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Palmottu natural analogue: A summary of the studies

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GTK

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ABSTRACT

Lasse Ahonen, Juha Kaija, Markku Paananen, Timo Ruskeeniemi, Veikko Hakkarainen, 2004. **Palmottu natural analogue: A summary of the studies.** Geological Survey of Finland, Report YST-121, 39 pages, 1 appendix. ISBN 951-690-896-9, ISSN 0783-3555.

This report is a review of reseach activities performed in the framework of Palmottu natural analogue studies. This uranium occurrence at Nummi-Pusula, Southern Finland has been a target of extensive studies since 1987, initially as a national analogue study 1996 – 2000, later on as an EU-financed international study 1996 – 2000. After that, up to 2003, Palmottu data has been used in an IAEA coordinated research project "The use of selected safety indicators in the assessment of radioactive waste disposal".

The report gives an extensive summary of the hydrogeological research conducted, with a special emphasis on the methodology used. A wide variety of borehole geophysical and hydrogeological methods was used during the study, and are described in the report. An areal geohydrological model, based mainly on map- and airborne survey data, is presented, as well as a three-dimensional structural model of the site.

Bedrock groundwater studies at Palmottu included development and use of new sampling methods. Swedish SKB sampling equipment was also used in the redox-potential measurements and groundwater sampling. A summary of the representative groundwater Eh-values is presented in the report. For uranium, distribution in different geological units were estimated. Isotope values of some samples indicate a potential contribution of glacial melt water.

Different modelling tasks were included in the Palmottu analogue study: modelling of speciation and solubility of dissolved elements, blind predictive modelling (BPM) with an intercomparison of results between different groups, and comparative transport modelling using different conceptual approaches.

A updated list of Palmottu-related publications and reports is included in the report.

Keywords: Natural analogue, uranium, transport, hydrogeology, redox potential,

TIIVISTELMÄ

Lasse Ahonen, Juha Kaija, Markku Paananen, Timo Ruskeeniemi, Veikko Hakkarainen, 2004. **Palmotun luonnonanalogiatutkimus: Yhteenveto tutkimuksista.** Geologian tutkimuskeskus, Tiedonanto YST-121, 39 sivua, 1 liite. ISBN 951-690-896-9, ISSN 0783-3555.

Tässä raportissa esitetään yleiskatsaus Palmotun luonnonanalogiahankkeen puitteissa tehtyyn tutkimustoimintaan. Palmotun uraaniesiintymä Nummi-Pusulassa on ollut laajojen tutkimusten kohteena vuodesta 1987, aluksi kansallisena luonnonanalogiahankkeena 1987 – 1995, sittemmin EU-rahoitteisena kansainvälisenä hankeena 1996 – 2000. Tämän jälkeen vuoteen 2003 Palmotun aineistoa on hyödynnetty IAEA:n koordinoimassa hankkeessa ”Valikoitujen turvallisuus-indikaattorien käyttö radioaktiivisen jätteen loppusijoituksen arvioinnissa”.

Raportissa esitetään yhteenveto suoritetusta hydrogeologisesta tutkimuksesta, erityisesti painottuen käytettyyn metodiikkaan. Tutkimuksessa hyödynnettiin laajasti kaira-reikägeofysiikan ja hydrogeologian menetelmiä, jotka kuvataan raportissa. Esitetään myös kartta- ja lentomittausaineistoon pääosin perustuva alueellinen geohydrologinen malli ja paikkakohtainen kolmiulotteinen hydrogeologinen rakennemalli.

Palmotun kalliopohjavesitutkimukseen kuului uusien näytteenottomenetelmien kehitys ja käyttöönotto. Redox-potentiaalin mittauksessa ja vesinäytteenotossa käytettiin myös ruotsalaista SKB-näytteenottolaitteistoa. Raportissa esitetään yhteenveto edustavista kalliopohjaveden Eh-arvoista. Uraanin osalta on arvioitu myös määrät eri geologisissa yksiköissä. Joidenkin vesinäytteiden isotooppiarvot viittaavat mahdolliseen edellisen jääkauden sulamisvesien vaikutukseen.

Palmotun analogiatutkimukseen sisältyi erilaisia mallinnustehtäviä: liuenneiden aineiden spesiaatio- ja liukoisuusmallinnus, ennustava mallinnus (BPM) eri ryhmien tulosten keskinäisenä vertailuna ja vertaileva kulkeutumismallinnus eri konseptuaalisia lähestymistapoja käyttäen.

Raporttiin sisältyy päivitetty luettelo Palmottuun liittyvistä julkaisuista ja raporteista.

