Palmottu (Finland)

Description: Palmottu is a small uranium occurrence in Nummi-Pusula, southern Finland, about 80 km north west of Helsinki. The site, named according to the adjacent small lake, was discovered 1979 by the Geological Survey of Finland (GTK) during a systematic airborne radiometric survey. Several small uranium occurrences were found during the exploration campaign, but Palmottu, as the most promising, was the only site targeted for extensive geological studies at the beginning of the 1980's. The ore exploration study included the drilling of 62 boreholes (total length 9100 m), of which the deepest reach a depth of about 250 m. However, the uranium deposit was estimated to be too small to be economic: about 1 Mt of U-bearing rock with an average grade of 0.1 % U (Räisänen 1989).

The Palmottu natural analogue study started in 1987 as a co-operative research project between GTK, the Laboratory of Engineering Geology and Geophysics, Helsinki University of Technology (HUT) and the Department of Radiochemistry, University of Helsinki (UHRAD). Later the team was complemented by the contribution of the Technical Research Centre (VTT). The study was mainly financed by the Ministry of Trade and Industry, and later by the Finnish Centre for Radiation and Nuclear Safety (STUK). The results of the Finnish studies are summarised in Suksi et al. 1992 and Blomqvist et al. 1995.

In 1996 the study grew to an international effort jointly funded by the European Commission, GTK, STUK, Svensk Kärnbränslehantering (SKB), Empresa Nacional de Residuos Radioactivos S.A (ENRESA), Centro de Investicationes Energéticas, Medioambientales y Tecnológicas (Ciemat) and Bureau de Récherches Géologiques et Minières (BRGM). The results of the two-phase research programme carried out between 1996 – 2000 are summarised in Blomqvist et al. 1998 and Blomqvist et al. 2000. Currently, only minor sampling activities and site maintenance are being undertaken.

Palmottu is situated within the Proterozoic (1.8 Ga) Svecofennian orogenic belt, at the marginal zone of a late-kinematic, microcline granite situated to the north of the site. The main rock types are garnet-bearing migmatitic mica gneiss (occasionally with cordierite) and pegmatitic granites varying in thickness from plate-like granitic bodies (up to 30 m) to narrow pegmatite veins. The mica gneiss represents a metamorphosed sedimentary unit of varying composition, now indicated by the compositional variation from garnet-cordierite rich to pyroxene-hornblende rich types. Intrusion of pegmatites is thought to be related to the emplacement of the nearby granite.

The Palmottu deposit represents a vein-type uranium mineralisation, rich in thorium (U/Th ratio about 2:1). Uranium is mainly hosted by biotite-bearing pegmatites, where the main U –mineral, uraninite, is typically associated with biotite accumulations. Uraninite grains are frequently surrounded by an alteration rim consisting mainly of uranium silicate (coffinite). The thickness of the discontinuous ore body varies from 1 to 15 metres. The strike of the deposit is parallel to the schistosity having a dip of about 70 – 80^{o} towards the south west, and the deposit partly underlies Lake Palmottu (Figure 1). Structurally, the U deposit is located on the southern limb of a large fold, near the crest. The fold has a near vertical, approximately east-west trending axial plane (Kuivamäki et al. 1990).

The aim of the Palmottu Natural Analogue study was a comprehensive characterisation of transport processes affecting uranium and thorium in a crystalline bedrock environment. Consequently, the study included a wide range of research activities at different scales.

Tectonic mapping and interpretation indicates that the site is situated in a local bedrock block of about 4 km^2. At the regional scale, the Palmottu block is slightly higher topographically than its surroundings. The site is situated near the Salpausselkä III, which is the last of the country-wide ice marginal formations. It was formed when the retreat of the continental ice sheet temporarily stopped about 10,000 years ago. Salpausselkä III is manifested by a wide and thick sand-gravel formation to the north west of Palmottu.

