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Hg in snow cover and snowmelt waters in high-sulfide tailing regions (Ursk tailing dump site, Kemerovo region, Russia)

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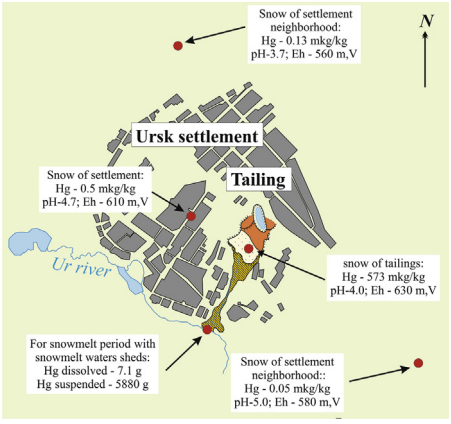
HIGHLIGHTS

- The tailings site corresponds to the high level of snow cover contamination with dust (247–480 mg/m^2 day).
- High value of the average daily yield of Hg (0.123–0.349 mg/m^2 day) was determined on the dumps and in the leakage flux.
- For snowmelt period, about 7 g of Hg in dissolved form (with colloids) and 5880 g as a suspension were taken to the river.

GRAPHICAL ABSTRACT



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ABSTRACT

Gold-bearing polymetallic Cu-Zn deposits of sulphur-pyrite ores were discovered in the Novo-Ursk region in the 1930s. The average content of mercury (Hg) was approximately 120 μg/g at the time.

A comprehensive study of Hg distribution in waste of metal ore enrichment industry was carried out in the cold season on the tailing dump site and in adjacent areas. Mercury concentration in among snow particulate, dissolved and colloid fractions was determined. The maximal Hg content in particulate fraction from the waste tailing site ranged 230–573 μg/g. Such indices as the frequency of aerosol dust deposition events per units of time and area, enrichment factor and the total load allowed to establish that the territory of the tailing waste dump site had a snow cover highly contaminated with dust deposited at a rate of 247–480 mg/(m^2·day). Adjacent areas could be considered as area with low Hg contamination rate with average deposition rate of 30 mg/(m^2·day). The elemental composition of the aerosol dust depositions was determined as well, which allowed to reveal the extent of enrichment waste dispersion throughout adjacent areas. The amount of Hg entering environment with snowmelt water discharge was estimated. As a result of snowmelting, in 2014 the nearest to the dump site

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hydrographic network got Hg as 7.1 g with colloids and as 5880 g as particles. The results obtained allowed to assess the degree of Hg contamination of areas under the impact of metal enrichment industry.

1. Introduction

Snow cover, having a quality to accumulate the contaminants that are absorbed on the surface of the crystals during their fallout, is used as an indicator of air pollution (Siudek et al., 2015; Wang et al., 2015; Garmanova and Larina, 2012). Snow cover pollution occurs in two stages: the first stage is pollution during snowflake formation in clouds, precipitation in the area, or wet deposition during snowfall. The second stage is pollution of already fallen snow after dry deposition of pollutants from the atmosphere or from the underlying soils and rocks (Tranter et al., 1986; Loseto et al., 2004; Larose et al., 2010). The substances accumulated in snow will be reallocated via various processes such as frost penetration and thawing during the winter season (Johannessen and Henriksen, 1978; Colbeck, 1989; Kuhn, 2001).

One of the toxicants, which can be transported by air or ground, is Hg. Sudden changes in environmental conditions (air temperature, relative humidity, insolation, cloudiness, pressure and wind direction) contribute to the rapid transformation of Hg in the atmosphere, after which it is immediately involved in a variety of chemical and photochemical reactions (Lalonde et al., 2002), which then leads to its displacement over great distances. This phenomenon is called the effect of “atmospheric Hg depletion” (AMDE). Thus, Hg can be deposited into the underlying surface and transformed into compounds that are even more toxic. This process is called “atmospheric transport” and is the reason Hg has spread in the global scale and accumulates different regions (Ariya et al., 2004; Skov et al., 2004). In the Arctic/Antarctic ecosystems, which lacks sources of contamination by heavy metals and toxic elements, up to 35 tons of Hg accumulates per year (Soerensen et al., 2016). AMDEs were originally hypothesized to drive large increases in oceanic Hg inputs, but subsequent work shows that approximately half of the deposited HgII is reemitted back to the atmosphere after photochemical reduction in the surface ocean-andsnowpack (Lalonde et al., 2002; Kirk et al., 2006; Dastoor et al., 2015). Rivers are a large source of total Hg to the Arctic Ocean, but prior modeling work has not evaluated their importance for MeHg cycling (Outridge et al., 2008; Fisher et al., 2012; Dastoor and Durnford, 2014; Zhang et al., 2015).

