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Early Atmospheric Pollution Recorded in the Sediments of High Mountain Lakes in the Vicinity of Mining Areas

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Abstract

Historic changes in metal pollution were investigated in sediment cores covering the last 200 to 500 years of four high alpine lakes in North Tyrol, Austria. The catchment areas of the lakes are located above timberline and consist of a high amount of bare rocks and fine grained scree. As a consequence, the geochemistry of these high alpine lakes is particularly sensitive to climate driven weathering rates. Therefore we distinguished atmospheric metal deposition from local inputs from the catchment area by normalizing trace metal concentrations to changes in conservative elements (i.e. Ti, Zr, Al). In all lakes, lead increased steadily over the last 100 to 150 years due to enhanced industrialization and usage of leaded gasoline. However, we observed even higher increases in lead in Mutterbergersee between 1570 and 1650 AD. This lake is located 15 km northwest form the Schneeberg/Monteneve mining area, that has been exploited for lead, copper, and zinc with earliest recording in 1237 and the strongest mining activities in the 15th and 16th century. Early atmospheric metal pollution was less distinct in the other lakes that were located in greater distance to minor mining areas: high concentrations in metals were observed in Gossenköllesee for lead and chrome around 1830, and in Brechsee for lead, arsenic, cadmium, and zinc in the second half of the 19th century. The lake located highest showed no distinct metal pollution prior to industrialization.

Introduction

Tyrol and the surrounding areas have been important mining areas over several centuries (Srbik, 1929; Exel, 1998). The historic mining activities have affected landscapes, society and also the environment (Prammer, 2007). Lake sediments offer a tool to go back in time and reconstruct changes in environmental conditions prior to the onset of monitoring. Sediment cores obtained from lakes can be sliced in thin layers. These single layers can be dated and analyzed for multiple parameters, ranging from geochemical, to mineralogical and biological proxies. The dated layers can then be interpreted in a chronological context. In this study, we compared dated sediment records from four remote high alpine lakes. We were particularly interested in deciphering traces of atmospheric metal pollution that are related to historic mining activities. Here we show the preliminary results of the lead stratigraphies.

Methods and Study Sites

Sediment cores have been obtained from the central part of four high alpine lakes with a modified Kajak corer (UWITEC). The cores have been sliced in 0.25 to 1 cm layers. The layers have been dated with 210Pb and 137Cs, which give a good result for the last 150 to 200 years. As the sediment accumulation rate was fairly constant, age of older core section was estimated based on a constant sedimentation rate. Consequently, ages older than 200 years are subjected to limited accuracy. Changes in geochemistry have been analyzed with WD-XRF, and ICP-MS resp., and mineralogical composition with XRD. All lakes are located above timberline on crystalline bedrock (containing micashists, granites, gneisses) in the Ötztaler Alps within 30km from each other (Tab. 1). They are not directly influenced by human activities but sheep are grazing in the catchments. We focused on lead because it is generally well preserved in the sediments as it is not affected by changes in Redox or pH. Furthermore, it can be measured very precisely.

Tab. 1: Information about study sites, core age and major metal enrichments

lake name	**altitude**	**max depth**	**size**	**Core**	**enrichment**
Mutterberger See (M)	2486 m	7.5 m	0.037 km²	ca. 500 yrs	Pb, As
Schwarzsee ob Sölden (S)	2796 m	18 m	0.035 km²	ca. 250 yrs	Pb, Zn
Brechsee (B)	2145 m	6.1 m	0.013 km²	ca. 250 yrs	Pb, Zn, Cu
Gossenköllesee (G)	2417 m	9.6 m	0.016 km²	ca. 800 yrs	Pb, Cr, Co, Ni

Results and Discussion

Lead (Pb) increased in the top sediment core sections in all lakes (Tab. 1, Fig. 1). In order to distinguish metal enrichment caused by changes in the catchment, esp. erosion, we normalized the Pb trends with titanium (lakes M, S, G), or aluminum (lake B; Ti not measured) resp. We observed the same increase both in Pb and normalized Pb trends. The most pronounced increase was visible since ca. 1880-1900 and is thus attributed to industrial activities and lead gasoline. Since the reduction of leaded gasoline in the 1970ies, a Pb decreased in all cores. In

