200 Years of Pb Deposition throughout the Czech Republic: Patterns and Sources

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Historical rates of Pb deposition were determined over the past 150−200 years for eight sites throughout the Czech Republic using ^{210}Pb-dated, *Sphagnum*-derived peat cores. Maximum historical Pb deposition was greater at sites in the northern and western parts of the Czech Republic than at sites in the southern part of the Czech Republic (peak values averaging 57, 21, and 16 mg m^{-2} yr^{-1}, respectively). Lead deposition patterns generally reflect increasing industrialization over the past 100−200 years, especially in the post-World War II era. For seven of the eight sites, maximum Pb deposition occurred between 1965 and 1992, corresponding to a period of peak production and burning of lignite coal. A decrease in Pb deposition rates since 1975−1980 was evident in seven of the sites. The most recent Pb deposition rates (1992), estimated from the uppermost peat core sections, averaged 32, 11, and 7 mg m^{-2} yr^{-1} for the northern, western, and southern sites, respectively, are higher than current Pb deposition in the eastern United States of 4 mg m^{-2} yr^{-1}. Lead deposition rates prior to Czech industrialization, estimated from the deepest dateable peat core sections, averaged 8, 5, and 1 mg m^{-2} yr^{-1} for the northern, western, and southern sites, respectively. Using acid-insoluble ash concentrations in peat and peat magnetic susceptibility determinations, we were able to identify past periods of elevated Pb deposition related to local mining of Pb-containing ore deposits at three of the sites and periods of elevated Pb deposition from fossil fuel combustion at five of the sites. Without stable Pb isotopic determinations, the importance of leaded gasoline-derived Pb could not be determined.

Introduction

Czechoslovakia had one of the more developed economies in Europe at the start of World War II, and its industry, especially in what is now the Czech Republic, was at one time more advanced than that of either Austria or Belgium (*1, 2*). As a legacy of 42 years of forced industrial expansion under a socialist regime, the Czech Republic has retained its status as one of the most air-polluted countries in the world (*3*−*5*). Although coal mining in northern Bohemia began in the 1860s, considerable air pollution began in 1945 with use of lignite coal for energy production for industrial purposes and home heating (Figure 1) (*3*). Lignite contains substantially greater ash (>33%) than more highly ranked coals (e.g., anthracite, the highest rank of coal, contains 5% or less) (*6*). Prior to German unification, the former East Germany and Czechoslovakia burned nearly one-third of the world's lignite, with little or no pollution control (*7*). As a result, both the former Czechoslovakia and other eastern bloc countries have been contaminated with numerous types of pollutants.

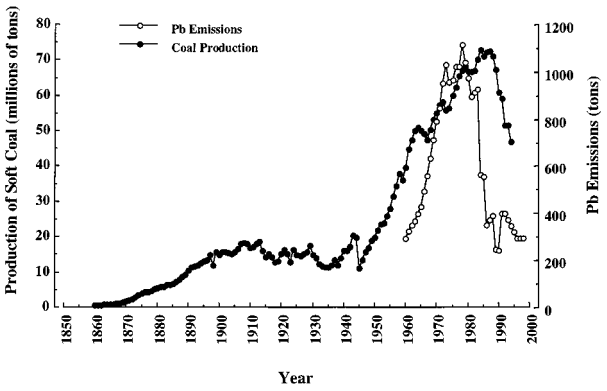

FIGURE 1. Coal production (millions of tonnes) for the Most Coal Basin in northern Bohemia and Pb emissions from gasoline combustion (tonnes) as a function of year in the former Bohemian Kingdom (pre-1918) Czechoslovakia (1918−1992), and the Czech Republic (1993-present); North Bohemian Mining Co., unpublished data.

Lead is one of many pollutants plaguing the environment of the Czech Republic. Lead is widely distributed primarily due to its emission from combustion of Pb-containing fuel (*8*). Lead is known to have a variety of effects on the human nervous and circulatory systems and is relatively toxic at low concentrations (*9, 10*). Prior to the Industrial Revolution in Europe and the United States, most (i.e., >90%) of the actively cycling native Pb in terrestrial ecosystems was in the soil reservoir, released mainly by natural weathering of parent rock (*9*). Since the Industrial Revolution, approximately 50% of total Pb produced from anthropogenic activities has been discharged into terrestrial and aquatic ecosystems (*11*). More than 95% of the Pb now within the biosphere is of anthropogenic origin (*12*). In the Czech Republic, Pb concentrations have exceeded drinking water standards on numerous occasions (*13*), while Pb concentrations in children's blood have exceeded 300−450 μg L^{-1}, which is more than three times the level certified as neurotoxic in the United States (*3, 13*). What is not known is the extent to which conditions in the past mimic present environmental degradation.

Monitoring programs can determine present-day release of pollutants into the atmosphere (*14*−*16*). However, data from such monitoring programs typically extend back into the past for a finite, and short, period of time. Documenting changes in atmospheric pollutant deposition that occurred prior to the initiation of modern monitoring programs requires historical reconstruction of environmental pollution (*4, 17*). Because pollutant monitoring under former socialist regimes often was either minimal or not allowed, historical reconstructions represent one of the only means by which fairly recent past environmental degradation in countries like the Czech Republic can be assessed. Therefore, at eight

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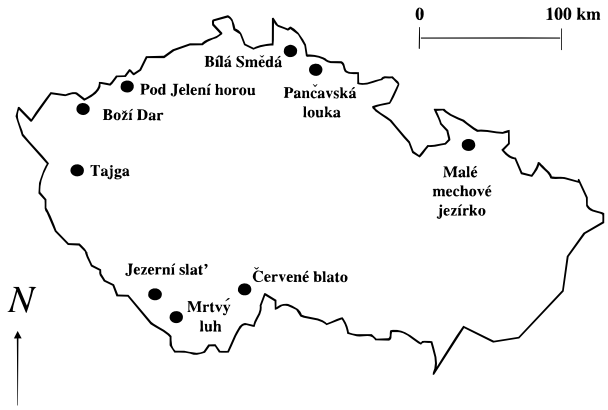

FIGURE 2. Field sites in the Czech Republic.

sites throughout the Czech Republic, we used ^{210}Pb-dated, *Sphagnum*-derived peat cores to determine the magnitude of Pb deposition over the past 200 years. In addition, we used acid-insoluble ash (AIA) contents of peat and magnetic susceptibility of peat as tools to distinguish among the major sources of Pb at these eight sites.

Materials and Methods

Sample Collection and Preparation. We selected eight *Sphagnum*-dominated peatlands located along the mountainous border regions within the Czech Republic (Figure 2) for determination of historical rates of Pb deposition. All sites are similar in topography and vegetation (ombrotrophic or weakly minerotrophic *Sphagnum*-dominated peatlands with either *Pinus mugo* or *Picea abies* as the dominant tree species). Surface water pH was measured using a standard pH meter. The water table was 1−10 cm below surface *Sphagnum* on the dates of peat core collection for all sites except Mrtvý luh where the water table was 40 cm below the surface. We collected one core from each site in the fall of 1992 using a 40-cm long, 10-cm diameter PVC cylinder. The PVC cylinders were sealed using rubber end caps with reinforced steel hose-clamps, returned to the laboratory, and frozen. Once frozen, we sectioned each peat core into 2-cm sections using a band saw, freeze-dried the peat sections to obtain dry weights and bulk densities, and homogenized each dried section using a mortar and pestle. Organic matter concentration was determined as percent mass lost during ignition at 550 °C.

