LA-ICP-MS analysis, high-resolution ICP-MS discrete analysis (~4.27 cm average sample resolution over the ca. 2000-year record), also conducted by CCI, independently revealed a dramatic drop in atmospheric Pb levels falling exactly within the period of the Black Death (1349-1353 C.E.), the greatest pandemic to ravage Eurasia in recorded history. Previous studies of atmospheric lead in low-resolution ice-core records available for the last two millennia did not document this same, sharp, multi-year decline to undetectable levels [*Hong et al.*, 1994, *Gabrieli et al.*, 2014]. Potential uncertainty in our layer-counted depth-age time scale was initially estimated to be less than 35 years at this interval, based on the lag generated by comparing our CG time series and previous annually dated CG time series [*Bohleber et al.*, 2013]. Further, we found that our Pb deposition record was in good agreement with shorter, multi-year resolution CG ice-core records for the period ca. 1650-2000 C.E. [*Schwikowski et al.*, 2004, *Gabrieli et al.*, 2014].

3 Results: Consilience of highly detailed historical evidence and the glaciochemical record

Remarkably, the drop in Pb concentration, captured by both the discrete ICP-MS and the continuous LA-ICP-MS methods, coincides with written historical evidence of the effects of the Black Death pandemic on European populations and metal production, and parallels data on similar downward trends in atmospheric CO_2 levels in the same period, due to population decline [*van Hoof et al.*, 2006]. The coincidence of the two independently derived time series (ice core and written record) and in particular the unique nature of the Pb drop in the ice core record at this confluence confirms the ice core dating of this event. The discrete Pb levels corresponding to the layers counted as years 1349-53 C.E. are the lowest in our record, and are much lower than levels documented in even the deepest CG layers, indicating that for at least the past two millennia human mining and smelting activities have been the originator of detectable lead pollution in the European continent.

Our findings are in sharp contrast with a consensus among policy makers and industry experts that ascribes a significant portion of pre-industrial atmospheric lead levels to natural, e.g. crustal or volcanic sources [*UNEP*, 2010, *Richardson et al.*, 2001]. The new measurements indicate that this consensus overestimates the contribution of such natural sources to current lead levels in the atmosphere. The location of the CG ice core in the heart of Europe provides a geographically specific signal. Whereas, for example, the first polar ice core detections of historic metal pollution were unable to distinguish clearly Roman and Chinese Empires' production areas, the new CG ice core's location is relatively close to the mining and smelting centers of western Europe from the historical beginnings of smelting activities to the present. The long-range transport necessary for heavy lead particles to reach and be trapped in polar ice is more difficult to interpret than the shorter distances between source and the Alpine core. Therefore, while long-range transport is necessary for heavy lead particles to be trapped in polar ice, the proximity of potential Pb sources offer a more precise, definitive, continuous and regionally specific signal.

Historical records show that massive mortalities in the spring and summer of 1349 C.E. halted metal production in all the major Pb-producing regions of western Europe (fig. 4, Table 1). During the pandemic, 30-50% of the European population died [*DeWitte et al.*, 2008]. Extensive archaeological investigation has recently estimated 45% in mortality in Eastern England [*Lewis et al.*, 2016], principally due to bubonic plague (*Yersinia pestis*), now definitively identified by genome sequencing [*Bos et al.*, 2011]. Throughout its ca. 2000-year record, the CG ice core shows levels of Pb significantly higher than those recorded for the Black Death.

The lowest Pb levels recorded in our study occurred during the Black Death (0.4 ng/L at 1353

C.E. in the high-resolution discrete ICP-MS, and below the limit of detection at 1351 C.E. in the ultra-high resolution LA-ICP-MS) and likely represent dispersal of Pb from the earth's crust, that is, as close to natural background Pb levels as were achieved in the full ca. 2000-year record. The new measurements significantly alter our understanding of atmospheric Pb pollution hitherto labeled as natural background and therefore assumed to be safe. Thus, they challenge the assumption that pre-industrial atmospheric Pb levels had no discernible effect on human physiology. These new data show that human activity has polluted European air almost uninterruptedly for the last ca. 2000 years. Only a devastating collapse in population and economic activity caused by pandemic disease reduced atmospheric pollution to what can now more accurately be termed "background" or natural levels. Pb crustal enrichment factors (EF_c, see also SI for potential volcanic EF influences and further discussion, fig. S4) evaluating the extent of anthropogenic soil contamination for the years corresponding to the Black Death corroborate our interpretation. They show a marked decline, reaching a value of 2.82 in the year 1352, the second lowest in the entire record (the past ca. 2000 years) with the lowest EF_c occurring in 1366, with a corresponding value of 2.36. The latter date corresponds very closely to the date range of a further plague pandemic between 1367-9, the impact of which is dramatically documented in the Halesowen manorial court rolls in the West Midlands of England [*Razi*, 1980].

