

# History of Atmospheric Lead Deposition Since 12,370 <sup>14</sup>C yr BP from a Peat Bog, Jura Mountains, Switzerland

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A continuous record of atmospheric lead since 12,370 carbon-14 years before the present (<sup>14</sup>C yr BP) is preserved in a Swiss peat bog. Enhanced fluxes caused by climate changes reached their maxima 10,590 <sup>14</sup>C yr BP (Younger Dryas) and 8230 <sup>14</sup>C yr BP. Soil erosion caused by forest clearing and agricultural tillage increased lead deposition after 5320 <sup>14</sup>C yr BP. Increasing lead/scandium and decreasing lead-206/lead-207 beginning 3000 <sup>14</sup>C yr BP indicate the beginning of lead pollution from mining and smelting, and anthropogenic sources have dominated lead emissions ever since. The greatest lead flux (15.7 milligrams per square meter per year in A.D. 1979) was 1570 times the natural, background value (0.01 milligram per square meter per year from 8030 to 5320 <sup>14</sup>C yr BP).

The history of atmospheric Pb pollution in Europe has its origins in antiquity (1, 2), but a complete, quantitative environmental record of Pb contamination from prehistory to the present has not yet been constructed. The European Greenland Ice-Core Project (GRIP) ice core drilled at Summit in remote Greenland has revealed evidence of hemispheric Pb contamination extending back three millennia to the time of ancient Mediterranean civilizations (3), but the quality of the sections recovered from the "brittle zone depth" (corresponding to ice deposited between 3500 and 7000 years ago) is too poor to allow reliable measurements of trace metals (3, 4). As a result, the natural fluxes of metals to the air and their response to the dynamic climate changes which characterize

the Holocene (5) remain poorly understood.

Peat bogs can be used as archives of atmospheric metal deposition (6). The surface layers in ombrotrophic bogs are hydrologically isolated from the influence of local groundwaters and surface waters and receive their inorganic solids exclusively by atmospheric deposition (7). Elevated Pb concentrations in peats dating from Roman times have been reported in bogs from many parts of Europe (8, 9). Isotopic studies have shown that Pb is effectively immobile in peat pro-

files (10), and comparative studies of peat bog and lake sediment records are in good agreement (11). A bog in northwestern Spain revealed 3000 years of Pb enrichments that are consistent with historical records of Pb mining in the Iberian Peninsula (12). In Switzerland, a peat bog provided a record of changing Pb concentrations for the entire Holocene (13). Here, we use that peat profile to reconstruct the changing rates of atmospheric Pb deposition and use the isotopic composition of Pb (14) to separate natural and anthropogenic sources.

Etang de la Gruère (EGR) in the Jura Mountains, Switzerland, is a raised, ombrotrophic bog (15) that consists of up to 650 cm of peat directly overlying lacustrine clay. The arboreal pollen record (16) indicates that the core represents the entire Holocene and part of the Late Glacial. Peat cores were collected, prepared, and analyzed for Pb and Sc (17). Age dates were obtained for the uppermost layers using <sup>210</sup>Pb analysis (18); deeper, older samples were dated using <sup>14</sup>C (Table 1).

## Pb and Sc concentrations and fluxes.

From the base of the peat profile at 650 cm (dated 12,370 <sup>14</sup>C yr BP) up to 145 cm (dated 3000 <sup>14</sup>C yr BP), Pb concentrations are proportional to those of Sc (Fig. 1), and the Pb/Sc ratios are generally in good agreement with crustal values (19). From 12,370 to 3000 <sup>14</sup>C yr BP, therefore, soil dust aerosols derived from rock weathering were the dominant source of atmospheric Pb.

The lowest concentrations of Pb and Sc were found in the peats between 405 and 235 cm, where Pb averages  $0.28 \pm 0.05 \mu\text{g/g}$  and

**Table 1.** Radiocarbon age dates of EGR peat samples. Peat samples were dated by <sup>14</sup>C decay counting using a procedure similar to that used for peats by Mook and Streuerman (59). The dried, powdered samples were pretreated with HCl-NaOH-HCl, then burned in a quartz glass tube in oxygen. The resulting CO<sub>2</sub> was purified, captured with liquid nitrogen, and subsequently reduced to methane with H<sub>2</sub> using a Ru catalyst at 300°C. The methane was counted in the underground laboratory (Physics Institute, University of Berne) for 70 hours. The ages are reported here as conventional <sup>14</sup>C years (<sup>14</sup>C yr BP) and as calibrated years BP (cal yr BP). Calibrated ages were calculated using CALIB REV, version 3.0.3 (60) and are reported here as intercepts or the range of intercepts (without standard deviations).