Lead-210 Dating. Approximately 3 g of dried, homogenized peat from each core section were mixed with 20 mL of a 1:1 acid solution (12 M HCl:16 M HNO_3, v/v), 1.0 mL of 30% H_2O_2 (to solubilize Pb and Po isotopes and to digest organic peat, respectively), and 1.0 mL of a ^{209}Po solution (0.37 Bq mL^{-1}) as a chemical yield tracer. The mixture was heated to 60 °C in a covered beaker and evaporated to dryness overnight. The following day, we added 5 mL of 12 M HCl plus 50 mL of distilled, deionized water, and boiled the samples for 3−5 min. The mixtures were filtered through Whatman no. 42 filter paper while warm, and the filtrate was evaporated to dryness. We added 1−2 mL of 12 M HCl to the residue and again evaporated to dryness, repeating this step three additional times. After the final evaporation, we moistened the residue with a few drops of 12 M HCl, then added 150 mL of 0.3 M HCl, and heated until warm. Between 0.3 and 0.5 g of ascorbic acid then was added to complex any soluble Fe; the solutions were stirred for 1 h with a magnetic bar. A 6.25 cm^2 silver disk (Krylon 7004 insulating varnish on one side to ensure isotope plating entirely on the other side) was placed into each solution and allowed to sit at 60 °C overnight to passively plate the Pb and Po isotopes from the digest solutions onto each silver disk. Subsequently, α-emission activity was measured on an EG&G ORTEC alpha spectrometer (model 576A).

The constant rate of supply (CRS) model (*18*) was used to calculate dates. Each date determined by ^{210}Pb-dating has an error term that comprises both counting error and propagated error associated with fitting the CRS model to the data. The magnitude of the error increases with depth (*19*). We discarded ^{210}Pb dates for depths where the error term exceeded the estimated number of years for a 2-cm peat core section.

Immobility of atmospherically deposited Pb is one major assumption in using ^{210}Pb dating of peat. In addition to past verification and corroboration of ^{210}Pb chronologies via pollen stratigraphy (cf. refs *20* and *21*), we have shown experimentally that Pb is largely immobile in *Sphagnum*-derived peat, even under conditions of a fluctuating water table, due to its intimate association with soil organic matter (*22*).

Lead Concentrations. Peat profiles were analyzed for total Pb content using a modified acid digestion procedure (*19*, *23*). To 0.5 g subsamples from each depth section we added 20 mL of 8 M HNO_3 along with 1−5 mL of 30% H_2O_2 and let the mixtures sit in covered beakers overnight. The following day we boiled the mixtures until one-half of the original volume remained, added 20 mL of 8 M HNO_3 and 1 mL of H_2O_2, and allowed the mixtures to sit overnight. The following day, we boiled the mixtures until one-half of the volume remained and then filtered through Whatman no. 42 ashless filter paper, retaining the filtrates for determination of Pb concentration.

Because HNO_3/H_2O_2 digestion may not destroy all of the solid phases that contain Pb (i.e., hydrous oxides of Fe, Al, and Mn), subsequent dry-ashing, to destroy solid Pb phases, and acid dissolution of the residue retained on the filter papers have been recommended (*23*). Therefore, the residue plus filter paper remaining after the HNO_3/H_2O_2 digestion was dry-ashed in a combustion furnace for 4 h at 550 °C. Dry-ashing after the initial acid digestion ensures that acid-soluble Pb complexes, such as $PbCl_2$, are not volatilized. After ashing, we added 10 mL of 8 M HNO_3 to the ash and heated for 1 h in a sand bath at 135 °C. The resulting mixtures were filtered through Whatman no. 42 ashless filter paper, and Pb concentrations were measured in the filtrates. To obtain total Pb for each depth section, we added the Pb recovered in the dry-ashing/acid dissolution step to the Pb recovered in the original HNO_3/H_2O_2 digestion step. Using this two-step procedure, we recovered between 2 and 39% of the total Pb in the dry-ash step (83% of the samples had <10% of the Pb in the dry-ash step; 15% of the samples had 11−20% of the Pb in the dry-ash step; 2% of the samples had >20% of the Pb in the dry-ash step).

The efficiency of the digestion procedure was determined by analyzing river sediment, certified for Pb concentration (Standard Reference Material no. 2407; U.S. National Institute of Standards and Technology − NIST). Recovery of Pb was complete using our digestion procedure [NIST value for Pb in river sediment was 161.0 μg g^{-1}; our Pb value was 165.3 $\pm$ 4.21 μg g^{-1} (n = 5; mean $\pm$ standard error)]. Concentrations of Pb in blanks did not exceed 1% of sample concentrations. We measured Pb concentrations in all final digest solutions using a Perkin-Elmer 1100B atomic absorption spectrophotometer with an air-acetylene flame.

Acid-Insoluble Ash (AIA). Insight into historical sources of Pb deposition may be obtained by examining the relationship between Pb and AIA contents of each peat core section (*20*). AIA consists mainly of refractory oxides and silicates (*24*, *25*). As these classes of compounds are basic components of soil, low quantities of AIA and small quantities of soil-associated Pb would be expected in remote peats as a result of long-range aeolian soil transport. Anthropogenic activities, such as coal or metal mining, could release much

larger quantities of AIA locally, with relatively higher Pb concentrations (*26*).

Acid-insoluble ash was determined on separate 0.5 g subsamples from each peat core section. We dry-ashed separate subsamples from each peat core section in porcelain crucibles at 550 °C for 4 h in a muffle furnace prior to hot acid digestion. Approximately 10 mL of aqua regia (12 M HCl:16 M HNO_3, 3:1 v/v) were added to the ashed samples, and the mixture was heated to 135 °C for 1 h. We filtered the digested mixture through Whatman no. 42 ashless filter paper, rinsing repeatedly. We placed the ashless filter paper along with its retained residue in a muffle furnace at 550 °C for 4 h and refer to the material remaining after this step as AIA (*20, 26*).

Magnetic Susceptibility. Another tool that may be used to interpret sources of Pb to these Czech peatlands is magnetic susceptibility (*20, 27, 28*). Mineral magnetic properties have been used as important environmental indicators of anthropogenic activity in terrestrial ecosystems, most notably for identifying coal combustion activity (*20, 29*). During coal combustion, but not petroleum combustion, hematite and magnetite minerals are produced and released to the atmosphere (*30*). Because magnetite and hematite have magnetic properties, the presence of these minerals in peat, as determined by magnetic susceptibility measurement, indicates that coal combustion products have been deposited to the surface of the bog. We note that the usefulness of magnetic susceptibility as an indicator of atmospherically deposited products of fossil fuel combustion may be restricted to near-surface, aerobic regions of a peat deposit. It is possible that under long periods of anoxic conditions, magnetic susceptibility may disappear with reduction and dissolution of hematite and magnetite minerals.