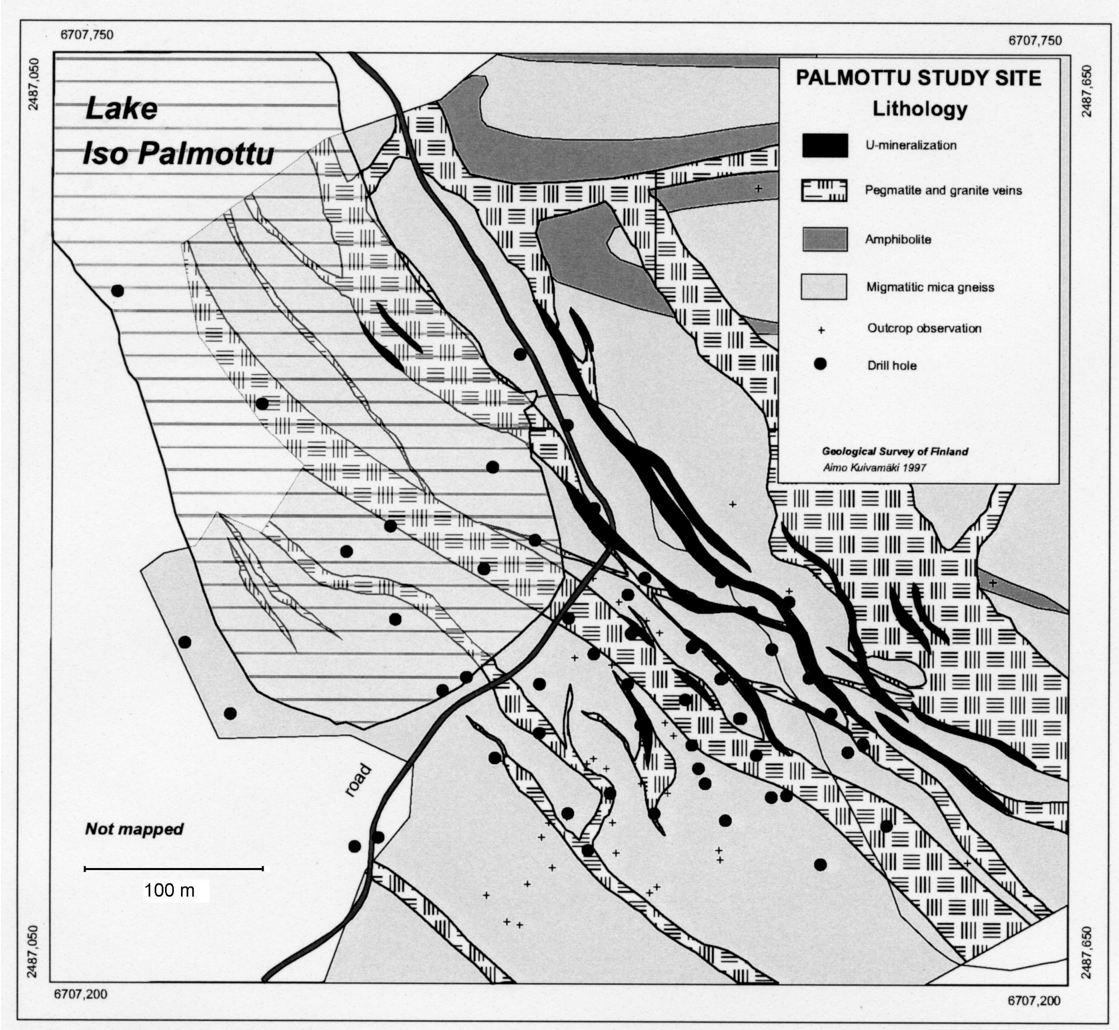

Figure 1: Detailed geological map of the Palmottu site.

The ice margin formation makes a significant contribution to the near-surface runoff pattern of the Lake Palmottu drainage area (about 1 km^2) , in which the flow directions are towards the lake. Surface discharge from the lake takes place towards the south along a brook marked by a small topographic depression ('Palmottu Brook Valley'), where the uranium deposit reaches the bedrock surface. The valley is covered by a relatively thin peat bog underlain by silt and sand. The peat contains a significant U inventory and high U concentrations were found in near-surface waters after rainfall (Blomqvist et al, 2000).

Groundwater investigations have been an important component of the analogue study. Almost 30 of the original exploration boreholes could be recovered and sampled, first by taking tube samples from the open holes, later by pumping from sealed fracture sections. The drill cores are stored at the central depot of GTK, and they have been carefully studied to determine both fracturing and lithology. Potential hydraulically conductive fractures were also located using electrical resistivity and temperature logging. Hydraulic testing included pumping and slug-tests of packered-off sections, 'spinner' tests of open bore holes, cross-hole and tracer tests. On the basis of the knowledge gained concerning the location of water-conducting fractures, most of the available boreholes were then permanently packered-off for long-term head monitoring and water sampling. During the project, new deep research boreholes (down to 400 m) were also drilled and the most important fractures in them isolated by packers.

