



Mobility and attenuation of arsenic in sulfide-rich mining wastes from the Czech Republic



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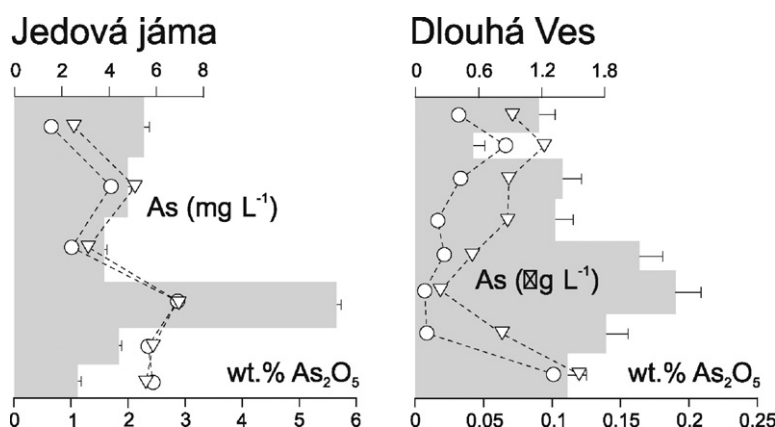
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HIGHLIGHTS

- Two 50 years-old sulfide-rich mining waste dumps were studied.
- Environmental stability of secondary arsenic mineral phases were assessed.
- Different modes of As binding are responsible for different As mobility.

GRAPHICAL ABSTRACT



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ABSTRACT

The mineralogical composition of mining wastes deposited in countless dumps around the world is the key factor that controls retention and release of pollutants. Here we report a multi-method data set combining mineralogical (X-ray diffraction, electron microprobe and Raman microspectrometry) and geochemical (sequential extraction and pore water analysis) methods to resolve As mobility in two 50-years-old mining waste dumps. Originally, all of the As in the mining wastes selected for the study was present as arsenopyrite and with time it has been replaced by secondary As phases. At Jedová jáma mining area, the most of As precipitated as X-ray amorphous ferric arsenate (HFA). Arsenic is also accumulated in the scorodite and Fe (hydr)oxide (with up to 3.2 wt.% As_2O_5) that is particularly represented by hematite. Mining wastes at Dlouhá Ves contain only trace amount of scorodite. Arsenic is primarily bound to Pb-jarosite and Fe (hydr)oxides (especially goethite) with up to 1.6 and 1.8 wt.% As_2O_5 , respectively. The pore water collected after rainfall events indicated high concentrations of As ($\sim 4600 \mu\text{g}\cdot\text{L}^{-1}$) at Jedová jáma, whereas aqueous As at Dlouhá Ves was negligible (up to $1.5 \mu\text{g}\cdot\text{L}^{-1}$). Highly mobile As at Jedová jáma is attributed to the dissolution of HFA and simultaneous precipitation of Fe (hydr)oxides under mildly acidic conditions (pH ~ 4.4); immobile As at Dlouhá Ves is due to the efficient adsorption on the Fe (hydr)oxides and hydroxosulfates under acidic pH of ~ 2.8 . Taken together, As mobility in the ferric

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arsenates-containing mining wastes may significantly vary. These wastes must be kept under acidic conditions or with high aqueous Fe(III) concentrations to prevent the release of As from incongruent dissolution of ferric arsenates.

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1. Introduction

Mining of base and precious metals results in the production of immense quantities of mining waste, mill tailings, and waste related to refining processes. In many cases, mining waste and mill tailings contain an abundance of sulfide minerals, which are of little economic value (mainly pyrite, pyrrhotite and arsenopyrite). Exposure of the sulfide minerals to atmosphere may lead to the formation of acid mine drainage and various secondary precipitates. Weathering processes of mine wastes also involve the remobilization of toxic elements that are released from the sulfides (e.g. As, Cu, Pb, Zn). The concentration of these elements in the pore solutions is mainly controlled by their precipitation together with ferric (hydr)oxides and (hydroxo)sulfates, and if concentrations are sufficiently high, the elements may also combine to form secondary arsenates, carbonates, oxides, silicates, etc. (Drahota and Filippi, 2009; Jambor et al., 2000; Jambor, 2003; Jamieson et al., 2015; Majzlan et al., 2014). Although these reactions can attenuate a large fraction of a hazardous element, dissolved concentrations may still be high depending on the solubility of a specific secondary mineral. Dissolved toxic elements can then be discharged into the surrounding soil and hydrological systems with potentially consequences for the biota (Foy et al., 1978; Hogsden and Harding, 2012; Kelly, 1999; Younger et al., 2002). Understanding the conditions that control the formation/dissolution of secondary products during oxidative alteration of sulfides thus facilitates assessments of potential health risks associated with such dissolution reactions and provides basis for remediation and prevention of off-site dispersion of metal-laden particles.