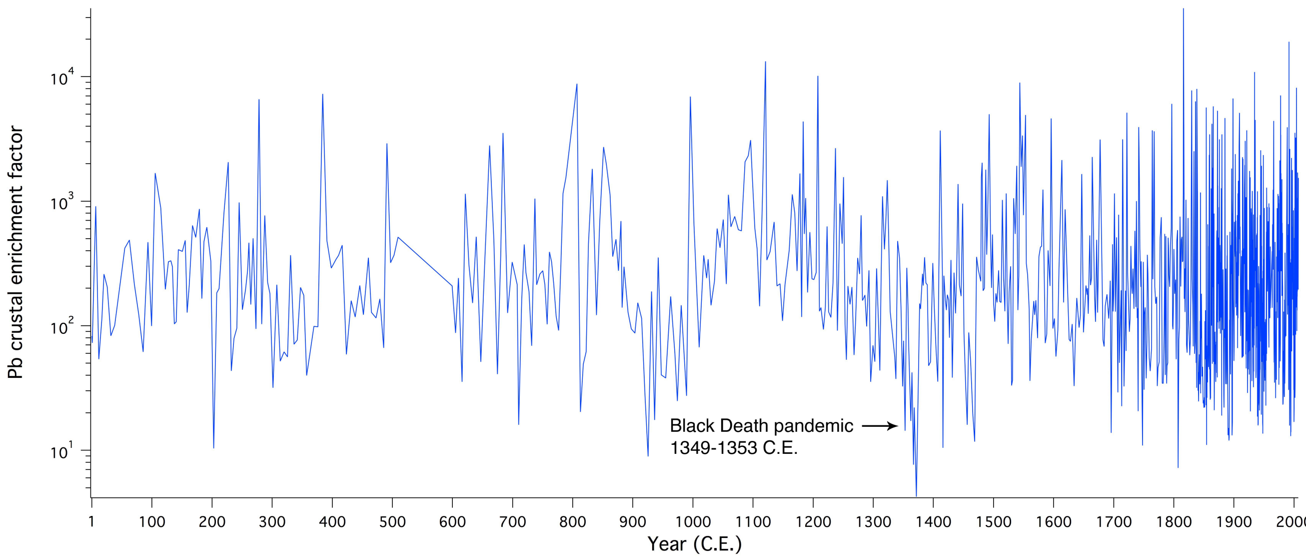

Fig. 3. Pb crustal enrichment factor (EF$_c$). The crustal enrichment factor calculations (using *Wedepohl*, 1995 dataset) are shown in SI. The graph covers the period ca. 1 - 2007 C.E. The Black Death drop marks the years 1349-1353 C.E. Values below 10 here are based on semi-quantitative calibration data.

In the Alpine region of Europe, high-level regional delivery and lower-level atmospheric circulation transport pollutants [*Schwikowski et al.*, 2004, *Gabrieli et al.*, 2014]. Modern atmospheric circulation patterns associated with the Azores High (fig. 4 and fig. S2) point to potential British, French and German sources of pollution transported to CG. Our record of the multi-year Black Death period is not associated with any anomalous atmospheric circulation patterns, based on Ca and Fe as crustal air mass proxies (fig. S3). Comparison with historical evidence from SoHP's geodatabase of climate events also presented no substantial change in observed climate patterns in the region at the time (Database S1). This leaves a dramatic decline, if not complete interruption of anthropogenic emissions of Pb at the time of the Black Death as the most likely dominant control, especially in light of documented Pb residence times in the troposphere, averaging a week to ten days [*Papastefanou*, 2006].

Historical documentary evidence—fiscal, legal and chronicle sources—shows that while Pb production in the Harz Mountains was already in severe decline in the 1330s, Britain dominated western European Pb production until the plague reached its regions of most labor-intensive mining between January and September 1349 C.E. (Table 1, Tables S2-5). We argue that British mines and smelting sites were the likely dominant source of Pb captured in the CG ice core at the time of the 1349-1353 C.E. collapse, since they were by far the principal producers in this period. Extensive and large-scale mining and smelting were largely constrained within the principal British lead producing regions by 1348, such as the High Peak District region of Derbyshire [*Blanchard*, 2005, Table S5], the Bere Ferrers mine in Devon [*Claughton*, 2010] and to a lesser extent at that time in the Yorkshire Dales and the hills of Shropshire and Flintshire [*Claughton et al.,* 2016]. Coincident location of both galena ore sources and woodland for fuel were the key factors governing the largest regional concentrations of these activities in Britain. The movement of the raw ore of metals such as iron by water is attested archaeologically, when coastal waters and shipping were immediately available, indicated from the mid thirteenth-century Magor Pill ship from the Welsh shore of the Bristol Channel [*Claughton et al.,* 2016*, Nayling,* 1998]. Dressed galena ore is recorded as having been paid by miners in the Peak District in the form of renders to local landowners for smelting, usually to the King or major aristocrats, from the twelfth century onwards but evidence of the movement of galena for smelting outside the Peak or other principal mining regions is currently lacking. Lead is only attested textually and archaeologically as having been moved inter-regionally and over long distances in its smelted form between the ninth and fourteenth centuries, over land and by water, as ingots or sheet [*Rieuwerts,* 1987, *Allen,* 2011, Kelly and Brooks*,* 2013]. The constraint of lead production to paramount mining and smelting regions in England is further demonstrated by specific traits within their regional economies. For example, the payment of rents and tithes to local landowners in dressed ore or smelted lead and the use of the metal as a medium of barter exchange [*Rieuwerts*, 1987, *Barnatt and Smith*, 2004, *Blanchard*, 2005, Table S5]. Other potential non-British sources of pollution, such as the French mines and woodland smelting sites west of CG, at Mont-Lozère, had already ceased activities by 1280 C.E. [*Baron et al.*, 2006]. Sardinia, the most significant Mediterranean Pb producer, was in deep decline already in the 1330s; moreover, the island lies outside the dominant atmospheric transportation pattern and had already been ravaged by plague in 1348 C.E. (fig. 4, Tables S2-5).

