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Lead Contamination of Subarctic Lakes and Its Response to Reduced Atmospheric Fallout: Can the Recovery Process Be Counteracted by the Ongoing Climate Change?

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Can a climate-triggered export of old contaminants from the soil alter the lead (Pb) contaminant burden of subarctic lakes? To address this question, we reconstructed the pollution history of three high latitude lakes situated in a region where a recent climatic shift has occurred. Dated sediment records were used as archives of past Pb inputs to the lakes, where the difference in the $^{206}Pb/^{207}Pb$ ratio between atmospheric contaminants ($^{206}Pb/^{207}Pb$ ratio <1.16) and geogenic Pb in the catchment soil ($^{206}Pb/^{207}Pb$ ratio >1.22) were used to trace fluxes of Pb contaminants. Lead contaminants were found in sediments deposited since Roman times. A significant export of Pb from the soil contaminant pool is indicated in two of the lakes surrounded by near-shore permafrost soils. Here, levels of Pb contaminants and $^{206}Pb/^{207}Pb$ ratios of sediments deposited after the 1970s appear not to have been strongly affected by the $\geq$90% reduction in atmospheric deposition rates and increasing $^{206}Pb/^{207}Pb$ ratios of atmospheric Pb since the 1990s. We concluded that soil processes stimulated by the ongoing climate change at high latitudes might work counteractive to efforts to reduce contaminant levels in subarctic lakes.

Introduction

The anthropogenic lead (Pb) contamination of the environment was recognized in the 1960s (*1*). Analysis of natural archives in remote regions, such as lake sediments, snow cores, and ice cores (*2–6*), reveals that even remote polar regions—previously considered to represent pristine conditions—were affected by Pb derived from anthropogenic emissions. As a result of improved environmental legislations, atmospheric Pb emissions decreased rapidly in the late 1970s, leading to decreasing levels of Pb in surface soils (*7*), vegetation (*8*), and lake sediments (*9–11*). Anthropogenic emissions decreased by >90% between the 1970s and the late 1990s (*8*), and the deposition rate at the end of the 1990s has reached preindustrial levels (*10*). Indeed, out-phasing of Pb from the atmosphere was a success from an environmental policy perspective; however, recently several assessment reports have suggested that the ongoing warming at high latitudes might stimulate the transport of atmospheric contaminants from catchment soils to surface waters as permafrost soil layers thaw (*12, 13*). Could such an accelerated export of contaminants increase levels of Pb contaminants in subarctic lakes despite reduced atmospheric inputs?

In forests, the main input of Pb contaminants to lakes occurs through direct atmospheric fallout on the lake surfaces with limited influence from the contaminants stored in the catchment soil (*9*). As a result, reconstructed pollution trends using lake sediments as archives mirror that of past atmospheric inputs (*10, 14*). For example, as a response to the >90% reduction in atmospheric fallout in Scandinavia (*8*), the concentration of Pb contaminants in boreal lake sediments (separated from natural Pb sources using stable Pb isotope analysis) has decreased to preindustrial levels (*10*). In boreal forests, the soil−water driven transport of atmospheric contaminants from upland and riparian soils to surface waters is a diffusive process that is unlikely to accelerate greatly in the future (*15*). In arctic regions, on the other hand, the prerequisites for the soil export of elements to surface waters may change dramatically in response to climatic change if it triggers thawing of previously permanently frozen soil layers. Such thawing might generate accelerated soil erosion, altered hydrological flow paths, increased runoff, and exposure of soluble compounds previously embedded in frozen layers. Studies in the U.K. have convincingly established that peat erosion can alter the Pb budget of aquatic conduits (*16*). Therefore, deepening of the active layer (upper soil that thaws during the summer) that triggers peat erosion could be a mechanism in which a warmer climate stimulates the transport of contaminants from the soil pool to arctic surface waters.