Magnetic susceptibility measurements were determined on separate 0.1−1.0 g subsamples of dried, homogenized peat from each core section using a Sapphire Magnetometer at the Paleomagnetic Laboratory, Department of Geology and Planetary Science, University of Pittsburgh, U.S.A.

Results and Discussion

An air pollution gradient exists within the Czech Republic such that the northern half is generally more polluted than the southern half with regard not only to SO_2 but also to NO_x and suspended particulate matter (*16*). The Czech Republic's heavy industries (i.e., petrochemicals, synthetic rubber, organics, plastics, detergents, household chemicals, and paints) are clumped in isolated geographic regions that are scattered throughout northern Bohemia (*2*). Northwest Bohemia, in particular, contains substantial industrial brown coal, or lignite deposits (notably the Most and Sokolov basins), which are part of the Black Triangle Region, well-known previously as the most air-polluted area in central Europe (*16*). Over 75% of the former Czechoslovakia's brown coal was mined in northwest Bohemia, and more than 50% of the coal mined in this region was burned locally (*16, 31*). Coal mining in this region increased from 14 Mt to more than 100 Mt between 1945 and 1987 (*3, 32*). More than 74 Mt of coal was mined from the Most basin, located in northern Bohemia, alone (Figure 1). During this time period, SO_2 concentrations increased concomitantly (*5*). Following the collapse of the former regime in 1989, SO_2 emissions decreased from 999 to 452 kt yr^{-1}; however, Norway spruce (*Picea abies*) forests continue to decline (*5, 32*). As of 1990, 100 000 ha (59%) of Norway spruce forests had died or were in the process of decline, and projections are for 64−70% of the total forested area to be affected by the year 2000 (*3*). The prevailing hypothesis for the major cause of forest decline is a direct effect of SO_2 (*5, 14, 33*).

With regard to SO_2, air pollution is greater in northwestern Bohemia than in northeastern Bohemia, and both are more air-polluted than southern Bohemia (*16*). The prevailing westerly winds also transport atmospheric pollutants originating from heavily industrialized regions in both the former East Germany and southern Poland, further aggravating air quality conditions in northwest Bohemia. According to monitoring data, of the total amount of SO_2 deposited, between 36 and 59% originates outside the Czech Republic's borders (*32*). Moreover, in 1985, the former Czechoslovakia imported significantly more sulfur and heavy metals than it exported throughout Europe (*34*). We wanted to determine if broad scale spatial patterns of atmospheric Pb deposition are consistent with the existing air pollution gradient. Specifically, we wanted to (1) determine historical rates of Pb deposition throughout the Czech Republic and to (2) determine sources of atmospheric Pb deposition using AIA and magnetic susceptibility as interpretive tools. Due to prevailing wind patterns and geographical positioning, we classified the eight *Sphagnum*-dominated peatland sites into three groups: the western group, which consists of Tajga, Boží Dar Bog, and Pod Jelení horou; the northern group, which consists of Bílá Smědá, Pančavská louka, and Malé mechové jezírko; and the southern group, which consists of Červené blato and Mrtvý luh.

Peat bulk density values for individual core depths ranged from 0.02 to 0.18 g cm^{-3}. Bulk density, averaged over all depth sections, differed among the eight sites ($p \leq 0.0001$; Table 1). High bulk densities for Tajga, Červené blato, and Mrtvý luh can be attributed to an influence of sedges and grasses in the peat at these sites, whereas at the other sites, the peat appeared to be exclusively *Sphagnum*-derived. Bulk density values are typical for *Sphagnum*-derived peat (cf. refs *35*−*37*).

Organic matter concentrations for individual core depths ranged from 73.6% to 98.8%, values that are typical of *Sphagnum*-derived peat (*35, 37*). Organic matter concentrations averaged over all depths differed among the eight sites ($p \leq 0.0001$, Table 1). Peat from four of the sites situated in the northern and western parts of the Czech Republic (Pod Jelení horou, Boží Dar Bog, Bílá Smědá, Pančavská louka) had somewhat lower mean organic matter concentrations than peat from the other four sites (Table 1). Both bulk density and organic matter concentrations generally decreased as pollution intensity increased from the southern Czech Republic heading north.

Lead concentrations for individual peat core depths ranged from 3 to 479 $\mu g\ g^{-1}$ (Table 2). Averaged across all depths, Pb concentrations differed among the eight sites ($p \leq 0.0001$, Table 1). The western and northern sites generally had the higher Pb concentrations than the southern sites (Tables 1 and 2), a pattern consistent with the known northwest/southeast pollution gradient.

Numerous peat profiles with Pb concentration data have been described for sites in both Europe and North America. For ombrotrophic or weakly minerotrophic bogs believed to have been largely isolated from anthropogenically elevated atmospheric Pb deposition, Pb concentrations in individual samples from a peat profile rarely exceed 150 $\mu g\ g^{-1}$ and commonly are much lower (e.g., refs *35* and *38*−*43*). However, when situated downwind of major industrialized areas, peat deposits may exhibit considerably elevated Pb concentrations. In the United States, for example, at Spruce Flats Bog in Pennsylvania (*19*) and at Cowles Bog in northern Indiana (*44*), maximum peat Pb concentrations were 179 and 199 $\mu g\ g^{-1}$, respectively, apparently reflecting Pb inputs via prevailing winds coming from the heavily industrialized Ohio River valley. Similarly, in the United Kingdom, 15 peat bogs were categorized as either "low background" (Pb concentrations of 32−155 $\mu g\ g^{-1}$) or "high background" (Pb concentrations of 481−800 $\mu g\ g^{-1}$) sites (*39*). Peat Pb concentrations increased as a function of the size of the human population of the

TABLE 1. Physical and Chemical Properties of Peat from the Eight Study Sites[a]

	site							
	Červené blato	Mrtvý luh	Tajga	Malé mechové jezírko	Boží Dar	Pod Jelení horou	Bílá Smědá	Pančavská louka
location	48°52′N 13°47′E	48°52′N 13°54′E	50°00′N 12°42′E	50°14′N	50°24′N 12°54′E	50°31N 13°10′E	50°50′N 15°18′E	50°46′N 15°30′E
elevation	440	740	940	780	980	890	980	1350
surface water pH	3.43	nd	3.69	3.45	4.21	3.73	5.37	4.90
bulk density	0.114[a] ±0.016	0.114[a] ±0.007	0.126[a] ±0.005	0.091[c,d] ±0.007	0.096[b] ±0.009	0.035[e] ±0.002	0.099[b,c] ±0.007	0.073[d] ±0.005
organic matter	96.5[a] ±0.5	95.6[a,b] ±0.7	95.3[a,b] ±0.3	93.9[a–c] ±1.2	90.8[c,d] ±1.9	91.6[c] ±1.0	81.1[e] ±1.3	88.6[d] ±1.3
Pb concentration	34.2[e] ±4.8	40.7[d,e] ±6.5	89.9[b,c] ±7.1	125.4[b,c] ±15.0	134.4[b] ±36.1	85.1[c,d] ±10.4	208.2[a] ±28.7	105.4[b,c] ±11.8
acid-insoluble ash	19.9[d] 4.3	21.0[d] ±6.1	33.6[c,d] ±2.8	55.9[a–c] ±9.1	24.7[d] ±7.9	59.5[a,b] ±10.0	40.1[b–d] ±7.2	72.7[a] ±12.9
magnetic susceptibility	0.00[b] ±0.0	2.2[b] ±1.0	1.0[b] ±1.5	0.1[b] ±0.1	2.2b[b] ±0.6	2.0[b] ±1.6	15.3[a] ±5.4	0.0[b] ±0.0

