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Key Points:
- There is effectively zero atmospheric Pb contamination in this region today
- Atmospheric Pb contamination has been in decline for decades
- Bogs are excellent archives of atmospheric Pb deposition

Supporting Information:
- Supporting Information S1
- Table S3

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Peat bogs in northern Alberta, Canada reveal decades of declining atmospheric Pb contamination

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Abstract Peat cores were collected from six bogs in northern Alberta to reconstruct changes in the atmospheric deposition of Pb, a valuable tracer of human activities. In each profile, the maximum Pb enrichment is found well below the surface. Radiometric age dating using three independent approaches (^{14}C measurements of plant macrofossils combined with the atmospheric bomb pulse curve, plus ^{210}Pb confirmed using the fallout radionuclides ^{137}Cs and ^{241}Am) showed that Pb contamination has been in decline for decades. Today, the surface layers of these bogs are comparable in composition to the "cleanest" peat samples ever found in the Northern Hemisphere, from a Swiss bog ~ 6000 to 9000 years old. The lack of contemporary Pb contamination in the Alberta bogs is testimony to successful international efforts of the past decades to reduce anthropogenic emissions of this potentially toxic metal to the atmosphere.

1. Introduction

Human activities have had a greater impact on the geochemical cycle of Pb than that of any other potentially toxic heavy metal [*Nriagu*, 1978]. Atmospheric Pb contamination is a global phenomenon [*Ng and Patterson*, 1981; *Boutron et al.,* 1994], the result of mining, smelting, refining, and metallurgical processing of Pb and other metal sulphides, in addition to the combustion of coal and other fossil fuels [*Shotyk and Le Roux*, 2005]. During the twentieth century, gasoline Pb additives were the largest single source of anthropogenic Pb to the atmosphere [*Wu and Boyle*, 1997], but Arctic ice cores provide remarkable records of hemispheric Pb contamination extending back in time more than three millennia [*Murozumi et al.,* 1969; *Rosman et al.,* 1997; *Zheng et al.,* 2007]. An ice core from Devon Island, Nunavut, Canada, for example, has provided a 15,000 year record of atmospheric Pb deposition, with evidence of Pb contamination dating back to the time of the Phoenicians, followed by the Greek and then Roman civilizations, with notable episodes of intense Pb emissions during the Medieval Period from silver mining in central Europe, then the Industrial Revolution, and later, from the introduction of leaded gasoline [*Zheng et al.,* 2007]. The use of gasoline Pb additives, first introduced in the U.S., in 1923 [*Nriagu*, 1990], peaked in 1970, when global production reached 250,000 tons per year [*Wu and Boyle*, 1997] and air Pb has been in decline ever since. High-resolution reconstructions of Pb concentrations [*McConnell et al.,* 2002] and stable Pb isotopes [*Shotyk et al.,* 2005] of polar ice representing the 19th and 20th centuries show that the elimination of leaded gasoline played a very important role in the reduction of the global Pb problem, but that other technological changes and improvements were also very important, given that Pb was emitted to the environment from many other sources. Lead in the atmosphere, therefore, is derived from a complex mixture of natural and anthropogenic sources varying in time and space. One of the challenges of environmental geochemistry is to develop ways and means of reconstructing the changing sources of atmospheric Pb and their rates of deposition to terrestrial and aquatic ecosystems [*Bindler et al.,* 2008; *Shotyk and Krachler*, 2010].

Ombrotrophic (i.e., rain-fed) peat bogs are also excellent archives of atmospheric Pb, providing the first complete, long-term record (15,000 years) of atmospheric Pb deposition in Europe: there, anthropogenic inputs have dominated continuously for more than 3000 years [*Shotyk et al.,* 1998; *Shotyk et al.,* 2001]. Detailed histories of atmospheric Pb contamination have since been reconstructed using peat cores from bogs across