The fate of Pb contaminants is easily traced in northern Fennoscandia given that anthropogenic airborne Pb has a ratio of 1.13−1.18, while naturally occurring soil minerals mainly contain Pb with a $^{206}Pb/^{207}Pb$ ratio >1.30 (*17*). The deposition history of Pb contaminants in Europe has been extensively studied using independent archives without any inputs from catchment sources, such as historical documents (*18*), stored herbarium samples (*19*), polar ice-cores (*4*), and peat bogs (*20*). Together these independent records suggest synchronous shifts in the atmospheric fallout of Pb across Europe; i.e., increasing deposition rates of Pb contaminants are evident in the Roman period (0 ± 200 AD) and Medieval period (1100 ± 200 AD) and progressively increasing deposition rates are evident during industrial times with a pollution peak in the 1970s (*21*). The detailed knowledge about long-term deposition trends of Pb in Europe makes it possible to evaluate to what extent the reconstructed contaminant history of subarctic lakes reflects atmospheric deposition trends or catchment soil export processes.

In this study, we reconstruct the contaminant history of three subarctic lakes with different catchment characteristics using stable Pb isotope analysis ($^{206}Pb/^{207}Pb$; $^{208}Pb/^{207}Pb$) of dated sediment sequences covering approximately the last 4000 years or so. Differences in reconstructed Pb contaminant trends in comparison with the European atmospheric pollution history are interpreted in the context of catchment inputs of Pb contaminants to the lakes. Special focus is put on the recovery of the lakes in the last few decades, a period

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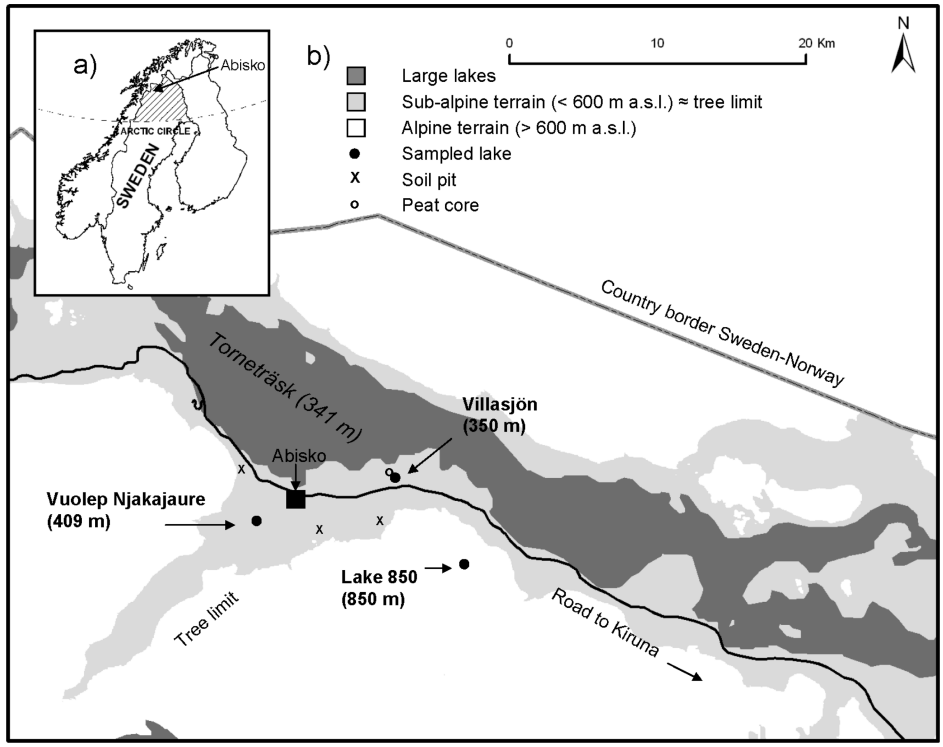

FIGURE 1. Map showing the studied lakes in the perspective of a) Scandinavia and b) the Abisko Valley, northern Sweden.

that is characterized by increasing temperatures and precipitation and a deepening of the active layer in the study area.

