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# A 150-year Record of Heavy Metals in the Varved Sediments of Lake Bolterskardet, Svalbard

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## Abstract

Laminated sediments from Bolterskardet Lake on Svalbard provide a new 150-year record of heavy metals in the Arctic. Independent data of  $^{137}\text{Cs}$  and  $^{210}\text{Pb}$  indicate that these laminations are annually deposited varves. The high sedimentation rate and varved sediments make Lake Bolterskardet a good site for studying history of heavy metal pollution in the region. A suite of heavy metals (Pb, As, Cd, Cu, Cr, Co, Ni, and Sn) were studied. The variations of Cu, Cr, Co, and Ni concentrations show an inverse pattern with the median grain size. It suggests that the particle size has a significant role in the accumulation and enrichment of heavy metals in the sediments. In the concentration profiles of studied heavy metals, only Pb concentrations show a significant increase from the lower parts to the upper parts of the core. Profiles of “total,” “lithogenic,” and “anthropogenic” Pb flux also show an increasing pattern. Anthropogenic Pb flux varies between  $0.1 \mu\text{g cm}^{-2} \text{yr}^{-1}$  and  $12.3 \mu\text{g cm}^{-2} \text{yr}^{-1}$ , with a mean value of  $2.4 \mu\text{g cm}^{-2} \text{yr}^{-1}$ . The anthropogenic Pb fluxes were relatively low at around  $0.7 \mu\text{g cm}^{-2} \text{yr}^{-1}$  prior to 1945, slowly increased after 1945, and reached a sidestep (between 1940s and 1970s) with mean value of  $1.8 \mu\text{g cm}^{-2} \text{yr}^{-1}$ . Second high value period was between 1980s and 1990s with mean value of  $5.9 \mu\text{g cm}^{-2} \text{yr}^{-1}$ . The variation of anthropogenic Pb fluxes before 1970 in Bolterskardet Lake coincides with the sulfate record and Pb concentration data from Greenland snow and ice, and the acidity concentration in the Svalbard ice cores. However, the anthropogenic Pb fluxes during the last two decades show a rapidly increasing trend. This regional pattern may suggest that local source is an important factor for heavy metal contamination in Svalbard, and the complex of long-range transport contaminations for regional and global process.

## Introduction

Growing concerns about heavy metal and organic pollution in the fragile Arctic ecosystems have led to numerous projects being carried out to study spatial and temporal trends of contaminants, and monitor the impact of human activities in the Arctic (e.g., Lockhart et al., 1995; Boutron et al., 1995; Äyrä et al., 1997; Braune et al., 1999; Macdonald et al., 2000; Shotyk et al., 2003; AMAP, 2004; Riget et al., 2004; Haack et al., 2004; Berg et al., 2004). On Svalbard, heavy metals have been studied from shelf sediments (Rognerud et al., 1998; Siegel et al., 2000), lacustrine sediments (Holte et al., 1996; Boyle et al., 2004; Rose et al., 2004), and animals (e.g., Fant et al., 2001; Derocher et al., 2003; Willeroider, 2003). It has been determined that heavy metal concentration in the surface sediments of Svalbard fjords is much higher than assumed background, presumably as a result of terrestrial water drainage of coal particles originating from local coal stores and industrial activities (Holte et al., 1996). Strongly anthropogenic influence was also illustrated by the examination of snow and cores (Simões and Zagorodnov, 2001; Isaksson et al., 2003). Simões and Zagorodnov (2001) concluded that Svalbard is one of the areas in the Arctic strongly affected by anthropogenic pollution. Recent studies suggested that the lakes on Svalbard are generally too insensitive to have recorded long-transported heavy metal pollution (Boyle et al., 2004).

In the industrialized area, lacustrine sediments can provide a quantitative record of atmospheric pollutants for unbiased estimation of pollution history (e.g., Norton and Kahl, 1991; Boyle and Birks, 1999; Shotyk et al., 2001; Arason and Fletcher, 2003; Abbott and Wolfe, 2003). However, it has been suggested that such techniques are

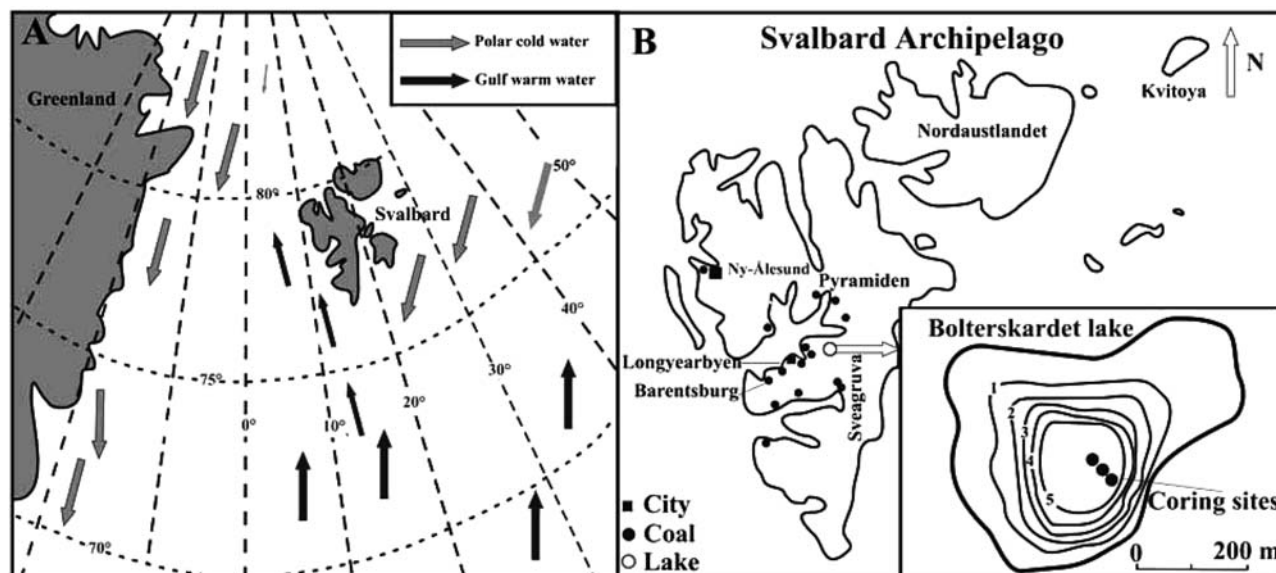
more problematic in regions where the atmospheric contamination signal is small, and sedimentation rate is low (Cornwell, 1986; Boyle et al., 2004). Additionally, Arctic lakes are very sensitive to climatic change, especially as increasing sedimentation rate following global warming would significantly dilute heavy metal's concentration in lacustrine sediments.

