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COMMENT
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This article is a comment on More et al. (2017) https://doi.org/10.1002/2017GH000064.

Key Points:

- Four decades of research have detailed the Pb pollution history in Europe, including background levels occurring >3,000 years ago
- Mining and metallurgy occurred in a wide range of locations resulting in local and regional histories within a common European narrative
- Colle Gnifetti record contains many interesting details, but further examination of the chemistry is needed to understand the Pb record

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Comment on "Next-Generation Ice Core Technology Reveals True Minimum Natural Levels of Lead (Pb) in the Atmosphere: Insights From the Black Death" by More et al.

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Abstract Over the past four decades numerous studies of lake sediment, marine sediment, and peat from sites in close proximity to mining or metallurgical centers and in remote locations have detailed local and regional histories of lead (Pb) pollution in Europe. Contrary to More et al.'s (2017, https://doi.org/10.1002/2017GH000064) claim that "previous assumptions about preindustrial "natural" background lead levels in the atmosphere have been misleading," these studies have clearly shown that true natural background conditions occurred more than 2,500 or even 3,500 years ago, and Pb pollution has proceeded uninterrupted since. The implications of this have been discussed within the context of environmental policy, for example, European Water Framework Directive. Though these records reflect a common European narrative of mining, metallurgy, and pollution, each reflects a combination of local and regional events, leading to differences in the timing and intensity of changes in each Pb record. No one record—ice or otherwise—fully represents the three millennia Pb pollution history in Europe. While the resolution of the ice record is impressive, there are questions about its interpretation. First, the authors discount local and regional Pb sources, whereas there is a close connection between the mining history in an area 40 km from the glacier and changes in a nearby lake Pb record; second, significant changes in ice chemistry cooccurring with the lowest Pb values are overlooked. A sharp increase in Ca/Fe ratios occurs precisely with the steepest Pb declines during the Black Death and mid-1400s, suggesting additional processes influencing the Pb record.

1. Four Decades of Research on Three Millennia of Pb Pollution in Western European

The analytical developments in ice core research are impressive, not least the ability to perform very high-resolution and even continuous trace-level analyses of ice cores that may result in "a million new environmental data points.". However, a million new data points may not necessarily result in new knowledge. While there are remarkable details in this new high-resolution analysis of a 2,000-year ice record from the Swiss-Italian Alps by More et al. (2017)—notably the precipitous drop in lead (Pb) at the time of the Black Death that is one focus of the paper—the broader context and wider implications are already well established. Phrases such as "reveals true natural levels," "contrary to widespread assumptions," "significantly alter our understanding," and "new data show that human activity has polluted European air for the last c. 2000 years" implicitly suggest the authors have uncovered something not previously known and lead to a few criticisms that I address below. The authors place too much emphasis on the historical story, but insufficiently on a complete analysis of their data and not at all on putting their data into the well-established framework of millennial Pb pollution in Europe. The many references I cite here represent only a sampling of this work.

It is unclear what the authors mean precisely by "widespread assumptions" or "alter[ing] our understanding" regarding natural Pb levels. The period of the Black Death is certainly the lowest point in this one ice record, but more than four decades of research involving analyses of lake-sediment, marine-sediment, and peat records from many sites across Western Europe has already established not only that human activities have contributed to atmospheric Pb pollution uninterruptedly over the past 2,000 years, but that it is necessary to go back 2,500 or even 3,500 years in order to establish background, prepollution conditions (Figures 1b–1f), which is beyond the 2,000-year time frame of the ice record. These studies have included sites in, for example, England, Switzerland, Sweden, Spain, France, Germany, Scotland, and Austria (Bindler et al., 1999; Brännvall et al., 1999, 2001; Breitenlechner et al., 2010; Cloy et al., 2008; Elbaz-Poulichet et al., 2011; Forel et al., 2010; Garcia-Alix et al., 2013; Jouffroy-Bapicot et al., 2007; Klaminder et al., 2003; Küttner et al., 2014;