[a] Values represent means ± standard errors averaged over all depth sections in each core. Different lower case letters indicate significant differences (ANOVA, randomized complete block design with depth as the blocked effect; $p < 0.05$, Duncan's Multiple Range Test for *a posteriori* comparisons). Elevation is in units of meters above sea level; Bulk density is in units of g cm^{-3}, organic matter is percent of total dry mass, Pb concentration is in units of μg g^{-1}, AIA concentration is in units of mg g^{-1}, and magnetic susceptibility is $\times 10^{-3}$ cgs kg^{-1}.

TABLE 2. Pb Concentrations in μg g^{-1} for Each 2 cm Section[a]

	site							
depth	Boží Dar	Bílá Smědá	Malé mechové jezírko	Pančavská Louka	Červené blato	Mrtvý luh	Tajga	Pod Jelení horou
0–2	11.38	55.58	55.93	21.75	14.45	12.88	88.90	17.75
2–4	27.79	70.88	79.48	64.05	21.05	22.05	110.85	33.45
4–6	41.64	100.48	132.43	92.45	28.55	42.55	109.90	66.45
6–8	93.24	115.48	184.98	93.75	29.20	80.20	102.40	83.90
8–10	171.4	167.13	181.83	154.95	32.95	73.25	128.80	95.90
10–12	478.9	239.03	188.43	136.95	66.60	70.25	109.30	124.95
12–14	233.7	290.13	159.43	77.25	67.15	43.10	92.05	120.05
14–16	138.9	312.68	171.18	88.85	41.25	37.20	87.08	139.60
16–18	140.3	297.48	135.48	106.90	31.75	31.90	71.30	107.90
18–20	108.6	313.88	99.33	161.60	31.15	32.70	73.93	75.90
20–22	90.49	265.58	82.53	137.60	24.90	23.47	66.00	76.80
22–24	78.73	269.63	33.68	128.20	20.85	18.25	39.25	78.80
24–26		249.68	70.48	109.95	17.05	21.55	27.48	
26–28		240.53	58.78	109.40	6.45	26.15	25.08	
28–30			32.38	83.60	5.45		21.95	
30–32			36.93	83.80	2.65		27.23	
32–34			37.73		4.30			

[a] Each peat core was variable in length as indicated by a blank.

largest town within 30 km of each site, suggesting an anthropogenic influence (*39*). In this sense, the substantially lower Pb concentrations in peat from the two southern bogs, Červené blato and Mrtvý luh, than in peat from the northern and western bogs (Tables 1 and 2) is consistent with an anthropogenic Pb deposition gradient, trending north to south within the Czech Republic. However, Pb concentration profiles must be regarded as a crude proxy for Pb deposition. Between-site differences in net *Sphagnum* primary production as well as depth-within-site differences in peat decomposition can influence peat Pb concentration profiles to an extent that they may not accurately reflect site differences or within-site past historical patterns in atmospheric Pb deposition.

To convert Pb concentration profiles in peat to historical Pb deposition profiles, we used peat bulk densities along with peat age/depth relationships, determined from ^{210}Pb-dating of the cores from the eight sites. Total unsupported ^{210}Pb inventories and estimated ^{210}Pb deposition values for the eight cores as well as surface unsupported ^{210}Pb concentrations (Table 3) were similar to values that have been reported for other ^{210}Pb-dated peat cores (e.g., refs *19*, *42*, and *45–49*).

TABLE 3. Unsupported ^{210}Pb Inventories, Surface Peat Concentrations, and Atmospheric Deposition Rates for Eight Peat Cores in the Czech Republic

site	total unsupported ^{210}Pb content (pCi cm^{-2})	unsupported ^{210}Pb concn at surf. (pCi g^{-1})	^{210}Pb supply rate (pCi cm^{-2} yr^{-1})
Bílá Smědá	22.941	7.551	0.704
Boží Dar Bog	11.907	9.560	0.366
Červené blato	18.935	8.974	0.581
Malé mechové J.	31.735	16.126	0.974
Mrtvý luh	12.597	7.970	0.387
Pančavská louka	18.040	10.506	0.554
Pod Jelení horou	11.352	10.898	0.349
Tajga	9.908	9.417	0.304