Here, we present heavy metals study of varved sediments from Lake Bolterskardet in Svalbard, and compared them to the published data from ice cores in Greenland and Svalbard and history records at the local site.

## Study Site

Svalbard is located at a crossroads of a branch of North Atlantic warm current and polar cold water, and the climate boundary of the polar front (Fig. 1). The variation of currents and shifts of the polar front would have a noticeable effect on Svalbard. These conditions make Svalbard a sensitive area for paleoenvironmental changes in the Arctic. Nevertheless, few studies have focused on high-resolution study, except for some important contributions on studying ice cores (Tarassov, 1992; Koerner, 1997; Pohjola et al., 2002; Isaksson et al., 2003) and recent lacustrine sediments (e.g., Birks et al., 2004).

Lake Bolterskardet ( $78^{\circ}06'\text{N}$ ,  $16^{\circ}01'\text{E}$ ) is located in the west of Spitsbergen, the largest island in Svalbard (Fig. 1). The lake is a topographically closed lake. It is surrounded by steep mountains with elevations of 700–900 m. The elevation of the lake is 385 m a.s.l. The lake has a surface area of  $0.16 \text{ km}^2$ , a catchment area of  $7.8 \text{ km}^2$ , and a maximum depth of ca. 6 m. The bedrock in the catchment is mainly



**FIGURE 1.** The location of Svalbard and coring sites. (A) Location of Svalbard and ocean currents (modified from Kempf, 1999). (B) Location of Bolterskardet Lake and coring sites (the interval of isobathic curve is 1 m).

composed of Tertiary siltstones and sandstones (Eldholm et al., 1984). We could not find any outcrop of coal in the catchment of Lake Bolterskardet. The nearest coal mine is located 10 km north of the lake (out of the catchment). There is little vegetation in the catchment area of the lake.

Compared to the data from Lake Linnévatnet on the coast of Spitsbergen, which is completely covered by ice from late September until early August or late July (Mangerud and Svendsen, 1990; Svendsen and Mangerud, 1997), we estimate the ice-free period might only be about one month in Lake Bolterskardet because of its high elevation.

## Methods

### SAMPLING

Three sediment cores were recovered from Bolterskardet Lake during August 2002 using a modified corer developed at the Institute of Geology and Geophysics, Chinese Academy of Sciences. The diameter of the corer is 70 mm. The coring sites are shown in Figure 1B (inset map). Steel tubes were used to connect the corer, slowly set in sediments. When we took out the core, a special device closed the corer. The water in the upper part of the coring tube was carefully removed using a hose and syringe. Considering high water content in the upper sediment of cores, the cores were kept vertical in the field until the surface water was evaporated by paper towels. In the laboratory, the cores were split, photographed, and logged. The lengths of the recovered cores (Cores S-385-B1, -B2, and -B3) are 46 cm, 69 cm, 41 cm, respectively. Based on initial inspection of the fresh split surface of cores, Core S-385-B2 (length: 69 cm) was chosen for this study. Half of the core was sampled at 1 cm intervals. The samples were freeze-dried to determine water content and dry bulk density. Magnetic susceptibility was measured with Bartington MS Meter. Grain size was analyzed by a laser size analyzer (Fritsch, Analysette-22).

### SEDIMENT CHRONOLOGY

#### Varve Counting

Sediment slabs (60 × 20 × 10 mm) were taken from the core. The sediment slabs were vacuum dried, penetrated with synthetic resin, and made into thin sections (Lamoureux, 1994; Chu et al., 2005). These

thin sections were microscopically examined for sedimentary laminations. Varves were identified and counted from thin sections using a Leitz light microscope.

#### Radiometric Dating

The freeze-dried samples were used for radionuclide measurement. Activity measurements of  $^{137}\text{Cs}$ ,  $^{210}\text{Pb}$ , and  $^{226}\text{Ra}$  were carried out by gamma spectrometry using a low-background well-type germanium detector (EGPC 100P-15R). Each sample was packed in a 15 mm polyethylene tube for 3 weeks of storage in sealed containers to allow radioactive equilibration (Hamilton et al., 1994; Ruiz-Fernández et al., 2003). Then the sample was counted for 48 h.  $^{210}\text{Pb}$  total was determined by gamma spectrometry via its energy at 46.5 keV. The short-lived  $^{226}\text{Ra}$  daughter nuclides  $^{214}\text{Pb}$  (241.9, 295.2, and 351.9 keV) and  $^{214}\text{Bi}$  (609.3 keV) were measured to determine supported  $^{210}\text{Pb}$  for the calculation of unsupported  $^{210}\text{Pb}$ .  $^{137}\text{Cs}$  was measured by its emissions at 662 keV. Radiometric dates were calculated from the  $^{210}\text{Pb}$  and  $^{137}\text{Cs}$  records using the CRS model (Appleby et al., 1986).