Europe [*Bränvall et al.,* 2001; *Klaminder et al.,* 2003; *Novák et al.,* 2003; *Monna et al.,* 2004; *Cloy et al.,* 2005, 2008; *Kylander et al.,* 2005; *Le Roux et al.,* 2005; *Bindler et al.,* 2008; *Farmer et al.,* 2015]. With appropriate methods for sample collection, handling, preparation, and age dating [*Givelet et al.,* 2004] peat cores have provided extremely detailed reconstructions of atmospheric Pb pollution histories, from Antiquity to the present day. The Lindow Moss near Manchester, for example, showed evidence of atmospheric Pb contamination beginning around 900 B.C., predating the arrival of the Romans in 50 B.C. and resulting from Iron Age Pb mining in England [*Le Roux et al.,* 2004]. Precisely dated peat cores from Denmark and the Faroe Islands showed that atmospheric Pb contamination began its decline 25 years before the introduction of unleaded gasoline [*Shotyk et al.,* 2003, 2005]. Given that rates of atmospheric Pb deposition obtained from living *Sphagnum* moss collected from bogs agree with data obtained from direct deposition measurements [*Kempter et al.,* 2010], rates of Pb accumulation obtained from peat cores appear to be reasonable reflections of past rates of atmospheric Pb deposition.

In contrast to the detailed reconstructions of atmospheric metal contamination in Europe, much less work has been undertaken in North America. Except for recent studies of atmospheric Hg deposition reconstructed using peat cores from Ontario [*Givelet et al.,* 2003] and Maine [*Roos-Barraclough et al.,* 2006; *Norton et al.,* 2015] there have been few studies of atmospheric Pb using peat cores from ombrotrophic bogs in North America [*Norton et al.,* 1997; *Benoit et al.,* 1998; *Weiss et al.,* 2002; *Kylander et al.,* 2009; *Pratte et al.,* 2013]. Careful age dating of peat cores from three bogs in southern Ontario showed that atmospheric Pb deposition went into decline toward the end of the 1950s [*Givelet et al.,* 2003], two decades before the introduction of unleaded gasoline in Canada, in 1976. Subsequent work on one of those cores using stable Pb isotopes showed that the predominant anthropogenic source of Pb in southern Ontario today is from the smelting and refining of metallic ores in northern Ontario and Quebec [*Shotyk and Krachler*, 2010]. In western Canada, there are no published peat bog reconstructions of atmospheric Pb deposition available for comparison.

Based on a recent survey of heavy metals using snowpack sampling, it was claimed that open pit mining and upgrading of the Athabasca Bituminous Sands (ABS) in northeastern Alberta (AB) is a significant source of atmospheric Pb along with many other chalcophile elements such as Ag, Cd, Sb, and Tl [*Kelly et al.,* 2010]. To test this hypothesis, *Sphagnum* moss was collected at three sites from each of 22 bogs in the vicinity of the ABS and total metal concentrations measured [*Shotyk et al.,* 2014]. *Sphagnum fuscum* is the dominant hummock-forming moss in peat bogs, and as such, it receives metals exclusively from the air [*Crum*, 1988]. This species is found in bogs across North America and Europe and has been used for many decades, particularly in Europe, for monitoring atmospheric heavy metal deposition [*Shotyk et al.,* 2015]. After correcting for differences in the abundance of mineral dust particles using Th, no significant contribution of anthropogenic Pb could be found in the moss samples from the ABS region [*Shotyk et al.,* 2014]. A subsequent study confirmed this result and, further, showed that metal concentrations in these moss samples were comparable to the contemporary "background" values for moss from central and northern Norway [*Shotyk et al.,* 1996a].

While moss samples provide an indication of the extent of contamination in contemporary atmospheric deposition, they provide no indication of the changing rates and sources of contaminants in the past. To fill this knowledge gap, peat cores were collected from five bogs in the vicinity of the open pit mines and bitumen upgraders in the ABS region, as well as a core from a bog 264 km upwind of this increasingly industrialized region. The results obtained from these cores are then compared with peat cores collected from ombrotrophic bogs in Europe to help put the results from the AB bogs into a global perspective.

2. Methods

Peat cores were collected from ombrotrophic (rain-fed) bogs in northern AB (Figure 1 and Figure S1): five sites are in the vicinity of open pit mines and upgrading facilities of ABS, and one site is far removed from this area and serves to provide a regional background signal. We provide the precise location of each coring site along with the distance (in km) from the midpoint to the two central upgraders (supporting information Table S1). Samples and certified reference materials were dissolved and trace metals determined using inductively coupled plasma-mass spectrometry (ICP-MS) in the ultraclean, metal-free SWAMP laboratory (supporting information Table S2). The paleobotanical stratigraphy of each site was reconstructed (supporting information Figure S2) and selected plant macrofossils age dated using ^{14}C. The pH of the pore waters and the ash content of the peats were used to identify the thickness of the ombrotrophic zone in each profile