Sample no.	Depth (centimeters)	Radiocarbon age ( <sup>14</sup> C yr BP)	Calibrated age (calendar year BP)	Lab no.
2f14	36–39	240 ± 20	289	B-6680
2f16	42–45	710 ± 20	660	B-6681
2f18	48–51	1,010 ± 30	930	B-6682
2f23	63–66	1,350 ± 50	1,281	B-6683
2f26	72–75	1,400 ± 50	1,297	B-6684
2f29	81–84	1,610 ± 50	1,517	B-6685
2f34, 2f35	96–102	2,110 ± 30	2,066–2,100	B-6459
2p5	140–150	3,000 ± 30	3,170–3,206	B-6651
2p13	220–230	5,320 ± 40	6,048–6,168	B-6652
2p16	250–260	5,790 ± 80	6,573–6,630	B-6653
2p32	410–420	8,030 ± 50	8,955	B-6935
2p34	430–440	8,230 ± 30	9,210	B-6654
2p36	450–460	8,520 ± 60	9,483	B-7071
2p42	510–520	9,320 ± 50	10,300–10,330	B-6936
2p46	550–560	10,590 ± 50	12,521	B-6655
2p49	580–590	11,440 ± 70	13,348	B-6937
2p54	630–640	12,000 ± 70	13,999	B-6938
2p55	640–650	12,370 ± 50	14,457	B-6656

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Sc  $0.07 \pm 0.02 \mu\text{g/g}$  (Fig. 1). The lowest rates of atmospheric Pb ( $0.010 \pm 0.002 \text{ mg/m}^2/\text{year}$ ) and Sc ( $0.003 \pm 0.001 \text{ mg/m}^2/\text{year}$ ) deposition, therefore, occurred between 8030 and 5320  $^{14}\text{C}$  yr BP (Fig. 2); these are the lowest concentrations and fluxes for the entire Holocene and are assumed to represent the "natural background" values.

Below 400 cm, there are two pronounced peaks in Pb and Sc concentrations (Fig. 1), centered at 435 cm (dated 8230  $^{14}\text{C}$  yr BP) and at 555 cm (dated 10,590  $^{14}\text{C}$  yr BP). The fluxes at these depths (Fig. 2) exceed the background rates by factors of 3.5 and 35, respectively. The highest rates of soil dust deposition experienced since the Late Glacial are recorded in the sample dated 10,590  $^{14}\text{C}$  yr BP; this corresponds to the Younger Dryas (YD), a period characterized by increased storminess, expansion of dry, dusty areas, and reduced vegetation cover (20, 21). In the U.S.-Greenland Ice Sheet Project (GISP2) ice core, this event lasted  $1300 \pm 70$  years and terminated  $11,640 \pm 250$  years ago (20). Our calibrated age of 12,521 cal yr BP (calibrated years before present) (Table 1), therefore, lies within the time frame for the YD indicated by the Greenland ice cores. Taking 585 and 515 cm as the beginning and end of the YD (Fig. 1), our radiocarbon ages for these points (11,440 and 9320  $^{14}\text{C}$  yr BP) are consistent with records from Swiss lake sediments giving the YD as 11,000 to 10,000  $^{14}\text{C}$  yr BP (22) and with other archives of climate

change from across Europe (23).

The younger peak in Pb and Sc at 435 cm (Fig. 1) is similar in age to the Vasset/Killian volcanic event from Massif Central, which was dated  $8230 \pm 140$   $^{14}\text{C}$  yr BP in sediments from a Swiss lake (24). However, an important period of climatic change also took place near this time (25).

At 225 cm (dated 5320  $^{14}\text{C}$  yr BP), the concentrations (Fig. 1) and fluxes (Fig. 2) of both Pb and Sc increased significantly, indicating a change in the rates of atmospheric deposition of soil dust. The timing of these changes is consistent with tree and cereal pollen records from the Jura Mountains marking the first signs of forest clearing and the beginning of plant cultivation around 6000  $^{14}\text{C}$  yr BP (26). These changes to the landscape would have been accompanied by soil tillage and enhanced rates of soil erosion, thereby promoting the creation and release of soil dust.