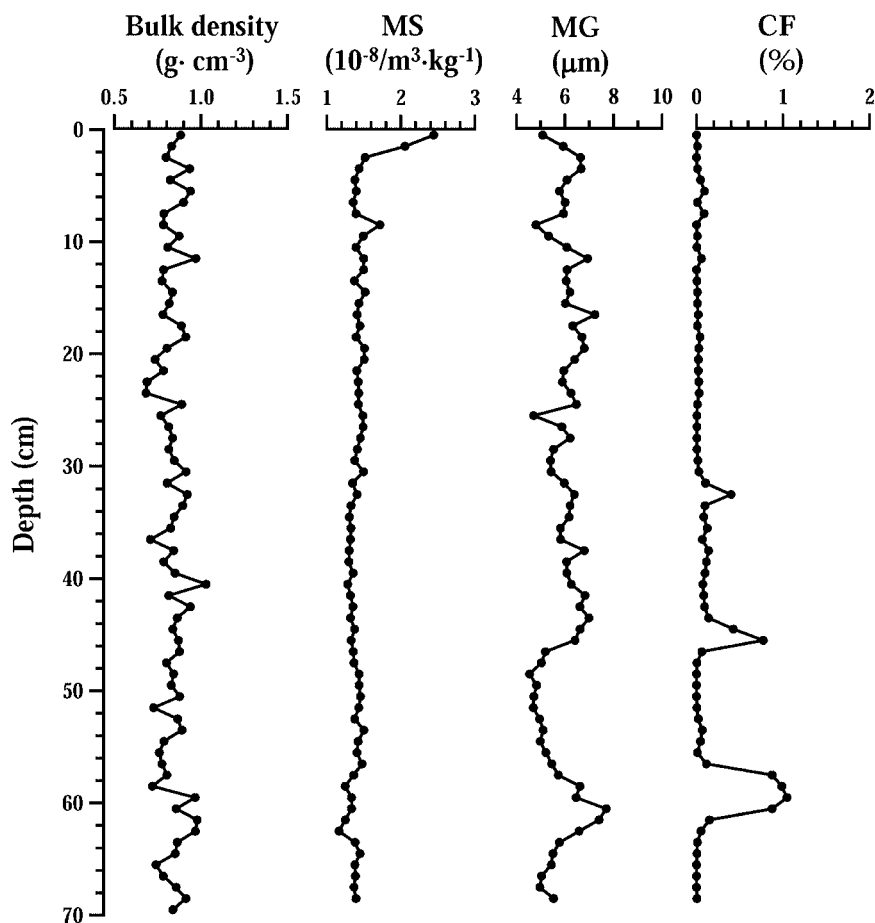
#### HEAVY METALS

Heavy metals were extracted from 0.1 g freeze-dried sediment with 25 mL, 1.58 M  $\text{HNO}_3$  by constant agitation at room temperature for 24 h (Abbott and Wolfe, 2003). This weak extraction procedure deliberately targets labile metals adsorbed to organic and inorganic surfaces, and not those associated with the mineralogy of sediment inorganic constituents (Abbott and Wolfe, 2003). Metal concentrations were determined using a VG-PQ-Excell inductively coupled plasma-mass spectrometer (ICP-MS). Detection limits are  $0.0001 \mu\text{g mL}^{-1}$  ( $\leq 0.0001$ ) for Cu, Pb, Co, Cd, and Sn, and between 0.0002 and  $0.0005 \mu\text{g mL}^{-1}$  for Cr, Ni, and As. The relative standard deviations for the elements are below 5% ( $n = 7$ ).

## Result and Discussion

### DESCRIPTION OF THE SEDIMENT CORE

Initial inspection of the fresh split surface of the cores revealed millimeter- to centimeter-scale, light- and dark-colored laminate couplets. There is a slumping layer between 15 cm and 23 cm. Figure 2



**FIGURE 2.** Down-core variations in bulk density, magnetic susceptibility (MS), median grain size (MG), and coarse (>63  $\mu\text{m}$ ) fractions (CF).

shows down-core variations in bulk density, magnetic susceptibility (MS), median grain size (MG), and coarse (>63  $\mu\text{m}$ ) fractions (CF). The sediments consist of fine-grained sizes ranging from fine sand to clay. There are very few coarse mineral grains (>63  $\mu\text{m}$ ) in the sediments (Fig. 2). It has been suggested that coarse sand and silt are distributed across the lake ice by eolian processes during the winter and spring (Lamoureux et al., 2002; Lewis et al., 2002). Absence of coarse sand-sized grains may indicate that mountains surrounding Bolter-skardet Lake greatly shield strong wind activity. The dry density of the samples falls between 0.68 and 1.0  $\text{g cm}^{-3}$ , with a mean value of 0.83  $\text{g cm}^{-3}$ . They appear to correlate with the variation of median grain size. With the exception of the upper part of the core, magnetic susceptibility shows less variation. The relative constancy of bulk density with depth suggests that no significant degree of compaction has occurred. The dry bulk density and fine grain sizes suggest the sediments have low permeability and that there was limited vertical mixing of pore waters.

We used standard methods (Battarbee et al., 2001) to check if there are diatoms and stomatocysts in the sediment. However, no diatoms or stomatocysts were found. There is also not enough pollen (above statistic amount) for a pollen study. The higher elevation and short ice-free period may greatly prevent algae survival in the lake.

#### SEDIMENT CHRONOLOGY

##### Clastic Varve

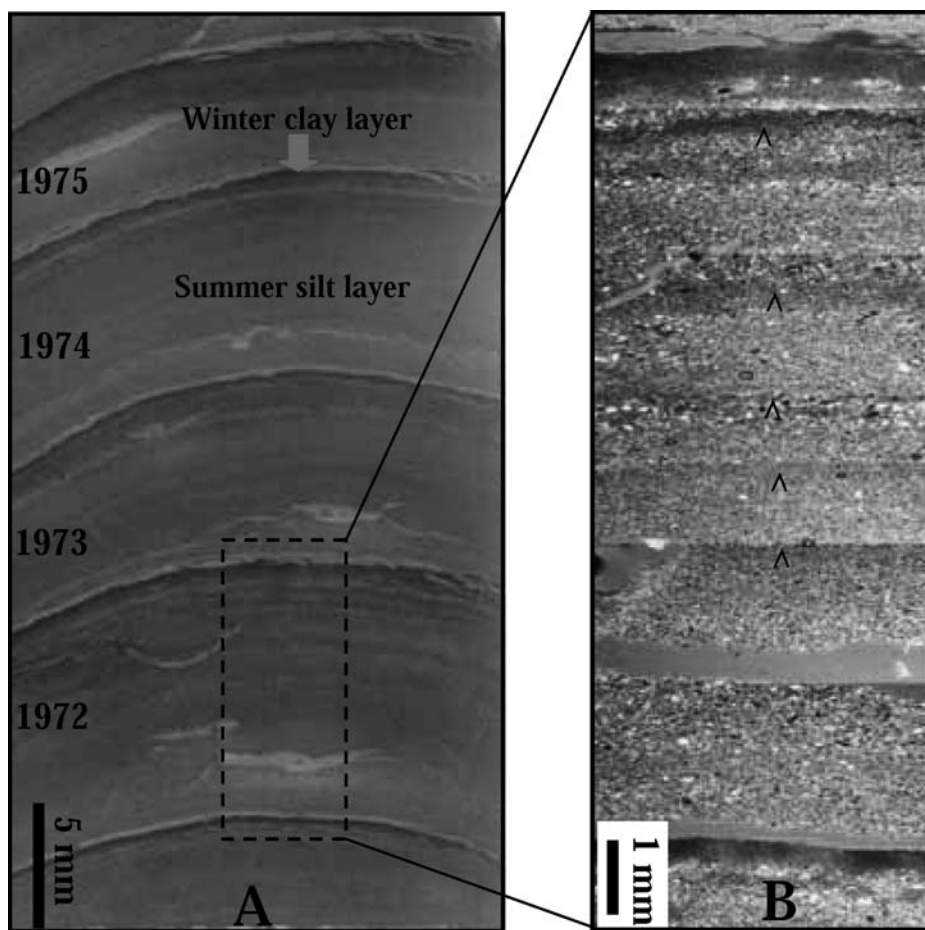
Clastic varves have been well defined in high Arctic areas by prior researchers (e.g., Hardy et al., 1996; Gajewski et al., 1997; Lamoureux, 1999; Hughen et al., 2000; Lamoureux et al., 2001; Moore et al., 2001; Lamoureux and Gilbert, 2004). It is generally recognized that the