As a result of the combined hydrogeological information, a site-scale structural pattern was established. The main fracture zones are near vertical and parallel to the schistosity, but strong indications regarding the existence of a sub-horizontal hydrostructure (H1) were also obtained.

Combination of the structural information with the data on groundwater chemistry allowed the construction of a general three dimensional hydrostructural model of the site (Figure 2). Near-surface groundwaters are typically of fresh Ca-HCO_3 type, while in deeper more stagnant conditions slightly saline (total dissolved solids up to 1.7 g/L) sodium sulphate and -chloride bearing water types prevail. The sulphate type seems to be associated with the uranium-rich rocks, whereas the chloride type represents the typical deeper groundwaters of the area outside the mineralisation. Stable isotope data indicate the presence of a glacial water component in the slightly saline waters.

Characterisation of the groundwater redox-conditions was considered to be one of the crucial tasks from the point of view of uranium behaviour. Relatively long term (about 1 day) continuous pumping and redox monitoring campaigns were carried out in packered-off sections of the old exploration drillholes. However, the most important results were obtained from the new research holes, because their larger diameter (56 mm) allowed the use of the SKB mobile field laboratory. With this equipment redox potential can be measured both down the hole ('in situ') and on the surface ('on line'). Both configurations are equipped with three different inert redox-sensitive electrodes (Pt, Au, carbon). Continuous monitoring of single fracture systems allowed measured to be obtained over several weeks.

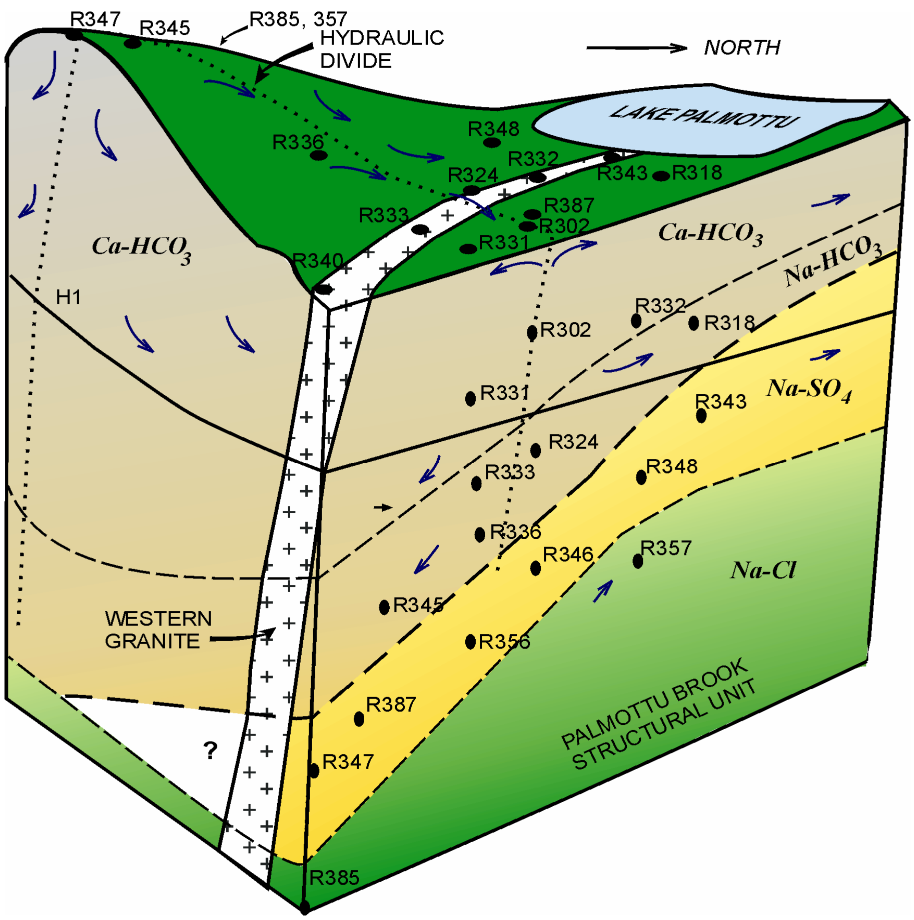

Figure 2: Hydrostructural model of Palmottu

The redox data were modelled by Cera et al. 1999. The results indicate the presence of an evolutionary redox sequence. Near-surface waters are clearly *oxic*, as indicated by the high redox potentials; oxygen depleted waters show Eh-values that match *iron-dominated* redox conditions, whereas the *sulphide* system seems to control redox potentials in the deepest and substantially reducing waters. There is also evidence for the impact of the uranium system on redox potentials in the mineralised zone (Ahonen et al. 1995). The role of uranium minerals in redox buffering was verified by studying the redox capacity of Palmottu uranium minerals in laboratory conditions.