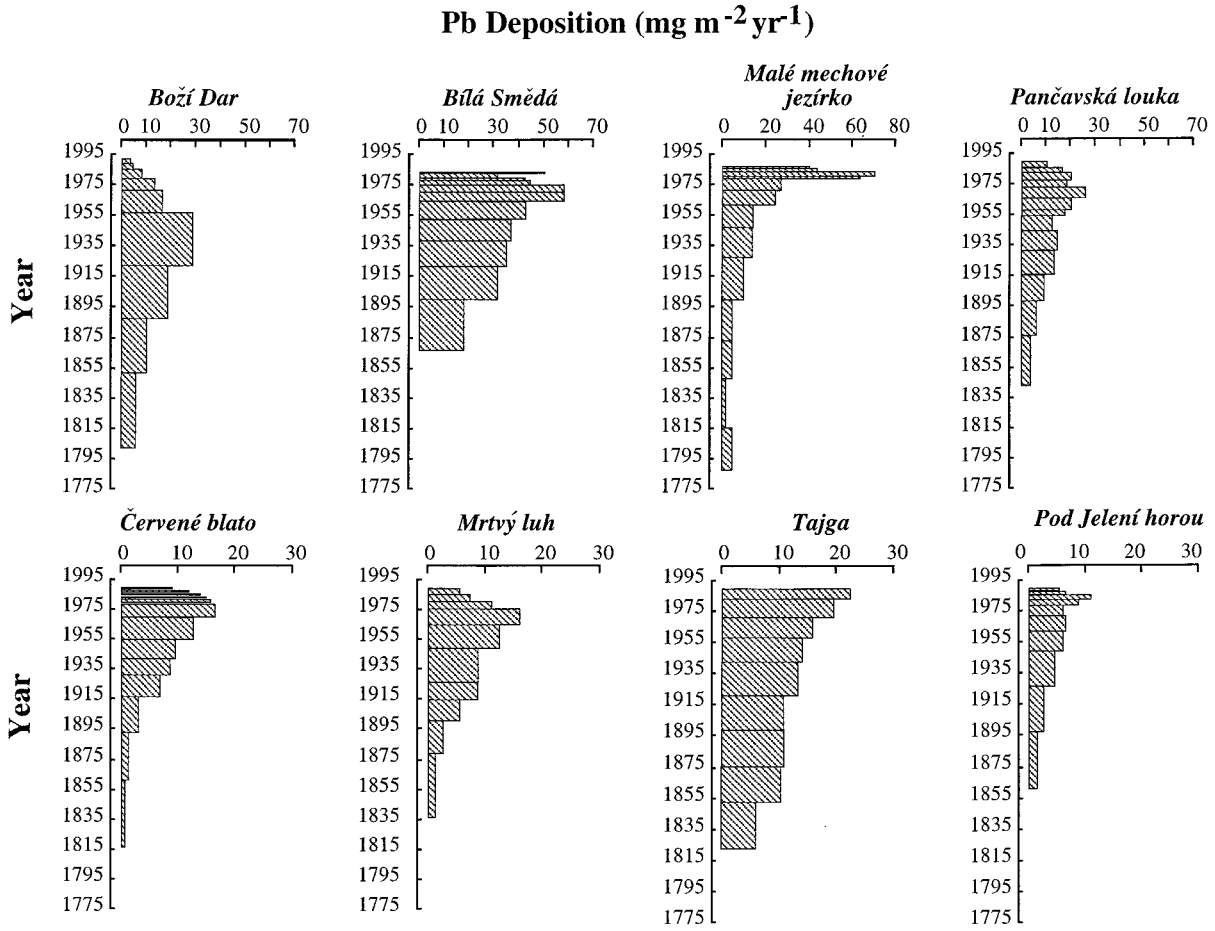

FIGURE 3. Lead deposition as a function of age for eight sites in the Czech Republic. Each horizontal bar represents a 2-cm depth section; depth sections are different widths to reflect variation in net peat accumulation over different time periods.

While Pb concentration profiles exhibit subsurface maxima (Table 2), Pb deposition profiles exhibit peaks closer to the surface (Figure 3), illustrating that Pb concentration data in peat provide a poor proxy for historical Pb deposition rates. Generally, peak Pb deposition (Figure 3) was greater at the northern sites (27, 59, and 71 mg m^{-2} yr^{-1} at Pančavská louka, Bílá Smědá, and Malé mechové jezirko, respectively) than at either the western (10, 23, and 29 mg m^{-2} yr^{-1} at Pod Jelení horou, Tajga, and Boží Dar bog, respectively) or southern sites (16 and 17 mg m^{-2} yr^{-1} at Červené blato and Mrtvý luh, respectively). The peak Pb deposition values for the northern sites represent some of the highest recent (post-Industrial Revolution) rates in the world (*17, 19, 39, 41, 42, 48, 50*).

Our data are constrained to the past 150−200 years because of the limitations of ^{210}Pb-dating. Within this time period, minimum Pb deposition was estimated for time periods represented by the deepest (or second deepest for Malé mechové jezirko) dateable peat sections. As with peak Pb deposition, minimum Pb deposition (Figure 3) generally was somewhat greater at the northern sites (2, 4, and 19 mg m^{-2} yr^{-1} at Malé mechové jezirko, Pančavská louka, and Bílá Smědá, respectively) than at either the western (2, 6, and 6 mg m^{-2} yr^{-1} at Pod Jelení horou, Tajga, and Boží Dar Bog, respectively) or southern sites (1 and 2 mg m^{-2} yr^{-1} at Červené blato and Mrtvý luh, respectively). Using ^{210}Pb-dating of a Swiss peat bog, Shotyk et al. (*17*) reported peak Pb deposition of 16 mg m^{-2} yr^{-1} (occurring in 1979), as contrasted with a preindustrialization background value of 1.3 mg m^{-2} yr^{-1}. Even this preindustrialization value, however, was much greater than what could be considered as a natural background value of 0.01 mg m^{-2} yr^{-1}, estimated for the period from 8030 to 5320 ^{14}C yr BP (*17*), reflecting a long-term history of Pb exploitation in Europe that predates the Industrial Revolution.

All sites except Boží Dar Bog exhibited peak Pb deposition between 1965 and 1992, a period that corresponds to maximum burning of lignite (Figure 1) (*3, 51*). Burning of lignite coal, especially in the Krušné hory Mountain region, began in the mid-1860s and sharply increased from 1963 to 1977, with maximum activity around 1988 (Figure 1) (*31, 33*).

At the Czech Hydrometeorological Institute's Košetice Observatory, current measured Pb deposition is greater in late fall and winter than in spring and summer (*52*). Elevated Pb deposition in winter is most likely attributable to burning of coal for domestic heating. The average Pb concentration of Czech lignite is 134 μg g^{-1} (*53*). Although coal combustion accounts for only a small proportion of total global anthropogenic Pb emissions, it occurs at over 50% of the rate of release by natural sources, its significance being masked only by the rate of leaded gasoline usage (*9, 53*). In regions such as the Czech Republic, where automobile numbers are low relative to western counterparts, yet heavily industrialized, Pb emissions via gasoline usage can be much lower than Pb emissions via coal combustion. In the Czech Republic, coal power plants produce 76% (i.e., 61 MWh) of the current Czech power production; these power plants are concentrated mainly in the brown coal basin of northern Bohemia (*16*).

At Boží Dar Bog, peak Pb deposition occurred between 1916 and 1954. The area in the vicinity of Boží Dar Bog contains several types of ore deposits (*55, 56*). While there is no active mining in the region today, many of these ore deposits have been exploited sporadically over the past 200 years. A Pb/Zn/Cu mine is located 1 km from Boží Dar Bog,

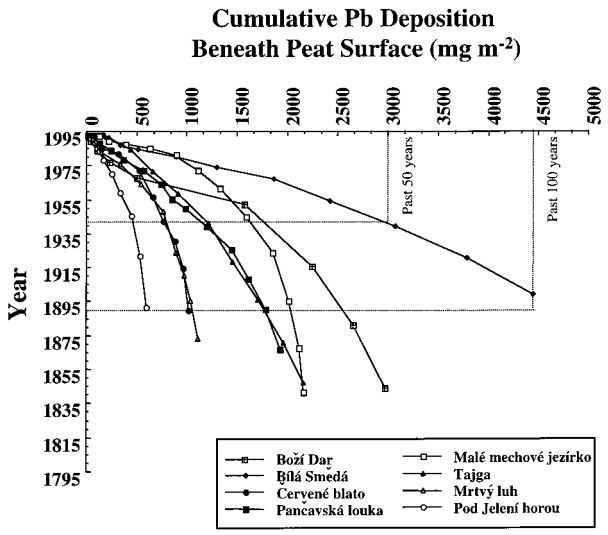

FIGURE 4. Cumulative Pb deposition (mg m^{-2}) beneath the peat surface as a function of year.

and small veins of galena (PbS) are known to have been mined by hand in the late 19th century and early 20th century (*55, 56*). Exploitation of these deposits may have resulted in maximum annual Pb deposition rates between 1916 and 1954 at Boží Dar Bog rather than after 1965 as is the case for the other seven sites. Lead concentration in Boží Dar peat during this same time interval approached 500 μg g^{-1} (Table 2).