Materials and Methods

Lake Characteristics. Basic characteristics of the sampled lakes are summarized in Supporting Information Table S1. Lake 850 (850 m.a.s.l.) and Vuolep Njakajaure (409 m.a.s.l.) are circumneutral lakes situated on the slopes of the Abisko Valley in northern Sweden (Figure 1a-b). The lakes receive input water from areas up to 1100 m.a.s.l., and their catchments are mainly characterized by mineral soils with heath and shrub vegetation or, in the case of Vuolep Njakajaure, also sparse subalpine forests (below ~600 m.a.s.l.) dominated by mountain birch (*Betula pubescens ssp. tortuosa*) and with scattered Scots pine (*Pinus sylvestris*). Villasjön is a slightly acidic lake situated in the same valley (350 m.a.s.l.) but partly surrounded by the Stordalen palsa mire complex. The lakes are situated in catchments with three types of permafrost morphology. Villasjön is surrounded by near-shore permafrost with a morphology of peat palsas (*22*), L. 850 has near-shore permafrost developed in mineral soils underlying shallow organic layers (*23*), and Vuolep Njakajaure has no obvious morphological soil features in the catchment indicative of permafrost (*24*).

In the valley bottom, the mean annual temperature is around −0.7 °C, and the mean annual precipitation is around 300 mm. Since 1978, the depth of the active layer has been monitored in the Abisko valley. This monitoring shows an average increase in the active layer between 1978 and 2006 of about 1 cm yr^{-1} and a recent phase (since the mid-1990s) of accelerated thawing (*25*). Between 1970 and 2000, an expansion of wet fen communities has been documented in mires underlain by permafrost (*26*). In addition, summer and annual precipitation have significantly increased (*27*). The valley has been relatively undisturbed by human activities until the opening of the railroad in the beginning of the 20th century and the construction of a road through the valley in 1983 (the railroad parallels the road shown in Figure 1).

Sediment Sampling and Analysis. Lake sediments were collected from L. 850 (1999), Villasjön (2005), and Vuolep Njakajaure (2007) using a gravity corer or a freeze corer for the upper parts and surface sediments (<80 cm) and a Russian peat corer for overlapping deeper sediment sections (Figure 1b.) In addition, a peat profile (sampled using a Russian peat corer and consisting of about 0.5 m of ombrotrophic peat and 0.5 m of minerotrophic peat) and podzolic soil profiles ($n = 3$; depth >70 cm) were sampled to constrain the isotopic composition of the peat and glacial deposits until (Figure 1b). All samples were dried at 105 °C for 24 h or freeze-dried before analysis. Lead concentrations and stable Pb isotope compositions (^{206}Pb and ^{207}Pb) of dried homogenized samples (0.1 g) were determined using ICP-MS (Perkin-Elmer model ELAN 6100) after a strong acid digestion (conc. HNO_3 + $HClO_4$, 10:1) for two to five hours at <130 °C in open Teflon vessels. Because this method does not recover all Pb in mineral-rich matrices, it is likely to underestimate the concentration of geogenic Pb in mineral soil samples. Concentrations of Pb were verified against the certified multielement standard, SPEX ICPMS-2 (SPEX CertiPrep Certified Reference materials). A ten-point calibration within the range of 0.5 to 75 ppb was used. Analyses of ^{206}Pb and ^{207}Pb were made using dwell times of 50 ms and were corrected empirically by repeated analysis of the NIST SRM 981 reference material (fractionation constant 1.2% per a.m.u.). The method was validated via the analysis of a certified lake sediment (IAEA SL-1), where a mean ± SD value of 35 ± 2 mg Pb kg^{-1} (certified 37 ± 8 mg Pb kg^{-1}) and a $^{206}Pb/^{207}Pb$ ratio of 1.213 ± 0.002 were obtained. The latter ratio agreed well with previously reported values of 1.214 ± 0.012 (*28*). Mass loss after ignition (LOI) of the sediment subsamples was determined after heating at 550 °C for four hours. Soil and peat data are presented as Supporting Information (Table S1).

Dating. The ^{210}Pb activity of the surface sediments of L. 850 and Vuolep Njakajaure was measured using gamma spectroscopy. The ^{210}Pb derived from atmospheric fallout (unsupported) was calculated by subtracting the ^{226}Ra activity

(supported) from the total ^{210}Pb activity for L. 850 or the activity of ^{210}Pb at depth (<30 cm) for Vuolep Njakajaure. The sediment sequences were dated using the constant rate of supply model and validated by ^{137}Cs activity of the cores according to standard procedures as described in detail elsewhere (*29*). AMS radiocarbon dates for plant macrofossils, bulk-sediment, or aquatic mosses have previously been published for L.850 (*30*) and for Villasjön (*31*).