Asiasanat: Luonnonanalogia, uraani, kulkeutuminen, hydrogeologia, hapetus-pelkistyspotentiaali

PREFACE

This report gives a summary of the results of the natural analogue studies of Palmottu during the years 1987–2003. The present work is based on the Finnish Centre for Radiation and Nuclear Safety (STUK) order 33/410/03. The contact person at STUK is Dr. Karl-Heinz Hellmuth.

Palmottu natural analogue study started in 1987 as a co-operative research project between GTK, Laboratory of engineering geology and geophysics of Helsinki University of Technology (HUT) and Department of Radiochemistry of the University of Helsinki (UHRAD). Later the team was complemented by the contribution of Technical Research Center (VTT). The study was mainly financed by the Ministry of Trade and Industry, and later by STUK.

In 1996 the study grow 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 Recherches Géologiques et Minières (BRGM).

The IAEA launched a Coordinated Research Project (CRP) "The use of selected safety indicators (concentrations; fluxes) in the assessment of radioactive waste disposal" for the period 1999-2003. The CRP’s objective was to contribute, through the development of international consensus, to the assessment of the long-term safety of radioactive waste disposal by means of additional safety indicators based on the observation of natural systems. Data from Palmottu was utilized in the study financed by STUK.

CONTENTS

1 INTRODUCTION

Treatment of high-activity nuclear waste from Finnish power plants is to be based on deep geological disposal. Even though the operative responsibility of the disposal is addressed to the waste producer, the controlling authorities and political decision makers have a key role in different steps of the licensing procedure. There must be a good scientific consensus indicating that the planned process is "in the overall good of the society". This is demonstrated in the Safety Case, which should be a consistent body of information, data and calculations showing that a competent level of safety is achieved.

The components of the Safety Case are – in addition to the Safety and Performance Assessment proper – the general scientific reasoning, as well as the tests and demonstrations of the system performance.

Natural analogues form a comprehensive group of studies of natural systems and sites, covering the behaviour of different materials and different processes expected in different types of repository concepts and designs. Aspects and outcomes of the natural analogue studies include both the testing of models and concepts and the demonstrations of the behaviour of natural systems. This report aims at giving an overview and summary of the results achieved in the Palmottu Natural analogue study.

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

Palmottu natural analogue study started in 1987 as a national co-operative research project between different organizations (Blomqvist et al. 1987). The progress of the study was reported in a series of annual reports, and reports summarizing the activities during several years periods (Suksi et al. 1992, Blomqvist et al. 1995, Kaija et al. 2000). Start of the EU-funded international research project was manifested in a substantial increase in research capacity. Results of the two-phase research program carried out between 1996 – 2000 are summarized in Blomqvist et al. 1998 and Blomqvist et al. 2000. The Palmottu Natural Analogue study aimed at a comprehensive characterisation of transport processes of uranium and thorium in crystalline bedrock environment. Consequently, a wide range of research in different scales was included.

The present report gives a comprehensive summary ot the research activity conducted within the Palmottu studies, emphasizing the methodological aspects and geosciences. A more focused aim of the report is to elucidate the usability of the methodological and site-related potential of Palmottu for future research. An up-to-date list of publications and reports related to the studies of the Palmottu site is given (Appendix).

2 GEOLOGICAL FRAMEWORK

2.1 Bedrock geology

Regional scale

Palmottu is situated within the Proterozoic Svecofennian orogenic belt, which formed about 1.8 billion years ago in the collision of continental plates leading to the rice of Svecofennian mountain belt on the plate contact. Nowadays the highly metamorphosed gneisses represent the roots of the ancient mountain belt. The original material of the rocks range from sand-type sediments settled down to water - now represented mainly by quartz feldspar gneiss - to volcanic rocks represented now by amphibolite. Mica gneiss is an aluminium-rich rock, being derived from clay-rich material. The currently exposed bedrock surface represents a product of erosional processes started after the orogeny. The exposed rocks were originally crystallized in depths of tens of kilometres, where molten magma crystallized forming the large granite body to the northwest of the Palmottu site at latest stage of the orogeny. The magma also mixed with the volcanic-sedimentary material, forming migmatites. Rock types of the Nummi-Pusula uranium province and of the Palmottu surroundings are shown in Figure 1. Tectonically the U-deposit is located on the southern limb near the crest of a large fold having near vertical, approximately east-west trending axial plane (Kuivamäki et al. 1991).