Across all sites, the most recently deposited peats, i.e., the upper few 2-cm sections, indicate recently decreasing Pb deposition. Nonetheless, Pb deposition rates calculated from the top 2-cm peat core sections continue to be somewhat greater in the northern part of the Czech Republic (11, 40, and 51 mg m^{-2} yr^{-1} at Pančavská louka, Malé mechové jezirko, and Bílá Smědá, respectively) than in either the western (4, 6, and 23 mg m^{-2} yr^{-1} at Boží Dar Bog, Pod Jelení horou, and Tajga, respectively) or southern (6 and 9 mg m^{-2} yr^{-1} at Mrtvý luh and Červené blato, respectively) parts of the Czech Republic (Figure 3). In 1993, the Czech Hydrometerological Institute reported annual wet Pb deposition values of 5−17 mg m^{-2} (*57*); in 1994−1995, Pb deposition of 2 mg m^{-2} yr^{-1} in bulk precipitation was reported for the Krušné hory mountain region (*16*). Our estimates of recent Pb deposition are similar to values obtained by direct measurement for five of the eight sites. However, higher values for three of the sites suggest localized areas of elevated deposition, especially in the northern and western parts of the country. Since we collected our peat cores, Pb deposition in the Czech Republic has continued to decrease, in part because of the introduction of new Czech cars using unleaded gasoline in 1993 (*58*). Annual average atmospheric Pb concentration determined by the Czech Hydrometerological Institute in 1992 (the year prior to the introduction of new automobiles, requiring unleaded gasoline) was 250 ng m^{-3} as compared to 1997 concentrations of 150 ng m^{-3} (*57*). Additionally, over the past decade, Pb mining in the Czech Republic has crashed. Mine Pb output decreased from 4.6 kt in 1989 to only 1.1 kt in 1992 (*59*). In 1993 and 1994 mining of ore deposits for Pb was discontinued in the Czech Republic (*60*).

Biotic Pb Exposure

Multiplying the amount of Pb deposited in each depth interval by the number of years represented by each depth interval and summing values over the entire peat profile reveal the total amount of Pb accumulated, per m^2 of peat surface area, over time (Figure 4). Generally, the northern and western sites, and in particular Boží Dar Bog and Bílá Smědá, have had greater accumulation of Pb over the past 50−100 years than the southern sites. Assuming cumulative Pb deposition over time can be used as a rough index of biotic exposure, a 50-year old organism that has lived in the towns of either Boží Dar or near Bílá Smědá since birth has been exposed to two or four times more Pb, respectively, than a 50-year old organism that has lived in Červené blato (Pb accumulation in peat: 1.6 or 3.1 g m^{-2} vs 0.8 g m^{-2}, respectively (Figure 4)). Similarly, a 100-year old organism living in either Boží Dar or near Bílá Smědá has been exposed to nine or 15 times more Pb, respectively, than a 100-year organism living near Mrtvý luh (2.7 or 4.4 vs 0.3 g m^{-2}, respectively).

Given the Czech propensity for family gardens and a fondness for rootcrops and mushrooming, biotic exposure of Pb in already highly polluted areas could be magnified through these activities. Lead concentration of fruiting bodies of edible mushrooms in the vicinity of a Pb smelter in central Bohemia was approximately 100 μg g^{-1} dry mass (*61*). Wild mushrooms are a popular delicacy in the Czech Republic, with normal intake at one meal ranging from 300 to 500 g of fresh mushrooms (approximately 30−50 g dry matter; (*61*)). Given the maximum Pb daily intake rate for a 60 kg individual of 460 μg d^{-1}, the acceptable intake of Pb could be exceeded in one meal prepared from mushrooms (*3, 61*). Lead exposure is one of many factors that could contribute to the poor health and low life expectancy (64.2 years for men in the Krušné hory region (*3, 62*)), especially in the most polluted regions of the Czech Republic.

Sources of Pb Deposition

Regressing Pb content against AIA content for each depth section within a peat core, along with peat magnetic susceptibility determinations and an understanding of local/regional disturbance history (notably mining activity that could enhance Pb deposition), may allow for the differentiation between background Pb deposited with soil-derived particles, Pb derived from fossil fuel combustion, gasoline-derived Pb, and particulate matter from dust generated by local mining activities (cf. ref *16*). In a plot of Pb content versus AIA content for a particular site, when all individual data points (corresponding to 2 cm depth intervals in this study) fall on or near the regression line (high R^2 value), the slope can be viewed as characterizing either regional background Pb deposition or a history of anthropogenically influenced Pb deposition from a source whose Pb/AIA ratio has remained constant over the time range spanned by the peat core. However, for sites that have received temporally discontinuous enhanced Pb deposition from anthropogenic sources (e.g., combustion of fossil fuels, including coal combustion, smelting, steel production; release of mining related dust; release of Pb from gasoline combustion), in a Pb/AIA plot, some plotted points should be situated well above the regional background regression line. These points should correspond to depth sections that are enriched in Pb relative to background values, reflecting specific time periods of enhanced Pb deposition from anthropogenic sources. If peat sections that are enriched in Pb also have measurable magnetic susceptibility, Pb deposition from fossil fuel combustion would be implicated. If peat sections that are enriched in Pb correspond to time periods of known local metal mining activity, Pb deposition from mining-derived dust would be implicated. Another potential source of enhanced Pb deposition is leaded gasoline, which was introduced into Czechoslovakia in 1920s. If inputs of Pb derived from leaded gasoline were substantial at any individual site, a steeper slope of the Pb/AIA regressions for those depth sections more recent than 1922 than for depth sections older than 1922 would be expected.