Expected Atmospheric Deposition Trends. The atmospheric pollution history in Europe, including the northern parts, has previously been summarized (*21*). Three atmospheric events have occurred synchronously over Europe in the past, and these atmospheric events (referred to as chronological markers) were located in the sediment sequences to evaluate the importance of previous atmospheric inputs of Pb contaminants to the lakes. The characteristics of these chronological markers as described by Renberg et al. (*21*) are summarized below.

1) The Roman peak in atmospheric pollution fallout (100 BC to 200 AD) is indicated by a transient increase in Pb concentrations and an associate decrease in ^{206}Pb/^{207}Pb ratio relative to background levels. This change in the ^{206}Pb/^{207}Pb ratio is a result of increased fallout of ancient pollution Pb due to Roman Pb production in southern Europe and is referred to as the Roman chronological marker.

2) In northern Sweden, the onset of the Medieval metal industry (1000 to 1200 AD) is characterized by increasing Pb concentrations occurring after the Roman chronological marker and the onset of a nearly continuous decrease in ^{206}Pb/^{207}Pb ratios. The observed trend results from increased atmospheric deposition of pollution-derived Pb following the onset of the Medieval mining industry in Europe and is referred to as the Medieval chronological marker.

3) The industrial peak in atmospheric Pb pollution (1970 to 1980) is indicated by the highest peak in the Pb concentration, co-occurring with or close to a minimum in the ^{206}Pb/^{207}Pb ratio of the sediment. This event is the result of maximum fallout of atmospheric Pb in the mid 1970s mainly due to the use of alkyl-lead additives in petrol. This chronological marker is referred to as the alkyl-lead chronological marker.

Results

Lead Concentrations and ^{206}Pb/^{207}Pb Ratios. The long sediment records (ca. 2000 years old) from Villasjön and L. 850 reveal that the Pb concentrations increase and the ^{206}Pb/^{207}Pb ratios decrease toward the surface sediment (Figures 2 and 3). Only the alpine lake (L. 850) exhibits an increasing Pb concentration and a decreasing ^{206}Pb/^{207}Pb ratio, trends that characterize the Roman chronological marker (see material and methods). The Roman marker is found at a depth of about 0.35 m (Figure 3). In the palsa mire lake (Villasjön), both Pb concentrations and ^{206}Pb/^{207}Pb ratios remain largely unaffected despite rapid sedimentation of eroded peat as indicated by the elevated macrofossil density of the sediments in unit 2 (Figure 2). Apparently, there are no obvious inputs of ancient contamination Pb.

Typical characteristics of the Medieval chronological marker are recorded around 30 and 16 cm in Villasjön and L. 850, respectively (Figures 2 and 3). However, in Villasjön the increase in Pb concentration occurs during a period characterized by increasing concentrations of plant macrofossils in the sediments, indicating inputs of Pb from eroding peat layers. Subsurface Pb concentration peaks in the range of 10−25 mg Pb kg^{-1}, sharing characteristics typical for the alkyl-lead chronological marker found in L. 850, Vuolep Njakajaure, and Villasjön at around 3, 14, and 7 cm, respectively (Figures 2 and 4). The near surface increase in Pb concentrations in Villasjön occurs at the level of maximum

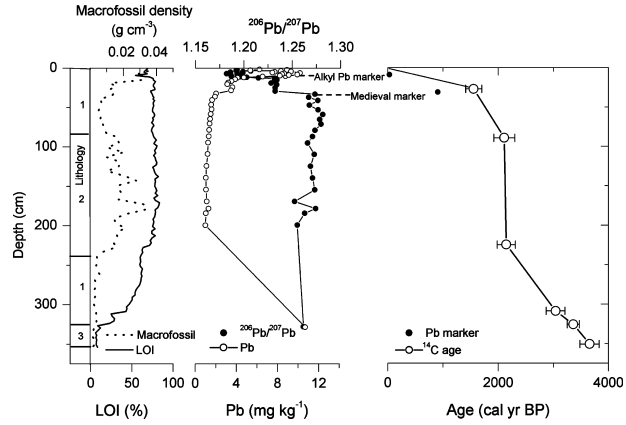

FIGURE 2. Organic matter content, macrofossil density, Pb concentrations, ^{206}Pb/^{207}Pb ratios, and inferred ages based on ^{210}Pb dating, stable Pb chronological markers, and radiocarbon dates of the sediment sequence from Villasjön. Numbers in the left panel refer to lithostratigraphic units where 1 is detritus gyttja, 2 is coarse detritus gyttja with abundant redeposited peat, and 3 is organic-rich silty clay.