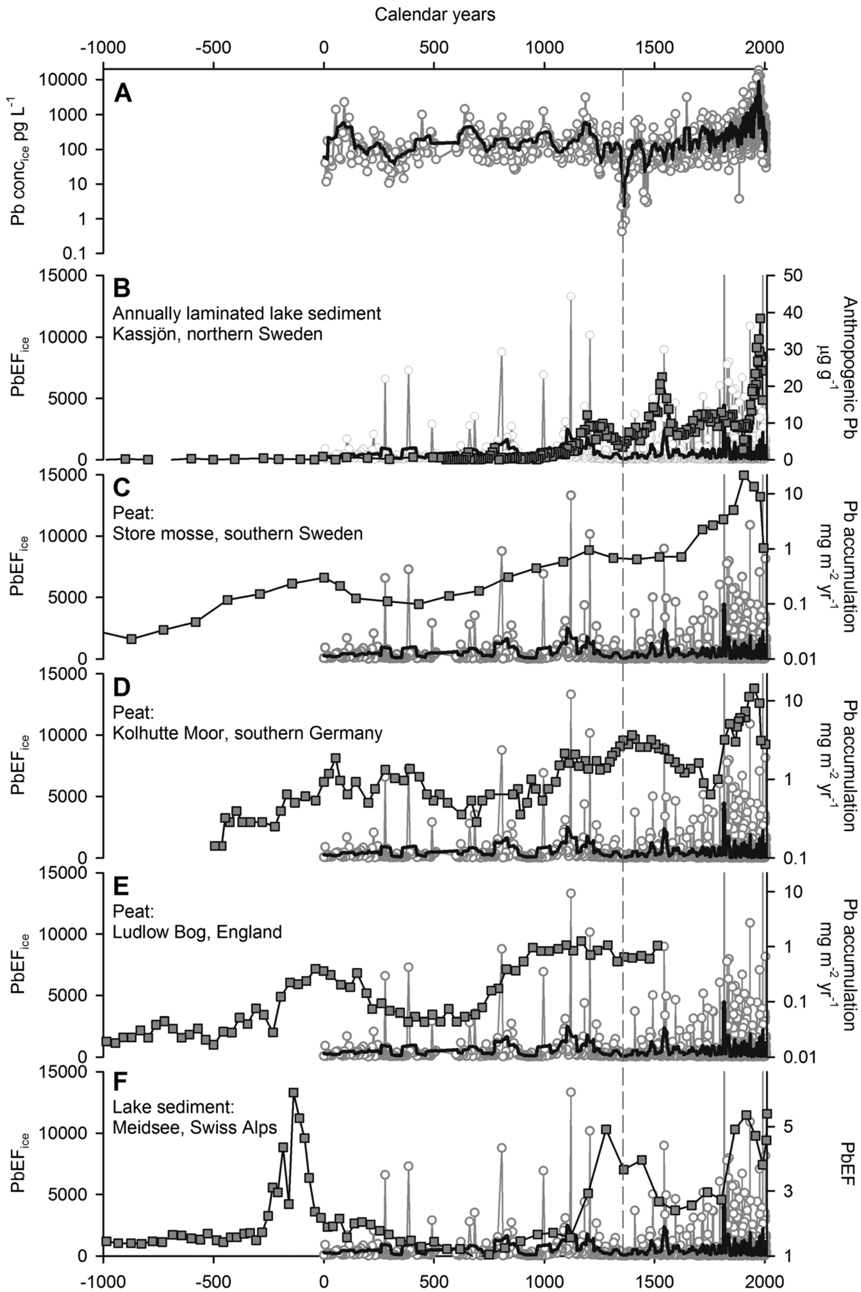

Figure 1. Comparison of the new, high-resolution, 2000-year ice record from the Swiss-Italian Alps with other lead records from Europe: (a) discrete lead concentration data from the Colle Gnifetti ice record (with superimposed 10-point running average), and comparisons of the Pb enrichment factor from the ice record (light grey circles and 10-point running average) in comparison to (b) annually laminated lake-sediment record from Kassjön, northern Sweden (Brännvall et al., 1999); (c) peat record from Store Mosse, southern Sweden; (d) peat record from Kolhütte Moor in the Black Forest, Germany (Le Roux et al., 2005); (e) peat record from Ludlow Bog, England (Le Roux et al., 2004); and (f) lake-sediment record from the alpine lake Meidsee, Switzerland (Thevenon et al., 2011), located about 35 km from Colle Gnifetti. The dashed vertical line corresponds to the peak of the Black Death, 1350 CE, discussed in detail by More et al. (2017) (note that the *x* axis units are different for the lead records).

Kylander et al., 2005; Le Roux et al., 2004, 2005; Leblanc et al., 2000; Lee & Tallis, 1973; Manteca et al., 2017; Martínez-Cortizas et al., 2002, 2016; Mil-Homens et al., 2016; Shotyk et al., 1998; Thevenon et al., 2011). This research in turn rests on the exploratory work by Clair Patterson and coworkers during the 1960–1980s of the historical dimensions of metal production (Murozumi et al., 1969; Patterson, 1965, 1971, 1972; Settle & Patterson, 1980).

This large body of work on environmental archives covering the Pb record in Europe is summarily addressed by More et al. (2017) in just five lines in the introduction, and surprisingly, thereafter, the results of the ice core analyses are never evaluated and discussed within the context of this work.

2. Assumptions of Natural Background Levels and Environmental Implications of Three Millennia of Pb Pollution

"Previous assumptions about preindustrial "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….

The new measurements significantly alter our understanding of atmospheric Pb pollution hitherto labeled as natural background and therefore assumed to be safe."