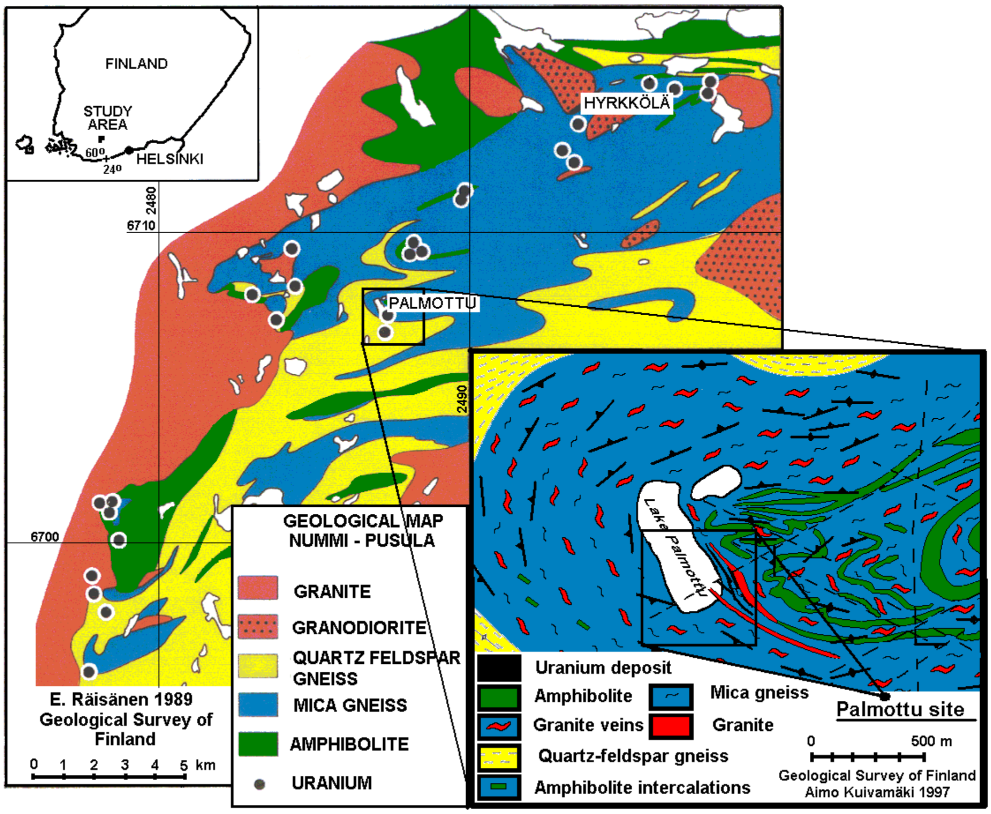

Figure 1. Geological map of the Nummi-Pusula area, southern Finland, showing the uranium discoveries. The insert to the right shows the geology of the surroundings of Palmottu. The map is compiled and modified from Räisänen 1989 and Kuivamäki et al. 1991.

Site scale

Main rock types of the Palmottu site are garnet-bearing migmatitic mica gneiss (occasionally with cordierite) and pegmatitic granites varying from thick (up to 30 m) plate-like granitic bodies to narrow pegmatite veins.

The Palmottu deposit represents a vein-type uranium mineralization, 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). Thickness of the discontinuous ore body varies from 1 to 15 meters. The strike of the deposit is parallel to the schistosity having a dip of about 70 – 80° towards southwest, and the deposit plunges towards the northwest being partially covered by the Lake Palmottu (Figure 2).