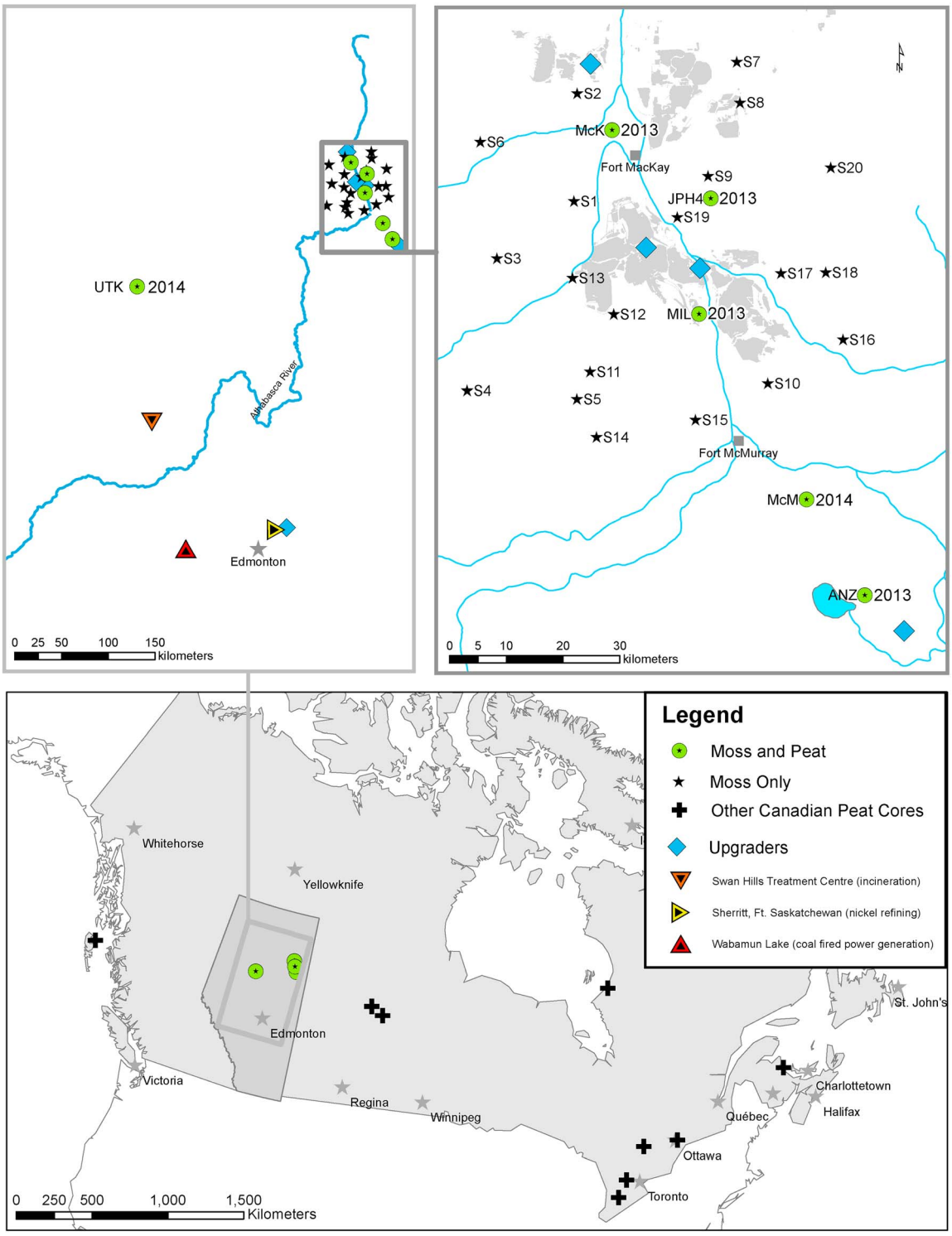

Figure 1. Map showing the locations of the peat cores described in the text (McK, JPH4, MIL, McM, ANZ and UTK) as well as the year of collection. Also shown for comparison, the locations of the moss samples collected from peat bogs of this region which were also measured for trace metals, including Pb and Th (*Shotyk et al.*, 2014). The shaded area in the map shown in the upper right corner refers to the locations of open pit bitumen mines. Also shown are the locations of the bitumen upgraders in the area of the Athabasca Bituminous Sands, namely Suncor, Syncrude, CNRL, and Nexen. The locations of coal-fired electricity generating stations near Lake Wabamun west of Edmonton, the bitumen upgrader east of Edmonton, the Ni refinery at Fort Saskatchewan, and the hazardous waste incinerator at Swan Hills, are also shown. The map of Canada shows the locations of peat cores collected in the past, but recently measured for Pb and Th in their surface layers [*Shotyk et al.*, 2014]