lamination couplets reflect seasonal variations of clastic input derived from glacier and snow melt.

In thin section under a petrologic microscope, laminae appear as rhythmic units of light-colored (silt) layers capped by a dark clay layer (Fig. 3). The silt layers are mainly composed of silt-sized particles (<20  $\mu\text{m}$ ), with occasional particles >63  $\mu\text{m}$ . The minerals of the silt are predominantly freshly eroded feldspars, quartz, and micas, consistent with the bedrock in the catchment. The silt layers are normally graded into fine clay, which in turn exhibits a sharp contact with the overlying silt layer. The dark clay layer is composed of fine particles smaller than 2  $\mu\text{m}$ ; seldom do any particles larger than 2  $\mu\text{m}$  occur (observed under highest magnification). Based on XRD results, dominant minerals in the clay layer are quartz, micas (muscovite, lepidolite), chlorite, and illite, reflecting a local origin. The silt layer thickness varies between 0.40 mm and 19 mm, whereas the clay layer is generally less than 1 mm. Two or more graded sub-rhythmites can be observed within a silt layer in some varves (Fig. 3B). Lamoureux (1999) suggested that sub-rhythmites may be due to two or more phases of snowmelt. The sub-rhythmites consist of silt and clay. It is easy to distinguish sub-rhythmites from the dark clay layer, which has a sharp interface and consists of pure clay.

##### Radiometric Dating

Independent data of  $^{137}\text{Cs}$  and  $^{210}\text{Pb}$  activities were used to verify the varve chronology in Core S-385-B2. Figure 4 shows  $^{137}\text{Cs}$  and unsupported  $^{210}\text{Pb}$  activities in Core S-385-B2. The highest  $^{137}\text{Cs}$  value of 48.5  $\text{Bq kg}^{-1}$  detected at 34 cm in the core is assumed to correlate to the 1963 maximum of emission due to nuclear bomb testing. Forty varves were counted in the upper 34 cm of the core, which is in good agreement with the  $^{137}\text{Cs}$  chronology. They indicate



**FIGURE 3. Photographs of varves. (A) Thin section photo shows varves in Lake Bolterskardet. (B) Micro-photographs under transmitted light, with gypsum plate. Varve appears as couplet of light silt layer and dark clay layer under a petrologic microscope. The silts grade upward into fine clay, then the clay layer sharply contacts with the silt layer in next layer. Some rhythmites (marked as >) can be found within silt layer.**

that the laminae couplets are annually deposited varves and can therefore be used to construct calendar-age chronologies. But, the  $^{137}\text{Cs}$  profile does not show the Chernobyl event, either this event is not a strong signal on Svalbard or undistinguishable for low activities of  $^{137}\text{Cs}$  in the region.

Although there is good agreement between the varve counts and 1963 highest  $^{137}\text{Cs}$  value, the Pb CRS model ages differ from the varve counts, especially in the 34–46 cm interval and low part of the core. It could be related to low levels of  $^{210}\text{Pb}$  in the sediments, and large variations of accumulation rates as suggested by Lamoureux (1999). In fact, the CRS age model is especially sensitive to changes in accumulation rates and often causes problems (Appleby and Oldfield, 1983; Appleby et al., 1986). The activities of  $^{137}\text{Cs}$  and  $^{210}\text{Pb}$  are lower than the values reported in other studies of Arctic lakes such as Lake Upper Soper (Hughen et al., 2000), Lake Nocolay (Lamoureux, 1999), Lake V1-V2 Greenland (Eriksson et al., 2004), and eight lakes in the coastal region of Svalbard (Appleby, 2004). The low activities of  $^{137}\text{Cs}$  and  $^{210}\text{Pb}$  in Lake Bolterskardet may be related to fast sedimentation rates, and low atmospheric  $^{210}\text{Pb}$  fluxes (the rainfall having an oceanic origin).

#### HEAVY METAL CONCENTRATION PROFILES

Profiles of heavy metal concentration are shown in Figure 6. Visual inspection of Figure 6 reveals that variations of Cu, Cr, Co, and Ni concentrations display a similar pattern, and have relatively large variations (Fig. 6). On the other hand, Pb, Cd, As, and Sn concentrations show a different pattern with less stratigraphic variations (Fig. 6). The variations of Cu, Cr, Co, and Ni concentrations show an inverse pattern with the median grain size. It suggests that the particle

size has a significant role in the enrichment of heavy metals in the sediments. Previous studies have observed that the heaviest enrichment of metals occurs in the finer grades (Padmalal et al., 1997; Song et al., 1999; Wang et al., 2003; Shah et al., 2005). Generally, with their very large surface area, very small particles have stronger ability to absorb heavy metals. On the other hand, Arctic dust favors a global or distant source for most of the dust, especially the fraction less than  $2\ \mu\text{m}$  (Darby et al., 1974).

