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REPLY

Key Points:
- Historical and archaeological evidence and context are crucially important in understanding past health, climate, and pollution data
- Geographic context and proximity to sources of lead (Pb) mining and smelting have a significant impact on the ice core record

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The Role of Historical Context in Understanding Past Climate, Pollution and Health Data in Trans-disciplinary Studies: Reply to Comments on More et al., 2017

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Abstract Understanding the context from which evidence emerges is of paramount importance in reaching robust conclusions in scientific inquiries. This is as true of the present as it is of the past. In a trans-disciplinary study such as More et al. (2017, https://doi.org/10.1002/2017GH000064) and many others appearing in this and similar journals, a proper analysis of context demands the use of historical evidence. This includes demographic, epidemiological, and socio-economic data—common in many studies of the impact of anthropogenic pollution on human health—and, as in this specific case, also geoarchaeological evidence. These records anchor climate and pollution data in the geographic and human circumstances of history, without which we lose a fundamental understanding of the data itself. This article addresses Hinkley (2018, https://doi.org/10.1002/2018GH000105) by highlighting the importance of context, focusing on the historical and archaeological evidence, and then discussing atmospheric deposition and circulation in the specific region of our study. Since many of the assertions in Bindler (2018, https://doi.org/10.1002/2018GH000135) are congruent with our findings and directly contradict Hinkley (2018), this reply refers to Bindler (2018), whenever appropriate, and indicates where our evidence diverges.

Plain Language Summary This article highlights the crucial importance of historical and archaeoscientific data in trans-disciplinary studies of planetary health, in the form of a reply to two comments to a previous article (More et al., 2017, https://doi.org/10.1002/2017GH000064). This reply emphasizes the crucial importance of geographic and historical context in assessing the significance and reach of scientific findings. The article also showcases the growing role of "Big-Data" scale data sets of demographic, epidemiological, and historical records for understanding how past pollution and climate data affected populations, especially when cutting-edge methods (e.g., laser ablation inductively coupled plasma mass spectrometry) in climate science provide an ever more detailed chronology of past crises and long-term pollution trends.

1. Historical and Archaeological Context

Both comments published above (Bindler, 2018; Hinkley, 2018) dismiss entirely half of the data presented in More et al. (2017), that is, the historical and archaeological evidence. Hinkley (2018) does so, curiously, by citing incorrect historical data. As submitted to us, Hinkley (2018), for instance, states that Roman legions spent their time collecting slaves to work their mines and as these efforts failed, so would mining operations, leading to his egregiously uninformed evaluation of Roman mining. In fact, historical evidence has shown that the mines were typically worked by private entrepreneurs or skilled workers on subcontract from the imperial government (Cuvigny, 1996; Hirt, 2010). Hinkley (2018) also makes a reference to the Rammelsberg mine, citing Georgius Agricola, an author who lived in the 1500s, more than five centuries after the event he claims to document (erroneously). The Rammelsberg mine (Harz mountains, Germany), in fact, began to be worked only in the 960s and reached large-scale production in the 990s (Blanchard, 2005; Spufford, 1988). The lead deposits in the peat record from the Harz show significant lead mining beginning only in the mid-late tenth century, reaching a peak in the twelfth and thirteenth centuries CE (Kempter & Frenzel, 2000). It is puzzling that Hinkley, (2018)—and indeed even Bindler (2018)—dismisses our

archaeological and documentary evidence to establish the provenance of lead; Hinkley (2018) resorts to limited and largely inaccurate historical citations to prove that the data do not reflect trends that the author expected to find. This kind of circular argument seems inconsistent with the best standards of scientific or historical inquiry.

