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A synthesis of lead isotopes in two millennia of European air

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Abstract

Four airborne particulate records from ombrotrophic peat bogs in southern Norway, extending back 300 years, have been measured for chronology, lead concentration, and lead isotope composition. Since southern Norway receives an airborne lead signal that accumulates emissions from the European continent, the trend in the four bog records can be used to correlate previously reported measurements from France, Switzerland, England, and Greenland that cover different ranges of time. When these are compiled, the integrated European record that emerges spans the last 2300 years of human influence on lead in the air over Europe and suggests human control of lead in airborne particulates over the entire period. From 366 BC through the first half of the 20th century, lead isotopic compositions in European air have fallen within the range of compositions in European ore bodies. Since 1950, isotopic compositions have been beyond the range in those ore body compositions and have fallen within the array of lead isotope compositions typical of gasoline from western industrial nations (a mixing line between US and Australian lead in gasoline). The overlap between the European record and the range in modern European air suggests an average isotopic composition of $^{206}Pb/^{207}Pb$ ca. 1.13 and of $^{208P}b/^{207}Pb$ ca. 2.41 in air over Europe during the last 20 years.

Keywords: Europe; lead; isotopes; bogs; atmosphere; transport; Norway

1. Introduction

The surface of the earth has been contaminated with the atmospheric emission of lead by human activities [1,2]. The long-term, global record of this contamination is documented by the elevation of lead concentrations in Greenland and Antarctic ice during the last 10,000 years [3–5], and the link to anthropogenic sources has been made by analyses of the isotopic composition of lead in an ice core from Greenland [5] and in Greenland and Antarctic snows [6,7].

In Europe, isotopic records of lead emissions are found in several countries but have not been synthesized into a coherent, long-term record for the continent. The existing records are either limited in temporal extent or have not been well correlated to each other. The most complete history has been measured for the lead concentration and $^{206}Pb/^{207}Pb$ isotopic composition of airborne particulates deposited in a bog core from Switzerland during the last 12,000 years [8]. The $^{206}Pb/^{207}Pb$ isotope ratios in this record are consistent with values in other records reported for the Roman period and during the last century, but the correlation has not been reported for $^{208}Pb/^{207}Pb$ ratios, which are useful for distin-

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guishing among possible lead sources [8]. Values for $^{208}Pb/^{207}Pb$ and $^{206}Pb/^{207}Pb$ have been reported for the upper 2200 years in the Swiss core [9], as well as for two other records from the last 200 years in sediment from France [10] and England [11], but these have not been compared to each other and have been considered to be either local or western European records. Finally, a chronology of lead concentration and isotopic composition from a Greenland ice core that documents lead emissions from the whole European continent has been compiled, but the record extends only to 1523 AD [5].

The research presented here assembles the first self-consistent record for Europe. By combining previous isotopic records with new results from an ombrotrophic bog core in Norway we compile a view of the last 2300 years. Since Norway receives an averaged airborne particulate signal from all of Europe, the Norwegian record can be used to extend previous chronologies to interpret a record for the whole continent.

2. Sampling and analytical techniques

Samples were taken from ombrotrophic peat bogs in order to provide a record of atmospheric deposition. Since ombrotrophic bogs receive all of their water and nutrients from the atmosphere and have negligible interaction with their substrate [12], they effectively serve as long-term traps of airborne particles. Bogs that showed down-core variations in lead concentration indicative of post-depositional lead mobilization were avoided in this study.

The location of the bogs in southern Norway (Fig. 1) allows them to preserve an averaged signal of European airborne lead emissions. Over the last two decades, surveys of lead concentrations in the soils and moss of southern Norway have concluded that the metals concentrations along the southern coast are attributed to emissions from the rest of the European continent [13–17]. An air trajectory analysis of air samples taken in 1978–1979 found that 80% of lead reaching southern Norway originated on the European continent [18]. Specifically, 50% of the airborne lead emissions were attributed to air masses from western Europe, 30% to eastern Europe (including European Russia), 7% to the North Atlantic and Arctic Oceans, and 13% to undetermined incoming air trajectories [18]. A follow-up study on samples collected in 1985–1986 found that although the overall concentration of lead and other metals in air was reduced, primarily due to emissions controls in western Europe, the origin of lead from western and eastern Europe was similar to the results for 1978–1979 [19]. Lead deposition from the atmosphere to bogs in this region, therefore, probably represents a European average.