In snow, Hg can be in both inorganic (Ferrari et al., 2002) and organic form (Marusczak et al., 2011). However, in the process of thawing, inorganic Hg can be transformed into the organic species - monomethylmercury (CH_3Hg^+) - a powerful neuro-toxicant, that accumulates in all trophic food chains (Fitzgerald et al., 2007), with concentrations that can be very high (AMAP, 2004). For example, in Arctic regions, without direct sources of pollution, high concentrations of Hg are detected in the liver and tissue of marine mammals and birds (up to 1.32 μg/g) (Wagemann et al., 1998; Campbell et al., 2005). Methylation occurs via biotic and abiotic processes. Biotic, or biological methylation, depends on the microbial activity and concentration of Hg (Barkay et al., 1997), whereas abiotic methylation depends on availability of the methyl donors, which are small organic molecules (methyl iodide and dimethylsulfide or acetate) (Hall et al., 1995; Cello et al., 2006; Fitzgerald et al., 2007; Hammerschmidt, 2007) and large organic components of dissolved organic matter (fulvic and humic acids) (Weber, 1993; Siciliano et al., 2005). Therefore, the snow cover chemistry affects both Hg forms and their transformations (Larose et al., 2010).

The nature of snow thawing and its subsequent formation of snow-melted water runoff is a multi-phase and multi-factor process, which is determined by a complex of physical and geographical conditions. The process of thawing is initiated by solar radiation and earth surface heating, and it, and depends on several factors, such as the amount of snow fall during the cold season; uniformity of its distribution over the area; amount of heat supplied to the territory during snowmelt; relief; wind direction and speed. (Tanasienko et al., 2009; Tanasienko and Chumbaev, 2010). Thus, the total surface runoff and eroded soil mass is defined by the climatic parameters, properties and state of the underlying surface (Colbeck, 1981; Daly and Wania, 2004; Lei and Wania, 2004).

During the winter season, Western Siberia soil freezes to a depth of 180–210 cm. By the beginning of the snowmelt period, in snow-free, the soil can thaw to a depth of 30 cm; the frozen layer is preserved below, which prevents filtration of snow-melted water coming from the surface. As a result, the erosion processes are actively developing in this area (Tanasienko et al., 2009). Artificial geosystems, formed during mining activities, are characterized by instability due to overburdened and steep slopes. The relief energy combined with abiotic factors give rise to the entire spectrum of deflation-erosion processes, which are tens and hundreds of times more intense than these processes under the natural conditions (Panikov et al., 1984). At low temperatures, a cryo-concentration of pore waters occur (Elberling, 2001; Ethier et al., 2012), leading to an increase in the intensity of matter destruction due to more intensive leaching by concentrated melted water in the snowmelts that follow. During the continuing snowmelt, the toxicants found in inside snow cover will migrate, with floodwaters, to the nearest hydrographic network, including surface water, sediments, soil and the underlying rocks. The area of their spread is much higher than the contours of the geochemical anomalies in the snow cover (Zosin et al., 2012).

Hg in snow evaporates from its surface, and this is an additional source of environmental pollution (Fain et al., 2007). In the air, Hg is available in three main species: Hg^0_{gas}, Hg_{inorg} and Hg_{org} (Agency for Toxic Substances …, 1999). Gaseous Hg (Hg^0) is the most common species in the atmosphere (Mitra, 1986). To a small extent, it is deposited in the underlying surface or washed out from the atmosphere by precipitation. In the atmosphere, Hg^0 is oxidized to Hg^{2+}, which quickly precipitates on the surface or is bound and removed from the atmosphere with aerosols. Typically, Hg^{2+} is called reactive gaseous Hg or gaseous oxidized Hg. Some examples include: HgO, $HgCl_2$, $HgBr_2$ and $Hg(OH)_2$ (Gustin and Jaffe, 2010). The small amounts of organic species of Hg are also found in the atmosphere and in the gas phase (Lin and Pehkonen, 1999). Together with the gaseous forms of Hg, some part of it is absorbed into the particles in the air (Xiu et al., 2009).

There are a few papers that consider the behaviour of Hg in snow and snowmelt waters; mostly they are focused on the study of the zones of influence of industrial enterprises in the cities (Dommergue et al., 2003; Larose et al., 2010; Jernelov and Wallin, 1973; Vasilenko, 1985; Talovskaya et al., 2014; Filimonenko et al., 2014). Investigation of the snow cover and snowmelt waters in