As seen in Figure 3, the uranium concentration in groundwaters is strongly dependent on the redox conditions and, thus, on the depth. Mobilisation of uranium takes place only in oxic near-surface conditions, where uranium concentrations up to 1000 µg/L were measured. Contrary to uranium, thorium concentrations remain below 0.05 µg in all conditions.

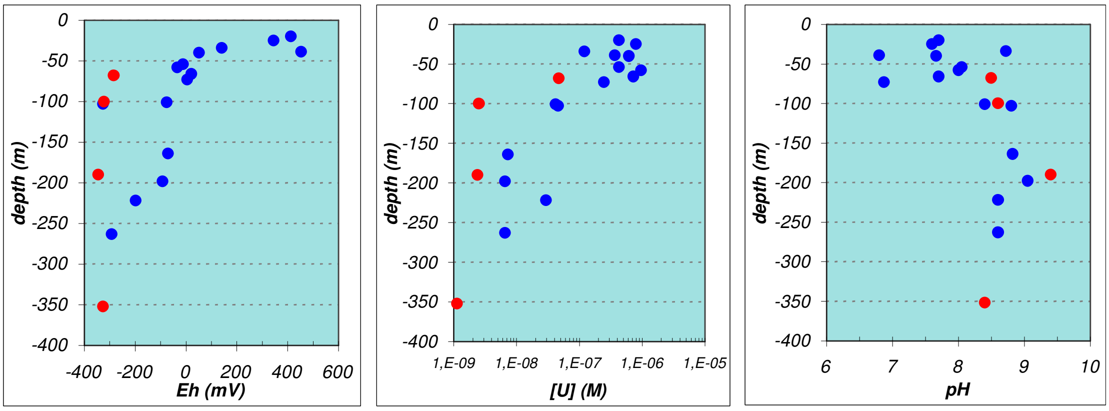

Figure 3: Redox conditions, uranium concentration and pH as a function of depth. Blue dots = data from eastern area, red dots = data from western area.

The Palmottu analogue study provided a good opportunity to test the applicability of predictive modelling tools. A blind predictive modelling exercise (BPM) was carried out to test conceptual understanding of the behaviour of selected trace and rare earth elements, for which comprehensive analytical data from both groundwaters and fracture minerals were available (Bruno et al 1999). The BPM was carried out as an inter-comparison exercise between modelling teams from QuantiSci (Spain), Ciemat and GTK-VTT. The results indicate that uranophane or soddyite (U(VI) silicates) are the most likely limiting phases controlling uranium concentrations under oxic conditions. Carbonate minerals and phosphates are important in controlling compositions of many trace elements and rare earths.

A quantitative description of radionuclide transport processes was the ultimate goal of the Palmottu study. Migration phenomena were investigated at both the process level (sorption, matrix diffusion, colloids and microbes) and by integrated modelling utilising data on the flow field, hydrogeochemistry and other transport-related processes.

The undisturbed inorganic colloid concentrations in Palmottu groundwaters are low: about 40 – 70 µg/L, consisting mainly of silica and iron hydroxide (Laaksoharju and Degueldre 1999). Measured concentrations of humic and fulvic colloids are about 5 µg/L and 3 µg/L, respectively (Kumpulainen & Vuorinen 1996). Total counts of bacteria (Most Probable Number MPN) are of the order of 10^5/mL. The main populations identified belong to acetogens, iron reducers and sulphate reducers. Microbes are effective consumers of oxidants in the Palmottu groundwater system. There are indications that microbes contribute to U(VI) reduction and immobilisation (Pedersen and Havemann 1999). Microbial biomass is not included in the colloid determinations.

The sorption properties of Palmottu rock material were studied using natural fracture surfaces. An *in situ* distribution coefficient (K_D) was determined by desorbing uranium from natural fracture surfaces (up to 25 mg/m^2) and comparing the value to the natural uranium concentration of corresponding groundwater. From a weight to volume ratio of 50 g/L, K_D-values up to about $2{\cdot}10^{-3}$ m^3/kg were derived (Suksi et al. 1999). Sequential extractions with different reagents show the presence of different sorption mechanisms having different binding strength. Easily soluble ‘labile’ uranium may contribute to more than 50 % of the sorbed fraction. Calcite is a typical fracture mineral in Palmottu and uranium was found associated to U-rich calcite samples.