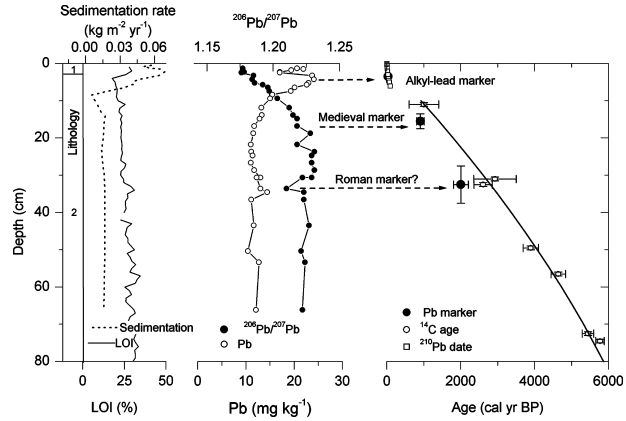

FIGURE 3. Organic matter content, sedimentation rates, Pb concentrations, ^{206}Pb/^{207}Pb ratios, and inferred ages based on ^{210}Pb dating, stable Pb chronological markers, and radiocarbon dates of the sediment sequence from L. 850. Note that the sedimentation rate is expressed as a 3-point running mean due to the uncertainty involved when calculating estimates for single levels. Numbers in the left panel refer to lithostratigraphic units where 1 is low density gyttja and 2 is high density gyttja.

concentration of macrofossils, providing further evidence of an input of Pb from the catchment sources.

The radiocarbon-based age of the medieval chronological marker (28−26 cm) in Villasjön (1700 to 1400 cal years BP at 2σ) is considerably older than the ^{14}C established age of 900 ± 150 years BP (Figure 2). Similarly, the radiocarbon age of the Roman chronological marker in L. 850 (2900 to 2300 cal years BP at 2σ) is older than the established age of 2200 to 1800 years BP (Figure 3). However, the age of the alkyl-lead chronological marker (1970 to 1980 AD) agrees well with the ^{210}Pb-inferred ages of the short cores from L. 850 and Vuolep Njakajaure (Figures 3 and 4).

In the sediments of all three lakes, indications of dynamic catchment-to-lake export processes are indicated by variable sedimentation rates or fluctuating concentrations of macrofossil (Figures 2 and 4). In Villasjön, a ~1.3 m section of the sediment sequence consisting mainly of reworked peat appears to have been deposited over a short period as indicated by the ^{14}C-dates. L. 850 and Vuolep Njakajaure exhibit increased sedimentation rates in the 20th century.

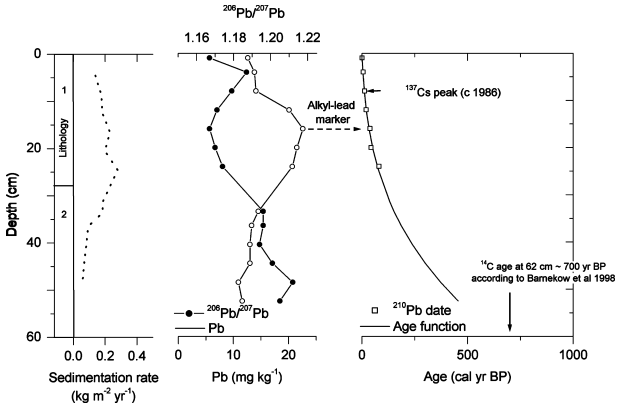

FIGURE 4. Sedimentation rates, Pb concentrations, $^{206}Pb/^{207}Pb$ ratios, and inferred ages based on ^{210}Pb dating, stable Pb chronological markers, and radiocarbon dates of the sediment sequence from Vuolep Njakajaure. Note that the sedimentation rate is expressed as a 3-point running mean due to the uncertainty involved when calculating estimates for single levels. Numbers in the left panel refer to lithostratigraphic units where 1 is low density gyttja and 2 is high density gyttja.

