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Antimony and arsenic speciation in the major waste dump of the abandoned Lojane Sb-As-Cr mine, North Macedonia

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We present the results of mineralogical and geochemical investigations of the solid waste dump material from the abandoned Lojane mine in the northern part of North Macedonia. The geologically unusual Lojane deposit was mined for Sb (stibnite), As (realgar) and Cr (chromite) until 1979. Its waste dumps and tailings were left without any proper rehabilitation. Hence, it represents an extensive source of antimony and arsenic pollution.

In April 2018 we took samples from the major waste dump near Vaksince village. This 70 m long and 20 m wide, relatively ore-rich dump lies parallel to the creek bed of the immediately adjacent Suva creek (NW-SE). In 2017 a Turkish exploration company had dug a ~1.6 m deep trench along the middle of the elongate dump. The samples were taken along this trench using a hand-drill tool (ca. 20 cm deep) and laterally along the exposed profiles.

X-ray fluorescence (XRF) spectroscopy analyses showed that the mean concentrations of Sb and As are 9950 ppm and 32004 ppm, respectively, but can locally rise to extreme values of 24 g/kg for Sb and 200 g/kg for As. Besides Sb and As, very high amounts of Ni (up to 4672 ppm) and Cr (up to 5136 ppm) have been measured.

A combination of powder X-ray diffraction, Raman spectroscopy and SEM-EDS analyses (of polished aliquots) showed that the main primary source of antimony is stibnite with up to 2 at.% of As, while the main primary sources of arsenic are realgar, gersdorffite (up to 9 at.% of Sb) and As-rich pyrite (up to 10 at.% of As). Additionally, minor amounts of Sb and As (up to 1.5 at.%) were detected in violarite ($\text{Fe}^{2+}\text{Ni}^{3+}_2\text{S}_4$) and greigite ($\text{Fe}^{2+}\text{Fe}^{3+}_2\text{S}_4$). Secondary phases of Sb and As are associated with amorphous iron oxyhydroxides (with up to 2 at.% Sb and up to 11 at.% As) and finely crystalline, porous and chemically inhomogeneous Ca-Fe-Sb-As-oxides/hydroxides belonging to the roméite group, with up to 7 at.% of As. Occasionally, dissolved Sb and As reprecipitated as senarmontite/valentinite, Sb_2O_3 , scorodite, $\text{FeAsO}_4 \cdot \text{H}_2\text{O}$ (up to 2 at.% of Sb), annabergite, $\text{Ni}_3(\text{AsO}_4)_2 \cdot 8\text{H}_2\text{O}$, hörnesite, $\text{Mg}_3(\text{AsO}_4)_2 \cdot 8\text{H}_2\text{O}$, and arseniosiderite, $\text{Ca}_2\text{Fe}_3(\text{AsO}_4)_3\text{O}_2 \cdot 3\text{H}_2\text{O}$. These observations are in good agreement with our prior study focussing on the secondary mineralogy of this and other dumps (Kolitsch et al., 2018).

Further phases confirmed, in approximate order of decreasing abundance are: dolomite, serpentine group-minerals, magnesite, gypsum, quartz, talc, vermiculite, clay minerals, micas (muscovite, phlogopite, annite), plagioclase (albite, anorthite), calcite, siderite, chromite, magnesiochromite, spinel, magnetite, vaesite, clinocllore, tremolite, diopside, fluorapatite, rutile, zircon, monazite-(Ce), hydroxylapatite(?), hydroniumjarosite, chamosite, hematite, manganese oxyhydroxides, bornite, chalcopyrite, cobaltite, galena and baryte.

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Kolitsch, U., Đorđević, T., Tasev, G., Serafimovski, T., Boev, I., Boev, B. (2018): Supergene mineralogy of the Lojane Sb-As-Cr deposit, Republic of Macedonia: Tracing the mobilization of toxic metals. *Geol. Maced.*, 32, 95-117.