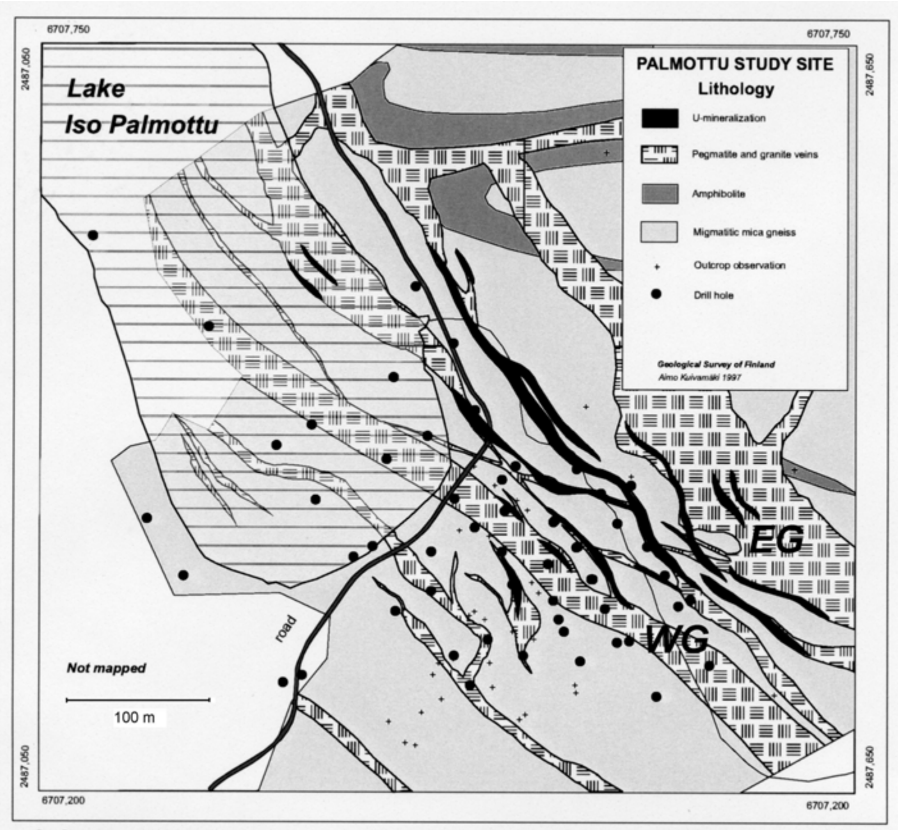

Figure 2. Detailed geological map of the Palmottu site (Kuivamäki 1991). The area shown is outlined in figure 1. The main granitic bodies, western granite and eastern granite are denoted by ***WG*** and ***EG***, respectively.

2.2 Tectonic framework

At the regional scale, the Palmottu U-deposit is located near the NW corner of a major rhomboidal block east of the Perniö granite. This block consists of Proterozoic Svecofennian schists, undergone three (Ploegsma & Westra 1990) or four (Kilpeläinen & Rastas 1990) Proterozoic deformation phases. The regional tectonic interpretation, based on aeromagnetic data, (Fig. 3; Kuivamäki et al 1991)) indicates that Palmottu is located within a lens-shaped block, bounded by major fracture/shear zones. To the NW of Palmottu, the major Johannislund shear zone can be detected, clearly controlling the occurrence of several U mineralisation units in Nummi-Pusula region (Kuivamäki et al. 1991).

In a local scale, interpretation of the fracture zones is based on elevation and geophysical (mainly airborne magnetic) data as well as outcrop observations. The interpreted features are also classified into four classes, according to their size and probable hydraulic transmissivity (Kuivamäki 1997).

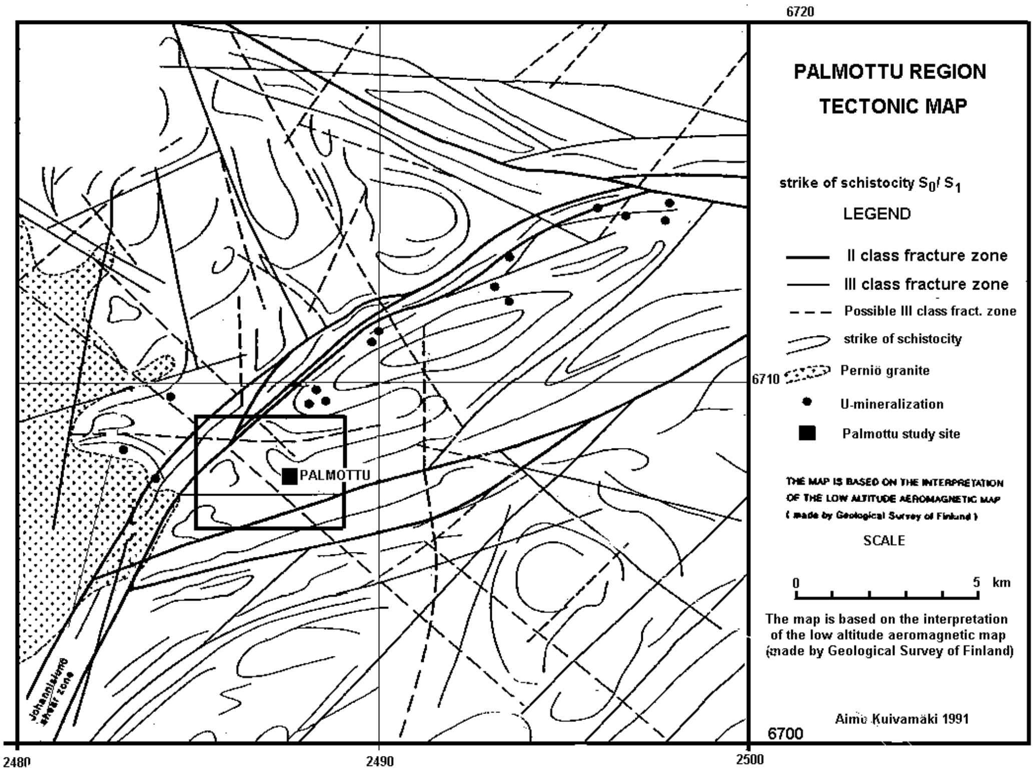

Figure 3. Tectonic framework of Palmottu region. Different observation scales used in the text can be identified in the picture: regional = the whole area; local = Palmottu surroundings indicated by open square (area of Fig. 5); site = borehole area (black square).