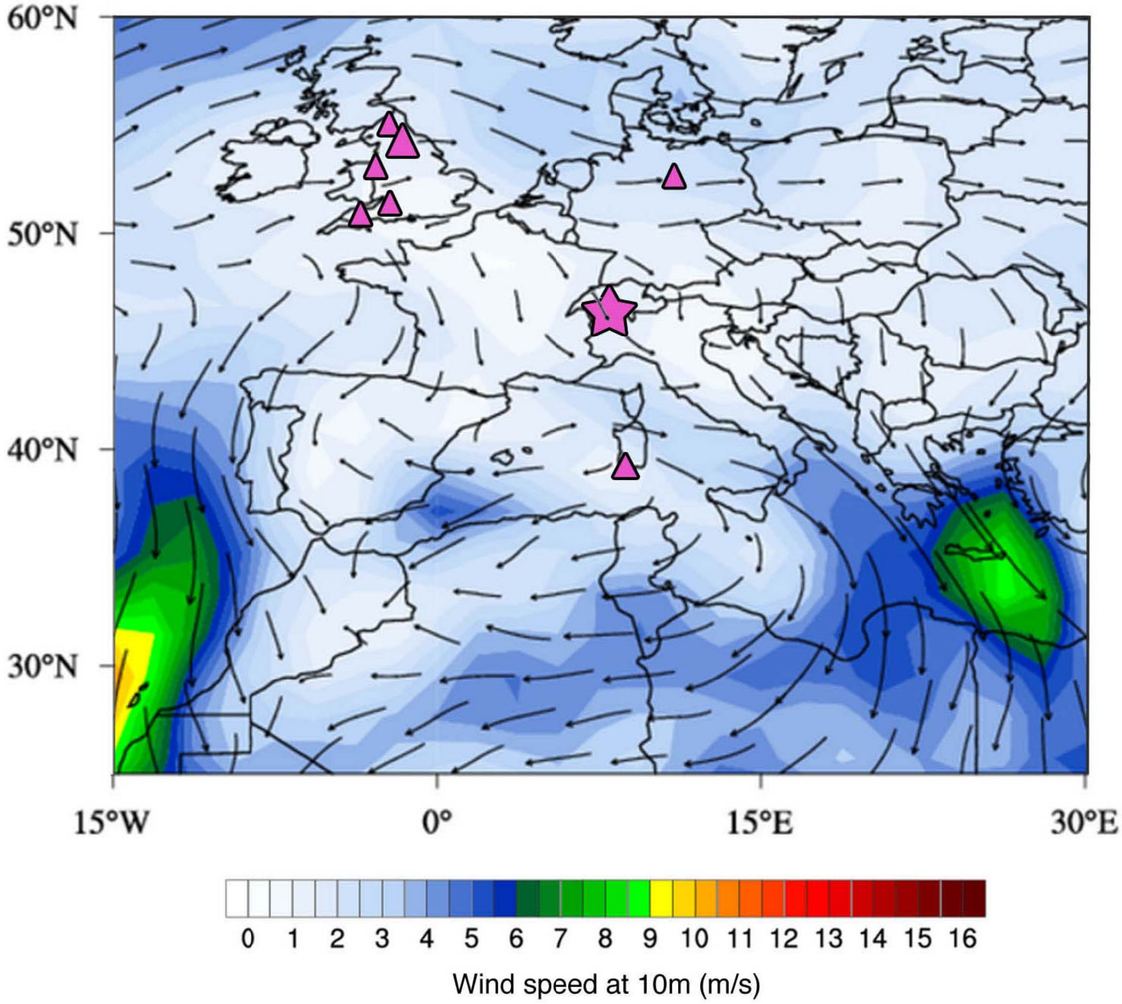

Fig. 4. Summer average atmospheric circulation (wind speed, m/s). NOAA/CIRES 20th-century reanalysis V2, JJA example 1984 is visualized using CCI's web-based Climate Reanalyzer using. The location of CG is highlighted with a star ( ) and major Pb/Ag mining centers with triangles ( ) 1347-1460 C.E. Size of triangle markers indicates approximate volume of production based on written sources.

The ultra-high-resolution CG data (fig. 5) show a steep progressive decline in Pb deposition, from layers dated 1349 C.E. to 1352 C.E., corresponding to the progression of the pandemic through different lead-producing areas. The arrival of the plague in the most productive British mining regions in the second half of 1349 C.E. corresponds to sub-annual LA-ICP-MS data points showing the beginning of the most severe drop in Pb concentration in the ice core. Table 1 summarizes the dates when the plague reached the British, German and Italian mining regions, and when mining and smelting operations were interrupted. Mining resumed sporadically and progressively from 1352 C.E. (Table S4), when some of the pre-plague mining sites reopened in Britain (High Peak District, in Derbyshire) along with new mines (North Yorkshire), but production levels fluctuated for a century due to the more limited demands of a population reduced by ca. 50%. There is no evidence of new mining or smelting in Sardinia until ca. 1420 C.E., nor in the Harz until the 1460s C.E. [*Blanchard*, 2005, *Dyer*, 2000].