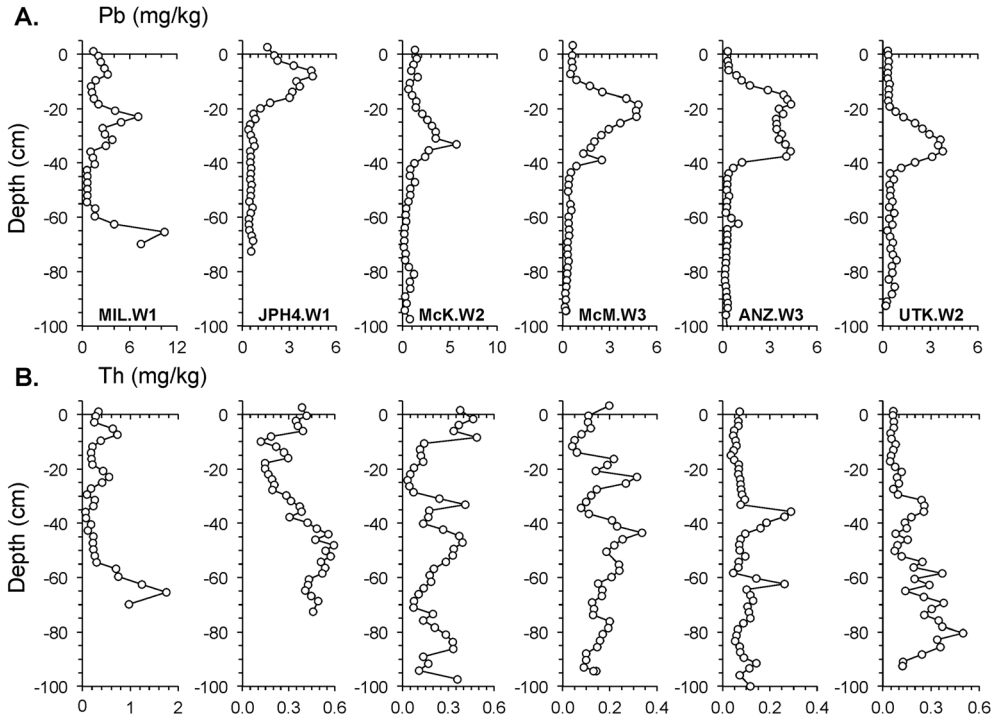

Figure 2. (A) Pb concentrations (mg/kg) in the six peat cores, listed in order of increasing distance from the mid-point between the two central bitumen upgraders. (B) Th concentrations (mg/kg).

(supporting information Figure S3). In addition to ^{14}C (supporting information Tables S3 and S4), all six peat cores were also dated using ^{210}Pb (CRS Model) determined using gamma spectrometry, and the ages confirmed with established chronostratigraphic markers (^{137}Cs and ^{241}Am). The degree of peat humification was evaluated using the main atomic ratios (C/N, H/C and O/C) of selected samples combined with stable isotopes ($\delta^{13}C$ and $\delta^{15}N$) and bulk density (supporting information Table S5). Analytical details are provided in the supporting information.