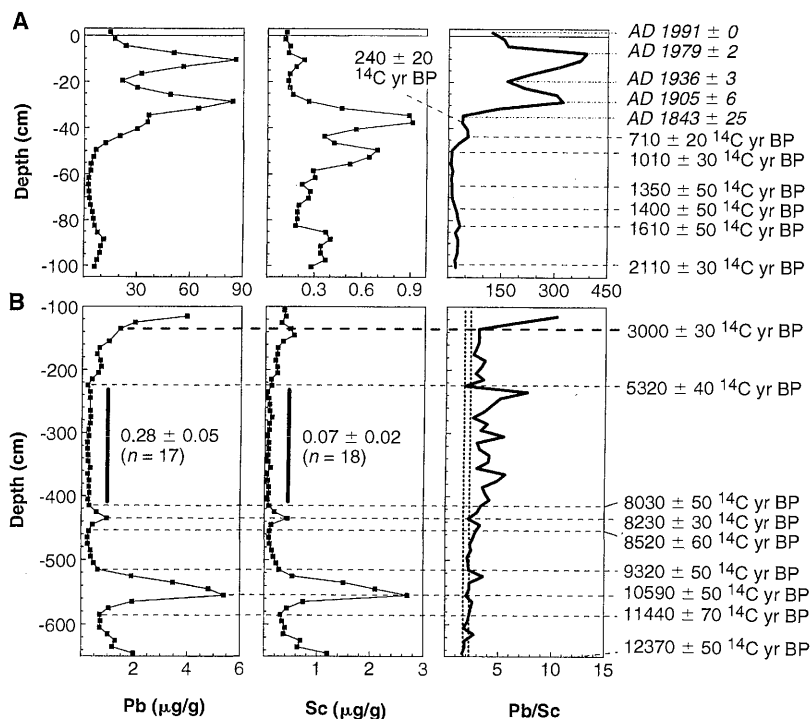
Above 145 cm (dated 3000  $^{14}\text{C}$  yr BP), the Pb concentrations (Fig. 1) and fluxes (Fig. 2) increase out of proportion with Sc. To explain these changes, therefore, a nonsilicate source of Pb is required. A peak in atmospheric Pb deposition was reached slightly after  $2110 \pm 30$   $^{14}\text{C}$  yr BP, when the flux was 37 times the background value. The sample containing the greatest Pb/Sc ratio during this period (8.6 times background) was dated 1610  $^{14}\text{C}$  yr BP (Fig. 1). Lead concentrations and fluxes declined afterward, but have ex-

ceeded the background values by at least nine times ever since.

The highest Pb/Sc ratios were in samples dated 710  $^{14}\text{C}$  yr BP, A.D. 1905, and A.D. 1979 (Fig. 1). Lead deposition reached  $10.1 \text{ mg/m}^2/\text{year}$  by A.D. 1905 (Fig. 2), for the first time exceeding the natural flux by a factor of more than 1000. The Pb flux has declined since A.D. 1979 ( $15.7 \text{ mg/m}^2/\text{year}$ ). The most recent Pb flux that we measured in the Jura bogs ( $8.55 \text{ mg/m}^2/\text{year}$ ) in A.D. 1991 is 855 times the natural, background value. However, the Pb/Sc ratio of this sample exceeds the background value by only 31.4 times (Fig. 1). Thus, while various sources of heavy-metal pollution (mining, refining, coal burning, gasoline Pb, and so on) are important Pb sources, soil dust deposition today remains an important component of the elevated Pb flux.

**Enrichment factors and isotopic composition of Pb.** We used the  $^{206}\text{Pb}/^{207}\text{Pb}$  ratios and the Pb enrichment factor (Pb EF) (27) to distinguish natural from anthropogenic sources of atmospheric Pb (Fig. 3). The mineral sediment underlying the peat (below 650 cm) has a  $^{206}\text{Pb}/^{207}\text{Pb}$  ratio of  $1.2165 \pm 0.0048$ . The deepest peat layer (2p55, at 640 to 650 cm) has a similar value ( $^{206}\text{Pb}/^{207}\text{Pb} = 1.2158 \pm 0.0002$ ). In contrast, the next higher peat sample (2p54 at 630 to 640 cm) has a significantly different ratio ( $1.2050 \pm 0.0002$ ), indicating that Pb in all highest layers of the bog are essentially unaffected by the basal mineral sediment. These data support earlier geochemical arguments (13) that the deepest peat layers of this bog are supplied with Pb exclusively by atmospheric deposition.