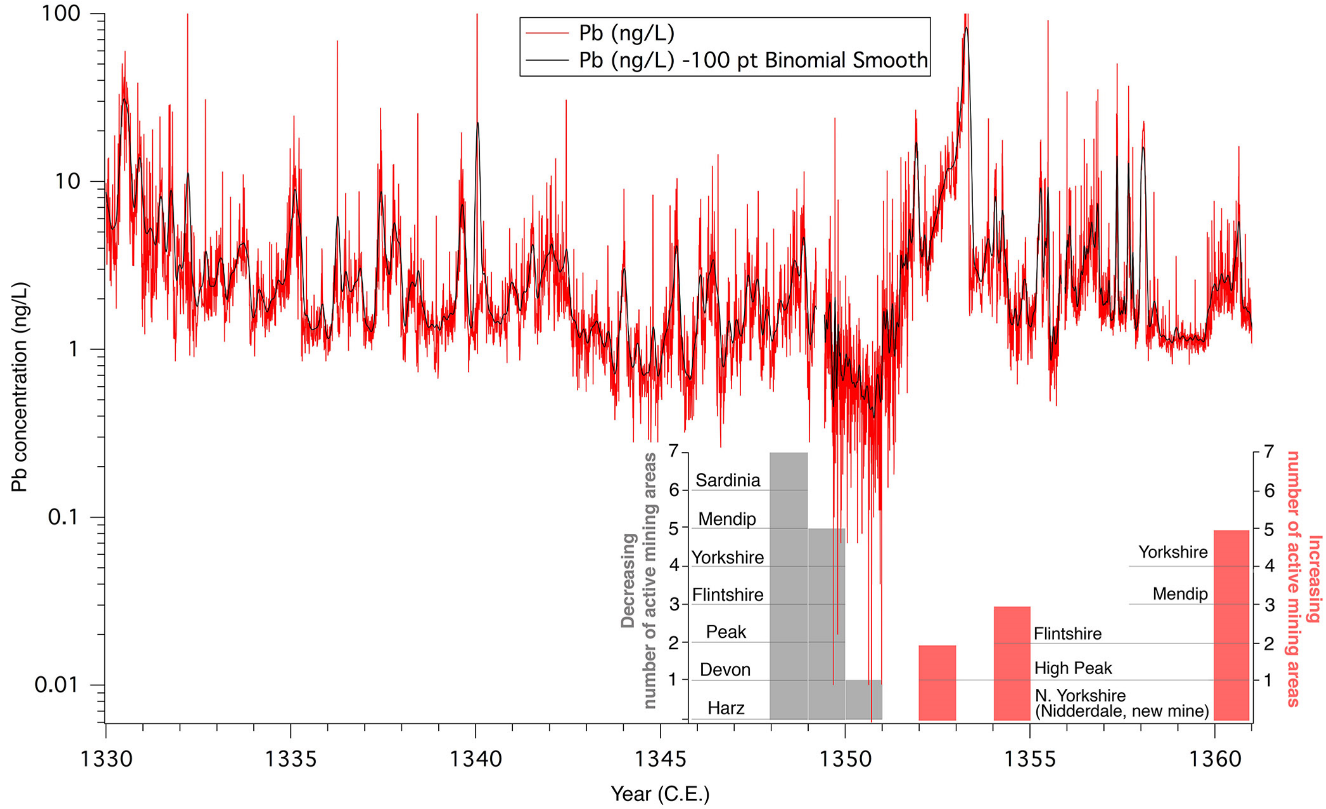

Fig. 5. Lead concentration in CG ice core, from ultra-high-resolution LA-ICP-MS, 1330-1360 C.E. (with an average of 279 measurements per year in 1349-1353). Grey histogram represents declining number of active major mining regions as they were progressively hit by the plague and ceased operations; red histogram represents number of mining regions resuming metal production, based on written sources. At present, there are no estimates of volume of aggregate metal production and thus the histograms reflect only regions that were active, not volume of Pb produced. Values below 1ng/L here are calculated using semi-quantitative calibration data. Smoothing (black line) is provided only as a visual aid, while the red plot presents the raw data. As shown in the methods section (SI), the LA-ICP-MS technique [*Sneed et al.*, 2015] measures total element concentration; spikes can thus be related to individual particles and/or storm event concentrations.

Table 1. Arrival of epidemic disease and cessation of operations in major Pb mining centers.

City/Region	Black Death arrives	Year Pb/Ag mining ceases
	BRITAIN	
Mendip	1348/1349	1340s
Devon	1349 March	1349
Flintshire	1349 June	1349/50
Derbyshire (Peak)	1349 May	1349-52
York	1349 May	
	GERMANY	
Harz (Goslar)		1350
Harz (Halberstadt)	1350 May	
Magdeburg	1350 May	

For details, see Tables S2-5. Dates adjusted to modern calendar, whenever appropriate (see SI for details).

In the high-resolution discrete CG Pb data (fig. 1), a second severe drop corresponds to the period 1460-65 C.E. Historical records show that British mining activities declined drastically at this time due to market oversupply, probably linked to another series of epidemics that affected Britain, as well as lower demand due to an economic downturn (Tables S2-4) [*Blanchard*, 2005, *Dyer*, 2000, *Nightingale*, 2005, *Gottfried*, 1977, *Hatcher*, 2003, *Creighton*, 1891]. Resurgence of Harz mining activities in the 1460s [*Bartels*, 2010] is not detected at CG, suggesting that German mines were either not a major contributor to Pb deposition at CG at that time, or that their emissions from smelting were relatively low. Pb crustal enrichment factors also reflect this second decline (Fig. 3)

The third lowest level of Pb deposition in the discrete ice core record corresponds to the year 1885. Mining activities slumped in that year due to the long-term economic collapse that affected Western countries in 1882-5 [*Brayshay* 1980]. A similar trend is observed in the United States in 1885, the year in which Pb production levels declined most severely in extant historical records dating back to the late 18th century [*Mushak*, 2011, *Brayshay*, 1980, *USGS*, 2013]. The most recent decrease in atmospheric Pb levels in Europe began in 1974. This decline reflects legislative efforts to phase out leaded fuel in Western countries, which resulted in decreased blood levels of Pb throughout Europe and the United States [*Schwikowski et al.*, 2004, *Strömberg et al.*, 2008, *Gabrieli et al.*, 2014].

