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The fate of Arsenic in the Carletti Spring System (Viterbo, Italy): a XAS speciation study

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A study aimed at unravelling arsenic (As) speciation in the carbonate sediments and the total suspended particulate (TSP) occurring at the Carletti spring system (CSS), part of the larger Bullicame (Viterbo, Central Italy) system, by means of XAS spectroscopy, has been undertaken.
In the CSS, As occurs as a geogenic anomaly, its mobility and speciation being likely affected by inorganic and/or microbiological processes. The determination of As speciation in the encrustation and TSP, highly diluted samples, is essential to validate the models on the fate of As in this "natural laboratory".

8 rock and 8 TSP samples were collected in the CSS, following relevant changes in temperature, distance from the spring and physicochemical features of the water/TSP/rock interfaces. All solid samples were analysed, without manipulation, by means of X-ray Absorption Spectroscopy at the As K edge in fluorescence mode and at low temperature (range 77-20 K). Experiments on rock and TSP samples were carried out at two beamlines (BM08 and BM26, respectively) at the ESRF facility.

The main results point to an almost constant As(III) over total As ratio over the whole spring system in the rock samples, As(III) being about 30% of the total Arsenic. Detailed investigation concerned with subsamples discriminated by colour (with reference to different abundances and speciation of co-localised microbiologic populations), finding no apparent changes in As speciation. TSP samples, analysed with a specific procedure due to their ultra-diluted nature, appear also constant and slightly enriched in As(III) with respect to the corresponding rocky samples. These data have to be compared to a water environment, which increases its oxidising potential and its pH while increasing the distance from the spring, as testified from the change of the As(III)/total As ratio from an initial 70% down to a final 40%.

This complex set of experimental results will be discussed on the light of two possible interpretive schemes, i.e. a kinetically constrained precipitation mechanism, and a microbiologically constrained change of As speciation, taking also into account of laboratory reference systems where the inorganic and biotic uptake of As(III) and As(V) were reproduced.

Primary authors: Ms MONTEGROSSI, Giordano (IGG-CNR); Dr VENTURI, Stefania (DST - Università di Firenze); Dr BARONI, Tommaso (DST - Università di Firenze); Dr CASENTINI, Barbara (IRSA-CNR); Dr FAZI, Stefano (IRSA-CNR); Prof. COSTAGLIOLA, Pilario (DST - Università di Firenze); Prof. RIMONDI, Valentina (DST - Università di Firenze); Dr PIACENTINI, Agnese (Università di Roma); Prof. MAZZONI, Cristina (Università di Roma); DI BENEDETTO, Francesco (Università degli Studi di Ferrara)

Presenter: DI BENEDETTO, Francesco (Università degli Studi di Ferrara)

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