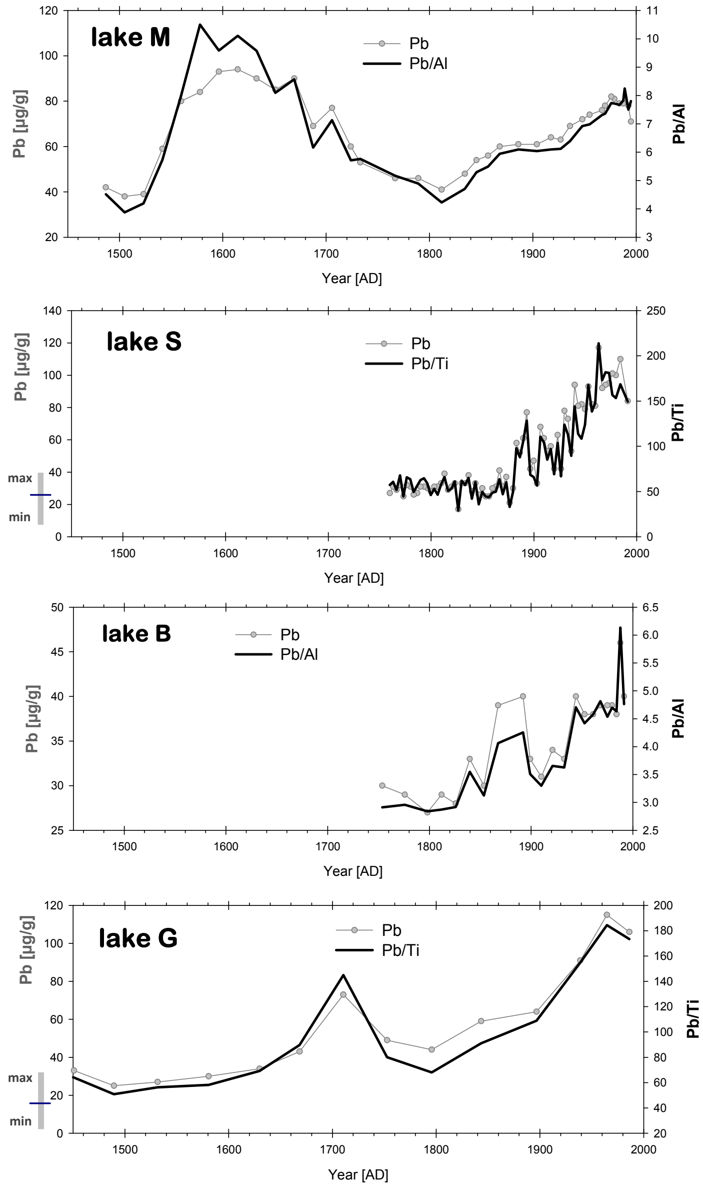

Fig. 1: Trends of lead concentrations and normalized lead in four high alpine lakes. Note the high pre-industrial lead peak in Lake M. For lakes S and G the range of Pb observed in rock samples from the catchment is indicated on the left y-axis.

Lake G a pre-industrial Pb peak was visible around 1700. However this peak was concurrent with a change in grain size thus pointing to a change in particulate input. The most interesting Pb peak occurred in Lake M between ca. 1580 and 1700. This peak was even higher than the industrial lead peak in the 20th century. Although dating is of limited accuracy for this core section, it was distinctly a pre-industrial lead peak. The Pb peak went together with an increase in As. The changes in lead were not caused by changes in the mineralogical composition. This is also visible in a PCA (principal component analyses) of the geochemical parameters including the minerals and organic parameters as explanatory variables which were projected passively into the ordination space (Fig. 2, see Leps & Smilaur, 2003): only 24% of the variation in geochemistry is explained by the mineralogical composition, an additional 26% is related to changes in organic parameters (C, N, P). None of the explanatory variables is related to lead.

A historic mining area, is located in the vicinity of Lake M, 15 km southeast across the mountain ridge. The peak of the mining area “Schneeberg - Monteneve” was between 1500 and 1600 (Srbik, 1929, esp. page 227). Despite protests of the local residents, the Fugger (illegally) started with smelting in 1535. They abandoned the mines in 1663. The lead peak in Lake M was observed around this period. This overlap together with the absence of other causes for lead input, esp. changes in erosion or mineralogical composition, points to atmospheric pollution as major source for the pre-industrial lead peak in Lake M. We currently test this hypothesis by analyzing the isotopic composition of lead ($^{206}Pb/^{207}Pb$) in selected sediment samples.