In the profiles, only Pb concentrations show a significant increase from the lower parts to the upper parts of the profile. The Pb concentrations vary between  $11.0\ \mu\text{g g}^{-1}$  and  $25.8\ \mu\text{g g}^{-1}$ , with a mean value of  $14.5\ \mu\text{g g}^{-1}$ . The Pb concentrations in the sediments of Lake Bolterskardet sediments are lower than the data from other lakes in Svalbard. For example, mean Pb concentrations are  $30.4\ \mu\text{g g}^{-1}$ ,  $24.4\ \mu\text{g g}^{-1}$ ,  $20.9\ \mu\text{g g}^{-1}$ ,  $28.6\ \mu\text{g g}^{-1}$ ,  $21.4\ \mu\text{g g}^{-1}$ , and  $43.2\ \mu\text{g g}^{-1}$  in Lake Ossian, Lake Ytertjørna, Lake Vassauga, Lake Daltjørna, Lake Tenndammen, and Lake Arresjøen, respectively (Boyle et al., 2004). The mean Pb concentration in this study and other lakes is greatly lower than the data ( $12\ \text{mg g}^{-1}$ ) from the Svalbard west shelf (Siegel et al., 2000), ( $26\ \text{mg g}^{-1}$ ) from Isfjorden near Longyearbyen (Siegel et al., 2000), ( $13\ \text{mg g}^{-1}$ ) from Kongsfjord (Skei, 1994; see in Siegel et al., 2000), and ( $20\ \text{mg g}^{-1}$ ) from the sediments from Longyearbyen harbor (Holte et al., 1994; see in Siegel et al., 2000). Considering large differences of Pb concentrations between lacustrine and marine sediments in the region of Svalbard, high lead contamination in the marine sediments may be transported considerably by ocean currents and ice (Gobeil et al., 2001), except for the factor of low sedimentary rates in marine. Previous studies have suggested that the concentration of heavy metals in sediments may be modified by some factors such as physical sediment mixing, diagenesis, organic matter

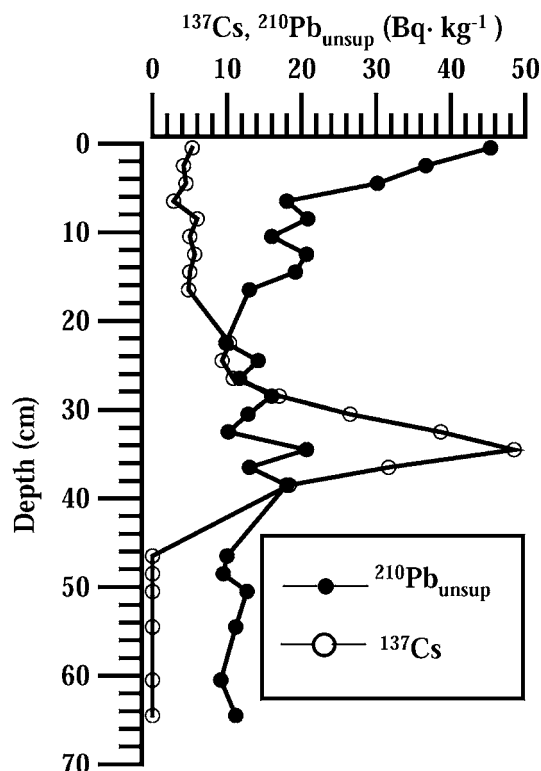


FIGURE 4.  $^{137}\text{Cs}$  and unsupported  $^{210}\text{Pb}$  activity versus depth for Core S-385-B2.

content, salinity, pH, redox condition, particle size, and sedimentation rate (Lottermoser et al., 1999; Boyle, 2001, 2004a; Wang et al., 2003). In this case, the varved sediments and lack of organic matter (no diatoms or chrysophytes) may greatly reduce the complexity of these processes. However, significant change of sedimentation rates may cause variations in heavy metal concentrations. High sedimentary rate can weaken or dilute the pollution signal relative to other factors, especially in regions where the atmospheric contamination signal is small (Boyle, 2004; Boyle et al., 2004). However, if sedimentary rate is very low, the pollution signal is confined to the top 1 to 2 sampling intervals (Boyle et al., 2004). In Lake Bolterskardet, the high sedimentary rate greatly helped us to obtain high resolution data.

#### LEAD FLUX

Heavy metal concentrations in lake sediments are strongly influenced by sedimentation rates, especially with regard to the increasing sedimentary rates observed in almost all varved lacustrine sediments in the Arctic with global warming (e.g., Hardy et al., 1996; Overpeck et al., 1997; Gajewski et al., 1997; Hughen et al., 2000; Jones et al., 2001; Lamoureux and Gilbert, 2004; Smith et al., 2004). The up-core increase in the concentration of any particular element might be explained either by an increase in its supply rate, or by a decrease in the supply rate of one or more of the other components (Boyle, 2004). Accumulation rate data could unambiguously distinguish these cases (Boyle, 2004). Previous studies also suggested that heavy metal concentrations can be separated into “lithogenic” and “anthropogenic” components using ratios to Al, Sc, or Ti (Shotyk et al., 1998, 2001, 2003, 2005; Boyle, 2004). Scandium has been suggested as a preferred reference element for estimating lithogenic components (Shotyk et al., 2001).

Here, we use Sc as an indicator of the concentration of lithogenic derived components supplied by rock weathering. The fluxes of

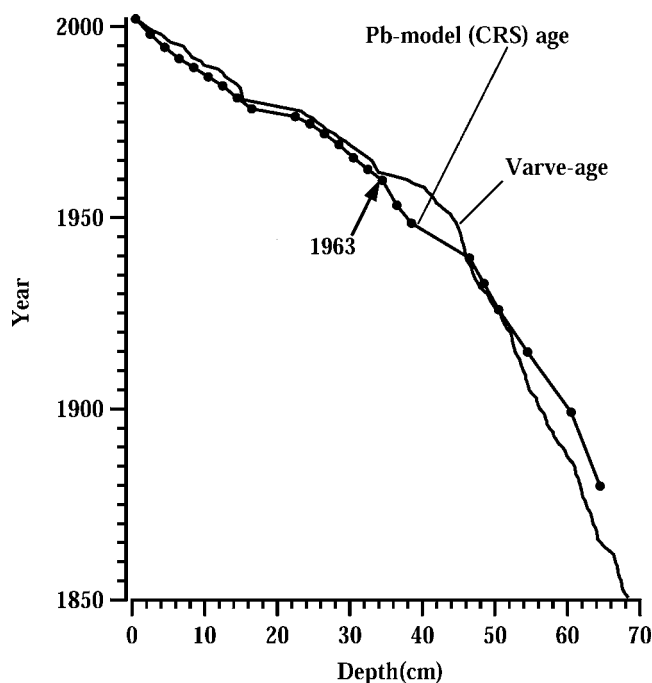


FIGURE 5. Varve ages and Pb-model (CRS) ages versus depth for Core S-385-B2.

