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The fate of Sb(V) in the Sb-polluted old mine area of Su Suergiu (SE-Sardinia, Italy): the role of secondary mopungite [$NaSb(OH)_6$]

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Antimony (Sb) is widely present in the environment as a consequence of both natural processes and human activities. Extremely high Sb concentrations can be found in present and past mine areas and in their surroundings, representing a serious environmental and toxicological threat.

The abandoned Sb mine of Su Suergiu (Sarrabus-Gerrei mining district, SE Sardinia, Italy), has been exploited from 1880 to 1980; the mined ore was dominated by stibnite (Sb_2S_3), which was processed in the smelter adjacent to the mine. After the closure of the complex, mining and metallurgical residues were dumped without intervention to mitigate their environmental impact. Waters draining Su Suergiu have high Sb concentration (up to 10^3 - 10^4 μgL^{-1}) and the contamination extends several km downstream reaching the Flumendosa River, the main river of south-eastern Sardinia, used in agriculture and domestic consumption.

The mineralogy of outcropping rocks, mine wastes and foundry slags from the mine area was investigated by X-Ray Powder Diffraction (XRPD) and Scanning Electron Microscopy - Energy Dispersive Spectroscopy (SEM-EDS) to understand the oxidation pathways and the role of secondary Sb-bearing minerals on Sb dispersion.

At Su Suergiu, the foundry slag heaps are the main contamination sources. Indeed, the residues of metallurgical processes, consisting of metallic Sb (Sb^0) and Sb_2O_3 (valentinite/sénarmontite) together with carbonates and NaAl-silicate hydrate, are exposed to surface environment and subjected to oxidation and weathering processes. The oxidation of Sb^0 and Sb^{3+} phases leads to the formation of Sb^{5+} that, at the slightly alkaline and oxidizing conditions of surface water draining the Su Suergiu area, is present in solution as dissolved $Sb(OH)_6^-$.

The local geochemical conditions of waters circulating within the foundry slag heaps, affected by the carbonates and Si-Na-Al phases, promote the precipitation of a rare secondary Sb mineral, namely mopungite $NaSb(OH)_6$. On the slag fragments, mopungite occurs as euhedral crystals (about 100 μm), alteration crusts and micro-aggregates filling the fractures; the different occurrences are likely due to the local availability of Na and water circulation.

Results indicate that mopungite is the last forming mineral in the oxidation paths of Sb phases and derives by a dissolution-precipitation process. Due to its relatively high solubility, mopungite act as

a temporary sink, whose effectiveness is conditioned by the hydrological regime and the water physicochemical conditions (T, pH, Eh, etc.).

At Su Suergiu foundry slag heaps the Sb mobility is mainly controlled by mopungite, whereas the role of Fe-bearing compounds, recognized as the main Sb binders in the most part of polluted sites worldwide, is here negligible.

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