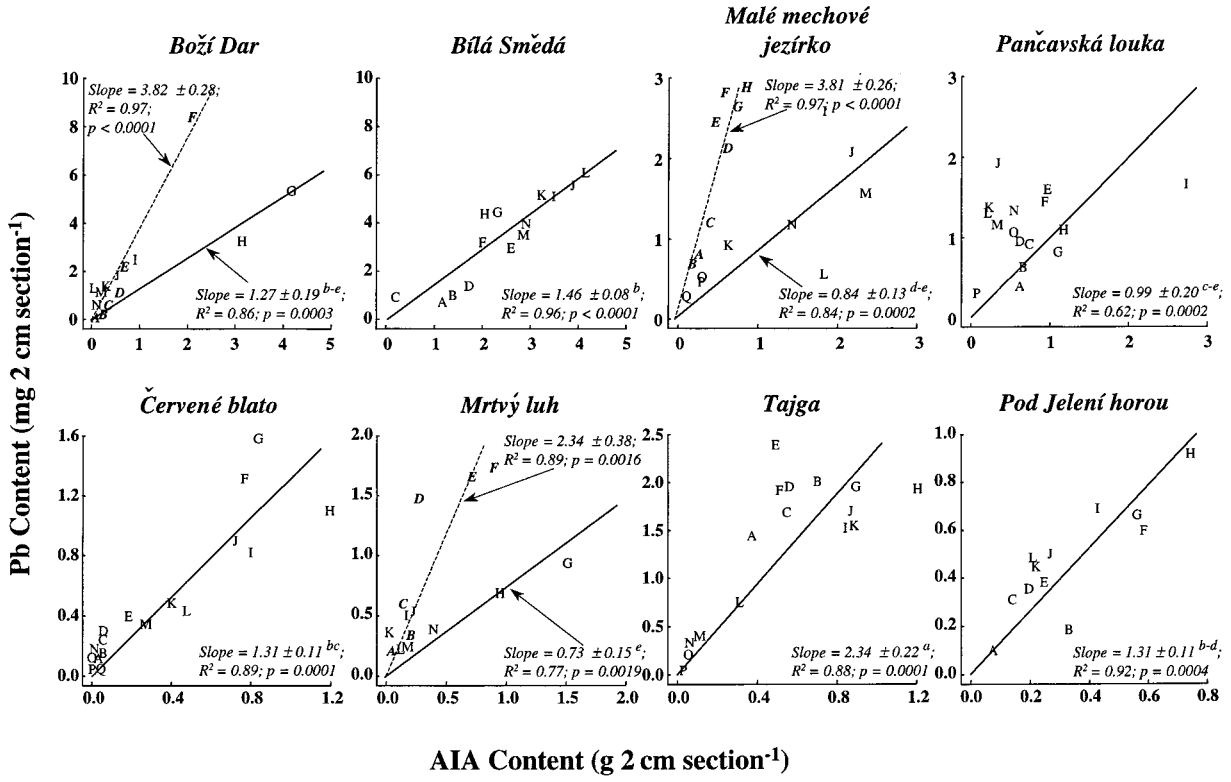

FIGURE 5. Pb content as a function of AIA content for 2-cm depth intervals for all eight sites. Data points are plotted as letters that correspond to individual depth sections with the letter A representing the youngest section. The two regression lines for Boží Dar Bog, Malé mechové jezírko, and Mrtvý luh represent the preleaded-gasoline era (1922 and older; solid line; italicized letters) and post-gasoline era (1923 and younger; dashed line). The remaining five sites are represented by overall regressions as they did not have significantly different pre-1922 and post-1922 slopes. For each regression, the slope ± standard error, R^2 value, and *p* value are given. Significantly different slopes (a posteriori Tukey's multiple comparisons of slopes; Zar, J. H. *Biostatistical Analysis*, 3rd ed.; Prentice-Hall: Upper Saddle River, NJ, 1996; pp 365−366) are represented by different lower case letters.

Using an analysis of covariance (test for homogeneity of slopes, regression lines restricted to pass through the origin (*63*)), only three sites had significantly different pre-1922 and post-1922 slopes: Boží Dar ($p \leq 0.0001$), Mrtvý luh, ($p = 0.0066$), and Malé mechové jezírko ($p = 0.0053$). In each of these three cases, the post-1922 slope was significantly greater than the pre-1922 slope (Figure 5), a result that would be consistent with leaded gasoline as a contributing source of Pb deposition at these three sites. The lack of significance between pre-1922 and post-1922 slopes for the remaining five sites suggests that leaded gasoline was not a major contributing source of Pb deposition at these sites (Figure 5). Analysis of covariance also indicated between-site differences in the slopes of the pre-1922 regression lines for these three sites and the overall regression lines for the remaining five sites ($p = 0.0024$) (Figure 5).

Differences among the eight sites with regard to the major sources of Pb deposition over the past 150−200 years are suggested from mean Pb and AIA concentrations (Table 1). The northern sites had the highest mean concentrations of both Pb and AIA, followed by the western sites, with the southern sites exhibiting the lowest mean concentrations of Pb and AIA (Table 1). These patterns are consistent with the previously established pollution gradient throughout the Czech Republic.

The two southern sites, Červené blato and Mrtvý luh, both situated at the cleaner end of the pollution gradient, differ with respect to the major source of Pb deposition. There is no evidence to suggest that Červené blato has been influenced by Pb from either gasoline (no separation into pre-1922 and post-1922 Pb/AIA regressions; Figure 5) or combustion of fossil fuels (absence of magnetic susceptibility in any peat section; Figure 6). For Mrtvý luh, there is a difference between

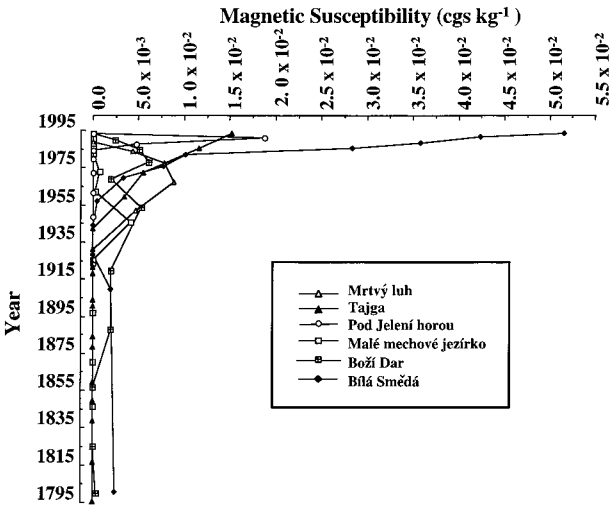

FIGURE 6. Magnetic susceptibility as a function of year for all six of the eight sites. None of the peat sections from the Červené blato or Pančavská louka cores had measurable magnetic susceptibility.

the pre-1922 and post-1922 Pb/AIA regression lines (Figure 5), but this difference may not represent a signal from leaded gasoline usage, especially considering the lack of such a signal at Červené blato. Measurable magnetic susceptibility in the Mrtvý luh peat core (Figure 6) occurs only in depth sections C−F (4−6 to 10−12 cm), i.e., those that contribute strongly to the separation of the post-1922 Pb/AIA regression from the pre-1922 regression (Figure 5). There is no evidence of metal mining activity within the vicinity of either Červené blato or Mrtvý luh within the time period spanned by the

peat cores. Therefore, the major source of anthropogenic Pb deposition at Mrtvy luh appears to be related to fossil fuel combustion, especially from 1924 to 1982. We also note that although Pb deposition has been low at both Červené blato and Mrtvý luh (Figures 3 and 4), the slope of the Pb/AIA regression line for Červené blato is significantly greater than the pre-1922 slope for Mrtvý luh (Figure 5), suggesting site differences in recent "background" Pb sources.

For the northern group of sites (Bílá Smědá, Pančavská Louka, and Malé mechové jezírko), different Pb/AIA relationships (Figure 5) suggest different sources of Pb deposition. At Bílá Smědá, Pb deposition has been high historically (Figures 3 and 4). The tight Pb/AIA regression line for Bílá Smědá probably reflects a constant anthropogenic Pb source over the time period spanned by the peat core. Considerable magnetic susceptibility of the peat, especially in the upper 14 cm of the core, indicates combustion of coal as a source of Pb deposition for at least the past 50 years (Figure 6). A brown coal/lignite deposit is located approximately 25 km west of the site (*64*). Pančavská louka exhibits the weakest relationship between Pb and AIA (lowest R^2 value of the eight sites), suggesting temporal variability in the sources of Pb deposition. Peat from Pančavská Louka contains no magnetic susceptibility, and there was no evidence for increased Pb deposition after 1922, implicating neither fossil fuel combustion nor leaded gasoline as major sources of Pb. However, Pančavská louka is located downwind from the Freiburg Ag mining district in Germany, one of the largest Pb polluters of the mining industries in Europe (*64*). The scatter surrounding the Pb/AIA regression for this site may be attributable, at least in part, to variability in Pb input from past sporadic mining.