Matrix diffusion of radionuclides along microfractures towards the rock matrix is a relatively slow process, but together with the sorption it acts as an effective sink or retardation mechanism. Matrix diffusion profiles of slabs sawn perpendicular to an active fracture zone show distinct compositional change towards the more intact rock. A typical feature is the formation of micro-scale redox front with more reducing conditions towards the more intact rock. Uranium series isotopes – if not in equilibrium -can be used in determining past geochemical events and their ages back to some hundred thousand years. Uranium series disequilibrium (USD) profiles of matrix diffusion sections indicated preferential mobilization of U-234. This was explained by the oxidation and mobilization of the isotope originally released by α-recoil.

Finally a migration modelling exercise was carried at Palmottu. Modelling study was done in steps: a 'blind' predictive stage and a stage that allowed calibration and model refinement. Modelling was carried out by several independent teams using different modelling tools. VTT used the FTRANS code, which includes rock matrix diffusion. Institut de Ciències de la Terra, CSIC, Barcelona used the RETRASO code (Salas and Ayora 1999), which couples flow, heat transport and geochemical processes and BRGM used the ALLAN-NEPTUNIX software package, which can be used to generate specific chemical simulators. A compartment type model emphasising geochemical processes was used by BRGM. A summary of the modelling tools is presented by Read et al. 1999.

The migration modelling focused on the eastern part of the study site, where hydraulic interference tests, tracer tests, head data and lithology point to a well defined hydrogeological system. The postulated migration route starts from unsaturated bedrock on a small hill and the groundwater evolves from rain water recharge to a uranium rich solution along the flow path. At a depth of about 100 metres conditions become substantially more reducing and deeper groundwaters are depleted in uranium. Mineralogical observations show that the fractures in the upper bedrock contain soluble, well crystallised U(VI) silicate (uranophane), which is considered to be both a source and sink for dissolved uranium in the shallow zone. Uranophane is a geologically young component in the system; USD-dating of samples from the eastern region indicates ages ranging from ~120 ka to very recent. Further study of the uranophane is on-going.

Recently, Palmottu has been the key site within the Finnish contribution to the IAEA project on natural geochemical concentrations and fluxes as safety indicators in the assessment of radioactive waste disposal. One conclusion from this study is that the U flux from the upper, oxidising zone is much higher (160 g/a) than that from the lower, reducing zone (0.002 g/a), but of the same order as the U flux caused by surficial bedrock erosion and chemical weathering (70 g/a) (Kaija et al, 2003). Uranium concentrations in Palmottu surface and groundwaters have also been compared to performance assessment predictions for a drinking water well. Such comparisons, in conjuction with an improved understanding of the processes that control these concentrations, are a crucial aid to increasing confidence in the predictions of safety analyses.(Read et al, 2002; Read, 2003). The quantity and quality of data available from Palmottu affords the site an unique role for enhancing our understanding of natural geochemical fluxes under the influences of climatic change.

Relevance: Palmottu provides a comprehensive set of process analogues of various aspects of radionuclide migration in crystalline bedrock. The thoroughly characterised geological and hydrogeological site allows comparison with corresponding data from the actual disposal site in Finland.

Position(s) in the matrix tables: Geosphere migration processes. Water-saturated crystalline bedrock.

Limitations: Only limited amount of hydrogeological and hydrogeochemical data from the actual nuclear waste disposal depths was obtained.

Quantitative information: Data on bedrock groundwater characteristics, including redox potentials, colloids and microbes. Transmissivity data on crystalline bedrock. 'In situ' K_d data. USD, detailed mineralogy, mineral chemistry and quantitative age determinations.

Uncertainties: Heterogeneity of the primary uranium source and the complexity of the hydrogeochemical system complicated the modelling of migration processes.

Time-scale: Geological, Quaternary and post-glacial/recent.

PA/safety case applications: Palmottu data were used in constructing TILA-99 solubility database (Ollila and Ahonen 1998).

Communication applications: None known

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Added value comments: Palmottu data has been used in defining natural safety indicators (IAEA coordinated research project).

Potential follow-up work: The indications of Late-Quaternary events (signs of glaciations and interglacials) provides good possibilities for further paleo-hydrogeological reseach.

Keywords: far-field, nuclide migration, pathways, matrix diffusion, redox front

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