Boom Clay (Belgium)

Description: The distributions of natural occurring REE, U and Th and their isotopes are considered as chemical analogues to critical radionuclides in radioactive waste inventories.

The mobility of these naturally occurring radionuclides in the Boom Clay in realistic long-term geological conditions has been investigated. The key assumption is that, in any geological medium, there is equilibrium between parent and daughter radionuclides provided the geological system remains undisturbed for several millions of years. A range of geochemical processes, such as chemical breakdown, erosion, precipitation of minerals from aqueous solutions by biological or inorganic processes, adsorption on clay minerals etc. can, however, cause isotopic fractioning leading to disequilibrium between isotopes.

A radiochemical study was performed on the Boom Clay solid phase using cores from the boreholes Mol-1, Doel-2b, Zoersel and HADES 2001/4. In order to analyse the liquid phase, pore water from the MORPHEUS piezometer (installed in the HADES 2001/4 borehole), was sampled.

The natural radioactivity of various U-Th isotopes (^{238}U, ^{235}U, ^{234}U, ^{232}Th, ^{230}Th, ^{228}Th) was measured and uranium-thorium series disequilibrium studies applied to determine the mobility/immobility of U and Th isotopes in the Boom Clay.

In the Boom Clay, Th shows a constant profile with depth into the clay, with an average concentration of 11 µg/g in the clay and a concentration below detection limit (< 0.5 µg/l) in the pore water. U has an average concentration of 4 µg/g throughout the Boom Clay; however, higher U concentrations (up to 14 µg/g) are present in the organic-rich layers at the base of the Putte Member, and in septarian nodule levels. In the pore water, U concentrations are generally less than 1 µg/l, but slightly higher values are measured in the organic-rich layers at the base of the Putte Member.

In the solid phases, $^{228}Th/^{232}Th$ isotope activity ratios are all equal to unity within the 2σ uncertainty, indicating that no fractionation has occurred between ^{232}Th and its daughter isotope ^{228}Th. The depth profile of the natural ^{238}U radioactivity follows the depth profile of the U concentration, i.e. a higher natural radioactivity at the base of the Putte Member. $^{234}U/^{238}U$ isotope activity ratio is always equal to 1 within the 2σ uncertainty, indicating a state of secular equilibrium between ^{238}U and its daughter isotope ^{234}U. In general, the $^{230}Th/^{234}U$ isotope activity ratio is also equal to 1 within the 2σ uncertainty, indicating that no fractionation has occurred between ^{234}U and its daughter isotope ^{230}Th. However, in one sample, the $^{230}Th/^{234}U$ isotope activity ratio is significantly larger than 1 within the 2σ uncertainty. This sample corresponds to the lower silt layer of the double band. This silt layer is very coarse, and hence a zone of higher permeability and thus potential higher mobility of species in solution. This probably caused the preferential leaching of uranium, as indicated by the $^{230}Th/^{234}U$ isotope activity ratio.

In the pore water, the $^{234}U/^{238}U$ activity ratios are indicative of radioactive disequilibrium, with $^{234}U/^{238}U$ activity ratios between 1 and 5. An excess of ^{234}U relative to ^{238}U, however, is a common observation in natural waters and is the result of α recoil.

Relevance: The mineralogical, geochemical and radiochemical characterisation of a clay formation allows a better understanding of the natural distribution and mobility of trace elements and radionuclides. It allows information to be obtained on the long-term behaviour of radionuclides in realistic geological disposal conditions and over geological time-periods relevant for the safety assessment.

Position(s) in the matrix tables: This natural analogue illustrates the radionuclide retardation in a mudrock at low temperature from a chemical view point.

Limitations*:* Only applicable for the Boom Clay, but transferability of core preservation and sample preparation techniques, measuring techniques and interpretation of data to other clay formations is possible.

Quantitative information: The study of natural occurring U and Th in an argillaceous formation and associated mobilisation/immobilisation processes make this study a valuable analogue investigation for evaluating real geological time and conditions, and support its use in a PA model.

Uncertainties: Assessment of the uncertainties associated with the qualitative and quantitative information from the study.

Time-scale: The time-scale of the analogue is geological (30 Ma).

PA/safety case applications: ONDRAF/NIRAS published the Safety Assessment and Feasibility Interim Report n° 2 (SAFIR-2) in 2001 which can be considered as a preliminary version of a safety case. The explicit use of results from analogue studies in this report was rather limited.

In section 11.5.3.2.4 "*Alternative safety and performance indicators*", various quantities (activity, concentrations, fluxes) calculated in the assessment were compared with data from a study on the occurrence of natural radionuclides in the host formation.

In section 11.5.5.3.2 "*Testing of models*", the study on the distribution of natural isotopes and rare earths in the Boom Clay was mentioned as an element that can be used to justify the migration parameters used in the assessment. The same section refers to the PHYMOL project in which the stable isotopes H-2 and O-18 and the radioactive isotope C-14 were measured and compared with results obtained with the hydrogeological model for the deep aquifers.

Communication applications: This natural analogue was used in a presentation about the Boom Clay as a host rock for the technical working groups of the MONA local partnership of (05/09/2002).

De Preter P., Marivoet J. & Minon J.-P. (1999). A deep radioactive waste repository in the Boom Clay: The long term safety functions and robust safety indicators. Proceedings ENS Topseal '99 "Commitment to the Future Environment", Volume I, 231-237.

NIRAS/ONDRAF (2001). Technical overview of the SAFIR 2 report. NIROND 2001-05E.

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References:

De Craen M., Delleuze D., Volckaert G., Sneyers A., Put M. (1998) The Boom Clay as natural analogue. Interim Report on the natural radioactivity of the Boom Clay NIRAS/ONDRAF contract CCHO-98/332, KNT 90 98 1042, 99/C072030/MDC/N-5.

De Craen M., Delleuze D., Volckaert G., Sneyers A., Put M. (1999) Behaviour of Natural Isotopes of the U and Th Decay Series in the Boom Clay: A Natural Analogue study. Proceedings ENS Topseal '99 "Commitment to the Future Environment", Volume II, 214-218.

De Craen M., Delleuze D., Volckaert G., Sneyers A., Put M. (2000) U-Th series disequilibrium studies on Boom Clay, a natural analogue study of radionuclide migration in argillaceous sediments. Földtani Közlöny 130/2, 219-228, Budapest.

De Craen M., Delleuze D., Volckaert G., Sneyers A., Put M. (2000) The Boom Clay as natural analogue. NIRAS/ONDRAF contract CCHO-98/332, KNT 90 98 1042, Final report to NIRAS/ONDRAF for the period 1997-1999, R-3444.

De Craen M., Wang L., Weetjens E. (2004) Natural evidence on the long-term behaviour of trace elements and radionuclides in the Boom Clay. NIRAS/ONDRAF contract CCHO-2000-773/00/00, KNT 90 018 1467, Final report to NIRAS/ONDRAF for the period 2000-2003, in prep.

NIRAS/ONDRAF (2001). Safety Assessment and Feasibility Interim Report n° 2.

Added value comments: None

Potential follow-up work: Detailed study of the mobility/immobility of U and Th isotopes in the upper part of the Boom Clay, in particular the transition to the aquifer.

Keywords: Boom Clay – uranium-thorium series disequilibrium studies

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