From 615 to 455 cm, the  $^{206}\text{Pb}/^{207}\text{Pb}$  ratio averages  $1.2045 \pm 0.0002$  ( $n = 12$ ), a value close to that for the average upper continental crust (28) and is assumed to represent the early Holocene. However, starting at 455 cm (dated 8520  $^{14}\text{C}$  yr BP), there is a shift toward significantly more radiogenic values (higher  $^{206}\text{Pb}/^{207}\text{Pb}$ ), extending over 40 to 50 cm (Fig. 3). The rocks from Massif Central have  $^{206}\text{Pb}/^{207}\text{Pb} = 1.20$  to 1.24 (29), similar in value to this part of the peat core. However, the duration of the shift in  $^{206}\text{Pb}/^{207}\text{Pb}$  in the core is greater than could be explained by the Vasset/Killian tephra (VKT) alone. For example, in Lake Soppensee of the Swiss plateau, the VKT is dated  $8230 \pm 140$   $^{14}\text{C}$  yr BP and the sample selected just above the VKT ash layer is dated  $8110 \pm 140$   $^{14}\text{C}$  yr BP (24). In contrast, the period of more radiogenic soil dust deposition at EGR extends from 455 cm (dated 8520  $^{14}\text{C}$  yr BP) to 395 cm. The peat at 395 cm is more recent than the sample at 415 cm (dated 8030  $^{14}\text{C}$  yr BP), so this period lasted longer than 500 years; given the average rates of peat accumulation in this section of the core, we estimate that it lasted approximately 750 years (30). Although the VKT



**Fig. 1.** Lead and Sc concentrations ( $\mu\text{g/g}$ ) and Pb/Sc ratios. Notice the difference in horizontal scales between core 2f (A) and 2p (B). Also shown are selected  $^{210}\text{Pb}$  age dates (18) and radiocarbon ages. The two vertical dotted lines in the Pb/Sc plot for core 2p (B) represent the values for Pb/Sc in the upper continental crust (19).

may be present in the peat core, it alone is unlikely to account for the vertically extensive shift in  $^{206}\text{Pb}/^{207}\text{Pb}$ . Another possible cause is the globally distributed cooling event that extended from ~9000 to 7800 calendar years ago (31), and was similar to the YD (dominated by cold, dry, dusty conditions). The greatest dust flux during this cool phase was one-tenth that of the YD (31). For comparison, in the EGR core, the dust flux at 8230  $^{14}\text{C}$  yr BP also was one-tenth of the maximum dust flux recorded during the YD (Fig. 2).

The mid-Holocene peats from 395 cm to 275 cm are significantly less radiogenic ( $^{206}\text{Pb}/^{207}\text{Pb} = 1.1994 \pm 0.0004$ ) than those of the early Holocene ( $1.2045 \pm 0.0002$ ; from 615 to 455 cm). Also, the mid-Holocene Pb/Sc ratios are generally greater than those of the early Holocene (Fig. 1), are in excess of crustal values, and are more variable. The elevated Pb/Sc ratios could have resulted from the natural enrichment of Pb in the fine fraction of soils during weathering, but this could not explain the shift in isotopic composition (Fig. 3). A more likely explanation is a change in the sources of soil dust to the bog. The mid-Holocene value for  $^{206}\text{Pb}/^{207}\text{Pb}$  is closer to the composition of Saharan dust (32).

At 145 cm (dated 3000  $^{14}\text{C}$  yr BP), the  $^{206}\text{Pb}/^{207}\text{Pb}$  ratios decrease and the Pb EF exceeds 2 for the first time (Fig. 3); all peat samples above this depth have  $^{206}\text{Pb}/^{207}\text{Pb} < 1.2$  and Pb EF  $\geq 2$ . In samples more recent than 3000  $^{14}\text{C}$  yr BP, therefore, Pb is enriched out of proportion with Sc, and the Pb is not sufficiently radiogenic to have been derived exclusively from soil dust: an additional, less radiogenic component was most likely supplied by Paleozoic and older Pb ores (33). The Pb EF and Pb isotope data can be explained by historical records of ancient Pb mining and long-range transport of aerosols from the Iberian Peninsula (34). A peat core from Galicia in northwestern Spain indicates a similar chronology of Pb enrichment (12), and Pb in Greenland ice dating from this time has been attributed to the same source (35).

Elevated Pb EFs dating from Roman times were found in the deepest sections of the 2f core, and the  $^{206}\text{Pb}/^{207}\text{Pb}$  ratios are all below 1.18 from 101 to 74 cm. At 74 cm (1400  $^{14}\text{C}$  yr BP), the Pb EF declines, and starting at 71 cm, the  $^{206}\text{Pb}/^{207}\text{Pb}$  ratios shift back toward more radiogenic values and exceed 1.18 for the first time since the beginning of the Roman Period. The period of greatest Roman mining was the late Republic and early Empire (400 B.C. to 37 A.D.), with production declining in the third century A.D. (36). By the early fifth century A.D., western Roman mining had collapsed (37). Although the Pb concentrations, Pb/Sc ratios, and Pb

isotope values from the EGR peat core certainly indicate a pronounced decline in Pb mining following the fall of Rome, the Pb/Sc ratios remained well above and the  $^{206}\text{Pb}/^{207}\text{Pb}$  ratios well below the values seen during the mid-Holocene (Fig. 3). Thus, while Pb contamination clearly declined with the fall of the western Roman Empire, atmospheric Pb pollution has been continuous from 3000  $^{14}\text{C}$  yr BP to the present.