Following this logic, Hinkley (2018) expects drops in atmospheric lead pollution during the Roman Empire and asserts that Figure 1 (in More et al., 2017) does not show the trends he anticipated, based on his erroneous and surprisingly limited historical citations. On the contrary, Figure 1 in our article shows, clearly, increases in atmospheric Pb deposits precisely at the economic peak of the Roman Empire, circa 50–150 CE, and other frequent increases and declines in subsequent centuries that are generally consistent with the latest historical and archaeological scholarship about the economic dynamism of the later Roman Empire (Harper, 2017; Kylander et al., 2005; Martínez Cortízas et al., 2013; McCormick et al., 2012; Sapart et al., 2012).

Hinkley (2018) seems not to reflect on the recent archaeological work that has identified the opening of new silver and lead mines in Merovingian Gaul (modern southwest France in the 600 CE), which tallies very well with our glaciochemical record, as will be again be manifest in the forthcoming work by our co-author, Christopher Loveluck (e.g., Mercier-Bion & Téreygeol, 2016; Téreygeol, 2016, 2007, 2010, 2013; More et al., 2017). From a historical perspective, the main difference between the new measurements represented in Figure 1 (in More et al., 2017) and the important early contributions from ice core and sediment studies that first identified historic changes in early civilizations' metal production from atmospheric lead depositions is the dramatically larger number and higher chronological resolution of the new measurements we provide (cf. Hong et al., 1994; Shotyk et al., 1998, cited in our article).

Bindler (2018) dismisses the insights inherent to the ultra-high-resolution, continuous record, presented in More et al. (2017), which offered for the first time an annual and even intra-annual assessment of changes in atmospheric pollution. On the contrary, Bindler (2018) insists on comparing our work to sediment studies whose resolution is at best decadal, while at the same time emphasizing the "importance of local or regional *histories*" of mining. None of the detailed historical and archaeological evidence presented in More et al. (2017)—which Bindler (2018) summarily and inexplicably dismisses—attests any lead mining activity, regional, local, or otherwise, in the vicinity of Colle Gnifetti (with 95% radiocarbon confidence), or in any other region influencing deposition at the site. The only remaining possibility is the mines of Great Britain, active on the eve of the Black Death pandemic. As any layperson may surmise, the decadal resolution of sediment records cannot shed light on yearly changes in mining activity. The five-year period of the Black Death (1349–1353), highlighted with intraannual, ultra-high resolution in More et al. (2017), is shorter than a decade, and thus cannot be captured by decadally resolved records such as the ones suggested by Bindler (2018). This warrants our statement, in More et al. (2017)—which Bindler (2018) objects to and reads out of context—that "new data show that human activity has polluted European air for the last c. 2000 years." Within the context of the last two millennia, even Bindler's (2018) own assertions indicate that our statement is correct to the best of current knowledge.

Archaeological and historical context has been, furthermore, the basis for critiques of isotopic provenance studies for the past 20 years. Hinkley (2018) insists that isotopic analyses of lead deposits are the only established method for determining provenance, when in fact the methodology has been, and continues to be under intense scrutiny. First, in multiple cases, even contiguous ore fields have been found to be isotopically nonhomogenous, indicating that ratios or signatures alone are not conclusive in establishing the provenance of a lead sample from the same ore field or geographic location (Pollard, 2008, 2009, 2017; Pernicka, 2014). Furthermore, there can also be considerable overlap between isotopic signatures of ore fields across western Europe, rendering them indistinguishable for sourcing purposes (Baron et al. 2009). This has prompted extensive, ongoing scientific debates, calling for increased attention to the archaeological context from which lead samples are retrieved. Modern samples cannot be compared with the isotopic values of archaeological lead—though they often are—because modern samples are often retrieved in modern contexts and at depths never reached by historical mining efforts (Baron et al., 2009; Budd et al., 1995a, 1995b, 1995c; Budd, Haggerty, et al., 1995; Budd et al., 1993, 1996; Ixer, 1999; Scaife et al., 1996). Once again, context is paramount.

