Spatial dispersion at a watershed scale of some mining-originated metals in various solid materials

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River sediments in the french Massif Central, the largest metallogenic area in France, are ones of the most mining-impacted. The study area belongs to the Loire River basin, the largest french basin. This study deals with a former Ag-Pb mining district (Miodet sub-basin, 30 km long and 100 km^2) located in the upstream part of the Loire catchment. The french Massif Central was the first area for Ag-Pb production in France. Mining activities in the studied district lasted between 1873 and 1901 and generated waste materials which constitute today dumps on ~ 24,600 m^2. Around 6000 t of Pb and 6 t of Ag were extracted during that period. Nowadays 100,000 m^3 of tailings are still exposed to atmospheric conditions, overhanging the Miodet River.

Both tailings, located upstream of the Miodet watershed, and sediments, collected all along the river, have been sampled to evaluate the spatial dispersion of various metals (especially As, Fe, Mn, Pb and Zn) at a basin scale, 100 years after the end of mining activities. In addition, the aim of this study is to evaluate the accumulation and stability of the metal host phases within the Miodet River as well as to estimate how the anthropogenic activities infer on metals dissemination. The use of complementary approaches (Fig. 1) allow determining the partitioning and the post-depositional redistribution (mobilization/sequestration) of metals and metalloids as well as to assess their mobility.

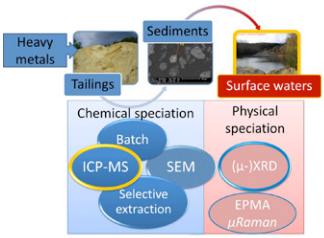

Figure 1: Diagram presenting the applied methodology.

^{34}S enrichment during chemotrophic sulfur oxidation by bacteria

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The three distinct proteobacteria *Paracoccus pantotrophus* (*Pp*), *Tetrathiobacter kashmirensis* (*Tk*) and *Thiomicrospira crunogena* (*Tc*) unanimously rendered kinetic enrichment of ^{34}S in the end product sulfate (SO_4^{2-}) (overall fractionation ranged between -4.6‰ and +5.8‰) during chemolithotrophic oxidation of thiosulfate ($S_2O_3^{2-}$) or tetrathionate ($S_4O_6^{2-}$). *Pp* uses the Sox multienzyme system to oxidize $S_2O_3^{2-}$ (but not $S_4O_6^{2-}$) to SO_4^{2-} without any intermediate formation. *Tk* oxidizes $S_4O_6^{2-}$ directly to SO_4^{2-}, but its $S_2O_3^{2-}$ oxidation proceeds via $S_4O_6^{2-}$ formation. *Tc*, in its turn, oxidizes $S_2O_3^{2-}$ to SO_4^{2-} by depositing extra-cellular elemental sulfur under low pH and oxygen. Molecular biology of sulfur oxidation in the last two species is ill-defined, even though both of them possess *sox* gene homologs in their genomes. All the studied processes were found to start with +ve Δ^{34}S values, subsequent to which (owing to Rayleigh fractionation) Δ^{34}S approached zero as δ^{34}S of produced SO_4^{2-} came close to δ^{34}S of the substrate $S_2O_3^{2-}/S_4O_6^{2-}$. In *Pp* and *Tc*, as the reactions progressed, $\Delta^{34}S_{\text{thiosulfate-sulfate}}$ values became –ve owing to continued ^{34}S enrichment in SO_4^{2-}. Apparent physiological disparities notwithstanding, all the three bacteria exhibited analogous ^{34}S fractionation kinetics during $S_2O_3^{2-}$ oxidation (and also $S_4O_6^{2-}$ oxidation in case of *Tk*), with ^{34}S enrichment rates observed during their peak sulfate-producing stages being almost identical. This indicated the potential involvement of identical S-S bond-breaking enzymes in all these processes. Concurrent proteomic analyses detected the hydrolase SoxB in the actively sulfate-producing cells of all three species. Inducible expression of *soxB* was also observed during $S_4O_6^{2-}$ oxidation by *Tk*, which was particularly interesting because the established Sox pathway has not yet accommodated $S_4O_6^{2-}$ as a substrate. Notably however, no other Sox protein except SoxB was detected in lithotrophically-growing *Tk* cells even after proteomic scanning of its entire peptide map. Instead, several other redox proteins were found over-expressed during $S_2O_3^{2-}/S_4O_6^{2-}$ dependent growth, thereby indicating that there is more to $S_4O_6^{2-}$oxidation than SoxB alone.

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