The lowest Pb/Sc ratios seen since the Roman Period are from ~65 to 50 cm, corresponding to ~1350 to 1010  $^{14}\text{C}$  yr BP. Even at this time, however, the Pb/Sc ratios were at least twice the preanthropogenic value, indicating that more than one-half of the Pb deposited on the bog surface was generated by ore mining. The  $^{206}\text{Pb}/^{207}\text{Pb}$  ratios in this interval average  $1.1847 \pm 0.0002$ , which is well below the preanthropogenic ratio of  $1.1994 \pm 0.0004$  (Fig. 3).

At 50 cm, Pb EFs increase and  $^{206}\text{Pb}/^{207}\text{Pb}$  ratios decrease (Fig. 3) until 44 cm, where the Pb EF reaches a peak value of 14.4. Sample 2f18 (50 cm) was dated 1010  $^{14}\text{C}$  yr BP and sample 2f16 (44 cm) at 710  $^{14}\text{C}$  yr BP; both dates are in good agreement with historical records of Medieval silver produc-

tion in Germany (38), when silver was obtained by mining Pb ores.

Above 32 cm (dated A.D. 1879), the Pb/Sc ratio increases rapidly, reaching a peak Pb EF of 82.3 at A.D. 1905 (Fig. 3). This period corresponds to the Second Industrial Revolution in Europe, which introduced great advances in smelting techniques and increased Pb production (39), fueled by burning coal. During this same interval,  $^{206}\text{Pb}/^{207}\text{Pb}$  ratios decrease rapidly, from  $1.1819 \pm 0.0003$  at 38 cm (210  $^{14}\text{C}$  yr BP) to  $1.1684 \pm 0.0004$  at 29 cm (A.D. 1905) (Fig. 3). A similar decrease in  $^{206}\text{Pb}/^{207}\text{Pb}$  ratios during the last century was reported for lake sediments in Belgium (40), archived herbage samples collected at the Rothamsted Experimental Station in England (41), and in lake sediments from Scotland (42) and Switzerland (43). Moreover, an identical trend (for the past 130 years) to the one we report for the EGR peat core has since been found, using herbarium specimens of *Sphagnum* moss collected since A.D. 1867 and in three other Swiss peat cores (44). Since the beginning of the 20th century, the ratios of  $^{206}\text{Pb}/^{207}\text{Pb}$  are so low that they cannot be attributed only to European Pb ores or coals (45); the most likely explanation is

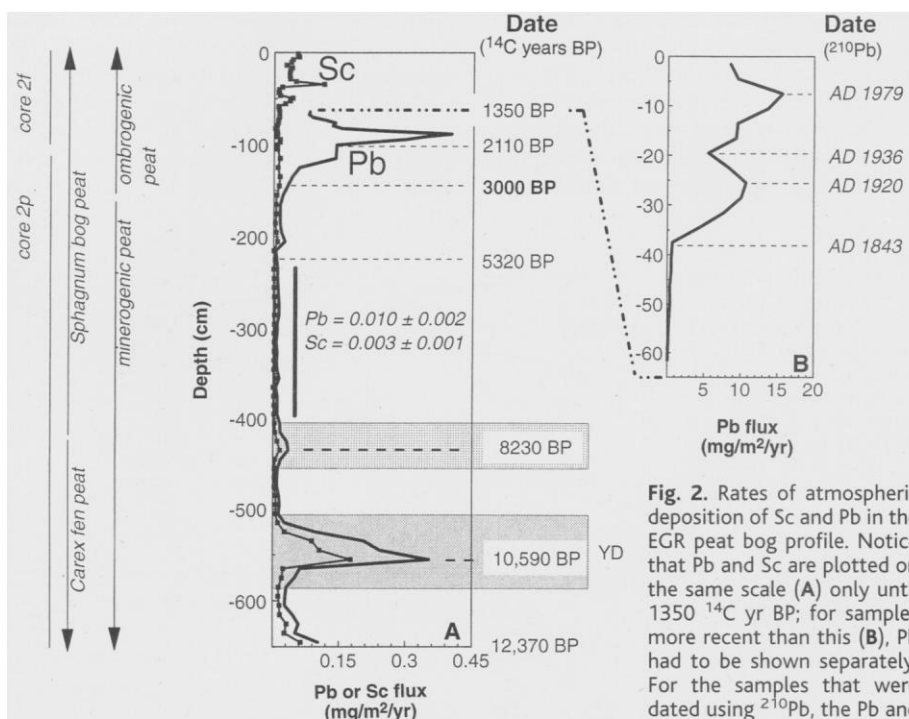


Fig. 2. Rates of atmospheric deposition of Sc and Pb in the EGR peat bog profile. Notice that Pb and Sc are plotted on the same scale (A) only until 1350  $^{14}\text{C}$  yr BP; for samples more recent than this (B), Pb had to be shown separately. For the samples that were dated using  $^{210}\text{Pb}$ , the Pb and Sc fluxes ( $\mu\text{g}/\text{cm}^2/\text{year}$ )