A substantial body of work has preceded the authors' assertion that the million new environmental data points "significantly alter our understanding" of the environmental and health implications of millennial Pb pollution. Whereas More et al. (2017) can only allude to the health and environmental implications of their ice core data, there have been studies that have explicitly explored these concepts. Again, Patterson (Settle & Patterson, 1980), as well as Jerome Nriagu (1983), paved the way into investigating the health dimensions of historical metal contamination. Since then a few studies have assessed the metal exposure of preindustrial populations, for example, the analysis of bones from an ancient mining site in Jordan (Pyatt et al., 2000) and tooth enamel in England spanning the period from 5500 to 300 BP (Budd et al., 2004).

It is similarly surprising when More et al. (2017) write, "Previous assumptions about preindustrial "natural" background lead levels in the atmosphere … have been misleading." In a landmark study, Renberg et al. (1994) opened with the statement, "Despite evidence from Greenland ice cores for pre industrial trace metal contamination it is commonly assumed that air pollution in remote areas is a recent problem caused by industrial activities … Here we report … that atmospheric lead deposition increased above background levels more than 2,600 years ago." Their final concluding sentence was, "We believe our investigation challenges the commonly held view of a 'clean' pre-industrial environment."

Since then, the concept of background conditions and the environmental implications of millennial changes in Pb deposition in Europe have been explicitly discussed, such as in regard to soil and sediment quality and the concept of natural reference conditions as well as how these data could inform present-day environmental management goals, for example, the European Water Framework Directive (Bindler et al., 2009, 2011). Two decades ago, for example, we modeled the concentration and storage of Pb in Swedish boreal forest soils over the past 5,500 years based on the Pb accumulation records from peat cores (Figure 1c; Bindler et al., 1999; Klaminder et al., 2003). Our objective had been to model a natural background concentration in the biologically active humus layer that we considered the proper reference point for ecotoxicological studies. A guiding tenet to that research was that "accumulation of Pb, and other heavy metals, has occurred over a long-term period, therefore, future environmental goals must be viewed in a long-term perspective."