3. Results and Discussion

The Pb concentrations in the top living layer of each core are very low (Figure 2a): the maximum (1.6 mg/kg at JPH4) is less than corresponding value for the Drizzle Bog (2.0 mg/kg) on Haida Gwai, a large island west of British Columbia (Figure 1). The Pb concentrations in the top layer at ANZ and UTK (0.3 mg/kg) are similar to the corresponding values for the "cleanest" peat samples ever found (0.28 ± 0.05 mg/kg, $n = 17$) in the Swiss bog "Etang de la Gruère" (EGR) and dating from 5320 to 8030 ^{14}C yr B.P. [*Shotyk et al.*, 1998]. In each bog, the Pb concentration profiles reveal a prominent peak below the surface, ranging in depth from ~ 10 cm at MIL to ~ 35 cm at UTK (Figure 2a). Part of the variation in Pb concentrations seen in any given peat profile will have been caused by anthropogenic, atmospheric deposition, but part may be due to differences in the abundance of mineral matter, reflecting natural variations in atmospheric deposition of mineral dusts, peat decomposition, and humification, or both (supporting information Figure S1). Thorium, a conservative, lithophile element which is easily measured using ICP-MS alongside Pb, can be used as an indicator of the abundance of mineral particles (Figure 2b). To correct the Pb concentrations for these variations, Pb was normalized to Th (Figure 3a). The Pb:Th ratios show maxima which are ~ 10 to 30 cm below the surface of each bog: this is a clear and unambiguous indication that the maximum extent of Pb contamination in atmospheric aerosols has been declining. Moreover, the Pb:Th ratios measured in the topmost layers of peat from these bogs (2.1 to 5.0) are comparable to the corresponding values for the cleanest peat samples ever found (2.3 to 4.6, average 3.4 ± 0.6, $n = 18$), namely those from the EGR bog in Switzerland [*Krachler and Shotyk*, 2004] and dating from 5320 to 8030 ^{14}C yr B.P. The crustal enrichment factor (EF) for Pb in the AB profiles was calculated by normalizing the Pb:Th ratio (Figure 3a) to the corresponding ratio for the Upper Continental Crust [*Wedepohl*, 1995].

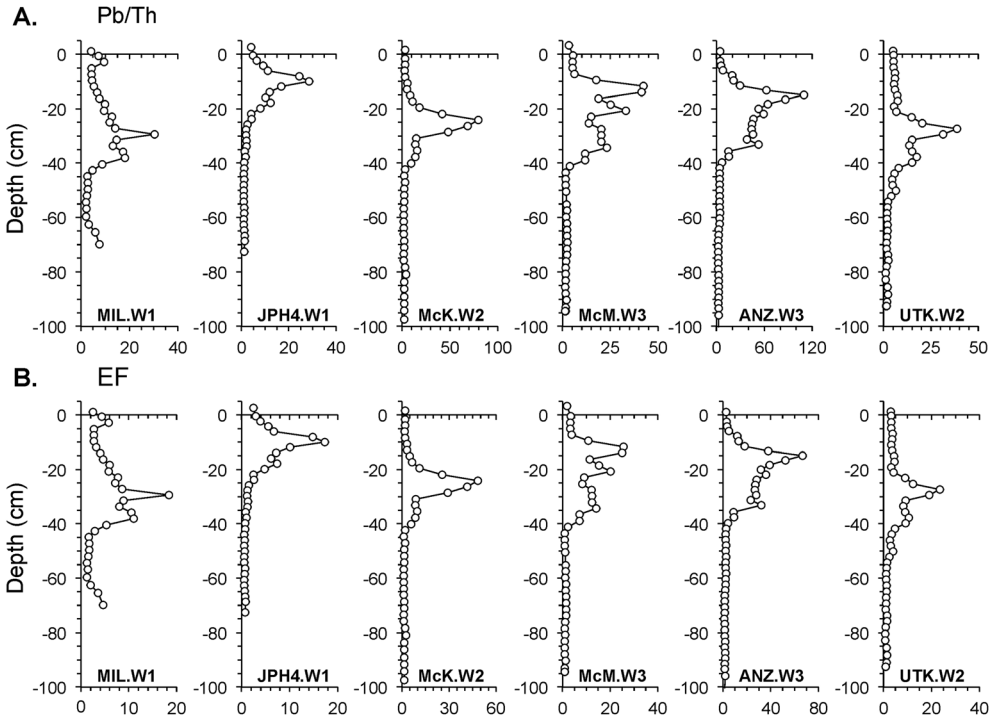

Figure 3. (A) Pb:Th for each of the six peat cores. (B) Pb Enrichment Factor, calculated using the Pb and Th concentrations for the Upper Continental Crust (*Wedepohl*, 1995).