Conclusion

In the four high alpine lakes lead clearly increased since the turn of the 19th century. This was attributed to industrial activities and usage of leaded gasoline. Lead decreased slightly in the most recent sediment layers due to the reduction of leaded gasoline. Early atmospheric metal pollution was only distinct in the lake located closest to a mining and smelting area. In the other lakes, pre industrial lead peaks have either not been captured within the relatively short sediment records (peak of mining in Tyrol in 15th to early 16th century), or might have been masked within the high geogenic background concentrations. Further analyses, including longer cores of Lake S and lead isotope analyses ($^{206}Pb/^{207}Pb$) will help to evaluate the extent of atmospheric pollution caused by early mining activitiess.

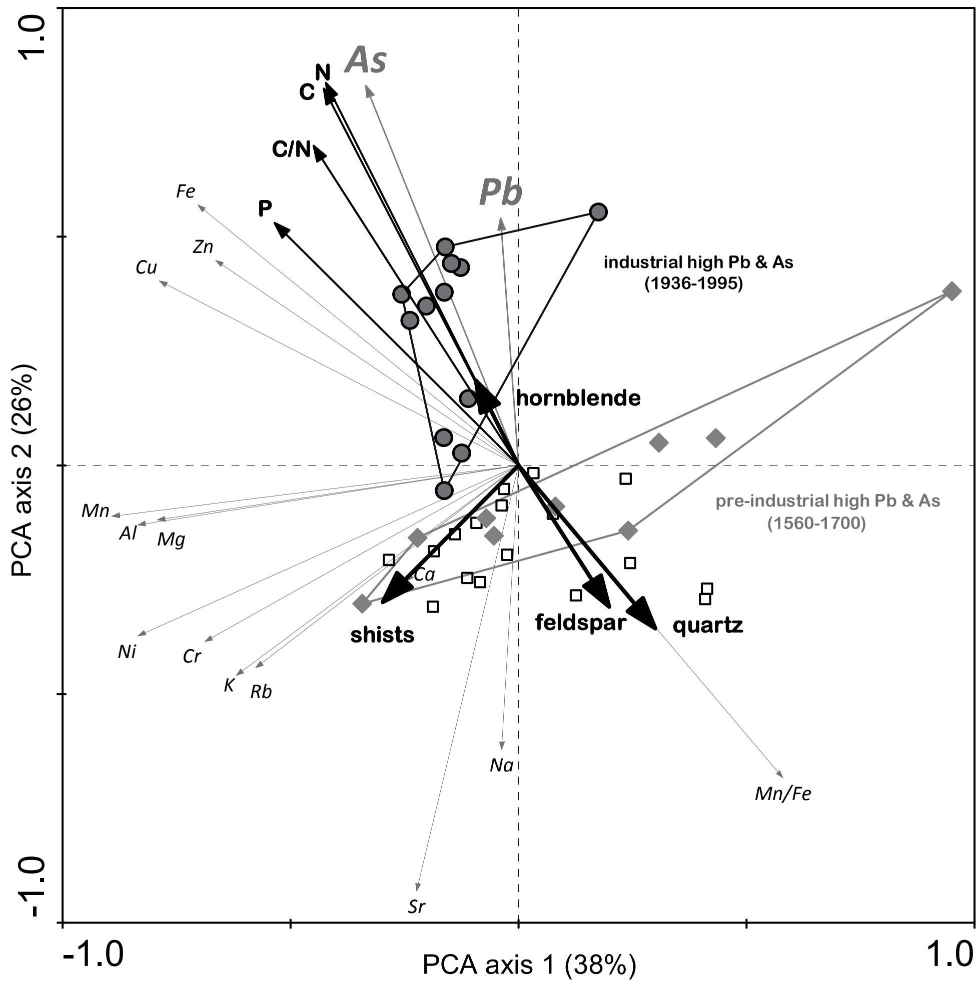

Figure 2: Principal component analysis (PCA) of geochemical parameters measured in lake M with environmental parameters (C, N, P, C/N, and feldspar, quartz, shists, hornblende) projected passively into the ordination space. Samples with high lead concentrations (> 70 µg/g) are enveloped: big grey diamonds = samples with high pre-industrial lead concenctration (ca. 1550-1700); big black dots = samples containing high industrial lead conc. (1930-1995). Thick black arrows: relation to mineralogical composition. Thin black arrows: relation to organic parameters. 24% and 26% of the variation in geochemical parameters can be explained by changes in minerals or organic parameters respectively.

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