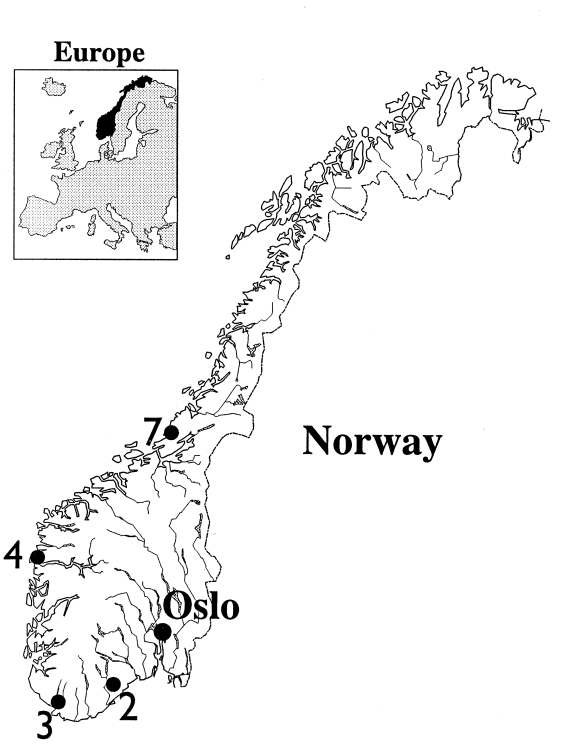

Fig. 1. A map of Norway showing locations of bog sample sites. Bog *2*, 58°57′N, 8°54′E. Bog *3*, 58°25′N, 6°48′E. Bog *4*, 61°19′N, 5°3′E. Bog *7*, 64°7′N, 10°30′E.

In order to reconstruct a long-term record of that deposition, cores were extracted, using trace-metal clean techniques [20], from four ombrotrophic peat bogs in southern Norway (Fig. 1). The sites were located in remote areas to preclude contamination from local industrial sources. The cores were age dated using standard ^{210}Pb techniques [21] and are >200 to >300 years old at their bottoms, based on the assumption that the accumulation rates stayed

constant below the depth where one could measure ^{210}Pb.

Aliquots, taken at 50 to 100 year intervals, were analyzed for lead concentrations (μg/g dry weight) by ICP–MS. Stable lead isotopic compositions (^{204}Pb, ^{206}Pb, ^{207}Pb, ^{208}Pb) of subsamples of those aliquots were then measured with a VG Sector 54/30 thermal ionization mass spectrometer (TIMS) that has been used in our previous work [22–24]. A fractionation correction of 0.0014 per atomic mass unit was derived from concurrent analyses of NIST SRM 981. During the period over which the ombrotrophic bog samples were analyzed, the reproducibility of the SRM 981 analyses was ±0.5% relative standard deviation. Errors on the lead isotope analyses are reported in the common fashion in Table 1, as ±2 standard error on the mean (the internal precision on the TIMS analysis).

3. Lead concentrations

Lead concentrations in the remote areas of southern Norway are elevated due to the long-range transport of emissions from the rest of Europe [15,17,19]. Over the past 20 years, analyses of airborne particulate lead deposited on a prevalent moss, *Hylocomium splendens*, have shown ten-fold greater lead concentrations along the southern coast of Norway compared to concentrations along the northern coast [16,17], and the surface soils of southern Norway contain lead concentrations that are 100 to 2000 times natural background levels [14,15]. The high lead concentrations in both moss and soil are attributed by principal component analysis to emissions from Europe [17–19].