Remarkably, the Pb EF at the surface of these bogs today ranges from only 1.9 to 2.5 (Figure 3b). A Pb EF = 1 indicates no Pb enrichment, relative to the Pb:Th ratio of crustal rocks, and an EF $\geq$ 2 may be considered the minimum EF to signal an additional, anthropogenic Pb source, over and above the natural Pb contribution to the atmosphere from soil-derived dust particles [*Shotyk et al.,* 1998; *Zheng et al.,* 2007]. In other words, the extent of Pb contamination in the topmost layers of the AB bogs is so small, and it is difficult to distinguish contemporary Pb to Th ratios from the natural background values. The Th concentrations were also used to calculate the natural concentrations of Pb in these profiles and total Pb to calculate the concentrations of anthropogenic Pb by difference: these calculations show that (i) in the past, natural Pb inputs were tiny, compared to the supply of Pb from human activities, (ii) that anthropogenic inputs have been in decline for decades, and (iii) that today, anthropogenic and natural Pb concentrations have converged (supporting information Figure S4).

3.1. Atmospheric Pb in AB, in Perspective

To put the data for the AB samples into context, the extent of Pb contamination in the top layers of these peat bogs today is no more than that found in the EGR peat bog from Switzerland in the sample dating from 3000 ^{14}C yr B.P. (3710 to 3210 cal yr B.P.): that sample was the first to have a Pb:Sc ratio double the natural background ratio [*Shotyk et al.,* 1998]. In the Swiss bog, the background ratio was defined as the average of peat samples (n = 17) dating from 5320 to 8030 ^{14}C yr B.P. which, in turn, were the lowest Pb:Sc (and Pb:Th) ratios found throughout the Holocene. In other words, the extent of Pb contamination being recorded by the peat bogs of northern AB today is no different than the intensity of atmospheric Pb contamination which occurred during the Iron Age of central Europe. Again, to view the Pb EF values from AB in the context of global atmospheric Pb contamination, the Pb EF in the Swiss peat samples dating from the Roman Period (1520 to 2100 cal yr B.P.) averages 10.3 (n = 7). So atmospheric Pb contamination in northern AB today is approximately 20% of that found in Switzerland during Roman times: this finding has special significance considering that most of the Pb production during Roman times was from mines in Spain and the U.K. [*Le Roux et al.,* 2004] and was therefore supplied to the Swiss bog by long-range atmospheric transport.

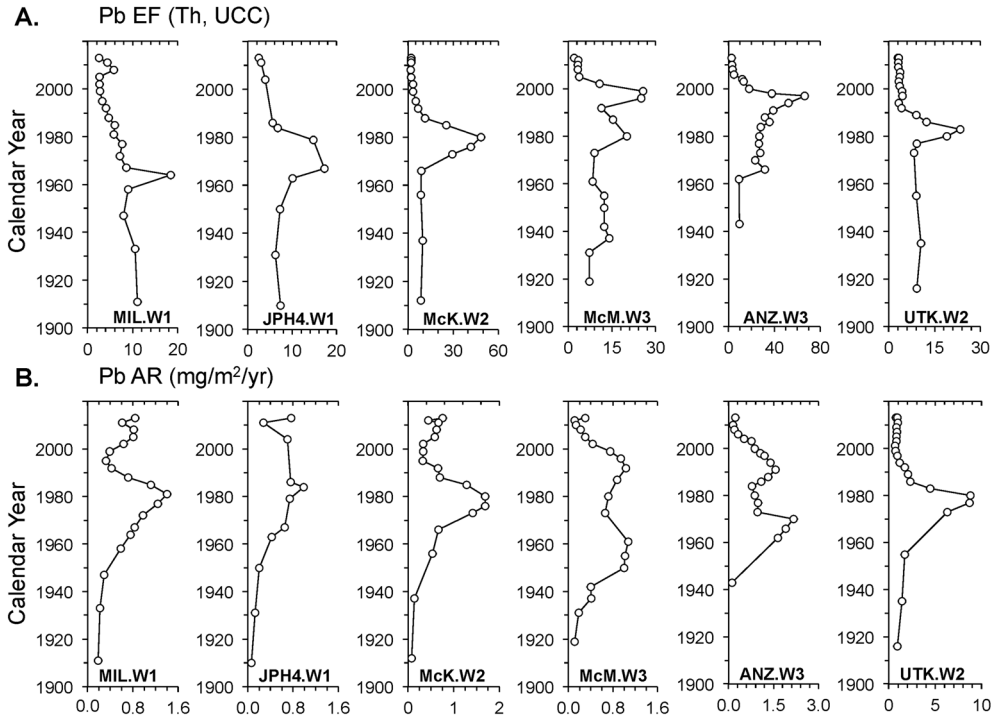

Figure 4. (A) Pb EF versus time. The age-depth models employed ^{210}Pb dating (CRS model, validated using ^{137}Cs and^{241}Am) along with ^{14}C age dates (supporting information Table S3) including those obtained using the atmospheric bomb pulse curve (B) Pb accumulation rates versus time. Lead accumulation rates were calculated as the product of peat accumulation rates (cm/yr), bulk density (g/cm^3), and Pb concentrations (μg/g).