Pančavská louka and Bílá Smědá are located in the same geographical region, but differ considerably with regard to both Pb deposition and major sources of Pb input. Short-range transport may play a greater role than long-range transport of polluted air masses at these two sites. Pančavská louka is located in the Krkonoše Mountains, which claim the highest elevation in the Czech Republic. At Bílá Smědá (elevation of 980 m asl), the considerable deposition of Pb from coal combustion is likely in the form of large fly ash particles, which may be removed from the atmosphere before reaching the high elevations at Pančavská louka (1350 m asl). This could explain why Pb deposition is greater at Bílá Smědá than at Pančavská louka. Major wind patterns are predominantly west to east; Bílá Smědá is located northwest of Pančavská louka. Pollution-related combustion products released around Bílá Smědá may be more likely transported toward Poland than toward Pančavská louka.

Malé mechové jezírko is the most easterly located of the northern sites; Pb deposition is intermediate between that at Pančavská louka and Bílá Smědá (Figure 4). Low peat magnetic susceptibility throughout the peat core indicates that combustion products have not contributed substantial quantities of Pb (Figure 6). Statistically separate Pb/AIA regressions for the pre-1922 and post-1992 periods (Figure 5) are more likely attributable to nearby mining activity than to leaded gasoline inputs. Within 50 km of Malé mechové jezírko, there are four polymetallic (Cu−Zn−Pb and Pb−Zn) deposits. Within 10 km south of Malé mechové jezírko is the Zlaté Hory deposit, which was opened in 1965 and was still being mined at the time of core collection (*64*). Also within 50 km south of the site are Horní Benesov and Horni Mesto Pb/Zn deposits, which have been recently mined (M. Novák, personal communication). Elevated Pb deposition at Malé mechové jezírko since 1965 (top six peat sections) is clearly evident (Figure 3); these peat sections have substantially enriched Pb relative to AIA contents (Figure 5).

Of the western sites (Boží Dar Bog, Pod jelení horou, Tajga), greatest cumulative Pb deposition is at Boží Dar Bog (Figure 4). Peat magnetic susceptibility (Figure 6) indicates a long history of fossil fuel combustion at this site. Although there are significantly different pre-1922 and post-1922 Pb/AIA regression lines, this difference does not suggest an influence of leaded gasoline, as the upper 8 cm of peat (since 1952) do not exhibit Pb enrichment (Figure 5). The steeper post-1922 regression line more likely is related to past mining activity. The peak in Pb deposition at Boží Dar Bog between 1916 and 1952 may be related to operation of a Pb/Zn/Cu mine located 1 km from Boží Dar Bog (*55*, *56*) (Figure 5; depth section F).

Despite its location in the mountainous regions of the western Czech Republic, Pod Jelení horou has the lowest cumulative Pb deposition of the eight sites, at least over the time period spanned by the peat core (Figure 4). There is no evidence of Pb inputs from leaded gasoline (Figure 5), and magnetic susceptibility is evident only in the upper 6 cm of peat (most recent 9 years) (Figure 6). The slope of the Pb/AIA regression for Pod Jelení horou is similar to that for the pre-1922 regression for Boží Dar Bog (Figure 5), which we have interpreted as reflecting a period of relatively low activity of metal mining in the western Czech Republic.

Plots of Pb per section versus AIA per section for Tajga show that the points corresponding to post-1896 ages (top six sections; letters A−F; Figure 5) are displaced above the regression lines, indicating a long history of inputs that are enriched in Pb. Peat magnetic susceptibility (Figure 6) indicates that coal combustion is most likely responsible for increasing Pb deposition at this site since 1958. Prior to 1958, however magnetic susceptibility is undetectable in peat, suggesting that coal combustion may not have been a major Pb source during this time period (Figures 5 and 6).

In conclusion, Pb deposition has been spatially and temporally variable in the Czech Republic. Consistent with the well documented general air pollution gradient from the northwestern to the southeastern Czech Republic, we documented generally lower rates of Pb deposition in the southern peat bog sites (Červené blato and Mrtvý luh) than in either western (Boží Dar Bog, Pod Jelení horou, and Tajga) or northern sites (Bílá Smědá, Pančavská louka, and Malé mechové jezírko). Lead deposition patterns at these eight sites generally reflect a pattern of increasing industrialization over the past 100−200 years, with evidence of a more rapid industrialization in the post-World War II era. Using Pb/AIA regressions and peat magnetic susceptibility determinations, we were able to identify periods of enhanced Pb deposition that could be ascribed to fossil fuel combustion (Mrtvý luh, Bílá Smědá, Boží Dar Bog, Pod Jelení horou, Tajga) and/or to mining of Pb-containing ore deposits (Pančavská louka, and Malé mechové jezírko, Boží Dar Bog). Although at three sites (Boží Dar Bog, Malé mechové jezírko, Mrtvý luh), a steeper post-1922 than pre-1922 Pb/AIA regression slope would be consistent with elevated Pb inputs from leaded gasoline, at each site there was strong evidence of enhanced post-1922 Pb deposition from fossil fuel combustion and/or local mining. Despite the usefulness of magnetic susceptibility and Pb/AIA regressions in interpreting past patterns of Pb deposition, stable Pb isotopic analyses are probably the only reliable way to isolate the influence of Pb deposition from leaded gasoline, especially in regions such as the Czech Republic where Pb deposition from fossil fuel combustion and from mining can be considerable.

The recent introduction of unleaded gasoline (as of October 1993 all new cars and all imported cars must have a catalyzer), along with declining Pb mining production, apparently are contributing to an ongoing reduction in Pb deposition throughout the Czech Republic. However, it will most likely take some time before rates of Pb deposition will reach pre-industrialization values. Prior to Czech industrialization, Pb deposition rates were similar to those recently measured in the eastern United States of 4.1 mg m^{-2} yr^{-1}

(*65*); yet in the recent past we have documented Pb deposition rates at locations within the Czech Republic that are as much as 20 times greater.

Under the former political regime, eastern and central Europe experienced considerable environmental degradation that has been poorly documented (*13*, *33*). For example, the oldest existing atmospheric Pb deposition data were collected as late as 1984, and these data were not included in the Czech Hydrometeorological reports until 10 years later in 1994 (Fiala, personal communication). The magnitude of historical atmospheric Pb deposition as well as of other pollutants in the Czech Republic (*66*), until recently, has been unknown. As a result, in the Czech Republic, historical reconstructions represent one of the only means by which heavy metal deposition in the past can be determined. Results from this study represent the only existing atmospherically Pb deposition data from the communist era and place current high rates of Pb deposition within a historical context as well as indicate the magnitude of pollution in the former Czechoslovakia.

Acknowledgments

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