4 Conclusions

Ultra-high-resolution measurements from the heart of Europe, combined with a densely documented historical and archaeological archive, usher in a new era in the detailed reconstruction of human interaction with the environment. Anticipating the forthcoming reduction of dating uncertainty in the deepest ice-core sections, the examination of pre-Black Death Pb deposition levels (fig. 1) points to intriguing areas of future research such as Europe's shift from gold to silver coinage with the opening of new Ag/Pb mines in France (Melle), between 640 and 680 C.E. Similarly, our new measurements of Pb deposition suggest that Europe's booming metal production ca. 1180-1220 C.E. (the highest pre-industrial Pb peak in our record) may have generated pollution levels rivaling those ca. 1650 C.E. Since previous research has correlated deposition levels to volume of emissions of sulfur, copper, uranium, arsenic, and lead in earlier ice cores, for example [*Schwikowski et al.*, 2004, *Mayewski et al.*, 1986], we expect that future research will elucidate whether deposition levels captured by the new ultra-high-resolution method can be correlated more precisely and quantitatively with historical volume of emissions and, potentially, of production levels. Our study also points to the need to explore possible connections between historic atmospheric Pb pollution and ecosystem health, including human fertility, intelligence, and behavior. Such trans-disciplinary research will represent a significant contribution in the planetary health field, in line with the aims outlined in *Amada et al.*, 2017.

In this paper we have mobilized more than a million new environmental data points using ICP-MS and LA-ICP-MS in conjunction with highly detailed historical records to show the devastating impact of the Black Death on European metal production, an insight into the pandemic's effect on human activity, demographics and population health. In the last ca. 2000 years, only two other instances (in the 1460s C.E. and in 1885 C.E.) even remotely approached Black Death Pb deposition levels, either due to economic decline or epidemic disease, or both. Our findings imply that what were once believed to be background Pb levels represent, in fact, a significant anthropogenic component of the atmosphere over the last ca. 2000 years. The sole exception was a four-year period at the time of the Black Death when atmospheric Pb pollution dropped to levels analytically undetectable by LA-ICP-MS. The geographic proximity to pollution sources and ultra-high resolution of the data presented here provide the most detailed, updated, regional record of European Pb pollution for the past two millennia, and indicate that manmade pollution has been and continues to be a major contributor to lead levels in the atmosphere. Current policies and industry consensus, based on the assumption that current Pb atmospheric levels contain a significant "natural" Pb contribution, are thus clearly misleading. The health implications of such anthropogenically elevated levels of Pb in the atmosphere need further investigation in light of these new data.

Author Contributions:

N.S., S.S., P.B. and M.H. conducted the sampling, analysis and annual layer counting. E.K. calculated enrichment factors. H.H. conducted radiocarbon analysis. P.A.M. and A.K. contributed climatological, glaciological and atmospheric circulation analysis and expertise. A.F.M., C.L. and M. McC. researched historical and current health aspects of Pb poisoning and mining, as well as historical epidemiology, archaeological and historical data. A.F.M. wrote the initial paper draft and all authors met to produce the final draft. All authors discussed the results and commented on the manuscript.