The cumulative mass of anthropogenic, atmospheric Pb which has been stored in the AB peat cores since industrialization began is approximately 5% of the values reported for the cleanest peat bogs we have studied in Switzerland or the remote Shetland and Faroe Islands, and only 10% of the value from the cleanest peat bogs studied in Europe by our team to date, from eastern Finland (supporting information Table S6). Compared to other peat bogs we have studied in Canada, the AB peat bogs contain no more than 5% of the anthropogenic Pb found in bogs from southern Ontario, 25% of that found in the bog from Point Escuminac National Park on the coast of New Brunswick, and only 70% of the Pb found in our cleanest peat core from northern Quebec. The inventories of anthropogenic Pb in the AB peat cores are within a factor of 4 of the cleanest peat bog we have found in Canada to date, from remote Haida Gwai which is west of the coast of British Columbia (supporting information Table S6). In fact, the average anthropogenic Pb inventory in the AB bogs is within a factor of 7 times the industrial Pb burden (7 mg/m^2) preserved in the past five centuries of ice from Greenland [*Hong et al.,* 1994].

3.2. Pb From the Industrial Development of Bituminous Sands?

Although it had been suggested that open pit mining and upgrading of bitumen in AB is a significant source of Pb and other heavy metals to the environment [*Kelly et al.,* 2010], the AB peat bogs provide no evidence of this. First, the Pb:Th ratio of the topmost peat layers of the bogs in the vicinity of the ABS mines and upgraders is actually slightly less than that of the remote site, UTK (Figure 3). Second, the Pb:Th ratios of the top layers of all of the peat cores are similar to the background values for the mid-Holocene [*Krachler and Shotyk*, 2004]. Third, the Pb:Th ratios have been in decline for decades. For example, consider the peat core from JPH-4 which is exclusively ombrotrophic and only 12 km from the midpoint between the two central upgraders: the maximum Pb EF value was found in peat dating from 1967, and Pb EF has been in decline ever since (Figure 4a). Thus, according to this peat profile, the extent of Pb contamination in atmospheric aerosols in the ABS region of northern Alberta began its continual decline when the industrial development of the bituminous sands was just beginning in 1967, with the opening of the Great Canadian Oil Sands. This finding is consistent with the results obtained from

sediment cores taken in the Peace-Athabasca Delta which yielded a chronology of Pb accumulation which was inversely related to oil production in the ABS region [*Wiklund et al.,* 2012].

To further illustrate the recent declines in atmospheric Pb contamination, the peat accumulation rates, bulk density, and Pb concentrations were used to calculate the Pb accumulation rates (Figure 4b). The maximum Pb accumulation rates (ARs) shown here (~ 1.5 $mg/m^2/yr$) are one third the maximum value recently reported for the Hietajärvi peat bog, a background site within the Patvinsuo National Park in eastern Finland [*Shotyk et al.,* 2016b], and one tenth the maximum value (15 $mg/m^2/yr$) reported for peat cores collected from the EGR bog in a rural setting of the Jura Mountains in Switzerland [*Shotyk et al.,* 1998]. Compared to more industrialized regions of Europe, the Pb ARs for the bogs in northern AB are dwarfed by the maximum values ($mg/m^2/yr$) reported for peat cores from bogs in northern Poland (16 [*De Vleeschouwer et al.,* 2009]), Scotland (60 [*Farmer et al.,* 2006]), Denmark (105 [*Shotyk et al.,* 2003]), England (147 [*Rothwell et al.,* 2010]), Sweden (150 [*Bindler*, 2011]), Germany (176 [*Hettwer et al.,* 2003]), and the Czech Republic (320 [*Mihaljevič et al.,* 2006]).

