



Interactions of arsenic with calcite surfaces revealed by in-situ nanoscale imaging

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Arsenic dissolved in water represents a key environmental and health challenge because several million people are under the threat of contamination. In calcareous environments calcite may play an important role in arsenic solubility and transfer in water. Arsenic-calcite interactions remain controversial, especially for As(III) which was proposed to be either incorporated as such, or as As(V) after oxidation. Here, we provide the first time-lapse in-situ study of calcite dissolution and growth in the presence of solutions with various amounts of As(III) or As(V). This was performed at room temperature and pH range 6-9 using a flow through cell connected to an atomic force microscope (AFM), to study the evolution of the (10-14) calcite cleavage surface morphology. Reaction products were then characterized by Raman spectroscopy. In parallel, co-precipitation experiments with either As(III) or As(V) were performed in batch reactors, and the speciation of arsenic in the resulting solids was studied by X-ray absorption spectroscopy (XAS). For As(V), AFM results showed that it interacts strongly with the calcite surface, and XAS results showed that As(V) was mostly incorporated in the calcite structure. For As(III), AFM results showed much less impact on calcite growth and dissolution and less incorporation was observed. This was confirmed by XAS results that indicate that As(III) was partly oxidized into As(V) before being incorporated into calcite and the resulting calcite contained 36% As(III) and 64% As(V). All these experimental results confirm that As(V) has a much stronger interaction with calcite than As(III) and that calcite may represent an important reservoir for arsenic in various geological environments.