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History of heavy metal accumulation in the Svalbard area: Distribution, origin and transport pathways☆

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ABSTRACT

In this study temporal changes of Pb, Zn, Cd and Cu concentrations were studied in 19 dated sediment cores collected from Svalbard fjords and the Barents Sea. The main aim was to study spatial and historical variations in heavy metal concentrations, deposition rates and sources in the context of different metal transport pathways. Metal concentrations ranged from 5.7 to 45.8 mg kg^{-1} for Pb, from 13.4 to 54.5 mg kg^{-1} for Cu, from 0.01 to 0.90 mg kg^{-1} for Cd and from 55.6 to 130.4 mg kg^{-1} for Zn. Some fjords were unpolluted by heavy metals while in others a clear signal of metal enrichment was found (outer Kongsfjorden, Hornsund, Adventfjorden). Large-scale processes such as atmospheric and oceanic transport were found to be important drivers of heavy metal contaminant distribution. The significance of global drivers varied among the fjords, due to coupling with local processes. Outer fjord parts, the most impacted by oceanic transport, were characterized by the excess $^{206}Pb/^{207}Pb$ values of ~1.17, while the inner basins were characterized by the excess $^{206}Pb/^{207}Pb$ of ~1.14 suggesting possible different importance of Pb sources (marine currents and atmospheric transport).

1. Introduction

Since the beginning of the industrial era (from ~1850) loading of metals to the environment due to human activities have increased nearly 10 times (Nriagu, 1996). These human activities intensified substantially after World War II. The Arctic has been recognized as a sink of contaminants emitted in the northern hemisphere (AMAP, 1998). A portion of the introduction of heavy metals to the Arctic environment comes from natural sources, e.g. rock weathering, while the remainder originates from several anthropogenic sources. The most important anthropogenic sources are long-range atmospheric transport of metals originating from industry, coal burning and transportation (Pacyna et al., 1984). Air mass tracking has indicated that metals present in the atmosphere over Svalbard have been emitted in Europe and Russia (Reimann and de Caritat, 2005). The system of ocean currents also transports heavy metals from western Europe northward (Gobeil et al., 2001). Drifting sea ice may also transport large amounts of contaminants trapped within it as it forms along the Siberian coasts (Pavlov et al., 2013). Apart from global contaminant pathways Svalbard can also receive contaminants from local sources, e.g. coal mining activities (Rose et al., 2004). In seawater, metals readily sorb onto sinking matter and deposit at the sea bed. Thus, sediments act as archives of the history of environmental contamination.

The heavy metal contamination of the Svalbard fjord sediments has been monitored by AMAP since the 1990s (e.g. AMAP, 1998). Most of the older (eg. Holte et al., 1996) and more recent (Lu et al., 2013; and Grotti et al., 2013) studies concentrate on surface sediments of selected fjords and report trace metal concentrations close to the natural background level. A better understanding of the contributions of different sources of heavy metals and their temporal variability in the Arctic is inevitable (Shotyk et al., 2005) especially in an era of climate change that affects the strength and directions of global processes (Macdonald et al., 2005).

Our aim was to study the spatial and historical variations in heavy metal concentrations, deposition rates and sources in seven fjords of the Svalbard archipelago and in the Barents Sea off Svalbard. These locations are characterized by different environmental conditions (e.g. influenced by different water masses, geological bedrock, human activity) and different transport pathways. The concentrations of different trace metals (Pb, Cd, Zn, Cu, Fe, Mn, Cr) were measured in ^{210}Pb-dated sediments to study changes in the metal concentrations within the last 150 years and to calculate

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metal deposition rates. The stable lead isotopic ratios ($^{206}Pb/^{207}Pb$, $^{208}Pb/^{206}Pb$) were used to identify the potential sources of Pb stored in Arctic sediments.

1.1. Study area

The Svalbard archipelago is located 400 miles north of Europe (from 74° to 81° N, Fig. 1). Fjords of western Svalbard are influenced by warm waters of Atlantic origin transported along the shelf break by the West Spitsbergen Current (WSC), a continuation of the Norwegian Atlantic Current (NwAC) (Walczowski and Piechura, 2006). Waters of cold Arctic origin flow to the south along the eastern part of the archipelago, with the East Spitsbergen Current, and partially continue over the shelf south and west of Svalbard with the Sørkapp Current (SC) (Fig. 1). The West Svalbard fjords are filled with a mixture of Arctic and Atlantic waters entering from the shelf as well as locally produced intermediate and winter-cooled water. The sources of freshwater in Svalbard fjords include direct input of glaciers melting, local precipitation, sea ice melt, and river runoff (Svendsen et al., 2002). Seven Svalbard fjords were selected for the study. Storfjorden is a large embayment located in south-eastern Svalbard. The dominant water mass is of Arctic origin as a sill at the fjord entrance significantly limits the inflow of Atlantic water. Hornsund is affected by cold waters from the SC and warm waters from the WSC. Adventfjorden, which is a little branch of Isfjorden, receives a mixture of WSC and shelf waters, and also a large input of freshwater from a glacial river. Kongsfjorden, located in northwest part of Svalbard, consists of two sub-basins. The inner part is strongly influenced by glacier outflow while the outer one is mostly filled by modified Atlantic water from WSC (Cottier et al., 2005). Magdalenefjorden and Smeerenburgfjorden are impacted by a mixture of Atlantic and sea ice melt waters entering from the shelf, and also by outflow from local glaciers. Rijpfjorden is a long fjord at northern Nordaustlandet, dominated by Arctic water of local origin. The Barents Sea is a complex system with large regional differences, being influenced from the south by warm and saline Atlantic water of the Norwegian Atlantic Current, and from the north by cold waters from the Arctic Ocean (Loeng, 1991).