Additionally, in the preindustrial period, ore, bars, ingots, and recycled lead from multiple sources—with different isotopic signatures—were often imported, mixed, and used during the smelting process in order

to increase fluidity, or for the process of cupellation, for example (Baron et al., 2009; Durali-Mueller et al., 2007; Pollard, 2008, 2009). The time span covered by More et al. (2017) falls within what historians have called the "Commercial Revolution" (circa 1200–1450), a period of rapidly intensifying trade networks across Eurasia and increased monetization, which required enormous amounts of silver, smelted from silver-lead ores (Blanchard, 2005; Laiou, 1997; Lopez, 1987; More, 2014; Spufford, 1988).

The possibility that, in this period, lead with various isotopic signatures could be imported, smelted, and mixed in the same furnaces increased dramatically, as new mines in central and eastern Europe began, in the thirteenth century, to provide both ore and bullion to flourishing commercial and minting centers in the west, which came to dominate Mediterranean trade (Blanchard, 2005; Lane, 1973; More, 2014; Spufford, 1988; Stahl, 2000). And finally, geoarcheologists have not yet excluded the possibility that fractionation was occurring during preindustrial smelting processes, which further complicates the use of isotopic analysis in identifying provenance (Budd et al., 1995a, 1995b, 1995c; Budd, Haggerty, et al., 1995; 1993, 1996; Pollard, 2008, 2009, 2017). On the contrary, several studies have shown that such smelting and resmelting processes can and did alter the isotopic signature of lead produced at the time (Baron et al., 2009, 2014; Durali-Mueller et al., 2007).

Hinkley (2018) also neglects to consider that isotopic analysis for Colle Gnifetti cores has been attempted only rarely (e.g., PhD thesis of Jacopo Gabrieli, 2008) and requires an entirely different sampling volume than that provided by our methodology. It is thus well beyond the scope of our study. As the full data sets provided with our article clearly show, the lead concentrations yielded by our methods are not sufficient for subannual isotopic analysis, which was thus never part of our trans-disciplinary article. Given the volume of ice necessary for isotopic provenance analysis, we would achieve at best decadal resolution and essentially nullify the major advantage of the laser ablation inductively coupled plasma mass spectrometry (LA-ICP-MS) method, that is, ultra-high-resolution chronological sampling. Additionally, the next-generation technology we showcased, applied for the first time on a European ice core, preserves the ice for future research (More et al., 2017; Spaulding et al., 2017; Sneed et al., 2015), while isotopic provenance analysis would require destruction of the core by melting of the ice. In light of the retreat of glaciers observed in the last two decades worldwide, the recovery and preservation of ice cores for future study are imperative.

Following established examples of multiple, analogous glaciochemical and sedimentary studies, we provided extensive archaeological and historical evidence to identify the most probable source regions of lead in our record. Decades of literature in toxicology (with which we begin the discussion in More et al., 2017) have called for ever lower lead pollution levels, including most recently the Lancet Commission on Pollution and Health, especially with regard to developing countries with less stringent lead pollution regulations (Landrigan et al., 2017). Lanphear et al. (2018) has shown that even extremely low levels of lead pollution have an enormous impact on human health and life expectancy—not only in children, as already established in the literature, but also in adults (Landrigan, 2018)—showing a remarkable correlation with increased rates of cardiovascular disease, increasing estimated lead-related deaths by an order of magnitude. In light of Lanphear's latest, alarming findings, the cumulative historical effect on human health and development of long-term lead pollution documented in More et al. (2017) remains to be assessed, but seems, at first glance, at least ominous. Our article is a contribution to the study of these pollution trends, with the highest-resolution record available, as well as a contribution to the environmental, economic, and health history of Eurasia and the Mediterranean.