The geographic variation and magnitude of lead concentrations in the top layers of the ombrotrophic bog cores (Table 1) is consistent with existing moss and soil data: 210–250 μg/g in southernmost Norway (bogs 2 and 3) relative to concentrations of 30–40 μg/g in bogs farther north (bogs 4 and 7). Like the soils and moss, the ombrotrophic peat bogs record the distribution of lead concentration attributed to airborne particulate lead emissions originating in Europe. Therefore, the ombrotrophic bogs can be considered to be particle traps averaging atmospheric emissions from Europe over time.

The temporal variation of lead concentrations in the ombrotrophic bogs suggests that they can record changes in lead emissions occurring over periods longer than 20 years, as illustrated by Fig. 2. It shows the increase in lead concentrations in the Norwegian peat bog cores over the last 300 years, which is consistent with the systematic increase in atmospheric lead emissions over Europe during the same time period [25]. However, short-term variations in lead concentrations in the bogs do not consistently

Table 1
Ages, lead concentrations, and lead isotopic compositions of Norwegian ombrotrophic peat bog cores

Sample No.	Pb (ppm)	Depth (cm)	Date (AD)	^{206}Pb/^{204}Pb	^{207}Pb/^{204}Pb	^{208}Pb/^{204}Pb	^{206}Pb/^{207}Pb	^{208}Pb/^{207}Pb
2-1	247	0–1	>1981	17.9697 ± 21	15.5856 ± 19	37.8932 ± 47	1.15402 ± 02	2.42772 ± 05
2-5	148	4–5	1940	18.1545 ± 17	15.5996 ± 14	38.1234 ± 37	1.16484 ± 02	2.44029 ± 04
2-15	57	14–16	~1800	18.2916 ± 23	15.6125 ± 20	38.2745 ± 50	1.17266 ± 02	2.44790 ± 04
2-19	3	30–35	<1700	18.3838 ± 22	15.6330 ± 20	38.4038 ± 57	1.17704 ± 02	2.45298 ± 10
3-1	208	0–1	>1981	17.8330 ± 14	15.6009 ± 11	37.8270 ± 33	1.14412 ± 02	2.42115 ± 07
3-5	149	4–5	1920	18.3262 ± 20	15.6459 ± 17	38.4119 ± 42	1.17239 ± 02	2.45151 ± 04
3-14	36	12–14	~1800	18.4036 ± 24	15.6573 ± 23	38.5031 ± 63	1.17647 ± 03	2.45545 ± 10
4-1	29	0–1	>1981	17.4624 ± 14	15.5554 ± 12	37.3261 ± 31	1.12363 ± 02	2.39603 ± 04
4-5	90	4–5	1960	17.8929 ± 16	15.5757 ± 14	37.8291 ± 36	1.14982 ± 02	2.42520 ± 04
4-14	49	12–14	1900	18.3957 ± 11	15.6346 ± 11	38.4528 ± 25	1.17766 ± 02	2.45583 ± 04
4-19	6	30–35	~1800	18.4257 ± 20	15.6387 ± 19	38.4868 ± 57	1.17927 ± 03	2.45746 ± 11
7-1	40	0–1	>1981	17.8282 ± 32	15.5721 ± 29	37.7196 ± 83	1.14593 ± 03	2.41873 ± 13
7-6	36	5–6	1950	18.3059 ± 18	15.6632 ± 16	38.4648 ± 45	1.16977 ± 03	2.45211 ± 09
7-15	10	14–16	1870	18.3304 ± 16	15.6182 ± 14	38.3415 ± 41	1.17469 ± 02	2.45129 ± 07

Errors on isotope measurements are shown as ±2 s.e. in the final two decimal places.

