



Mechanisms of low-temperature vapor-gas streams formation from sulfide mine waste

Svetlana Bortnikova^a, Nataliya Yurkevich^{a,*}, Anna Devyatova^a, Olga Saeva^a, Olga Shuvaeva^b, Alexei Makas^a, Mikhail Troshkov^a, Natalya Abrosimova^a, Maksim Kirillov^c, Tatyana Korneeva^a, Tatyana Kremleva^d, Nikolay Fefilov^d, Gulnara Shigabaeva^d

^a Trofimuk Institute of Petroleum Geology and Geophysics, Siberian Branch of the Russian Academy of Sciences, Koptug Ave. 3, 630090 Novosibirsk, Russia

^b Nikolaev Institute of Inorganic Chemistry, Siberian Branch of Russian Academy of Sciences, 3, Lavrentiev Avenue, Novosibirsk 630090, Russia

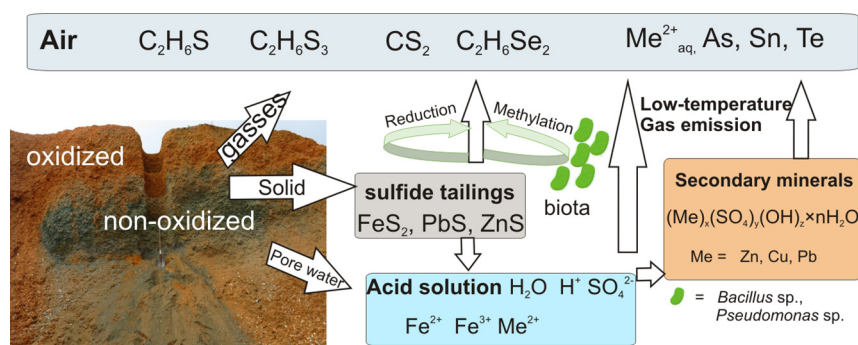
^c Sobolev Institute of Geology and Mineralogy, Siberian Branch of the Russian Academy of Sciences, Koptug Ave. 3, 630090 Novosibirsk, Russia

^d Tyumen State University, Volodarskogo str, 6, 625003 Tyumen, Russia

HIGHLIGHTS

- Sulfide wastes are a source of trace elements, organic gases, and S_{6-8} in the air.
- Metals and metalloids migrate in a vapor-gas stream under ambient conditions.
- Secondary crystalline hydrates are sources of elements in the gas streams.
- Biochemical methylation contributes to the transport of As, Se, S, and Te.
- Low-temperature migration of metals into true gasses is a new direction of research.

GRAPHICAL ABSTRACT



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ABSTRACT

This paper presents experimental data that revealed the potential for chemical element transport by low-temperature vapor-gas streams. The study was conducted on sulfide waste heap sites located in the Kemerovo region, Russia. Condensates of vapor-gas streams were collected and analyzed in the air above the waste heaps and during laboratory experiments using samplers specially designed for this purpose. The gas streams from a waste heaps are complex mixtures consisting of water vapor, sulfur- and selenium-containing compounds (sulfur dioxide SO₂, dimethyl sulfide C₂H₆S, carbon disulfide CS₂, dimethyl disulfide C₂H₆S₂, dimethyl selenide C₂H₆Se, and dimethyl diselenide C₂H₆Se₂), elemental sulfur (S₆, S₇, and S₈) and various chemical elements, including rock-forming elements (Ca, Mg, Na, K, Si, Fe, Al, and Mn), metals (Cu, Zn, Pb, Ni, and Sn), and metalloids (As, Te, and Sb). The main sources of chemical elements in the gas streams are unstable secondary minerals associated with crystalline hydrates: gypsum CaSO₄ × 0.5H₂O, sideronatrite Na₂Fe(SO₄)₂(OH) × 3H₂O, serpierite CaCu₃Zn(SO₄)₂(OH)₆ × 3H₂O, and copiapite (Mg,Zn,Fe²⁺+Fe³⁺)₄(SO₄)₆(OH)₂ × 20H₂O that formed during the oxidation of sulfide minerals. Some of the elements come from pore waters that are acidic, highly mineralized solutions. The mechanism of element migration from the pore waters is as follows: the water vapor phase transports elements in the form of aqueous ions, but complexed species (such as MeSO₄(aq), MeCl(aq), Me(OH)⁺, etc.) remain in the salt residue. A significant contribution to the processes of transformation and transport of elements is

* Corresponding author.