were calculated as the product of the metal concentration ( $\mu\text{g}/\text{g}$ ) and dry mass accumulation rates ( $\text{g}/\text{cm}^2/\text{year}$ ) taken from the constant rate of supply (CRS) model (78). For older samples, which were dated using  $^{14}\text{C}$ , the fluxes for individual samples were calculated as  $I = C \times A \times \rho$ , where  $C$  = metal concentration ( $\mu\text{g}/\text{g}$ ),  $A$  = incremental net rate of peat accumulation ( $\text{cm}/\text{year}$ ), and  $\rho$  = peat bulk density ( $\text{g}/\text{cm}^3$ ). Selected  $^{14}\text{C}$  age dates (Table 1) defined the depth increments; net accumulation rates ranged from  $1.28$  to  $7.22 \times 10^{-2}$   $\text{cm}/\text{year}$ ; for comparison, the average peat accumulation rate for the entire profile is  $5.25 \times 10^{-2}$   $\text{cm}/\text{year}$ . The background fluxes are  $10.3 \pm 2.3$  Pb  $\mu\text{g}/\text{m}^2/\text{year}$  and  $2.7 \pm 1.0$  Sc  $\mu\text{g}/\text{m}^2/\text{year}$ , but for convenience, the fluxes are plotted in units of  $\text{mg}/\text{m}^2/\text{year}$ . The transition from ombrogenic to minerogenic peat takes place at 155 cm (13), but nonatmospheric sources of Pb are not important to the Pb inventory of the minerogenic peats, which extend from 155 to 645 cm.

the introduction of Australian Pb ores to Europe. Australian Pb ores from the Broken Hill mine have ratios of  $^{206}\text{Pb}/^{207}\text{Pb} < 1.04$  (33) and were first imported into England in 1826; by the middle of the 19th century, British ores had become uneconomic and were effectively replaced by Australian ones (39).

The distinct peak of Pb/Sc ratio at A.D. 1905 (Fig. 1) is seen not only at EGR, but also in three other Swiss bogs (44). Many lake sediments in Switzerland also contain elevated Pb concentrations at depths corresponding to the first few decades of the 20th century (46).

Starting at 20 cm (dated A.D. 1936), there is another pronounced increase in Pb concentrations and a further sharp decline in the  $^{206}\text{Pb}/^{207}\text{Pb}$  ratios. The largest (and also the most recent) peak in Pb/Sc at EGR was dated A.D. 1979 with Pb EF = 99.1 (Fig. 3). The isotopic composition of Pb in this sample (2f5, with  $^{206}\text{Pb}/^{207}\text{Pb} = 1.1492 \pm 0.0002$  and  $^{206}\text{Pb}/^{204}\text{Pb} = 17.876 \pm 0.022$ ) is consistent with the gasoline lead (47) used in Berne, Switzerland, in the 1970s ( $^{206}\text{Pb}/^{207}\text{Pb} = 1.14$  and  $^{206}\text{Pb}/^{204}\text{Pb} = 17.83$ ). The rapid rise in Pb/Sc from 1936 to 1979, therefore, is attributable to gasoline Pb that was introduced into Switzerland in 1947 (48). The isotopic signatures of these recent peat samples are compatible with values for leaded gasoline in France and the United Kingdom (49); these, in turn, reflect the variety of ore deposits that were used to manufacture gasoline Pb additives, including the most important mines in Australia and Canada (50).

Since its introduction, gasoline Pb has dominated atmospheric Pb emissions in Europe, reaching a maximum between A.D. 1975 and 1982 (51).

The most recent sample analyzed (dated A.D. 1991) shows a decrease in Pb EF, a reduced rate of atmospheric Pb deposition, and a shift in isotopic composition back toward more radiogenic values (Figs. 1 through 3); similar patterns are seen in other Swiss bogs (44). Chemical and isotopic analyses of Pb in western European aerosols (52) show a similar trend during the past 20 years, consistent with the gradual elimination of leaded gasoline.