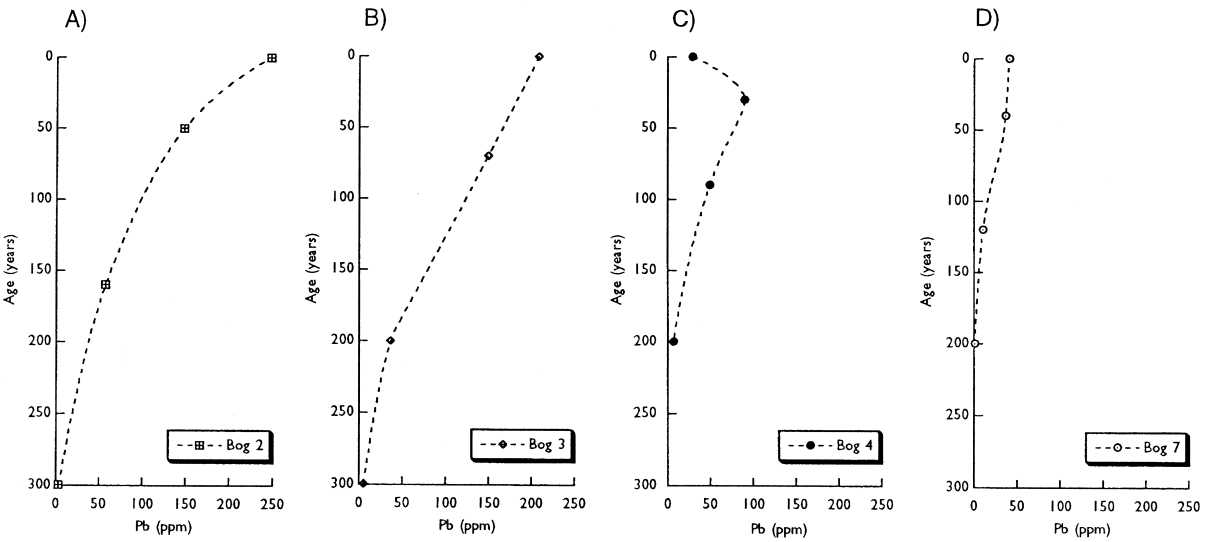

Fig. 2. An age vs. lead concentration plot of Norwegian ombrotrophic bog samples.

exhibit the recent decrease observed in *H. splendens* since the 1980s [16,17].

That decline, which corresponds to the reduction in the use of lead additives in gasoline in Europe [1], has also been observed in adjacent North Atlantic surface waters [26,27] and Greenland ice cores [3,25]. The absence of a decrease in lead concentrations in three of four of the ombrotrophic peat cores is attributed to the averaging of atmospheric inputs over one or two decades in peat bog deposits. Consequently, the bogs do not appear useful for resolving trends in atmospheric fluxes on a sub-decadal scale.

4. Isotopic compositions

The lead isotopic compositions of the Norwegian ombrotrophic bogs have changed through time. The oldest and least-contaminated deposits, from the late 15th to 19th centuries, have $^{207}Pb/^{204}Pb$ ratios >15.61, while local bedrock ratios are ≤ 15.59 [28]. The difference between the bog and bedrock ratios (0.13% relative) is well outside of the analytical error on the bog analyses (<0.02%, 2 s.e.). This suggests that the bogs are recording atmospheric deposition with no measurable influence from local bedrock.

As the bogs become younger their $^{206}Pb/^{207}Pb$ and $^{208}Pb/^{207}Pb$ ratios become lower (Fig. 3). There is overlap in isotopic composition between the two age groups older than 1950, but there is no overlap between the most recent deposits (1991 to 1960) and the older ones. Taken together, the Norwegian bog data define a linear trend ($^{206}Pb/^{207}Pb = -1.02 + 0.90 \times {}^{208}Pb/^{207}Pb$; $r^2 = 0.99$).

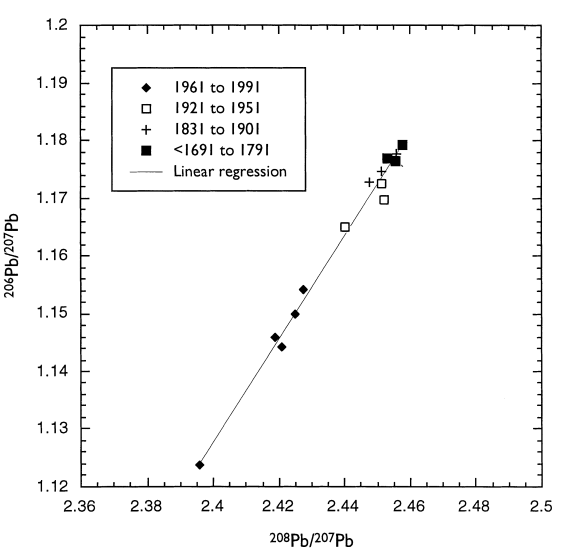

Fig. 3. A lead isotope plot of ombrotrophic peat bog core samples from southern Norway. The line represents a least-squares linear regression through all data points. The age dates on the core samples were determined by ^{210}Pb (see text).