3.3. Environmental Significance of Diminishing Atmospheric Pb Contamination

Lead poisoning was known in antiquity, having been described in the earliest writings on papyrus, parchment, copper, and clay [*Lessler*, 1988; *Hernberg*, 2000; *Needleman*, 2004]. This knowledge, however, did not prevent the introduction of Pb additives to gasoline. Since the first discovery of Pb contamination along roadways in Vancouver, British Columbia by *Warren and Delevault* [1960], the environmental impacts of gasoline Pb additives have generated a voluminous scientific literature: according to *Nriagu* [1990], in 1978 alone there were more than 5000 papers and reports on virtually every aspect of Pb in the environment. Despite claims from industry that it was a "gift from God" and "essential to civilization" [*Rosner and Markowitz*, 1985], the addition of tetraethyl Pb to gasoline was controversial from the start, partly because 15 workers died and 300 became insane, due to occupational exposure when production first began in American factories [*Needleman*, 1997, 1998, 2000; *Nriagu*, 1998]. Decades later, an international survey would show strong linear correlations between blood Pb concentrations and gasoline Pb concentrations in cities worldwide, but thankfully blood Pb concentrations began their decline soon after the introduction of unleaded gasoline in 1976 [*Thomas*, 1995; *Thomas et al.,* 1999].

The environmental history of this fascinating element continues to unfold, with evidence of atmospheric Pb contamination dating from the Bronze Age having been found in peat bogs from central Romania [*Grüters*, 2010], the Czech Republic [*Veron et al.,* 2014], and most recently from the Iberian Peninsula [*Martínez Cortizas et al.,* 2016]. Environmental Pb contamination in Egypt has an even longer history [*Véron et al.,* 2006, 2013]. Pioneering work on skeletal Pb concentrations of ancient Peruvians by Clair Patterson [*Ericson et al.,* 1979; *Settle and Patterson*, 1980] suggested that Pb exposure by modern humans dwarfs natural levels and has allowed blood Pb concentrations of unimpacted humans to be estimated [*Smith and Flegal*, 1992, 1995]. Human dental tooth enamel from archeological sites across Britain and Ireland since the Neolithic illustrate the connection between Pb use and body Pb burdens [*Montgomery et al.,* 2010], with values from the Roman Period up to 10,000 times natural levels; these individuals would have had blood Pb concentrations corresponding to "very severe" poisoning. Although blood Pb concentrations have widely declined [*Petit et al.,* 2015], there is no known safe level and this measure is now "seen by society as evidence of its commitment to its own health" [*Amaya et al.,* 2010].

The report of an additional, new source of Pb to the environment, from the industrial development of ABS in northern AB [*Kelly et al.,* 2010], raised considerable concern among First Nations communities who feared for the safety and security of their indigenous food supply (primarily berries, fish, and meat) and therefore also for their traditional lifestyle. In fact, the declining concentrations and enrichments of Pb seen in the peat bogs of northern AB are consistent with declines found in peat cores from Ontario [*Givelet et al.,* 2003; *Shotyk and Krachler*, 2010; *Pratte et al.,* 2013] as well as Manitoba [*Shotyk et al.,* 2016c], northern Quebec, and coastal British Columbia (*W. Shotyk*, unpublished data, 2005 and 2007). Viewed in this light and considering the pronounced declines in the extent of atmospheric Pb contamination found in bogs across Europe [*Shotyk et al.,* 2001, 2003, 2005, and many other authors], it would appear that the battle to end global atmospheric Pb contamination has been largely successful, at least in Europe and North America.

4. Conclusion

Peat cores from five bogs in northern Alberta show that atmospheric Pb contamination has been in decline for decades and has reached the point where, today, it is difficult to detect any Pb contamination, relative to background levels, in the topmost layers of these ecosystems. Although it had been suggested that the industrial development of the Athabasca Bituminous Sands is a significant source of Pb to the environment, in fact, the extent of atmospheric Pb contamination has been in decline more or less since the industry began and despite its rapid expansion.

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as well as table for precise sectioning of frozen peat cores; sample collection and preparation; preparation and measurement of samples for ^{137}Cs, ^{210}Pb, and ^{241}Am using ultralow background gamma spectrometry, including QA/QC. R.P. creation of maps showing all relevant geographic features and locations of bogs from this and previous studies, including both peat and moss only collection sites. B.S. design of calibration standards for ^{137}Cs, ^{210}Pb, and ^{241}Am using ultralow background gamma spectrometry; design of analytical protocols for these fallout radionuclides in peat including QA/QC program; calibration of these instruments and training of Noernberg. S.vB. responsible for plant macrofossil stratigraphy and selection/identification of samples for ^{14}C age dating using AMS. C.Z. peat core collection and preparation, and peat humification. The authors declare no competing financial interests.

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