Acknowledgments

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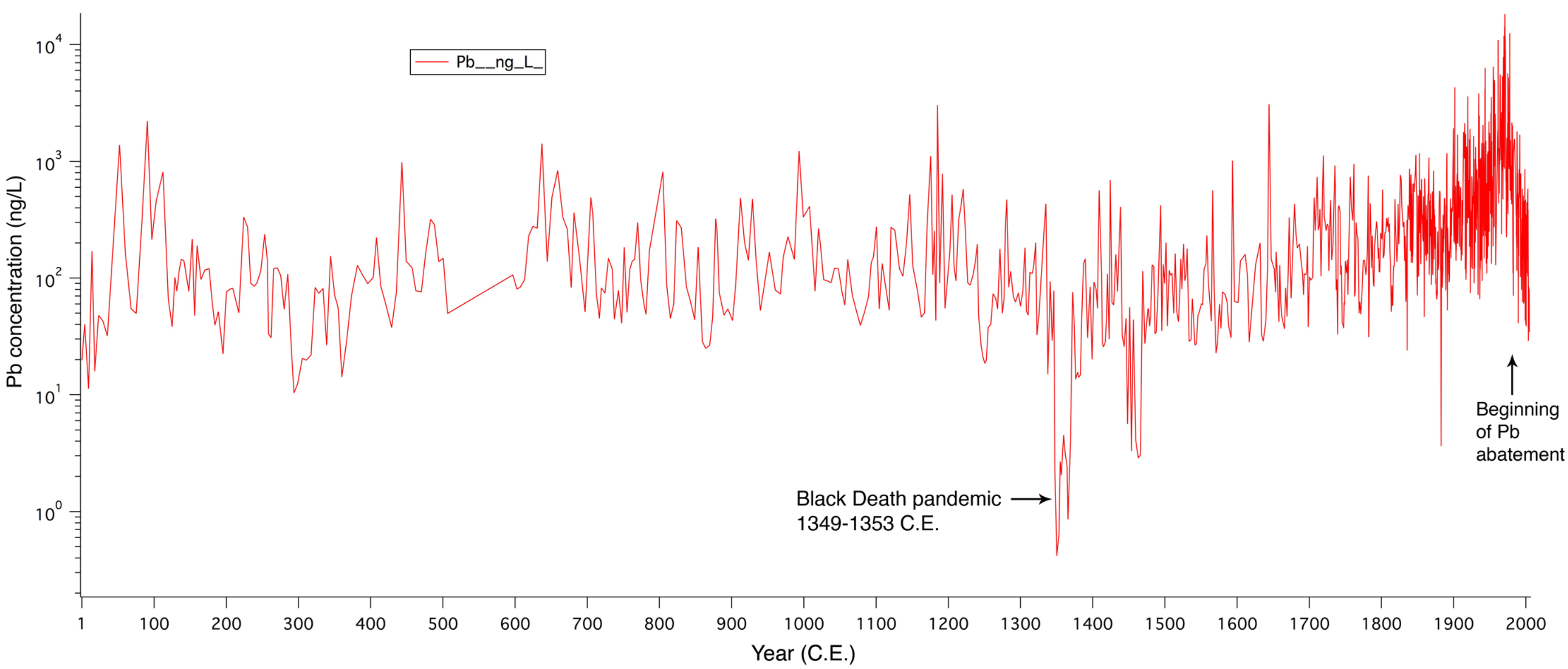