Hinkley (2018) claims in multiple statements that the trends observed in our study—a sharp decline in lead levels caused by economic and demographic collapse—have not been observed in other literature. The primary support provided for this argument is the author's claim that the lead levels observed in Matsumoto & Hinkley (2001)—notably for Antarctica—apply to the European context (more than 8,000 miles away, in another hemisphere), imply natural origin and have not been challenged by more localized studies with higher geographic proximity. We reject all these claims as incorrect. One need only read Bindler (2018)—in complete agreement with us on this point—to conclude that Hinkley's (2018) claims are flawed.

Many recent studies have linked human activity with rising lead air pollution levels in the preindustrial period. In our article we specifically indicate that: "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 ~1250 to 900 B.C.E. to the present, with periods of only moderate decline." This and other paragraphs in our article—in agreement with Bindler (2018)—cite ice-core, lake sediment, and peat-core studies, as Hinkley (2018) requests, where lead fluctuations are linked to human activity (Bindler, 2011; Bindler et al., 1999; Brännvall et al., 1999; Forel et al., 2010; García-Alix et al., 2013; Klaminder et al., 2006, 2003; Le Roux et al., 2004; McConnell et al., 2018; Renberg et al., 2009; Shotyk et al., 1998). Again, for a comprehensive review of this data, one only need refer to Bindler (2018) above.

Bindler's (2018) claim that we did not cite this data or that we argue that there was no evidence of Iron-Age or Roman mining—and that we should have emphasized the work of Renberg et al. (1994), among other collaborators of Bindler's—is incorrect. We cited Renberg, Bindler, and Brannvall (2001) (a more recent publication) and Nriagu (1983), among several others, as landmark studies. The additional, usually older, literature discussed by Bindler (2018) was omitted for reasons of space, which limited us to the most comprehensive and representative publications. When More et al. (2017) mentioned "previous assumptions about preindustrial 'natural' levels"—to which Bindler objects—this referred to policies and reports by international agencies, such as United Nations Environment Programme using Richardson et al. (2001) (all cited), where, in fact, such erroneous assumptions are to be found. Despite the decades of literature proving the contrary, national and international agencies continue to operate under those assumptions. One of the aims of More et al. (2017) was in fact to correct this, with the highest-resolution record in existence, and in the spirit of the literature that Bindler, and we, cited.

Declines in lead deposition during epidemics and widespread environmental crises are reflected in these publications, which have the advantage of much higher geographic proximity to major sites of European lead mining and smelting, especially when compared to evidence from Antarctica. That same geographic proximity affords the Colle Gnifetti record much higher sensitivity. Would Hinkley (2018) accept European ice cores as a climate or pollution proxy for South America of reliability equal to Antarctic records, for example? It is unclear why the reverse (Antarctic ice cores as a proxy of reliability equal to European cores for Europe) should be acceptable. Once again, context is crucial.

Unlike Bindler (2018), Hinkley (2018) explicitly concedes that, "Peat bogs preserve a less finely resolved time stratigraphic record than ice cores but their records are robust over appropriate time spans." The key words here are "appropriate time spans." The application of LA-ICP-MS to the Colle Gnifetti ice core—the major innovation described in our article—produces ultra-high-resolution records: 288 measurements for Pb within the year 1349 CE, for example, versus nine data points for the 73,000-year record presented in Matsumoto and Hinkley (2001), the most recent of which dates from circa 700 CE or *six centuries before* the salient period discussed in our study. Thus, our method allows for detection of Pb fluctuations or trends on much smaller time spans (years, seasons, or even single deposition events) than any previous measurements of sedimentary cores. Yearly fluctuations could go undetected in lower-resolution stratigraphic records, such as peat bogs, whose chronological resolution is much lower (i.e., decades or centuries).

Methodological innovations aside, the fact remains that comparing our European ice core record with data from the southern hemisphere (Antarctica) in the premodern period ignores historical reality, that is, that the overwhelming majority of lead mining and smelting in the period of the Black Death (1349–53 CE) occurred in the northern hemisphere. The proximity of the Colle Gnifetti ice core to the centers of lead mining and smelting is not comparable to the Antarctic context, which was much farther (by several orders of magnitude, in another hemisphere) from European lead mining and smelting centers.

