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Accumulation of atmospheric radionuclides and heavy metals in cryoconite holes on an Arctic glacier

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HIGHLIGHTS

- Extremely high radioactivity was found in Arctic cryoconites.
- Cryoconites retain and concentrate airborne radionuclides on glacier surfaces.
- Heavy metals in cryoconites exceed natural background.
- Strong anthropogenic contribution to heavy metal deposition in the Arctic was noted.

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ABSTRACT

Surface of glaciers is covered by mineral and organic dust, together with microorganisms forming cryoconite granules. Despite fact that glaciers and ice sheets constitute significance part of land surface, reservoir of freshwater, and sites of high biological production, the knowledge on the cryoconite granules still remain unsatisfactory. This study presents information on radionuclide and heavy metal contents in cryoconites. Cryoconites collected from the Hans Glacier in SW Spitsbergen reveal high activity concentrations of anthropogenic (^{238,239,240}Pu, ^{137}Cs, ^{90}Sr) and natural (^{210}Pb) radionuclides. The ^{238}Pu/$^{239+240}$Pu activity ratios in these cryoconites significantly exceed the mean global fallout ratio (0.025). The ^{238}Pu/$^{239+240}$Pu ranged from 0.064 to 0.118. The $^{239+240}$Pu/^{137}Cs varied from 0.011 ± 0.003 to 0.030 ± 0.007. Such activity ratios as observed in these cryoconites were significantly higher than the values characterizing global fallout, pointing to possible contributions of these radionuclides from other sources. Heavy metals (Pb, Cd, Cu, Zn, Fe, and Mn) in cryoconites exceed both UCC concentrations and local rocks' concentrations, particularly for cadmium. The concentration ratios of stable lead isotopes (^{206}Pb/^{207}Pb, ^{208}Pb/^{206}Pb) were determined to discriminate between the natural and anthropogenic sources of Pb in cryoconites and to confirm the strong anthropogenic contribution to heavy metal deposition in the Arctic. In investigated cryoconite holes, two groups of invertebrates, both extremophiles, Tardigrada and Rotifera were detected. Our study indicate that cryoconites are aggregates of mineral and organic substances on surfaces of glaciers are able to accumulate large amounts of airborne pollutants bound to extracellular polymeric substances secreted by microorganisms.

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1. Introduction

Polar regions suffer the most pronounced environmental changes due to global climate change (e.g. Karcher et al., 2010; Macdonald et al., 2005). The Arctic receives contaminants by long-range global transport (AMAP, 1997, 2004). It is important to understand the polar ecosystems' exposure to products of human activity because the number of additional contamination sources may rise in future due to global climate changes. One example is the increase of air temperatures that results in melting of ice and permafrost and the subsequent release of contaminants and

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potential pathogens accumulated over many years (Hodson, 2014; Edwards, 2015; Cook et al., 2015a). The atmospheric circulation pattern and the precipitation intensity are also expected to change, which also may escalate contamination transport and elution from the atmosphere (Macdonald et al., 2005; Schiedek et al., 2007; Pouch and Zaborska, 2015).

Polar regions, the Arctic in particular, are very vulnerable to contamination transported from middle latitudes by atmospheric circulation, marine currents, rivers and sea ice (Karcher et al., 2010; Macdonald et al., 2005; Zaborska and Carroll, 2010). Heavy metals from fossil fuel combustion and mining are the major contaminant group of growing concern in the Arctic (Gobeil et al., 2001; AMAP, 1997). The Arctic region is also especially vulnerable to radioactive contamination originating from global (Dahlgaard, 1995; Føyn and Sværen, 1997; Karcher et al., 2010; Zaborska et al., 2010) and local sources (Pogrebov et al., 1997; Smith et al., 2000). The principal sources of these anthropogenic radionuclides in the terrestrial Arctic are global (stratospheric) fallout from atmospheric nuclear tests and local (tropospheric) fallout from nuclear tests conducted at Novaya Zemlya (Salbu, 2001; UNSCEAR, 2000). Until now, the research studies of contamination of the European Arctic by radionuclides and heavy metals concentrated on the marine environment because the number of contamination sources is higher there than in the terrestrial environment (e.g. AMAP, 1997; Bazzano et al., 2015; Heldal et al., 2002; Lu et al., 2013; Macdonald et al., 2005; Zaborska et al., 2010). Little is known particularly on contamination of the terrestrial environment by radionuclides; data are available for a few sites (Dowdall et al., 2003; Gwynn et al., 2004a,b; Łokas et al., 2013a,b; 2014).

Cryoconites granules consist of a mixture of mineral particles, organic substances and living microorganisms, common among them being the filamentous cyanobacteria (Wharton et al., 1985; Takeuchi et al., 2001, 2010; Cook et al., 2015a; Kaczmarek et al., 2016; Zawierucha et al., 2015). Because of low albedo (Takeuchi, 2002), cryoconites facilitate melting of ice leading to formation of holes on the glacier surface (Gribbon, 1979; Podgorny and Grenfell, 1996; MacDonell and Fitzsimons, 2008; Cook et al., 2010). The cryoconite holes are less frequent in steep ablation-prone parts of glaciers where, due to the high volumes and energy of run-off, they cannot develop or have a short lifespan, especially those in the Arctic (Gribbon, 1979; Takeuchi et al., 2000; MacDonell and Fitzsimons, 2008, 2012). Once they develop, the cryoconites and cryoconite holes themselves contribute to glacier ablation (Fountain et al., 2004, 2008; Bøggild et al., 2010; MacDonell and Fitzsimons, 2008, 2012). Current changes in the Arctic environment may result in release of cryoconite-bound contamination to marine biota and their transfer along the trophic chain (e.g. DiGregorio et al., 1978).