**History of atmospheric Pb deposition in Europe since 12,370  $^{14}\text{C}$  yr BP.** The peat profile at EGR reveals 10 major periods in the history of atmospheric Pb deposition (Fig. 3). Until 3000  $^{14}\text{C}$  yr BP, soil dust was the single most important source of Pb to the bog. The dust was probably generated over a very wide area, with most particles smaller than 5  $\mu\text{m}$  in diameter and transported up to several thousand km (53). Starting at 8520  $^{14}\text{C}$  yr BP, there was a change in dust sources. The most likely source of more radiogenic mineral dust would be the Archaean rocks of the Scandinavian shield: sediments derived from these rocks in Sweden show background  $^{206}\text{Pb}/^{207}\text{Pb}$  ratios of 1.3 to 1.35 (11). Scandinavia was still under ice cover by 9500  $^{14}\text{C}$  yr BP, but became free of ice by 8600  $^{14}\text{C}$  yr BP (54). If only 7% of the soil dust to the bog was supplied by Scandinavian rock flour with  $^{206}\text{Pb}/^{207}\text{Pb} = 1.35$ , this would

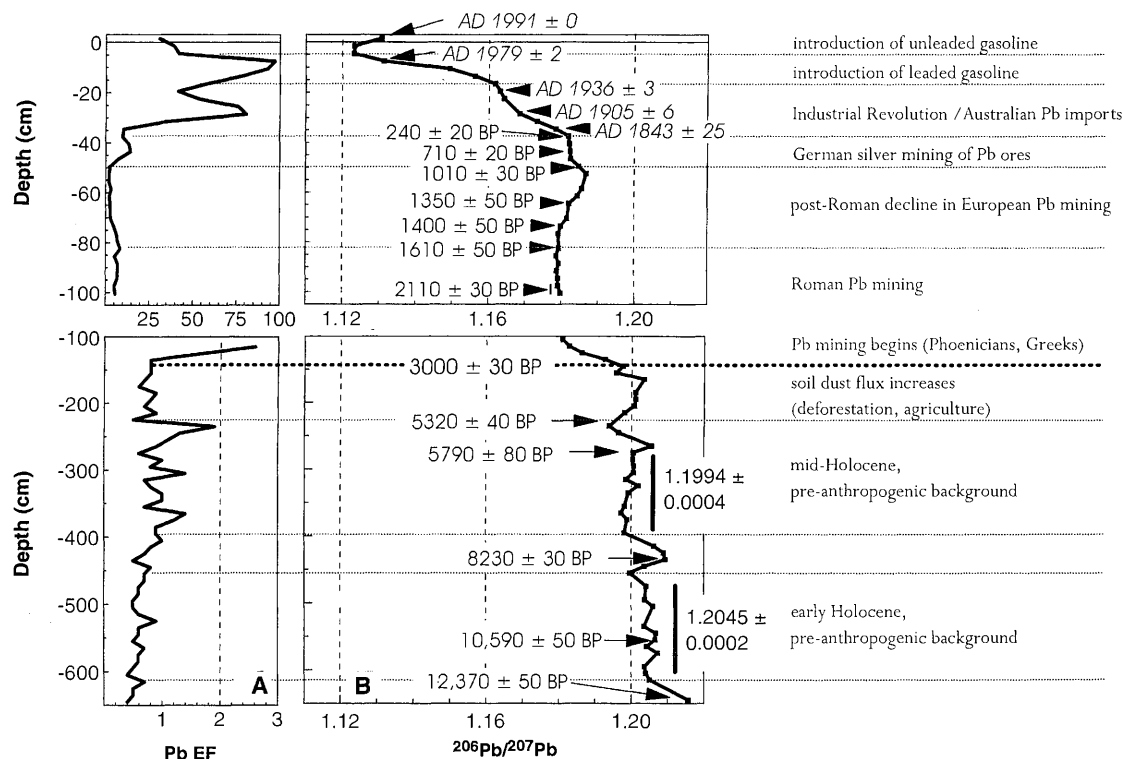
have shifted the  $^{206}\text{Pb}/^{207}\text{Pb}$  ratios of the peats from 1.20 to 1.21, as seen in Fig. 3. After  $\sim 7700$   $^{14}\text{C}$  yr BP (30), the  $^{206}\text{Pb}/^{207}\text{Pb}$  ratios were lower, indicating that Sahara dust was the predominant Pb source. These changes are consistent with the expansion of vegetation across Scandinavia at this time (54)—which would have cut off the supply of radiogenic soil dust—and with the Saharan climate change from savannah to desert when humidity began to decline at 7500  $^{14}\text{C}$  yr BP (55).

The decline in  $^{206}\text{Pb}/^{207}\text{Pb}$  ratios and the increase in Pb EFs starting at 3000  $^{14}\text{C}$  yr BP (Fig. 3) show that anthropogenic sources have dominated atmospheric Pb emissions in Europe ever since.