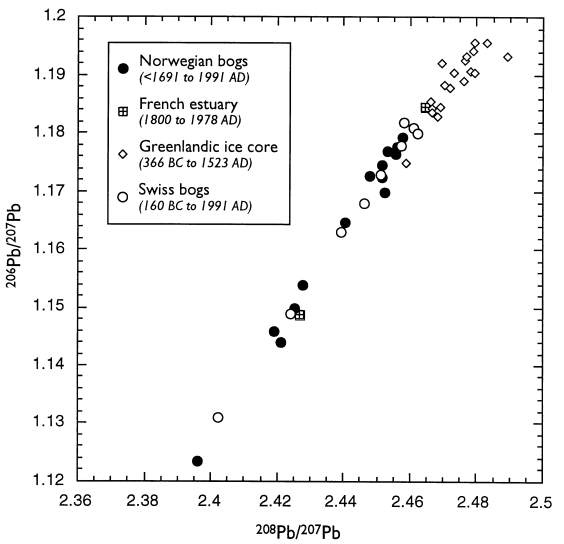

Fig. 4. A comparison of ombrotrophic peat bog lead isotopes with three other records from Europe and Greenland. The French estuary data are from Elbaz-Poulichet et al. [10], the Swiss data are from Shotyk et al. [9], and the Greenland data are from Rosman et al. [5]. A fourth record from England [11] overlaps all of these records but data are more scattered due to a correction applied for terrestrial lead inputs. The English data are therefore omitted from the plot for clarity.

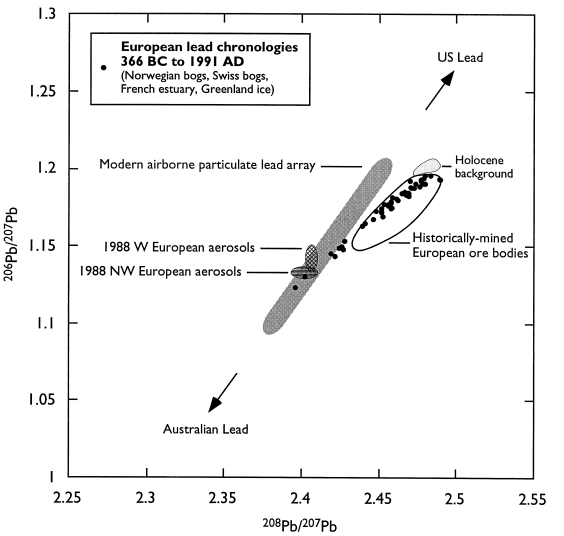

Fig. 5. A comparison of European lead chronologies with possible sources of airborne lead. The field for the Holocene background is defined from data in Greenland ice ranging in age from 7313 BC to 680 BC [5]. Data for the array of lead isotope compositions of gasoline from western industrial nations are taken from the isotopic composition of contemporary measurements in Antarctic [29] and Greenland snows [6,7] and samples of North Atlantic [27] and Antarctic surface waters [24]. The field for the array encloses the area of highest overlap of data — more than 90% of data points are contained within the field, and there is no coherent, differing pattern among the outliers. Data for the European lead trends are from sediments in France [10], an ombrotrophic peat bog in Switzerland [9], an ice core from Greenland [5], and the ombrotrophic peat bogs from southern Norway in this study. The fields for aerosol data are from sampling conducted in 1988 [30]. The field of European ore body compositions encompasses analyses from mines in Algiers, Austria, England, Italy, Germany, Greece, Hungary, Morocco, Poland, Spain, the USSR, and Yugoslavia [5,30]. US lead is the composition from the tri-state mines ($^{206}Pb/^{207}Pb = 1.33–1.39$) [34], the dominant source of lead mined in the US since about the late 1960s or early 1970s [35–37]. Australian lead is the composition from the Broken Hill mines ($^{206}Pb/^{207}Pb = 1.04$) [34].