2. Atmospheric Circulation and Partitioning of Sources: The Importance of Geographic Proximity and Context

Hinkley (2018) emphasizes the importance of enrichment factor evaluations in assessing the source of the lead emissions reported in More et al. (2017). We agree that enrichment factors are a valuable tool, and in fact, we reported them in Figures 3 and S4 of our article; how they are derived is discussed in the supporting information. Hinkley (2018) argues that we would need to provide enrichment factors relative to other metals associated with volcanism, and not sulfur. This line of argument implies that volcanic source contribution

can always be distinguished from other sources without accounting for complex regional natural emission sources, transport, or deposition regimes.

Such assumptions may perhaps have value where conventional low-resolution sampling techniques are concerned—such as those used to evaluate decadal-scale atmospheric concentration trends, as has been practiced in previous decades—but we observed large inconsistencies in using this approach for a high-resolution time series that likely captures single atmospheric storm-type events. We provided details of how enrichment factors were calculated (see the supporting information). We used lead, titanium, and sulfur concentrations in the upper continental crust and estimates of global sulfur emissions from volcanoes to calculate regional, natural Pb background levels. We used titanium in our enrichment factor analysis as a dust marker, as previously done by Wagenbach et al. (1996) for alpine cores.

Based on lead deposition records from Antarctica (published in the author's earlier work), Hinkley (2018) argues that volcanic or otherwise non-anthropogenic lead emissions dominate glaciochemical records throughout the globe and throughout history (the adjective "worldwide" is repeated multiple times in his 2001 article and 2018 comment). Here again, context is very important. The residence time of Pb in the atmosphere (10–14 days at most; Papastefanou, 2006; Poet et al., 1972; Settle & Patterson, 1991) and the distance between Antarctica and major mining areas in Europe (no less than 8,000 miles) make it exceedingly unlikely that anthropogenic Pb would reach Antarctic ice in amounts comparable to what we see at Colle Gnifetti, located a few hundred miles from major premodern European mining and smelting operations. This renders Antarctic ice cores a much less accurate proxy of European lead mining in the period in question. On the contrary, regionally produced volcanic Pb from the southern hemisphere could reach Antarctica much more readily. This partially explains the predominantly volcanic lead signal measured in the referenced Antarctic ice core, in Hinkley (2018), which—it must be mentioned—was recovered in close proximity to active volcanoes, that is, Mt. Erebus, Mt. Melbourne, and the Pleiades.

Furthermore, the pattern of economic development and the scale and history of metal production generating lead pollution are completely different in the southern hemisphere, fed by the much lower scale and age of metallurgical activity of southern Africa, Australia, and Pre-Columbian south America. The scale of these activities stands in stark contrast to the northern hemisphere, with the Roman and Islamic Empires, China, and India, all of which contributed to deposition (Maddison, 2003; Nef, 1987; Petersen, 2010; Pomeranz, 2000; Schwikowski et al., 2004). One of the central points of our article is that the geographic proximity of the Colle Gnifetti ice core to major European mining areas affords a regional specificity that no other ice cores have allowed. This has been highlighted by previous literature (e.g., Schwikowski et al., 2004), motivating the recovery of ice cores from this site. For a review of other, lower resolution, regional records showing this same pattern, once again, one may simply refer to Bindler (2018) above.