**Comparison with the ice core Pb record from Greenland.** Our results are in agreement with the Greenland GRIP ice core Pb record for the past three millennia (3): both sets of archives document the effects of Roman Pb mining, Medieval German silver production, and the Industrial Revolution. In addition, the peat bog record is consistent with the recent changes in Pb concentrations seen in Greenland snow (56). However, there are also some important differences between the two archives (57).

Today, atmospheric Pb fluxes are decreasing, and the  $^{206}\text{Pb}/^{207}\text{Pb}$  ratios are moving back toward more radiogenic values typical of soil dust derived from crustal weathering. These changes are a testimony to the success of relatively recent efforts (within the past few decades) to reduce atmospheric Pb emis-

**Fig. 3. (A)** Pb EF calculated as the ratio of Pb/Sc in the peats, normalized to the background value (27). An EF < 2 indicates that the sample is not enriched, relative to background (13). **(B)** Isotopic composition of Pb summarized as  $^{206}\text{Pb}/^{207}\text{Pb}$  and the chronology of atmospheric Pb deposition since 12,370  $^{14}\text{C}$  yr BP. The heavy, horizontal dashed line at 3000  $^{14}\text{C}$  yr BP separates those sections of the peat profile where the dominant Pb source is soil dust (samples predating this time) versus those where ore Pb predominated (samples postdating this time).



sions from industrial sources and the gradual removal of Pb from gasoline. The ratio of  $^{206}\text{Pb}/^{207}\text{Pb}$  in aerosols deposited in A.D. 1991 ( $1.1307 \pm 0.0002$ ) is, however, very far from the preanthropogenic values seen during the middle of the Holocene ( $1.1994 \pm 0.0004$ ). Also, the rates of atmospheric Pb deposition today are still several hundred times higher than the natural, background flux.

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- for by soil dust and the other half by Pb mining. Sample 2p2 has a  $^{206}\text{Pb}/^{207}\text{Pb} = 1.1827$  and its age lies between 3000 (145 cm, core 2p) and 2110  $^{14}\text{C}$  yr BP (96 to 102 cm, core 2f). If we assume that the background soil dust signature at this time was  $^{206}\text{Pb}/^{207}\text{Pb} = 1.1999$  (Fig. 3), we can calculate the isotopic composition of the Pb ores by solving for  $x$ :  $1.1827 = 0.5 (1.1999) + 0.5 (x)$ . Using this simple approach, we calculated that the Pb ores must have had  $^{206}\text{Pb}/^{207}\text{Pb} = 1.1655$ . According to J. O. Nriagu (2), mining in the Iberian Peninsula accounted for 37% of the Pb that was produced during the Iron Age (1200 to 50 B.C.), making it the most important Pb mining area of its time. Ores from these mines are known to have  $^{206}\text{Pb}/^{207}\text{Pb}$  values between 1.1722 and 1.1619 (58). The single most important ore body from this area is Rio Tinto, and the galenas from this ore range from  $^{206}\text{Pb}/^{207}\text{Pb} = 1.1632$  to 1.1639 [C. Pomies, A. Cocherie, C. Guerrot, E. Marcoux, J. Lancelot, *Chem. Geol.* **144**, 137 (1998)].
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## Distinct Mechanism for Antidepressant Activity by Blockade of Central Substance P Receptors

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The localization of substance P in brain regions that coordinate stress responses and receive convergent monoaminergic innervation suggested that substance P antagonists might have psychotherapeutic properties. Like clinically used antidepressant and anxiolytic drugs, substance P antagonists suppressed isolation-induced vocalizations in guinea pigs. In a placebo-controlled trial in patients with moderate to severe major depression, robust antidepressant effects of the substance P antagonist MK-869 were consistently observed. In preclinical studies, substance P antagonists did not interact with monoamine systems in the manner seen with established antidepressant drugs. These findings suggest that substance P may play an important role in psychiatric disorders.

The development of new drugs to treat depression has been severely constrained by a poor understanding of the pathophysiology of this disease and of the mechanisms by which drugs that augment monoamine function alleviate its symptoms. The predictive validity of many preclinical assays is also limited by an inability to model psychiatric disease in animals. However, there is a pressing need for improved antidepressant therapies, given the considerable prevalence, morbidity, and

mortality of depressive disorders, the incomplete efficacy of currently available drugs in many patients, and the potentially distressing adverse effects of existing therapies (1).

**Localization of substance P in brain: Evidence for an involvement in the response to stress.** Substance P is the most abundant neurokinin in the mammalian central nervous system (CNS). Mapping studies indicate that the substance P-preferring neurokinin-1 (NK<sub>1</sub>) receptor is highly expressed