The lead isotopic compositions of the Norwegian ombrotrophic bogs overlap three other chronologies of the composition of lead isotopes in air over Europe (Fig. 4). Records exist from sediments in France [10] and Britain [11], an ombrotrophic peat bog in Switzerland [9], and an ice core from Greenland at a depth in the core during which human-generated lead signals are attributed to Europe [5]. The British sediment data [11] cover a period from ca. 1750 to 1970 AD; the French sediment data [10] extend from 1800 to 1978 AD; the Swiss bog data [9] extend from 160 BC to 1991 AD (with significant gaps before 1843 AD); and the Greenland ice core data [5] extend from 366 BC to 1523 AD. The British sediment record overlaps with the others but is not plotted in Fig. 4 since corrections for terrigenous inputs to the isotopic compositions of the sediments create scatter [11].

From 366 BC to 1991 AD the lead isotopic compositions of the four previous data sets on the European continent and Greenland are co-linear ($^{206}Pb/^{207}Pb = -0.86 + 0.83 \times {}^{208}Pb/^{207}Pb$; $r^2 = 0.98$). All four records overlap with the chronology from southern Norway, which collects airborne lead from the rest of Europe, suggesting that all of the chronologies can be synthesized together to represent the long-term trend in lead isotope compositions in the air over Europe: an integrated European record. All points in the synthesized record fall below the Northern Hemispheric Holocene background lead isotopic composition (see Fig. 5 caption).

The integrated European record overlaps two primary sources of lead. From 366 BC until ca. 1950 AD the record falls within the range of lead isotope compositions of historically mined European ore bodies. All deposits younger than 1950 in the record contain lead isotope compositions that plot beyond the range found in ore bodies and extend into the modern range of globally distributed lead in airborne particulates. This modern airborne particulate array is derived from the isotopic composition of contemporary measurements in Antarctic [29] and Greenland snows [6,7] and samples of North Atlantic [27] and Antarctic surface waters [24] (see Fig. 5 caption). The modern lead array appears to be dominated by the binary mixture of industrial lead from two sources. These are the United States and Australia, which have been the primary suppliers of lead additives used in gasoline over the past 70 years [24]. The dominance of leaded gasoline emissions in the last half of the century is seen in the consistency of the modern lead array with the isotopic compositions of recent (1988) European aerosols [30] (Fig. 5).

5. Discussion

Although the overall correlation between the integrated European record of atmospheric deposition and the dominant anthropogenic and natural sources suggests control of atmospheric lead by humans over the entire time period from 366 BC to the present, the record is not complete in detail. The natural background lead composition is known from only one location, mining records are intermittent, and the sub-decadal trends in modern times are poorly resolved.

A reference value for the natural background lead isotope composition is not available at each sampling site in Europe. The field for the natural background composition in Fig. 5 encloses values that are almost invariant over a period of 7000 years in a Greenland ice core, but data from Greenland do not strictly represent local backgrounds for the records in Norway, Switzerland, France, and England. Although the $^{206}Pb/^{207}Pb$ ratios in the mid-Holocene sections of the 12,000 year Swiss core [8] are consistent with the $^{206}Pb/^{207}Pb$ in the Holocene background from Greenland [5], a firm correlation is difficult to make without reported $^{208}Pb/^{207}Pb$ values in the Swiss core.

