Interaction of selenite with separated Boom Clay fractions – adsorption vs. reduction

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Boom Clay (BC) is currently considered as a possible host formation for geological disposal of radioactive waste in Belgium and the Netherlands. Migration of ^{79}Se, a critical radionuclide due to its relatively long half-life and high mobility, can be influenced by interactions with the clay material. These interactions depend on the Se redox state and include sorption and redox transformations. In particular the reduction of relatively mobile, oxidized Se species such as SeO_3^{2-} or SeO_4^{-2} is of interest as in its reduced state Se forms sparsely soluble phases (e.g. Se^0, FeSe, $FeSe_2$). Regarding the interaction with Se, BC contains various reactive phases such as clay minerals, pyrite, siderite and organic matter.

In this study, we compared the reaction between Se(IV) and BC material with that of its individual size fractions: clay (<2µm) and silt (2-63 µm). Size fractionation was performed to separate the contribution of different BC constituents to the sorption and reduction of Se(IV). In batch reactions, BC material was suspended in 1:1 diluted seawater to which 50 and 150 µM Se(IV) was added. The progress of the reactions was followed by monitoring the composition of the solution and by analyzing solids after various reaction times by X-ray absorption near-edge structure (XANES) analysis.

Concentrations of dissolved Se decreased faster in experiments containing the clay-size fraction compared to those with the silt fraction. XANES analysis indicated that initially most of the solid-bound Se was in the form of Se(IV) and the fraction of Se(0) increased over time. That is, adsorption of Se(IV) accounted for the fast removal of Se from solution which became subsequently reduced to Se(0) at a slower pace. In contrast, adsorption of Se(IV) was only minor in the silt fraction experiments and reduction to Se(0) was the predominant mechanism of Se removal from solution. Although removal of Se(IV) from solution was faster with the clay-fraction, reduction of Se(IV) was more efficient with the silt fraction. This suggests that adsorption of Se(IV) to clay minerals contributes to the retardation of Se(IV) but might delay its reduction to Se(0).