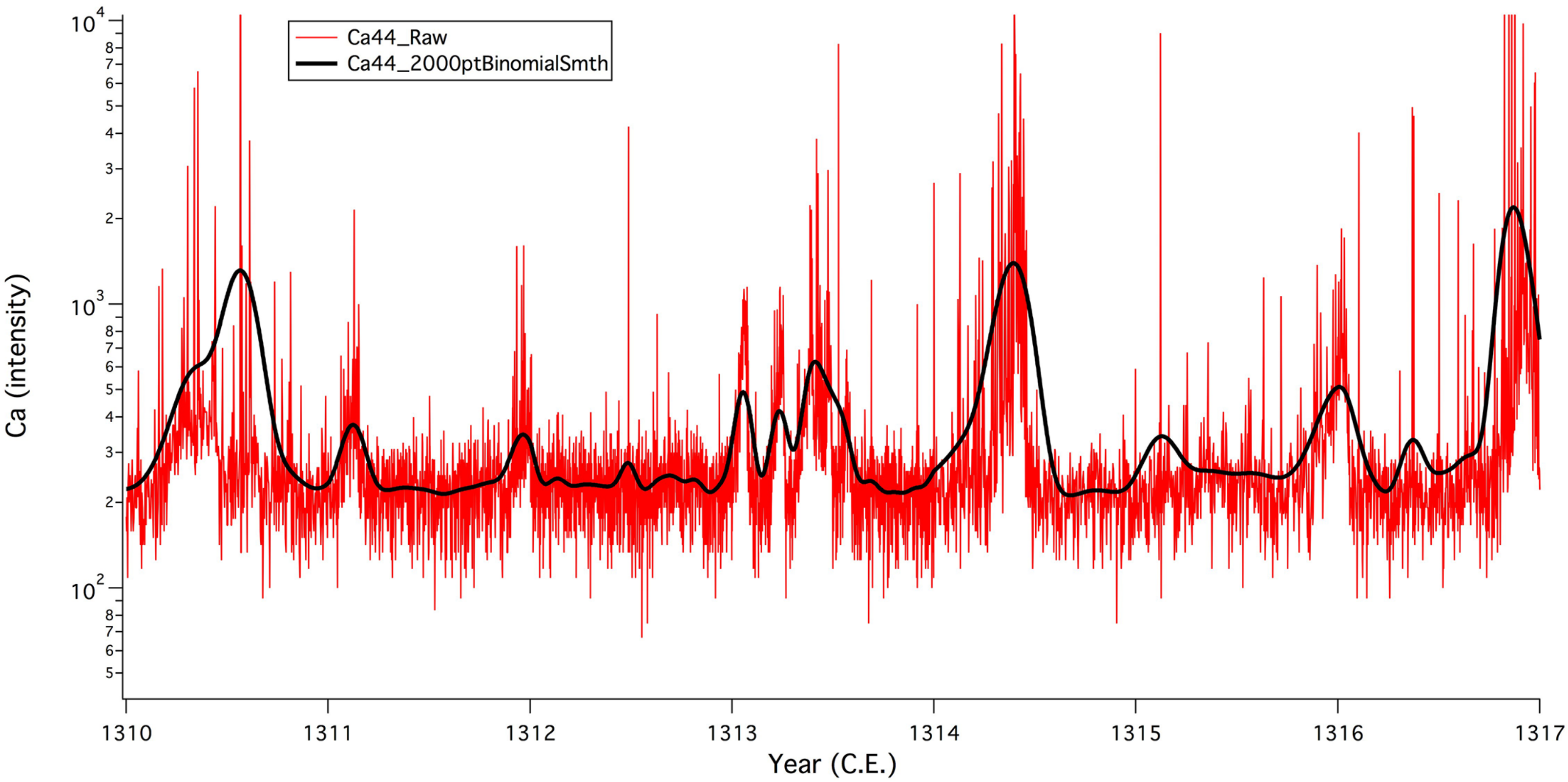
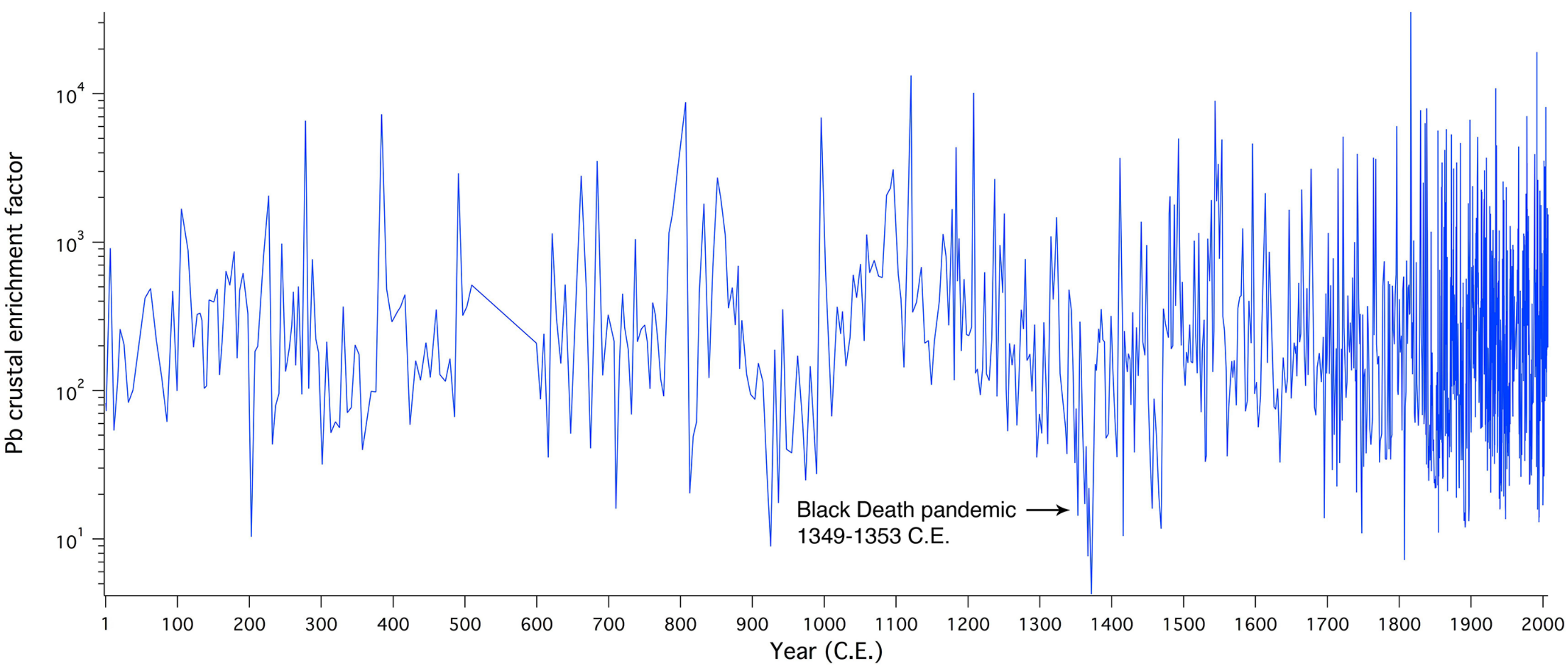
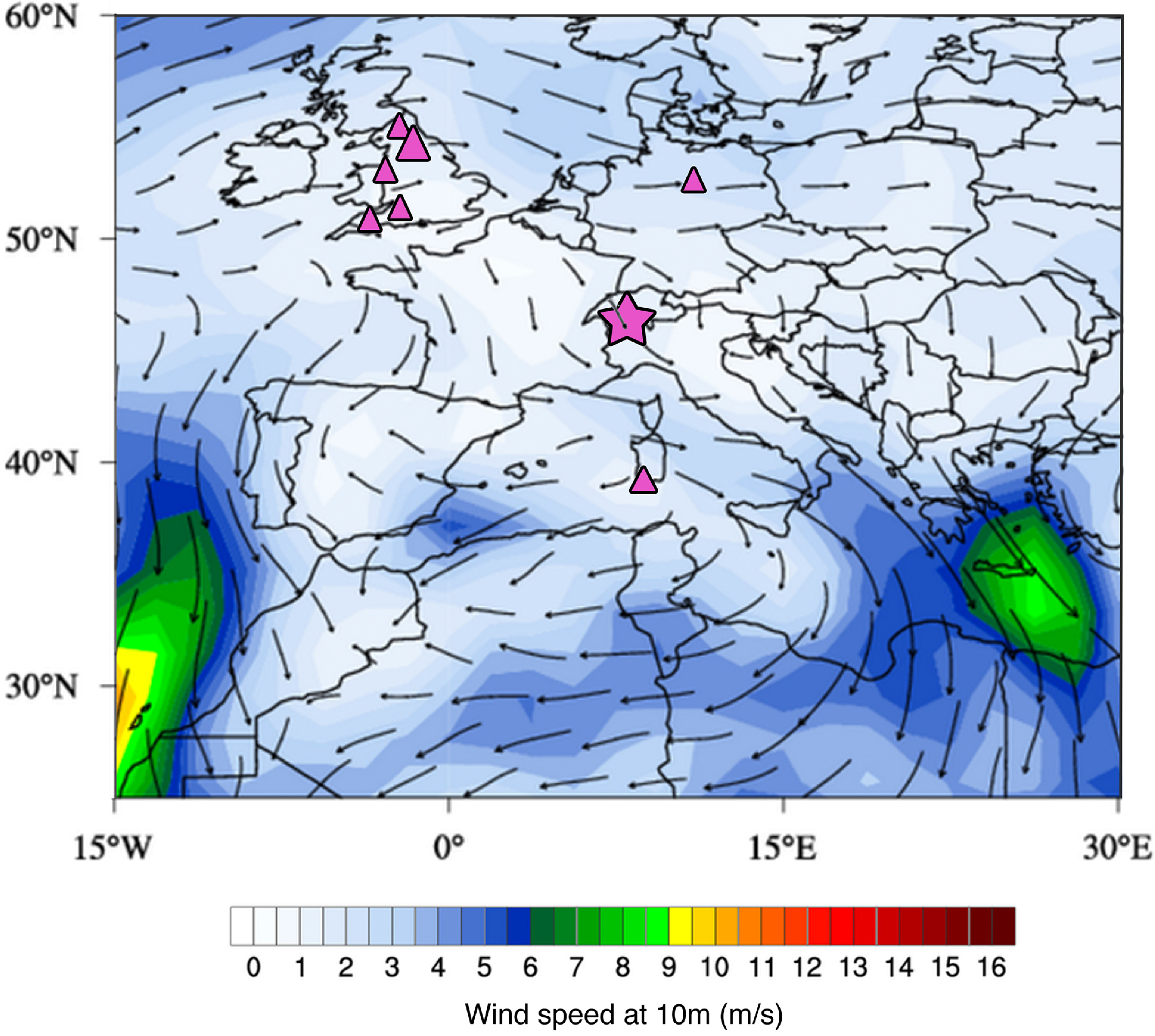