Among the European records with published $^{208}Pb/^{207}Pb$ ratios, the oldest measured layer (from 160 BC in a Swiss ombrotrophic bog) was deposited over 400 years after human influence on the lead in air over Europe is documented. By 160 BC, tropospheric lead concentrations had been elevated by Greek and Roman mining to four to ten times above natural background concentrations [8,31,32]. The lowest concentration in Greenland ice that postdates the onset of Greek mining occurs ca. 400 AD when Roman mines were near exhaustion and the empire was disintegrating. During this period the lead concentration in the ice remained 30% higher than background. As far away as Greenland the lead concentration in the troposphere was above background from ca. 550 BC onward. The compiled European records are all at least 3000 km closer to the ore processing operations, so none should be expected to exhibit a natural background lead isotope composition during the time span for which complete isotopic data exist, indicating that lead in the air over Europe has not had a natural background lead isotope signature in over 2300 years. This conclusion is corroborated in the 12,000 year Swiss bog record which records a shift in $^{206}Pb/^{207}Pb$ beginning at 3000 BP [8].

The first 2250 years of the synthesized record of lead in European air appears to be dominated by emissions from ore processing. The influence of ore processing is suggested by the complete overlap in the $^{206}Pb/^{207}Pb$ and $^{208}Pb/^{207}Pb$ of the lead compositions in the European record with the European ore body compositions, but other sources of anthropogenic lead emissions to the troposphere, coal burning being the most significant, are not considered in Fig. 5. Although coal burning also releases lead into the air, it is not likely to be strongly expressed in the airborne lead isotopic composition. In a detailed study of global tropospheric lead emissions, the relative contribution to airborne lead from ore processing in 1988 was found to be 80% relative to all coal burning activities [33]. The primary reason for the relative dominance of ore processing is the typically 10 to 100 thousand fold higher concentration of lead in ore bodies compared to coal.

Since this contrast is independent of ore processing technology, the relative dominance of ore processing in the past can be inferred.

We have not attempted to address the change through time in atmospheric inputs from the various European mining districts. The early part of the record has been shown to be dominated by Spanish mines producing silver for the Roman Empire [5], but the documentation of mining outputs after the fall of the Roman Empire and the depositional chronologies of the records that we have synthesized here are not known in enough detail to allow much further discussion. Moreover, the limited data on analyses of the lead isotope compositions of ore bodies may not be representative, since the composition of the ore mined from each deposit is also presumed to shift through time.

Although ores accounted for 80% of emissions relative to coal burning in 1988, mobile sources accounted for over 80% of the total emissions [33]. The transition to the dominance of leaded gasoline emissions is evident in the integrated European record. After 1950, none of the deposits contain lead isotopes that overlap the ore body field. Lead began to be added to gasoline in the 1920s, but the initially small number of cars and the limited temporal sensitivity of the concentration records combined to delay the onset of the signal [20]. In the most recent analyses from the integrated European record, the isotopic composition of airborne lead is on a trend that intersects western and northwestern European aerosols measured in 1988 [30]. These aerosol values, in turn, largely overlap the range of compositions typical in average emissions from western industrial nations that have been controlled by the mix of Australian and US lead historically added to gasoline. The convergence of the three groups is on a value of $^{206}Pb/^{207}Pb$ ca. 1.13 and $^{208}Pb/^{207}Pb$ ca. 2.41, which we propose as an estimate of the average lead isotopic composition in the air over Europe in the last 20 years.

6. Conclusions

The Norwegian ombrotrophic bog cores have allowed a history of the overall changes in lead composition in the air over Europe to be constructed. The integrated record compiled here extends the study of a Greenland ice core by Rosman et al. [5] to include measurements from the 17th through 20th centuries, and complements the 12,000 year Swiss bog record [8] by including $^{208}Pb/^{207}Pb$ measurements and correlation with other European chronologies. The lead isotope compositions in the integrated record overlap with the compositions of anthropogenic emissions for the last 2300 years and are very different from measurements of natural background lead. From the onset of Greek and Roman mining until the 20th century the European record appears to be controlled by processing of European ore bodies. Gasoline is the most prevalent atmospheric lead source from the 1950s onward, and the transition from previous inputs to these emissions can be seen across Europe in the integrated record. At present, a phaseout of leaded gasoline is nearing completion, and the lead isotopic signature in the air over Europe is likely to be different than it was in 1991. The incessant emission of lead into the environment, however, ensures that anthropogenic lead will continue to overwhelm the very small amounts that occur naturally in the air. ***[CL]***

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