E-mail addresses: BortnikovaSB@ipgg.sbras.ru (S. Bortnikova), YurkevichNV@ipgg.sbras.ru (N. Yurkevich), DevyatovaAY@ipgg.sbras.ru (A. Devyatova), SaevaOP@ipgg.sbras.ru (O. Saeva), olga@niic.nsc.ru (O. Shuvaeva), makasal@ipgg.sbras.ru (A. Makas), troshkovml@ipgg.sbras.ru (M. Troshkov), AbrosimovaNA@ipgg.sbras.ru (N. Abrosimova), Kirillovm@igm.nsc.ru (M. Kirillov), KorneevaTV@ipgg.sbras.ru (T. Korneeva), t.a.kremleva@utmn.ru (T. Kremleva), fefilov@mail.ru (N. Fefilov), g.n.shigabaeva@utmn.ru (G. Shigabaeva).

made by biochemical methylation reactions, which occur in the presence of bacteria producers of methyl groups and are accompanied by the formation of volatile compounds of arsenic, selenium, sulfur, and tellurium.

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1. Introduction

There are numerous natural and anthropogenic sources of atmospheric particulates (Talovskaya et al., 2017; Zhao et al., 2018; Tadros et al., 2018; Shevko et al., 2018) including mining operations (Csavina et al., 2012; Yurkevich et al., 2015). In addition, an assessment on the global health impacts of contaminants in the environment identified metals and metalloids, such as Hg, Pb, As, and Cr, as some of the most toxic pollutants (McCartor and Becker, 2010).

Atmospheric dust and aerosols occur in three main size ranges: ultrafine particles (UFP or Aitken, $<0.1 \mu\text{m}$), accumulative mode ($0.1\text{--}1 \mu\text{m}$) and coarse mode (or dust, $>1 \mu\text{m}$), as is discussed in detail by Seinfeld and Pandis (2006). Contaminants commonly associated with particulate matter (PM) from mining operations are usually most concentrated in the finer particle size fraction ($<2 \mu\text{m}$), which travels greater distance in the environment and poses greater potential risks than coarser particles (e.g., Ravi et al., 2011). Compared with the number of studies on water, soil, and biota pollution, only a few studies explicitly focus on the transport of metals and metalloids in dust and aerosols from mining operations, despite the potential environmental and health risks associated with this specific transport pathway (Csavina et al., 2012). The transport of volatile compounds composed of metals and metalloids from mining materials has not been extensively studied. One of the reasons is a very complicated procedure of sampling, and methodological difficulties in direct elemental analysis of gases. Nevertheless, atmospheric transport of metal and metalloid contaminants has important health and environmental implications and, therefore, should be clarified (Csavina et al., 2012).

Studies of air pollution by the mining and metallurgical industries are mainly devoted to the removal of dust from the surface of tailings (Boyd et al., 2009; Šerbula et al., 2015; Stovern et al., 2016, 2014; Maseki et al., 2017; Bisquert et al., 2017), composition of aerosols with different sizes (fine and ultrafine sizes), atmospheric particulate matter (Csavina et al., 2014; Sanchez de la Campa et al., 2015; González-Castanedo et al., 2014), nanometer-scale in size particles (Dall'Osto et al., 2018), and sulfur dioxide emissions (Šerbula et al., 2015). Detailed analysis of sources, forms of element migration, and the extent of atmospheric pollution in the vicinity of mining operations were reported in the review of Csavina et al. (2012). Atmospheric aerosols in the vicinity of mining operations contain higher concentrations of arsenic, lead, and cadmium than would be expected from natural background values. For these elements, maximum concentrations occur in particles with diameters of $<0.32 \mu\text{m}$ (Csavina et al., 2011). Current directives only regulate some metals and metalloids in PM₁₀ in ambient air and do not contain any legal references concerning the chemical composition of UFP. UFP are respired deep into the lungs where they can be transported directly to the blood stream (Krombach et al., 1997; Park and Wexler, 2008; Valiulis et al., 2008). Therefore, determining the chemical composition of UFP is crucial in quantifying the potential deleterious effects on human health and the environment.