anthropogenic heavy metals are calculated as follows (Shotyk et al., 2001, 2005):

$$[\text{Pb flux}]_{\text{anthropogenic}} = [\text{Pb flux}]_{\text{total}} - [\text{Pb flux}]_{\text{lithogenic}} \quad (1)$$

$$[\text{Pb flux}]_{\text{total}} = [\text{Pb concentration}]_{\text{total}} \times [\text{dry density}] \times [\text{SR}] \quad (2)$$

$$[\text{Pb}]_{\text{lithogenic}} = [\text{Sc concentration}]_{\text{sample}} \times [\text{Pb/Sc}]_{\text{upper continental crust}} \times [\text{dry density}] \times [\text{SR}] \quad (3)$$

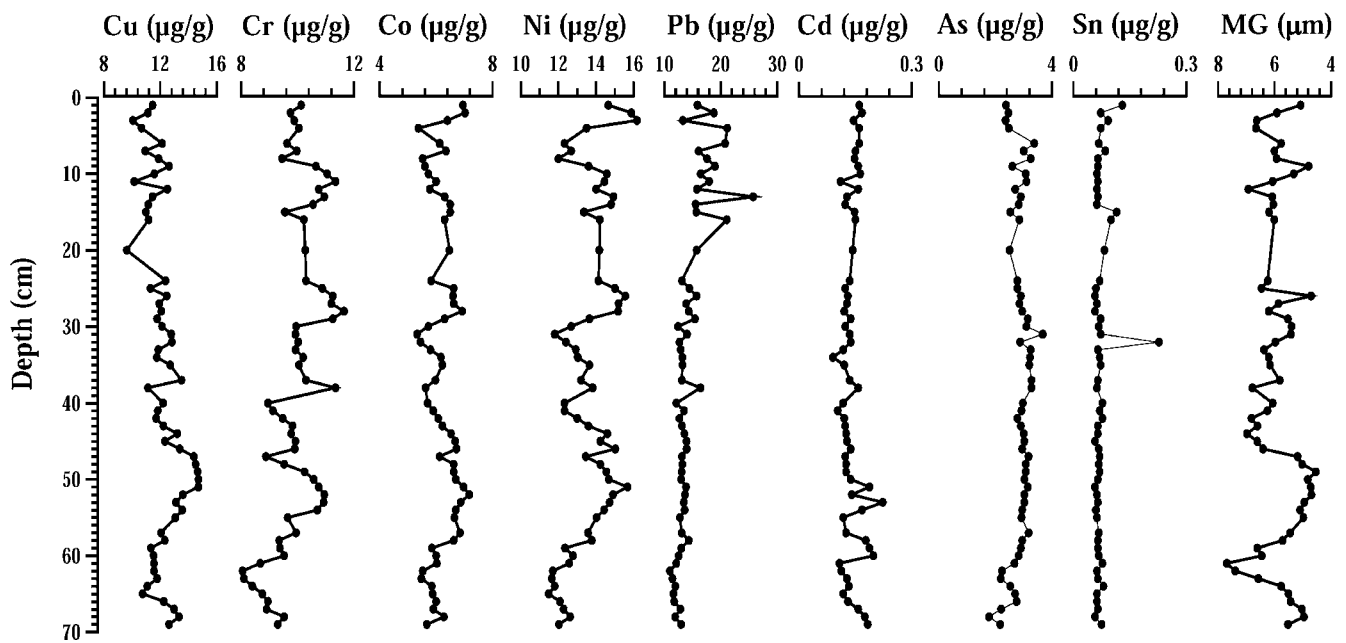
where  $[\text{Pb/Sc}]_{\text{upper continental crust}}$  (2.43) is taken from upper continental crust (Wedepohl, 1995). This value is slightly lower than the average Pb/Sc value (3.07) calculated from 60 to 69 cm in the Core S-385-B2, and the “background” value (Pb/Sc = 3.9) in EGR peat bog (Shotyk et al., 1998). SR is sedimentation rates and calculated from weighted varve thickness for each sample.

Profiles of “total,” “lithogenic,” and “anthropogenic” Pb flux are shown in Figure 7. They show an increasing pattern in the profiles. Total Pb flux varies between  $2.1 \mu\text{g cm}^{-2} \text{yr}^{-1}$  and  $29.1 \mu\text{g cm}^{-2} \text{yr}^{-1}$ , with mean values of  $8.4 \mu\text{g cm}^{-2} \text{yr}^{-1}$ . Lithogenic Pb flux varies between  $1.4 \mu\text{g cm}^{-2} \text{yr}^{-1}$  and  $21.1 \mu\text{g cm}^{-2} \text{yr}^{-1}$ , with mean values of  $5.9 \mu\text{g cm}^{-2} \text{yr}^{-1}$ . Anthropogenic Pb flux varies between  $0.1 \mu\text{g cm}^{-2} \text{yr}^{-1}$  and  $12.3 \mu\text{g cm}^{-2} \text{yr}^{-1}$ , with mean values of  $2.4 \mu\text{g cm}^{-2} \text{yr}^{-1}$ . The anthropogenic Pb fluxes were relatively low at around  $0.7 \mu\text{g cm}^{-2} \text{yr}^{-1}$  prior to 1945, slowly increased after 1945, reached a sidestep (between 1940s and 1970s) with a mean value of  $1.8 \mu\text{g cm}^{-2} \text{yr}^{-1}$ . Second highest value period was between 1980s and 1990s with mean value of  $5.9 \mu\text{g cm}^{-2} \text{yr}^{-1}$ .

#### ANTHROPOGENIC INFLUENCE

In the Arctic, pollutants were often ascribed mainly to long-range sources from atmospheric and sea ice transportation. However, in Lake Bolterskardet, it is reasonable to speculate that local sources (especially for coal mining activity and coal combustion in Long-yearbyen) also contribute pollutants in the sediments.