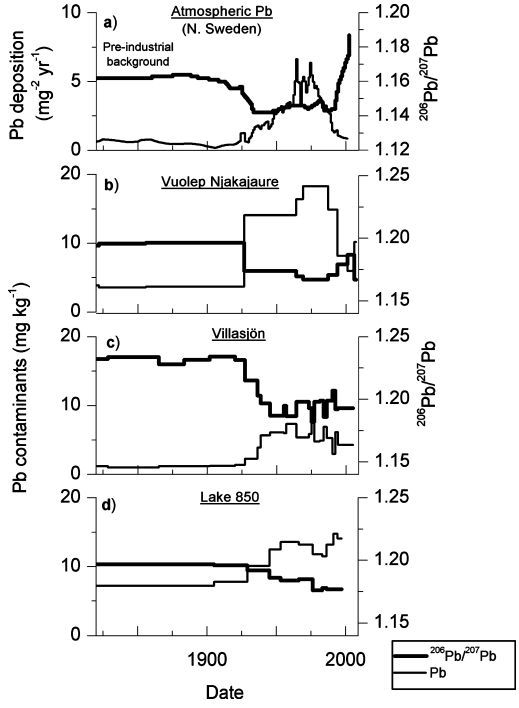

FIGURE 5. a) The $^{206}Pb/^{207}Pb$ ratio of forest mosses collected at various dates from sites in northern Sweden and preserved in herbarium collections (32) serving as a reference of the $^{206}Pb/^{207}Pb$ ratio of atmospheric Pb. The $^{206}Pb/^{207}Pb$ ratio of the sediments and the concentration of Pb contaminants in b) Vuolep Njakajaure, c) Villasjön, and d) L. 850. Note the approximate time-scale of the Villasjön record (see the text for explanations).

Concentrations of Pb contaminants in the sediments can be calculated assuming mixing between geogenic sources in the catchment and atmospheric Pb (Supporting Information eq 1). The approximate temporal changes in Pb contaminant concentrations during the last centuries in the sediments and their relation to atmospheric inputs are shown in Figure 5a-d. Due to the apparent problem in establishing a chronology based on radiocarbon dates in Villasjön (Figure 2), the chronology of this sediment sequence has been derived through cross-correlation with the other two dated sequences that used the rapid increase in Pb contaminants at 13 cm as a marker for ~1920 and the peak concentration around 7 cm as a marker for 1975 (the alkyl Pb marker). Consequently, only the major trends in this record should be interpreted, and absolute dates indicated in Figure 2 should be treated with caution. In contrary to atmospheric trends in the $^{206}Pb/^{207}Pb$ ratio in northern Sweden (Figure 5a), the $^{206}Pb/^{207}Pb$ ratio of none of the studied lake sediment sequences (Figure 5b-d) have returned to pre-1900 values. Yet, Vuolep Njakajaure shows an increasing $^{206}Pb/^{207}Pb$ ratio trend after the 1970s, similar to the atmospheric trend if the uppermost sample is neglected. In addition, this lake also has a Pb contaminant concentration trend (calculated using the mixing $^{206}Pb/^{207}Pb$ model) that follows the decreasing deposition trend (Figure 5a) if the upper sample is neglected. The Pb contaminant level in the sediments of the other two lakes, on the other hand, has not returned to pre-1900 levels as expected from the decreasing atmospheric fallout. In fact, the Pb contaminant concentration in L. 850 appears to be increasing in sediments deposited since the middle of the 1980s.

Discussion

Lead-containing minerals in the soils surrounding the studied lakes have a $^{206}Pb/^{207}Pb$ ratio typically around 1.25 as indicated by data obtained on old sediments and deeper mineral soil layers (Figures 2−4; Supporting Information; Table S1). Despite this natural constraint, the surface sediments of the studied lakes exhibit a $^{206}Pb/^{207}Pb$ ratio mainly between 1.16 and 1.19. This indicates additional input of Pb contaminants from the atmosphere having a $^{206}Pb/^{207}Pb$ ratio typically between 1.13 and 1.18 in northern Sweden (*32*). Clearly, atmospheric Pb contaminants have significantly affected the Pb burden of all the studied lakes. Indeed, this result is not surprising given that atmospheric Pb contaminants have been deposited at high northern latitudes since the onset of the ancient mining era in Europe about 3500 years ago (*4*).