The objective of our study was to measure and characterize UFP emitted in vapor-gas streams from sulfide waste heaps. Although there is literature discussing UFP containing metals and metalloids in hot vapor from smelting and slag dump (Banic et al., 2006), the possibility of chemical element transport in gas streams at low temperatures (ambient air) has not been explored.

Our previous work established that air streams over sulfide tailings are complex mixtures of sulfur-containing gases that are capable of carrying many chemical elements (Bortnikova et al., 2018, 2017, 2016). The obtained results raise questions about possible sources of low-

temperature gas streams, mechanisms of migration and comparative mobility of chemical elements. A series of field and laboratory experiments was carried out to collect condensates of the vapor-gas mixture separated from the sulfide waste heap and solutions corresponding to the composition of the pore waters of the waste.

The purpose of this work is i) to evaluate the possible sources and mechanisms of chemical element transport by low-temperature gas flows; and ii) to determine the composition of gases and trace contaminants in the air over sulfide waste heap.

2. Study area

The Belokluch waste heaps are located in the west of the Kemerovo region, Salair Ridge: 54.463501°N , 85.386201°E (Fig. 1S). The heaps originated in the 1930s and stored the wastes of the cyanidation of the weathering ores of the Belokluch deposit (Ursk ore field).

The Ursk ore field consists of dacite porphyrites, tuffs, quartz-albite-chlorite shales, lying among limestones. The ore field includes the Novo-Ursk, Beloklyuchev and Samoilo deposits and a number of mineralizations, among which 11 ore bodies with pyrite-polymetallic ore have been identified and explored. The structure, morphology, and composition of the ore bodies, the sequence of mineral formation, and the history of mining have been studied and described by many researchers; thus, a brief description of the ore field is given in this paper (Bolgov, 1937; Zerkalov, 1959; Kovalev, 1969; Distanov, 1977). The main sulfide minerals in the ores of the Ursk field are pyrite, arsenopyrite, sphalerite, chalcopyrite, tetrahedrite, and galena. The vein minerals present are quartz, sericite, barite, calcite, and gypsum. The weathered-oxidized profile was mined in two of the three deposits of the Ursk ore field in the 1930s: Novo-Ursk and Beloklyuch. The upper parts of the ore bodies (loose quartz-barite and quartz-pyrite material) were mined, and gold was extracted by cyanidation. Processed ore waste was stored as heaps. Investigation of the low-temperature vapor-gas streams was performed on the waste heaps of processed ore of the Belokluch deposit (Fig. 1S).

3. Methods

3.1. Field sampling

During a field campaign in 2017, Belokluch waste heap was excavated to reveal a 5 m vertical section and sampled in detail. Excavations were made using a shovel to dig through the heap to obtain a detailed record of layering in different parts of the waste pile. The cross section was sampled at intervals based on visual lithological boundaries, reflecting changes in the mineral composition and the degree of oxidation (Fig. 1S). Three parallel samples were taken from each interval. One-liter samples were collected from one parallel sample for pore water squeezing and kept tightly packed in polyethylene bags to prevent contact with air and the evaporation of pore waters. The second was collected for chemical analyses and the third one for laboratory experiments.

In addition, 3 samples of waste (OX-1, OX-2, NOX-1) of 0.5 l were collected and packed «in situ» in a hermetical sterile glass tins for GC/MS (gas chromatography/mass spectrometry) analysis of vapor phase.

The sites on the waste heap for collecting condensates were selected to be similar to the obtained geochemical composition of the samples after laboratory studies.

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