Figure 8 shows the history record and the anthropogenic Pb flux in Lake Bolterskardet. On Svalbard, coal mining operations began in

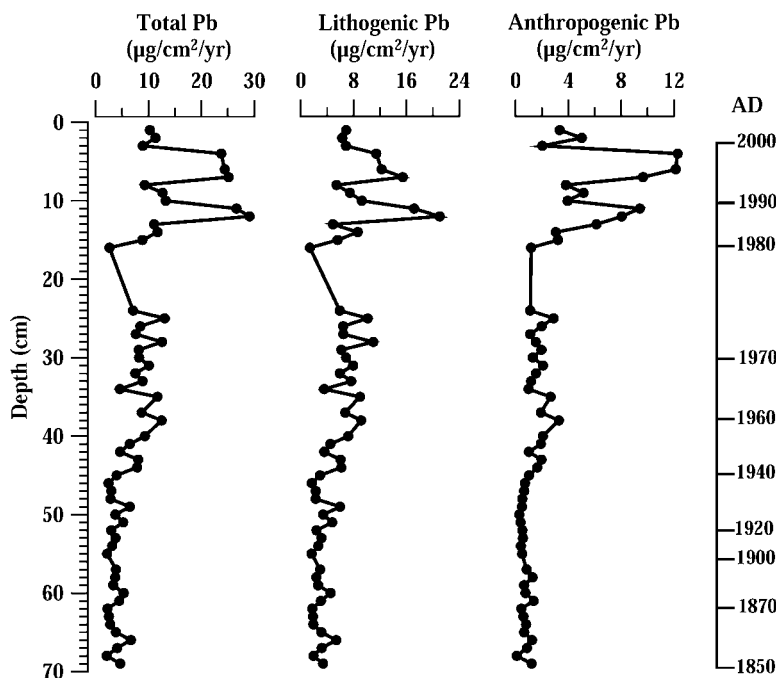


**FIGURE 6.** Profiles of heavy metals (Cu, Cr, Co, Ni, Pb, Cd, As, and Sn) concentration and median grain size (MG). Right side shows time scale from varve counting.

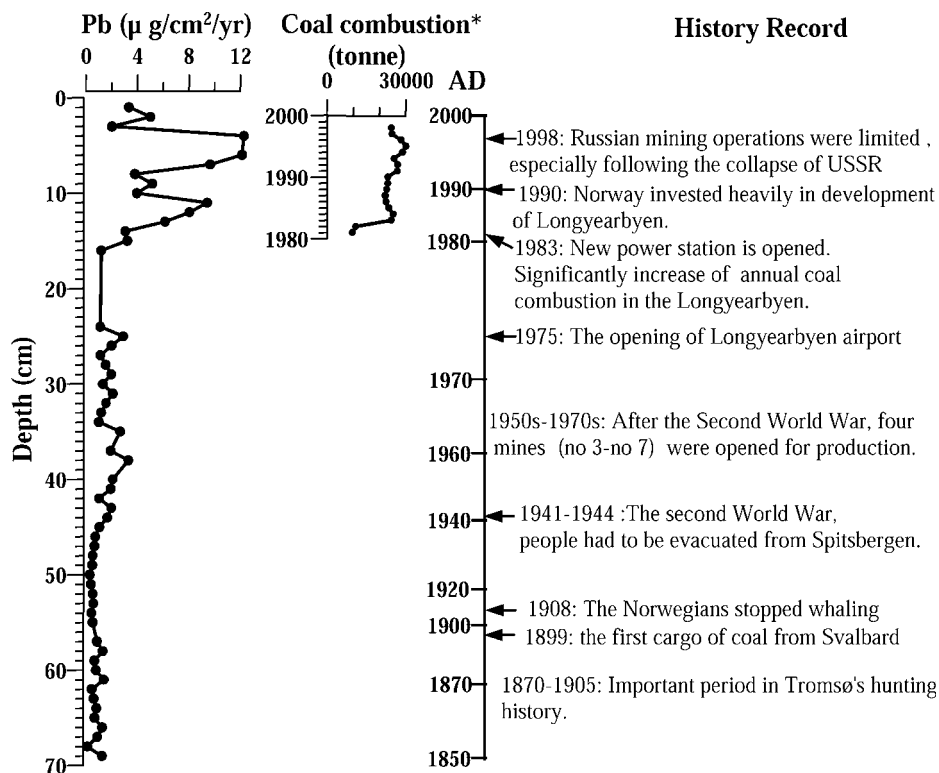
the early 20th century, steadily increased since the late 1920s, and temporarily ceased during the Second World War. In Longyearbyen, a rapid increase in production occurred between 1960 and 1980 (Svalbard Statistics, 2003). Annual coal combustion in Longyearbyen quickly increased from ca. 10,000 tonnes in 1981 to 25,000 tonnes in 1984 (Fig. 8) (Rose et al., 2004). Further, seasonal variations of coal combustion and ash production for the Longyearbyen power station (Rose et al., 2004) show a similar pattern with observed data (e.g.,  $\text{SO}_4$ , trace metals) in Ny-Ålesund (Jaworowski, 1989; Berg et al., 1996; see in Rose et al., 2004). It is suggested that the coal combustion is an important source for pollution on Svalbard. The anthropogenic Pb fluxes in Lake Bolterskardet generally coincide with the historical

record. Although mining activities in Longyearbyen are limited after 1990s, increase of coal combustion in the Longyearbyen power station should have an important role in the high anthropogenic Pb fluxes in the sediments of the lake during the last two decades.

Figure 9 shows diagrams of sulfate record from the ice core in southern Greenland (Mayewski et al., 1990), heavy metals in central Greenland snow and ice (Candelone et al., 1995), acidity concentration in the Austfonna and Høghetta ice cores on Svalbard (Simões and Zagorodnov, 2001), and anthropogenic Pb flux in this study. Although Greenland and Svalbard are under different atmospheric circulation regime, the variations of anthropogenic Pb fluxes before 1970 in Bolterskardet Lake show a similar pattern with both the data from



