



A long-running in situ experiment in clay: 12 years of the Bitumen–Nitrate–Clay interaction experiment at Mont Terri rock laboratory

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Abstract. In various countries such as Belgium and Germany, deep clay formations are investigated as potential host rock to dispose of radioactive waste. Intermediate-level long-lived bituminised waste, produced from spent fuel reprocessing, is currently foreseen to be disposed of in such a deep repository. This type of waste typically contains large amounts of salts embedded in an organic matrix, of which NaNO_3 is the most prevalent. NaNO_3 , along with radionuclides immobilised in the bitumen matrix, will slowly dissolve and leach out into the surrounding host rock. The transport and reactivity of nitrate in a deep clay formation, in the presence and absence of electron donors coming from bituminised waste (i.e. acetate, H_2), has been studied extensively inside anoxic and water-saturated chambers in the clay in the Bitumen–Nitrate–Clay interaction (BN) experiment. This in situ experiment in Opalinus clay is located in the Mont Terri rock laboratory (Switzerland). The effect of a nitrate plume on redox-sensitive radionuclides is also studied with stable selenium in selenate form as a proxy for ^{79}Se , an important fission product for the long-term dose to humans.

The BN experiment consists of a vertical borehole containing three packed-off intervals, in contact with the surrounding clay through cylindrical sintered stainless steel filter screens. The intervals are saturated with artificial Opalinus clay pore water at a pH between 7.5 and 8, containing all major ions at concentrations specific to the location of the BN experiment but without natural organic matter. Each interval is connected to a water circulation system with a gear pump, a flow meter and water sampling containers. This ensures a chemically well-mixed solution, constant water flow and easy sampling of the interval solutions under anoxic conditions. Water samples are regularly taken to assess the chemical composition and the microbial population in the intervals. An online UV spectrophotometer and pH and redox electrodes are installed in the water circuit of the intervals, to continuously monitor the nitrate and nitrite concentrations, pH and Eh. A given solution can be injected into the intervals to follow up redox reactions affecting nitrate and selenate in situ, in the presence or absence of additional electron donors.

From 2011 to 2019, several injections with nitrate were performed in the intervals to investigate the biogeochemical reactivity of nitrate in the borehole and clay. These tests showed microbial reduction of nitrate, using electron donors from Opalinus clay (e.g. dissolved organic matter or pyrite). Nitrite and nitrogenous gases were formed. Pulses of electron donors (acetate or H_2) boosted the microbial activity and nitrate reduction rate. From 2019 until now, injection tests with selenate, with and without nitrate, have been performed. These tests showed

clearly that the presence of nitrate, a stronger oxidiser, inhibited the microbial reduction of selenate. In the absence of nitrate, the microbial population was able to reduce selenate to selenite and more reduced Se species. The results obtained in this in situ experiment will be summarised in this work, addressing questions concerning the effect of biogeochemical perturbations of the clay on the migration of redox-sensitive radionuclides.