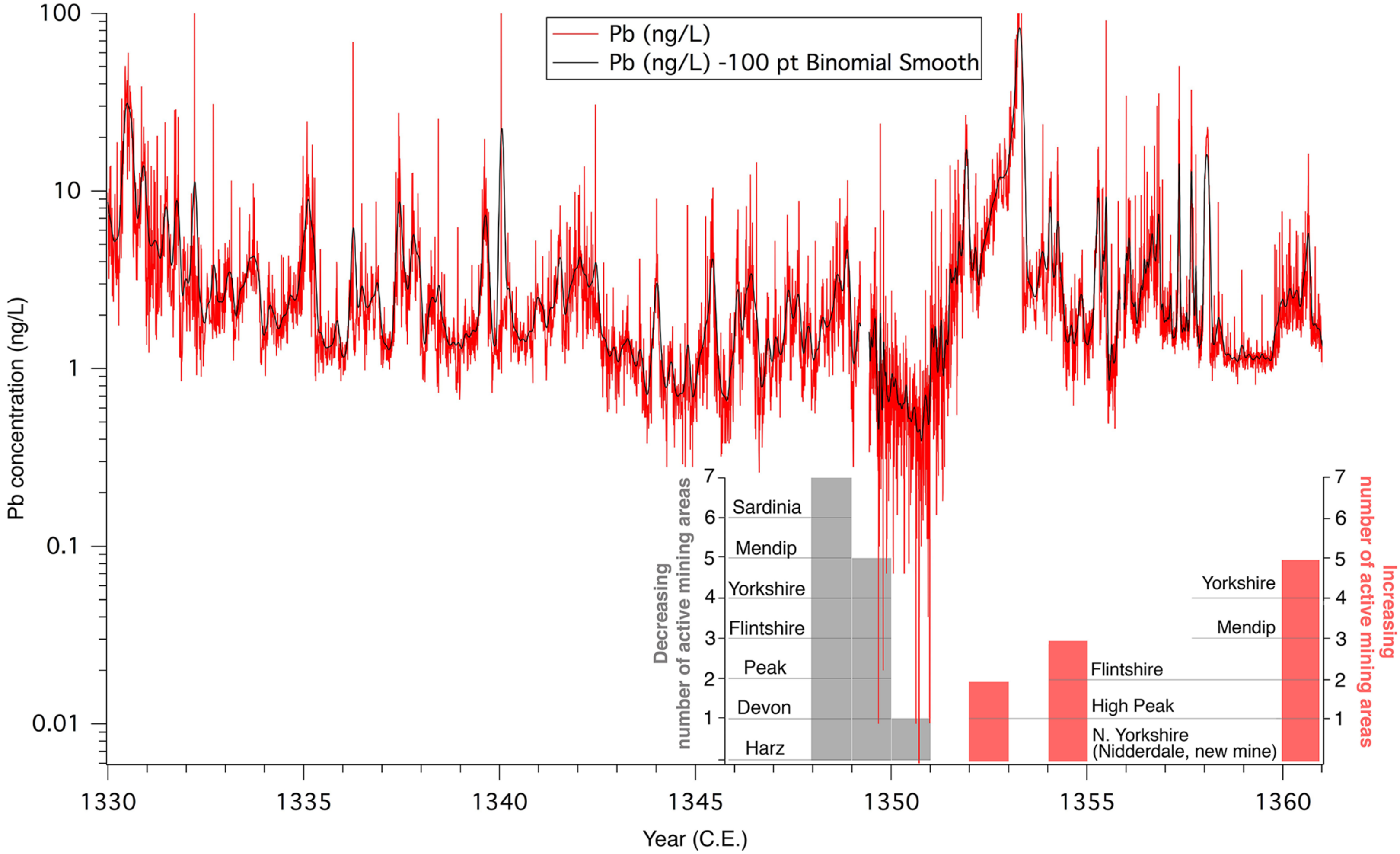
Pb__ng_L_
Pb concentration (ng/L)
Year (C.E.)
Black Death pandemic
1349-1353 C.E.
Beginning
of Pb
abatement


Ca44_Raw
Ca44_2000ptBinomialSmth
Ca (intensity)
Year (C.E.)
1310
1311
1312
1313
1314
1315
1316
1317



Pb crustal enrichment factor
10^4
10^3
10^2
10^1
Black Death pandemic
1349-1353 C.E.
1 100 200 300 400 500 600 700 800 900 1000 1100 1200 1300 1400 1500 1600 1700 1800 1900 2000
Year (C.E.)


60°N
50°N
40°N
30°N
15°W
0°
15°E
30°E
0
1
2
3
4
5
6
7
8
9
10
11
12
13
14
15
16
Wind speed at 10m (m/s)



Pb (ng/L)
Pb (ng/L) -100 pt Binomial Smooth
Pb concentration (ng/L)
Year (C.E.)
Decreasing number of active mining areas
Increasing number of active mining areas
Sardinia
Mendip
Yorkshire
Flintshire
Peak
Devon
Harz
Flintshire
High Peak
N. Yorkshire (Nidderdale, new mine)
Yorkshire
Mendip