Svalbard is influenced by inflow of air from the eastern and south-eastern directions, thus air masses reaching the fjords originate mainly from Europe and Russia (Reimann and de Caritat, 2005; Isaksen et al., 2016). Air mass transport varies seasonally and is influenced by Arctic Oscillation (AO). The largest transport of air masses and contaminants to the Arctic occurs in the winter (80% of south-north transport). During winters characterized by a positive AO index the air masses and airborne contaminants are transported to the Arctic mainly from Europe and eastern North America. Under a negative AO index the transport of air masses from the direction of Eurasia prevails. In the summer, northward transport of air masses from low latitudes weakens and the air masses originate from North America and Europe (Macdonald et al., 2005).

The largest natural sources of mineral particles containing metals to the marine bottom of the fjords are local rocks eroded by the glaciers, airborne particles from local rock weathering and rivers draining soils from the local catchment. Generally, Svalbard is built from the Caledonian Fold belt comprising mainly Precambrian rocks of the Hecla Hoek formation (Magdalenafjorden, Smeerenburgfjorden and Rijpfjorden; Hjelle, 1993). Eastern Kongsfjorden is built of Devonian iron oxide-rich Old Red formation deposits. The other parts of the fjord are formed by Proterozoic metamorphosed rocks, Paleozoic sedimentary rocks and coal deposits. Mezozoic

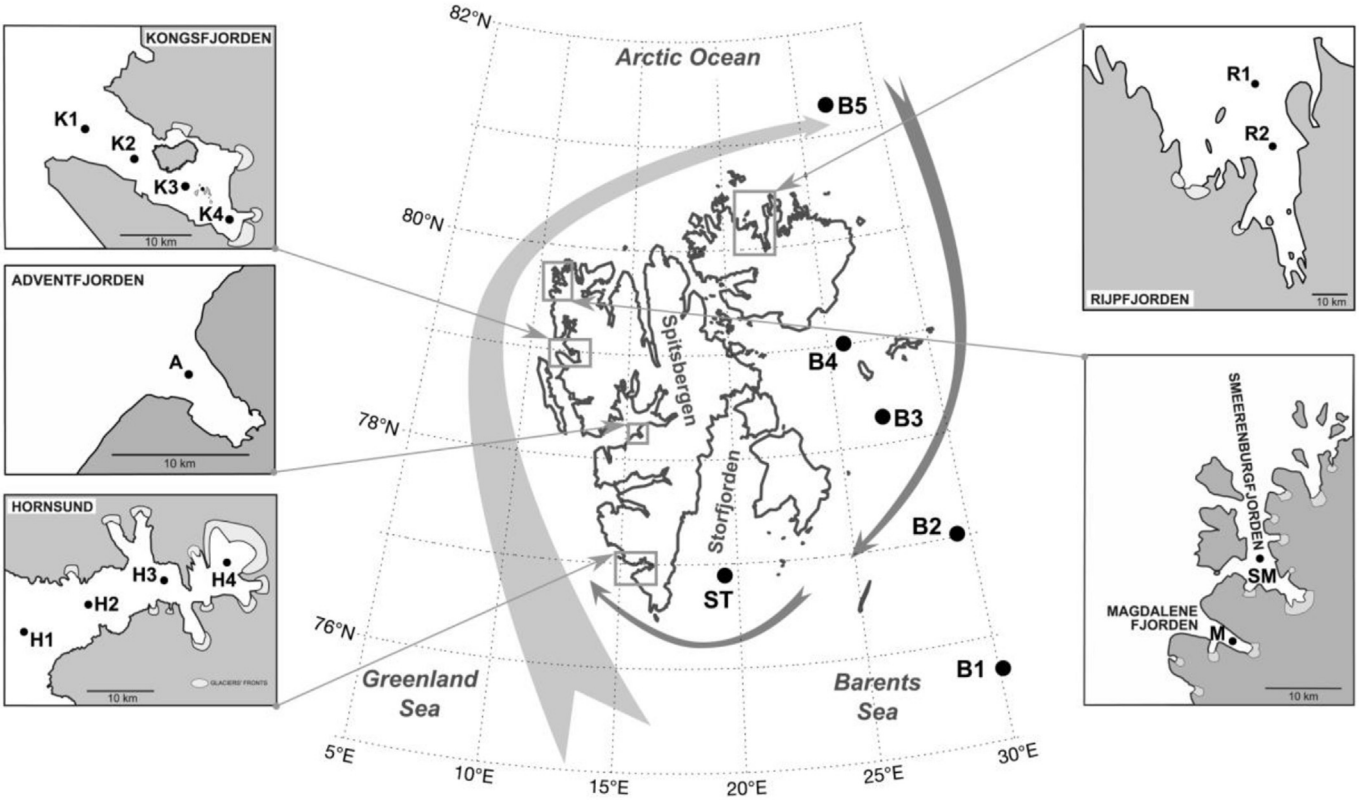

Fig. 1. The map of a study region with black dots indicating 19 sampling stations: Storfjorden (ST), Hornsund (H1, H2, H3, H4), Adventfjorden (A), Kongsfjorden (K1, K2, K3, K4), Magdalenefjorden (M), Smeerenburgfjorden (SM), Rijpfjorden (R1, R2) and the Barents Sea (B1, B2, B3, B4, B5). The warm West Spitsbergen Current inflow is marked by light grey arrow while the cold Arctic waters of East Spitsbergen and Sørkapp Currents are marked by dark grey arrows.