Increasing $^{206}Pb/^{207}Pb$ ratios of atmospheric Pb have been noted in northern Scandinavia after the 1980s (*32*), trends that are similar in other parts of Europe (*19*). Such temporal trends in combination with reduced deposition rates of atmospheric Pb contaminants have caused the $^{206}Pb/^{207}Pb$ ratio of lake sediments deposited after 1990 to increase significantly (*21, 33*). Interestingly, the $^{206}Pb/^{207}Pb$ ratios of the subarctic lake sediments do not increase after the 1990s for Villasjön and L. 850, and the lowest ratio is found at the surface sediments of Vuolep Njakajaure (Figure 5b-d). The reduction in Pb contaminant concentration in these two former lakes is much lower than the >90% reduction in atmospheric fallout during the past four decades in northern Sweden (*8*). Hence, the weak recovery in the $^{206}Pb/^{207}Pb$ ratio and Pb contaminant concentrations suggests that catchment export processes of previous deposited atmospheric contaminants have a significant impact on the contaminant burden of at least two of the lakes. A $^{206}Pb/^{207}Pb$ vs $^{208}Pb/^{207}Pb$ plot of the sediments further confirms this by showing that the isotopic composition of the surface sediments cannot be explained without including mixing of Pb deposited prior 1990 (Supporting Information; Figure S1).

Peat macrofossils in the sediments of Villasjön provide compelling evidence for a significant export of organic matter from the catchment to this lake (Figure 2). This is also indicated by the discrepancy between ages inferred from radiocarbon dates and the Pb chronological markers. The anomalously old radiocarbon dates obtained for bulk sediments and aquatic mosses in L. 850 and Villasjön, respectively, are probably related to redeposition of organic matter from catchment soils and significant reservoir effects, i.e., old soil-derived carbon has been reassimilated by the dated submerged aquatic vegetation in Villasjön. Similar effects of these

processes have previously been reported by Barnekow et al. (*24*) based on comparison of radiocarbon dates obtained on bulk sediment samples and macroscopic plant remains in lake sediments from the study area.

Lead is mainly cotransported to aquatic systems along with organic compounds (*16, 34*). However, before the 20th century the export of organic matter from catchment soils did not generate a significant flux of Pb contaminants given the low inventory of contaminants in the soils at this time, a finding that may explain why the reconstructed contaminant trends largely mirror that of the atmospheric fallout. For example, about 2000 years ago peat erosion around Villasjön released organic matter from nearly 'lead-free' peat layers having Pb concentrations down to 0.6 mg kg^{-1} as indicated by levels found in deep peat layers in the Stordalen mire today (Supporting Information; Table S1), totally suppressing the Pb contaminant signal in the sediments from ancient emission into the lake rather than amplifying it (Figure 2). On the other hand, peat erosion and leaching of dissolved organic compounds during the last decades release organic matter enriched in Pb as a result of contamination during the industrial era, processes that have proved important for contaminant fluxes in aquatic systems at lower latitudes (*16*). Within the 1.3 m thick sediment layer consisting of reworked ancient peat, about 160 mg Pb m^{-2} has accumulated that is derived mainly from geogenic sources. If such an export event occurred in the contemporary environment, it may generate an up to three times higher flux considering that the surface peat now contains about three times more Pb as a result of atmospheric contamination (Supporting Information; Table S1). Hence, a catchment release of this magnitude could more than double the inventories (~160 mg Pb m^{-2}) of Pb contaminants currently in the sediments that have accumulated over the last millennium.