In the Czech Republic, extensive past mining operations caused severe pollutions. Copious amounts of mining, processing and metallurgical waste were generated during this period and the remediation of these sites proceeds slowly or not at all. The goal of the present study is to understand the processes controlling the migration and sequestration of Cu, Cd, Pb, Zn and, in particular, the mobility of As in sulfide-rich mining waste dumps. To achieve this goal, we have investigated two 50-years-old sulfide-rich mining waste dumps at different mining sites of the Czech Republic. The two sites were actively mined during the last century: one site for As (Jedová jáma, Vejprty) and the other one for Ag, Pb, Zn (Dlouhá Ves). The primary sources of As and heavy metals at these sites are sulfide minerals (mainly arsenopyrite, pyrite, sphalerite and galena), but the alteration processes affecting the sulfides and related mobilization of the released hazardous elements at these sites have not yet been established. In a previous work (Filippi et al., 2015) and here, we combined detailed mineralogical investigation of secondary phases with studies of pore water chemistry that allowed us to identify major scavengers of hazardous elements in the mining waste and to determine the mechanisms controlling elemental mobility.

2. Study sites

The Jedová jáma mine (*Giftschacht* in German, English translation *Poison Shaft*) is located in the eastern part of the historical Vejprty ore deposit in the Krušné hory (Erzgebirge) Mts. (Saxothuringian domain of the Variscan belt of Europe, Bohemian Massif). The subject of exploitation at the Jedová jáma mine were Palaeozoic micaschists and quartzites intensively mineralized by arsenopyrite. The As mineralization is characterized by quartz-dominated gangue with minor phyllosilicate minerals, fluorite, hematite and extensive hydrothermal alteration. Arsenopyrite is the dominant sulfide forming impregnations to massive

accumulations in the ore; pyrite is less frequent. Exploitation of hydrothermal As mineralization at the Jedová jáma mine dates back to the early 19th century and then proceeded during short period between 1924 and 1925 (Velebil and Přebil, 2012). Extensive fluorite exploration in the mine in 1964 generated mining waste that has been spread in a relatively thin and flat layer (up to 1 m) around two ventilation shafts covering approximately 900 m² (Fig. 1). This material was subject of our environmental study.

The polymetallic deposit at Dlouhá Ves is located in the south-eastern part of the historical Havlíčkův Brod ore district (mined from the 13th century) in the Bohemian-Moravian Highlands (Moldanubian zone, Bohemian Massif) (Fig. 1). Hydrothermal activity produced veins with abundant Fe-rich sphalerite and less common arsenopyrite, chalcopyrite, galena, pyrite and pyrrhotite in the quartz-dominated gangue (Hak and Novák, 1973). The hydrothermal Ag–Pb–Zn mineralization at Dlouhá Ves was mined since 1957, with a peak period between 1964 and 1966. Mining activity at that time has resulted in the production of approximately 20,000 t of ore and deposition of several mining waste dumps (Fig. 1) with the height up to 5 m and the area of approximately one hectare, and variable As and Pb contents (from 0.03 to 3.67 wt.% As and from 0.06 to 4.91 wt.% Pb) (Kocourková et al., 2011). The primary sulfides have mostly been oxidized and Fe (hydr)oxides and the minerals of the jarosite-beudantite group are currently the major As carriers in the dump (Kocourková et al., 2011). Ferric arsenates (amorphous ferric arsenate, kaňkite and scorodite) and water-soluble sulfates (alunogen, epsomite, gypsum, halotrichite, melanterite, rhomboclase and rozenite) were identified as minor to accessory secondary phases (Houzar et al., 2011; Kocourková et al., 2011).

The Jedová jáma at the Vejprty is located in a mountainous region (an altitude of 830 m above sea level). The average annual temperature is 4 °C, and the total precipitation averages 900 mm. Dlouhá Ves, located at a lower altitude (490 m above sea level), is characterized by higher average temperature (7 °C) but lower precipitation (approximately 650 mm).

3. Materials and methods

3.1. Materials

At each of the two mining sites, samples were collected from excavation pits in order to gain insight into the stratification of the mining waste dump, to compare vertical zoning, and to collect samples from the interior of the dump. Solid samples were collected and composited along a profile for discrete depth intervals (10 to 30 cm) on the basis of morphological transitions (color, consistency). The solids were collected using a stainless steel hand trowel, stored in sealed plastic bags and transported to the laboratory. In the laboratory, the samples were air-dried and sieved to obtain the <2 mm fraction. Wet sieving was avoided due to the potential loss of soluble As-hosting phases. For bulk chemical analyses, a split subsample was ground in an agate mortar to analytical fineness. The residue of the <2 mm fraction was used for mineralogical and physico-chemical characterization. Each sample was examined with a binocular light microscope to separate different grains for X-ray powder diffraction analysis. The heavy fractions of the samples were concentrated in 1,1,2,2-tetrabromoethane (specific gravity of 2.93 g·cm⁻³). Selected samples and heavy-grain concentrates were prepared for electron microprobe analysis and Raman microspectrometry analysis in the form of polished sections.

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