Isotope ratios of radionuclides found in Alpine cryoconites point to global fallout as one of the sources of their radioactive contamination. These observations indicate that cryoconite material may persist on glaciers for periods as long as 40 years (Bossew et al., 2007; Tieber et al., 2009). Occurrences of cryoconite-derived material with high radionuclide contents in the glacier forefront indicate that the cryoconite granules can be retained on the glacier surface and deposited at the terminus after ice melts out (Łokas et al., 2014).

The objectives of this study are the following: (1) to evaluate the level of contamination by artificial (^{238,239,240}Pu, ^{137}Cs, ^{90}Sr) and natural (^{210}Pb) radionuclides and trace metals (Pb, Cd, Cu, Zn, Fe, Mn) in cryoconites collected from the surface of tidewater Hans Glacier in south-west Spitsbergen and (2) to distinguish the sources of contamination based on artificial radionuclides ratios (^{238}Pu/$^{239+240}$Pu, $^{239+240}$Pu/^{137}Cs, ^{137}Cs/^{90}Sr and $^{239+240}$Pu/^{90}Sr) and Pb isotopic composition (^{206}Pb/^{207}Pb, ^{208}Pb/^{206}Pb). This study is the first attempt to characterize contamination of an Arctic glacier with both the radioactive and non-radioactive metallic elements. Additionally, discussion on the influence of radionuclides and heavy metals on glacier invertebrates with highlights of knowledge gaps is presented.

2. Materials and methods

2.1. Study area and sampling

The Svalbard archipelago is located in the European Arctic between latitudes 74° N and 81° N and longitudes 10° E and 35° E. The archipelago is located close to the confluence of ocean currents and air masses with different thermal characteristics (Humlum et al., 2007), therefore it is one of the most climatically sensitive regions in the world (Rogers et al., 2005). Glaciers and ice caps cover 59% (36.6 km^2) of the 61.7 km^2 area of the Svalbard archipelago (Hagen et al., 2003). Hornsund fjord is located in the southern part of Spitsbergen (Wedel Jarlsberg Land). The fjord is dominated by rugged mountains with elevations exceeding 600 m above sea level (a.s.l.) and coastal plains with marine terraces covered with rich tundra vegetation, dominated during summer by running water and mass wasting (Nawrot, 2011). The climate of Hornsund is influenced by the ocean. However, climatic conditions are variable and are characterized by low temperatures, low precipitation and foehn winds (Migała et al., 2008). The average annual temperature in this area is −3.8 °C (Migała and Wawrzyniak, 2013), and the soils in the northern coast are lithosols and frost-deformed regosols (Skiba, 2013). The sampling was conducted in August 2011 and July 2014 on the Hans Glacier. This is a glacier located on the northern coast of Hornsund, within the limits of South Spitsbergen National Park (Fig. 1).

Hans Glacier is a tidewater, polythermal glacier, about 16 km long, with a 1.3 km wide calving front, an area of 57 km^2, maximum thickness 400 m and surface slopes below 2°. The speed of the Hans Glacier surface at a distance of 0.5 km from the ice cliff is 60 m a^{-1}. The mean calving speed is ca. 250 m^{-1} and annual calving flux is up to 22 × 10^6 m^3 (Jania et al., 1996; Szafraniec, 2002). The front of Hansbreen has been retreating since the end of the 19th century. The total retreat from 1900 until 2008 is about 2.7 km along the central flowline.

Cryoconite samples were collected in 2011 (11, 12, 13) manually, stored in plastic boxes and preserved in 96% ethyl alcohol. Samples (1–10) were collected in 2014 with disposable plastic Pasteur pipettes from the bottom of selected cryoconite holes and transferred to 15 cm^3 plastic test tubes. After collection, the samples were preserved in 96% ethyl alcohol. The depth of cryoconite holes was measured on site with a ruler. The area of each cryoconite was calculated from calibrated photographic documentation using the Olympus cellSens Entry 1.11 software (Table 1). Two samples (14, 15) were collected in 2014 manually, transferred to 30 cm^3 plastic test tubes and not preserved in alcohol. Samples were evaporated and dried at 105 °C in the laboratory. Samples were generally very fine-grained and sufficiently homogeneous that no further treatment was required. The organic matter contents were determined by loss on ignition at 550 °C.

2.2. Radiometric analysis

Samples were analysed by gamma (^{137}Cs, ^{210}Pb), alpha (^{238}Pu, $^{239+240}$Pu) and beta (^{90}Sr) spectrometry. For the ^{137}Cs and ^{210}Pb analyses, samples were packed into 100 mL polypropylene cylindrical containers and were measured using high-resolution gamma spectrometry with a planar HPGe (high-purity germanium) detector with a composite foil window made of carbon fibre which

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