Geographic proximity, coupled with new, ultra-high-resolution LA-ICP-MS analysis enable us to detect the anthropogenic contribution to lead pollution with a level of chronological precision (annual or sub-annual) never achieved before. The extensive archaeological and historical evidence provided, in conjunction with a study of atmospheric circulation and analysis of other glaciochemical indicators (Ca, Fe, etc.), point to British mines as the most likely source of Pb emissions. To say that this is not so—as Hinkley (2018) and Bindler (2018) surmise—is to make the unlikely argument that by coincidence, a constant volcanic source of Pb continued for hundreds of years but happened to stop depositing Pb on Colle Gnifetti at the precise moment and for the precise duration of the Black Death pandemic's worst demographic collapse (1348–53 CE), and again, by coincidence, during the pandemic of 1460–65 CE, and again during the economic downturn of 1882–85 CE. The historical evidence is a central part of this study and cannot be ignored simply to fit paradigms and methodologies that ignore historical reality and scientific advancement in interdisciplinary research.

3. Dust Transport to Colle Gnifetti

Perplexing assertions in Hinkley (2018) are not limited to the misuse or dismissal of historical and archaeological data but extend to the context of European glaciers, as exemplified by this statement: "Most of the Colle Gnefetti [sic] samples are not dusty, and not especially variable in their dust content." This is incorrect, misleading, and even contradicts another assertion in Hinkley (2018), where it is stated that: "continental Eurasian and North American glaciers, although closer to Europe, may be too dusty to provide sensitive
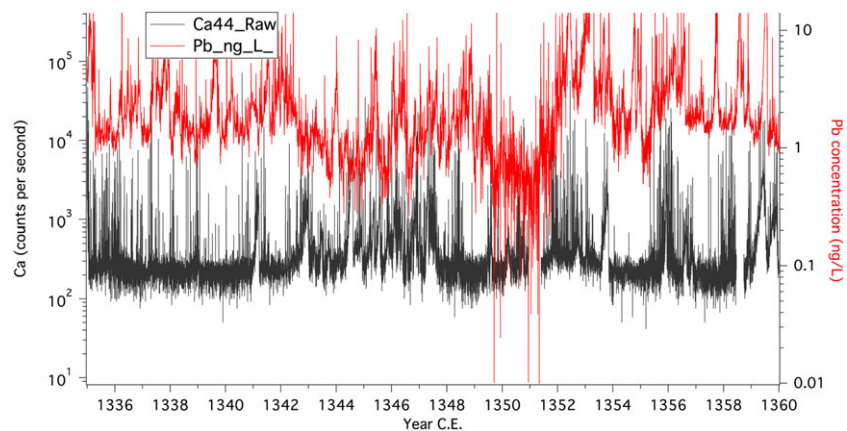
Figure 1. Ultra-high-resolution calcium and lead records from the Colle Gnifetti ice core obtained via laser ablation inductively coupled plasma mass spectrometry.

baseline information about amounts of trace metals." Thus, in Hinkley (2018), our records seem either dusty or not depending on the author's need to dismiss our data. Colle Gnifetti cores are known to be subject to seasonal Saharan dust transport (Prodi & Fea, 1979; Schwikowski et al., 2004; Wagenbach et al., 1996; Wagenbach, Bohleber, & Preunkert, 2012; Wagenbach & Geis, 1989), as Bindler (2018) also confirms. A brief review of the relevant literature regarding this site would easily show this. Dust profiles for nearby (shallower) cores have been published and are cited in our article. Saharan dust profiles have been used for years as temporal markers for ice core chronologies.

One of the data sets published with our article is, in fact, an ultra-high-resolution LA-ICP-MS record of calcium, which shows Saharan dust variability over a period of two millennia. The calcium variability observed at the time of the Black Death pandemic does not show an interruption or decline in dust transport lasting from 1349 to 1353 CE (Figure 1), thereby indicating that Pb deposition from dust did not vary to such an extent that it could explain the trend observed. Bindler (2018) noted this pattern. Anthropogenic sources, on the other hand, clearly ceased producing lead pollution in this period, as shown in the extensive archaeological and documentary evidence provided in More et al. (2017). Articles currently in preparation by our group show the same trend for other mined metals.