We later synthesized these and other data from studies of peat and lake sediments, forest soils, and vegetation, expressly linking preindustrial Pb pollution with contemporary biogeochemical cycling (Bindler, 2011; Bindler et al., 2011, 2008; Klaminder et al., 2006; Renberg et al., 2009). But a point of interest in the ice data is that, whereas Patterson (1965) estimated—later supported by analyses of peat records—there had been a 1,000-fold increase in Pb in the atmosphere, the new ice Pb concentrations vary as much as 44,000-fold.

3. European Narrative, but Local and Regional Sources of Historical Pb Pollution

"[O]ur new measurements of Pb deposition suggest that Europe's booming metal production ~1180–1220 C.E. (the highest preindustrial Pb peak in our record) may have generated pollution levels rivaling those ~1650 C.E."

"We argue that British mines and smelting sites were the likely dominant source of Pb"

While there is a common historical narrative to mining and metallurgy in Europe over the past several millennia—and thus also metal pollution (Renberg et al., 2001)—the authors should be cautious in suggesting that one ice record represents European pollution history as a whole, although ultimately they tie events in England to changes in the ice record. When the many Pb records across Western Europe are compared, the specific details such as the timing and intensity differ greatly between sites, indicating the importance of local or regional histories. For example, in Figures 1b–1f there are substantial differences among the records in the timing and intensity of the Roman Pb peak, the onset of a medieval increase in Pb, and the relative importance of each period. One notable comparison is the Ludlow Bog record from England (Figure 1e), which declines over a longer period including the Black Death, but not to background levels, which have not occurred at the site since 1000 BCE.

It is surprising that the authors discuss the alpine ice record mainly in terms of English Pb sources and discount all of the mining areas to be found in close proximity to the Alps. This would include the extensive mining and metallurgy that took place in the Vosges Mountains in eastern France, where Pb concentrations increase from the late tenth century (Forel et al., 2010; Jouffroy-Bapicot et al., 2007; Mariet et al., 2016), as well as many areas in the Alps (Breitenlechner et al., 2014; Cattin et al., 2011; Py et al., 2014). That Pb levels do not return to background at any point in any of these regional records, which indicates that despite the rise and fall of mining and metallurgy at various time points, pollution never ceased.

Another comparison of interest is the Holocene sediment record from Meidsee, a Swiss alpine lake located only ~35 km to the north of Colle Gnifetti. In the Meidsee record the largest Pb enrichment is centered on 140 BCE, which precedes the start of the ice record, with an increase of almost equal magnitude in the Middle Ages (Figure 1f; Thevenon et al., 2011). These peaks coincide with periods of active silver mining and minting that took place in the Wallis (Valais) region (Guénette-Beck et al., 2009), which is only 20 km to the west of Meidsee and 40 km northwest of Colle Gnifetti. The Pb isotopic compositions of the Wallis ores and artefacts plot on the mixing line for the Meidsee sediment record. This history not only fits well with the Meidsee sediment record but might also be relevant for the ice record. Separate from this, Pb isotope data from previous work from Colle Gnifetti also point toward regional Pb sources (Gabrielli, 2008).

4. Exploring Details in the Ice Record

The high-resolution analyses by More et al. (2017) have the potential to add some interesting details to an already extensive body of work on historical Pb pollution in Europe, but most of the details in this high-resolution record remain unexplored. This is not only in regard to examining the record in light of the many available Pb records (Figures 1b–1f) but also in examining the details of the Pb record with regard to the chemistry of the ice itself. One of the important advances in the past two decades in the study of ancient pollution has been the increased scope of the analyses that extend beyond Pb concentrations and enrichment factors. An analysis of Pb within a larger geochemical (and often also pollen) context can provide a more complete understanding of the changes in Pb and the important drivers behind them (e.g., Currás et al., 2012; Karlsson et al., 2015; Küttner et al., 2014).

