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Speciation and natural attenuation of arsenic in a shallow subsurface of a natural geochemical anomaly

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The mobility of subsurface arsenic is controlled by sorption, precipitation, and dissolution processes that are tied directly to coupled redox reactions with more abundant, but spatially variable, iron and sulphur species. It is generally accepted that microbial activities play a critical role in the release/attenuation of arsenic, with iron-, sulphur- and arsenic-reducing bacteria stimulated by organic carbon implicated. The area of natural As-Au geochemical anomaly at Mokrsko (central Czech Republic) is well known to have high concentrations of arsenic present in soil and regolith (up to 5690 mg/kg), and surface-, ground- and pore-waters (up to 7.76 mg/L).

Chemical and mineralogical characterization of vertical and lateral changes in shallow subsurface speciation and pore water chemistry showed that two different mechanisms for arsenic attenuation were active: first, arsenopyrite oxidation in the deep regolith by the influx of the oxygenated water resulted in the formation of scorodite, pharmacosiderite, arseniosiderite and As-bearing ferric oxyhydroxides; second, sulphate and As(V) reduction in the shallow hyporheic zone of the stream, resulting in formation of arsenic sulphide, provided attenuation during vertical and lateral infiltration of surface water and shallow groundwater, respectively. This study demonstrates that nanocrystalline realgar can form in shallow (>0.2 m), sulphate-reducing sediment at ambient conditions. Since the precipitation of realgar was not consistent with thermodynamic modelling of the pore-water, precipitation of realgar was attributed to sulphate-reducing microenvironments developed on the surface of organic matter particles. The microbial populations potentially catalysing arsenic and sulphate reduction were characterized by arrA, dsrB and 16S rDNA clone libraries sequencing. Although broad prokaryotic diversity was observed in both environments, the sediment was enriched in alpha proteobacteria, Myxococcales, Firmicutes, TM7 and OP11, and the regolith was enriched in delta proteobacteria, Chloroflexi, Gemmatimonadetes and Acidobacteria. Members of candidate groups BRC1, OD1, SPAM and WS3 and a single Thaumarchaeote were found only in regolith. The diversity of arrA and dsrB was obviously higher in the sediment (11 and 10 different paralogs, respectively) than in the regolith (4 and 7 paralogs, respectively). Anaerobic respiration of sulphate and arsenate is thus common in both environments, but in sediment the group of bacteria possesing this metabolism was more diversified and probably more common.