4. Calcium, Iron, Sodium, and Chlorine Variabilities

Hinkley (2018) goes on to state that we do not discuss Ca and Fe variabilities in our article; this is also incorrect. On page 216 of More et al. (2017) we stated: "Our record of the multiyear Black Death period is not associated with any anomalous atmospheric circulation patterns, based on Ca and Fe as crustal air mass proxies (Figure S3)." This statement clearly conveys that dominant dust transport and prevailing atmospheric circulation did not change, even as the Pb deposition did change. Admitting his unfamiliarity with the site, Bindler (2018) questions this deposition pattern, which is clearly addressed in extant literature on Colle Gnifetti (Prodi & Fea, 1979; Schwikowski et al., 2004; Wagenbach et al., 2012, 1996; Wagenbach & Geis, 1989). The fact that ICP-MS Ca and Fe decline during the Black Death simply shows that the deposition pattern at the time was not affected by a Saharan dust event (south to north) but reflected a NW to SE pattern consistent with the Iceland Low Pressure System, as described in Figure 4 in More et al. (2017). This is also supported by an increase in Na and Cl deposition, as shown below in Figure 2. Hinkley (2018) even comments on this sea salt deposition data at Colle Gnifetti, published for the first time in the present reply, and not in the original article (More et al., 2017). Sea salt will be the subject of publications we are preparing, which confirm our findings, as in Figure 2 below, where we actually detected an increase in Na and Cl deposition in the years in which we observe the beginning of the most significant decline in lead pollution, due to the Black Death pandemic (1349–53 CE). If prevailing wind patterns from the Atlantic Ocean (as controlled by the Icelandic Low Pressure System) had been disrupted for five years—a possibility raised by Bindler (2018), who seems to ignore the atmospheric circulation evidence provided, via climate ReAnalyzer™—causing a decline in lead deposition, we agree with Hinkley (2018) that we would see a decline in sodium and chlorine deposition as well. As Figure 2 shows, this did not occur during the years of the Black Death pandemic.
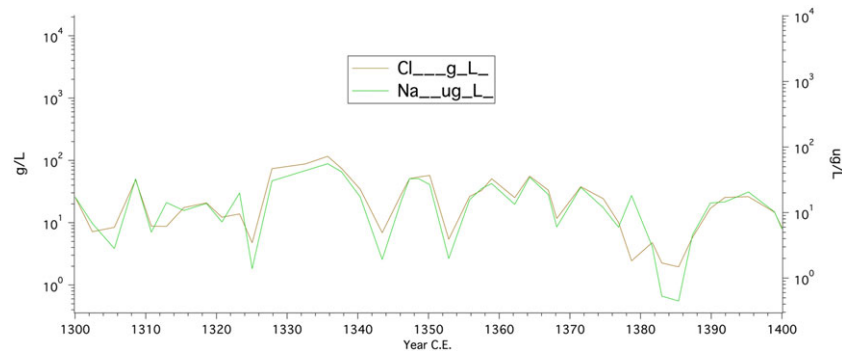
Figure 2. Chlorine and sodium time series (1300–1400 CE) developed using discrete meltwater samples from the Colle Gnifetti ice core, as measured by ion chromatography and inductively coupled plasma mass spectrometry, respectively.

5. Conclusion

We heartily agree with Hinkley (2018) and Bindler (2018) that more data are what are needed, and we would very much welcome it, if other geoscience researchers would integrate extensive archaeological and historical research within their projects to the same extent as More et al. (2017). Trans-disciplinary endeavors are answering this call with multiple, independent but consilient data sets that begin to describe changes in the environment and human and ecosystem health with ever increasing detail. Indispensable and deeply revealing, in this effort, are new records that describe localized conditions—geographic, human, environmental, climatic, and epidemiological—in the past and present. These new records supply the context, historical, or current, which is indispensable to understand any set of evidence, in an effort to "educate a global public about implications of their decisions on Planetary Health/GeoHealth," as Amalia Almada et al. eloquently wrote in these pages only a few months ago (Almada et al., 2017).

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