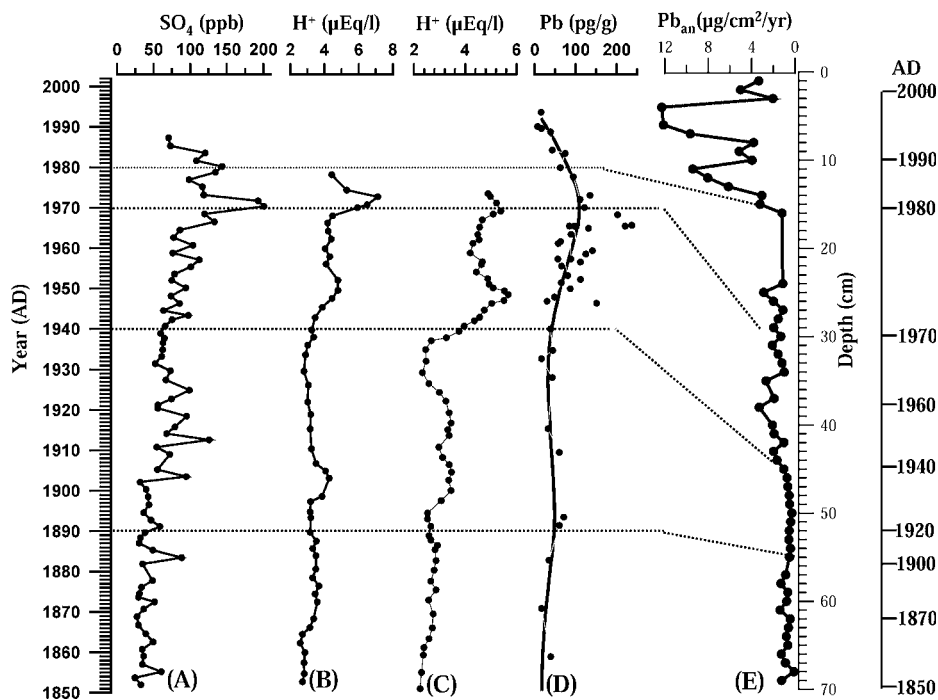
**FIGURE 7.** Profiles of "total," "lithogenic," and "anthropogenic" Pb fluxes in Lake Bolterskardet. Right side shows time scale from varve counting.



**FIGURE 8.** Anthropogenic Pb flux in Lake Bolterskardet, annual coal combustion in Longyearbyen, and historical record in Svalbard. \*The data of annual coal combustion (tonnes) for Longyearbyen is from Rose et al. (2004).

Greenland and the data from Svalbard. However, both the sulfate record and heavy metal data in the Greenland show a decreasing trend during the last two decades. Data from other studies in the Arctic also suggested a decreasing trend in contamination in recent two decades, which was explained as the reduction in total U.S. Pb emissions (e.g., Boutron et al., 1991; Shotyk et al., 2003; McConnell et al., 2002). Discrepancy in the pollution record from the two sites may partly be explained as the different atmospheric circulation between Greenland and Svalbard. Greenland is strongly affected by pollutant emissions from North America (Kahl et al., 1997), whereas Svalbard is affected

by long-distance transport of contaminants from industrial areas, including eastern and western Europe and Canada (Staebler et al., 1999; Goto-Azuma and Koerner, 2001). It suggests the complex of long-range transport contaminants in the regional and global scale in the Arctic. On the other hand, local human activities (coal combustion) may be also an important source of heavy metal contamination since Longyearbyen has quickly developed, including as a travel site in the last two decades. Recent increase of contaminants in Svalbard has been demonstrated in previous studies. For example, on the basis that heavy metal concentration in the surface sediments of Svalbard fjords



**FIGURE 9.** Comparison diagrams of sulfate and Pb concentration in the Greenland snow and ice, acidity concentration in the ice cores of Svalbard, and Pb flux in this study. (A) The data are from Mayewski et al. (1990, [ftp://ncdc.noaa.gov/pub/data/paleo/icecore/antarctica/taylor/tdchem](http://ncdc.noaa.gov/pub/data/paleo/icecore/antarctica/taylor/tdchem)). (B) Acidity concentration in the Austfonna ice cores on Svalbard (Simões and Zagorodnov, 2001). (C) Acidity concentration in the Høghetta ice cores on Svalbard (Fujii et al., 1990; Simões and Zagorodnov, 2001). (D) Pb concentration in central Greenland snow and ice (Candelone et al., 1995), which also combines the Pb data from Boutron et al. (1991) and Murozumi et al. (1969). (E) This study.



is much higher than assumed background, Holte et al. (1996) suggested it may be a result of terrestrial water drainage of coal particles originating from local coal stores and industrial activities. The sulfate record from ice cores in Svalbard also supported important coal mining activities (Isaksson et al., 2003). Based on the study of polycyclic aromatic hydrocarbon (PAH) compounds and polychlorinated biphenyl (PCB) in lacustrine sediments, Rose et al. (2004) concluded that PHA may mainly come from local sources. Our result confirmed previous suggestions that Svalbard is strongly affected by anthropogenic pollution (Rose et al., 2004). The record of heavy metals in the sediments is likely to be a combination of both long-range and local sources.

## Conclusions

Laminated sediments from Bolterskardet Lake are interpreted to be varves on the basis of thin section observation and independent data of  $^{137}\text{Cs}$  and  $^{210}\text{Pb}$ . The high sedimentation rate and varved sediments make Lake Bolterskardet a good site for studying history of heavy metal pollution in the region. The variations of Cu, Cr, Co, and Ni concentrations show an inverse pattern with the median grain size. It suggested that the particle size has a significant role in the accumulation and enrichments of heavy metals.

In the profiles of studied heavy metals, only Pb concentrations show a significant increase from the lower parts to the upper parts of the profile. Anthropogenic Pb flux also shows an increasing pattern in the profile. The anthropogenic Pb fluxes were relatively low at around  $0.7 \mu\text{g cm}^{-2} \text{yr}^{-1}$  prior to 1945, slowly increased after 1945, and reached a sidestep (between 1940s and 1970s) with mean value of  $1.8 \mu\text{g cm}^{-2} \text{yr}^{-1}$ . Second high value period was between 1980s and 1990s with mean value of  $5.9 \mu\text{g cm}^{-2} \text{yr}^{-1}$ . The variation of anthropogenic Pb fluxes before 1970 in Lake Bolterskardet shows a pattern similar to both the data in Greenland and the data in Svalbard. However, a quickly increased trend of anthropogenic Pb fluxes has been observed during the last two decades in Bolterskardet Lake. It suggests that local human activities (e.g., coal combustion) may have an important role for the increase of the anthropogenic Pb flux in the recent two decades. The record of heavy metals in the sediments is likely to be a combination of both long-range sources and local origin.

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