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Rapid arsenite oxidation by jacobsite under anoxic conditions: The role of divalent cations

MAREIKE LACINA[1], SYLVAIN BOUCHET[2] AND **LAUREL THOMAS ARRIGO**[1]

[1]University of Neuchâtel
[2]Federal Institute of Metrology (METAS)

Arsenic (As) is a toxic trace metalloid often found in its inorganic forms (As^{III}/As^{V}) at high concentrations in mining affected soils. As^{V} is less toxic and easily immobilized by poorly crystalline iron (Fe) and manganese (Mn) oxides while As^{III} is more toxic and mobile. Iron oxides are known to oxidize As^{III} to As^{V} in the presence of aqueous Fe^{II} under anoxic conditions [1], whereas aqueous Fe^{II} inhibits the oxidation capacity of Mn oxides [2]. In nature, Mn and Fe often substitute for one another. However, the reactivity of mixed minerals towards As under anoxic conditions and how other cations commonly present in the soil solution affect the As oxidation capacity remains largely unknown.

We studied arsenite oxidation during sorption to jacobsite ($(Mn^{II},Mn^{III})Fe^{III}{}_2O_4$) by reacting 1g/L jacobsite with 26 µM As^{III} at pH 7 under anoxic conditions in the presence and absence of 1mM aqueous Fe^{II}, Mn^{II} or Ca^{II} to investigate the role of competing divalent cations. Aqueous samples were collected to measure the As and cation concentrations with ICP-MS. The oxidation state of As, Fe and Mn in the solid phase was measured with their respective K-edge XANES over time.

After 6h, the Fe^{II}-reacted treatment removed As^{III} more efficiently from solution (99%) than the other treatments (average 85%). Results of the solid phase analysis showed the near complete oxidation of As^{III} to As^{V} within 7d in the cation-free (97% As^{V}) and Ca^{II}-reacted treatment (93% As^{V}). In comparison, As^{III} oxidation was slower for jacobsite reacted with Mn^{II} (83% As^{V} after 7d), but still near complete after 28d (96% As^{V}). Reacting jacobsite with aqueous Fe^{II} inhibited As oxidation (40% As^{V}), possibly caused by the Fe^{II}-induced reduction of structural Mn^{III}.

Our results demonstrate the strong As sorption capacity of jacobsite under anoxic conditions. However, the presence of other redox-active elements such as Fe may hinder the oxidation of arsenite on the jacobsite surface. These results have implications for remediation approaches seeking to remove As from contaminated soil.

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