Importantly, the export of Pb contaminants from the catchment soil pool has historically been small, a finding that is indicated by i) temporal contaminant trends in the sediments that largely follow the atmospheric pollution history in Europe and ii) sediment inventories of Pb contaminants that overlap with those of upland soils and peat profiles exposed solely to atmospheric fallout (Supporting Information; Table S2). However, as previously stated there is a significant impact of catchment-derived Pb contaminants after the 1980s in the sediment records of Villasjön and L. 850 as indicated by temporal trends in the $^{206}Pb/^{207}Pb$ ratio and Pb contaminant concentrations (Figure 5c-d) that deviate from that of the known atmospheric pollution history (Figure 5a). Studies from the boreal region suggest limited catchment export of Pb contaminants during the last three decades as indicated by a rapid change in the $^{206}Pb/^{207}Pb$ ratio and Pb concentrations of the sediments after the 1980s (*21*), i.e., trends similar to those recorded in Vuolep Njakajaure with the exception of the uppermost sample. It appears, therefore, that the catchment export of Pb contaminants has been more profound during the last decades in subarctic lakes surrounded by permafrost soils than boreal or subalpine birch-forest lakes.

In Villasjön, one obvious driver of the catchment export of Pb contaminants is thermokarst erosion. Accelerated thawing of permafrost in the study area has been documented since the mid-1990s (25), and severe peat erosion since the 1970s has been observed within the Villasjön catchment as result of differential settling of mire hummocks previously uplifted by permafrost heave (*22*). Therefore, the weak recovery in Villasjön is likely due to inputs of Pb contaminants derived from collapsing peat layers where wetter conditions facilitate the lateral transport to the lakes. Erosion of near-shore soils containing permafrost during periods of climatic deterioration has previously been suggested to be the main sediment source to L. 850 during the Holocene (*23*). Therefore, it seems plausible that the recent input of Pb contaminants from the catchment soils observed at L. 850 is stimulated by altered biogeochemical conditions in near-shore soils due to subsidence of thawing permafrost soils, conditions that are similar in Villasjön where more organic-rich soils prevail. The increasing Pb contaminant concentrations also follow after a period of increased soil erosion as indicated by increased sedimentation rates (Figure 3), which further supports this hypothesis.

The subarctic soils contains a legacy of atmospheric Pb contaminants deposited over millennia. The latest assessment report on climate change by the Intergovernmental Panel on Climate Change (*12*) hypothesizes that climate change may accelerate the transport of contaminants from soil to surface waters in the arctic. Our study cannot provide a causal relationship between the recent climate warming in the study area and the observed increasing influence of Pb contaminants from catchment soils since the 1970s. However, there are several indirect lines of evidence that suggest that a warmer climate stimulates the soil export rate of contaminants to lakes in the area. A recently published mass-balance calculation along thermokarst erosion gradients at Stordalen suggests that a substantial amount of Hg contaminants accumulated in peat can be mobilized as the thawing and degradation of frozen and uplifted areas proceeds (*35*). Our retrospective data showing a limited recovery in the Pb contaminant input to the nearby lake Villasjön due to significant peat erosion in combination with the findings of the former study demonstrates that thermokarst erosion can accelerate the input of contaminants to subarctic lakes. In systems similar to Stordalen where climatic variables such as increased air temperatures, summer precipitation, and snow depth had a documented stimulating effect on peat erosion and leaching of organic matter (*22, 27*), a climate-triggered release of contaminants seems plausible. As previously shown, the release of contaminants from the soil pool could likely be even more pronounced than observed in these lakes, considering the rapid sedimentation of more than 1 m of eroded peat material in the ancient the sediments of Villasjön (Figure 2). If such export is repeated in the near future, it could generate an unprecedented flux to this lake.

It is important to stress that our inferred coupling between climate change and increased export rates of contaminants from catchment soils to lakes is tentative. However, the findings presented serve as a rationale for future studies that assess processes in which climate change may stimulate soil export processes at a larger regional scale or assess eventual incorporation of contaminants released from thawing permafrost soils into arctic and subarctic ecosystems.

Acknowledgments

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Supporting Information Available

Tabulated data regarding basic lake characteristics and calculated inventories of Pb contaminants in sediments, upland soils and peat profiles from the area. Also shown are $^{206}Pb/^{207}Pb$ and $^{208}Pb/^{207}Pb$ ratios of the sediments in relation to atmospheric and geogenic Pb sources. This information is available free of charge via the Internet at http://pubs.acs.org/.

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