For the ice data, one noteworthy detail in the supporting information is that the exponential declines in Pb concentrations during the Black Death and again during the mid-1400s occur over the same exact time frame as an approximately tenfold increase in the Ca/Fe ratio (Pearson's correlation for Ca/Fe versus ln[Pb] is -0.66, $p = 0.01$; Figures 2a and 2b). Being insufficiently familiar with the complexity of ice chemistry, I can only speculate on the significance of this change in Ca/Fe, but it suggests that there may be processes beyond only pollution that control the Pb record in the ice. Based on the individual concentration records of Ca and Fe (Ca being enriched, for example, in Saharan dust events), the authors preclude changes in atmospheric circulation patterns; however, the pronounced change in the Ca/Fe ratio suggests a change in precipitation source regions for the glacier, whereby the lowest Pb values in the ice may reflect periods with air masses originating from relatively cleaner source regions. Observationally, these two periods of elevated Ca/Fe coincide with increased flood frequency in the French Alps (Wilhelm et al., 2013), and with minima in solar insolation (Steinhilber et al., 2009)—periods of climate change linked to plague outbreaks (Schmid et al., 2015). (Addendum: more recently published data from this Colle Gnifetti ice record by some of the coauthors, Bohleber et al., 2018, provide a stronger indication that climate exerted a strong influence on the Pb

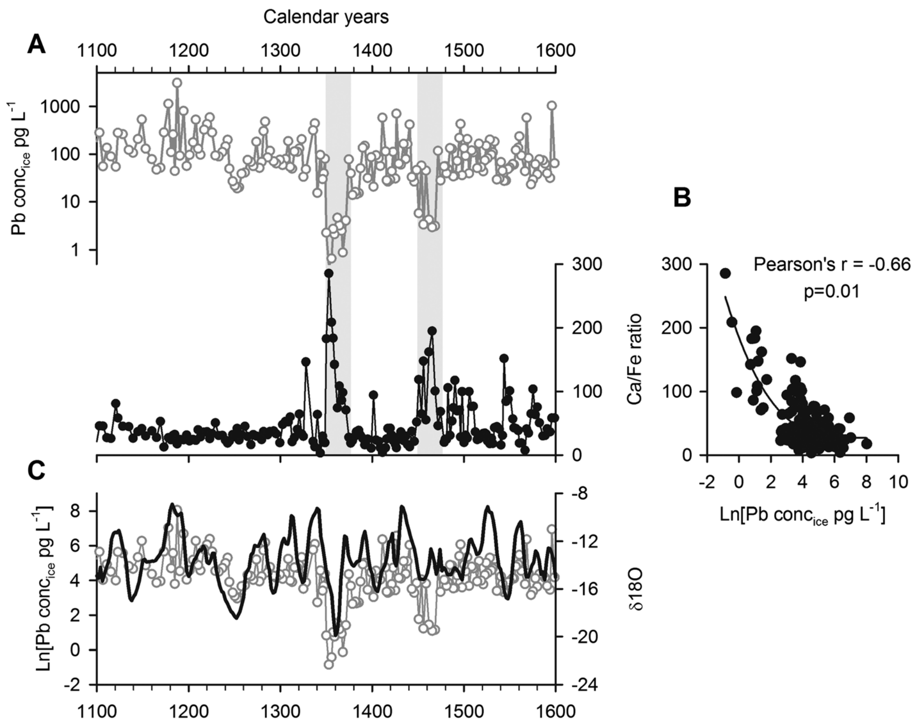

Figure 2. Pb concentrations and Ca/Fe ratios in the Colle Gnifetti ice record: (a) plotted for the period 1100–1600 CE (time interval provided in supplementary data) and (b) scatterplot of ln[Pb] concentration versus Ca/Fe. (c**)** New data from the ice record for δ^{18}O (Bohleber et al., 2018) and Pb, which adds support for a climate imprint on the Pb record.

record, indicated by the strong covariation between δ^{18}O and Pb; Figure 2c.) Careful evaluation of the ice Pb record within a larger geochemical data set derived from the high-resolution continuous analyses not only may yield more valuable insights on pollution transport and deposition at high elevations in the Alps but also may contribute further to identifying the origin of air masses during historical periods.

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