2.3 Quaternary deposits and flow routes

Quaternary Ice Ages finished the Finnish landscape. Unsorted basal till formed in the bottom of the moving ice sheet, while sorted sediments, e.g. gravel, sand, silt and clay settled from water.

Palmottu 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 continental ice sheet temporarily stopped about 10 000 years ago. Salpausselkä III is manifested by a wide and thick sand-gravel formation on the northwest of Palmottu. Figure 4 shows the distribution of Quaternary deposits around Palmottu, shown is also a schematic depiction of the ice margin retreated to the Salpausselkä III position. During the deglaciation, meltwaters from the delta formation passed the Palmottu site and may have been filtrated into the fractures of the bedrock. Due to its slightly elevated position, Palmottu block remained on land, when it was uncovered below the ice sheet.

The ice margin formation has a significant contribution to the present near-surface runoff pattern of the drainage area (about 1 km^2) of the lake Palmottu, in which the present flow directions are towards the lake. Surficial discharge from the lake takes place towards the south along a brook running along a small topographic depression (‘Palmottu Brook Valley’), where the uranium deposit reach the bedrock surface. The valley is covered by a relatively thin peat bog underlain by silt and sand. Typical to uranium is its strong tendency to enrich in peat. At Palmottu, uranium concentration in peat comparable to those observed at other peat bogs near U-sources, while the near-surface water does not show any elevated uranium concentration.

Figure 5 shows the near-surface hydrological principles of the Palmottu area. The whole bedrock-block around Palmottu is topographically slightly higher than the surroundings (Fig. 4). Regional flow of the area runs from the large Salpausselkä III sand formation northwest of Palmottu toward the low clay-dominated area to the south.

The drainage basin of the Lake Palmottu (Hakkarainen et al. 1991) is situated within the Palmottu sub-block (Central Palmottu block). The annual precipitation within the drainage basin is about 0.5 million cubic meters. However, the north-western sand formation evidently also has an important contribution to the water balance of Lake Palmottu (blue arrows).

The Central Palmottu block of ca. 2 x 2 kilometer is bounded by major (class II) fracture zones. The block is cut by several more local (class III) and less significant class IV zones with various directions. The drilling site, located to the SSW of Lake Palmottu, is bordered by two NNW – SSE trending zones in the east and the west and a NE – SW trending feature in the south. The main direction of class IV fracture zones at or near the drilling site is NW –SE.

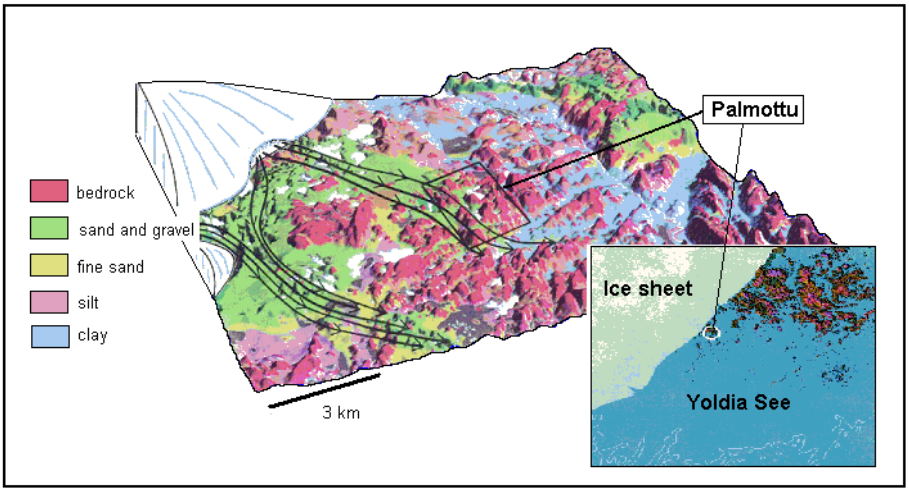

Figure 4. Quaternary deposits around Palmottu. Retreating ice sheet is depicted in the upper left corner. Flow lines show the main routes of meltwater discharge. The insert at the lower right corner visualizes the position of the Palmottu site during retreat of the ice sheet.

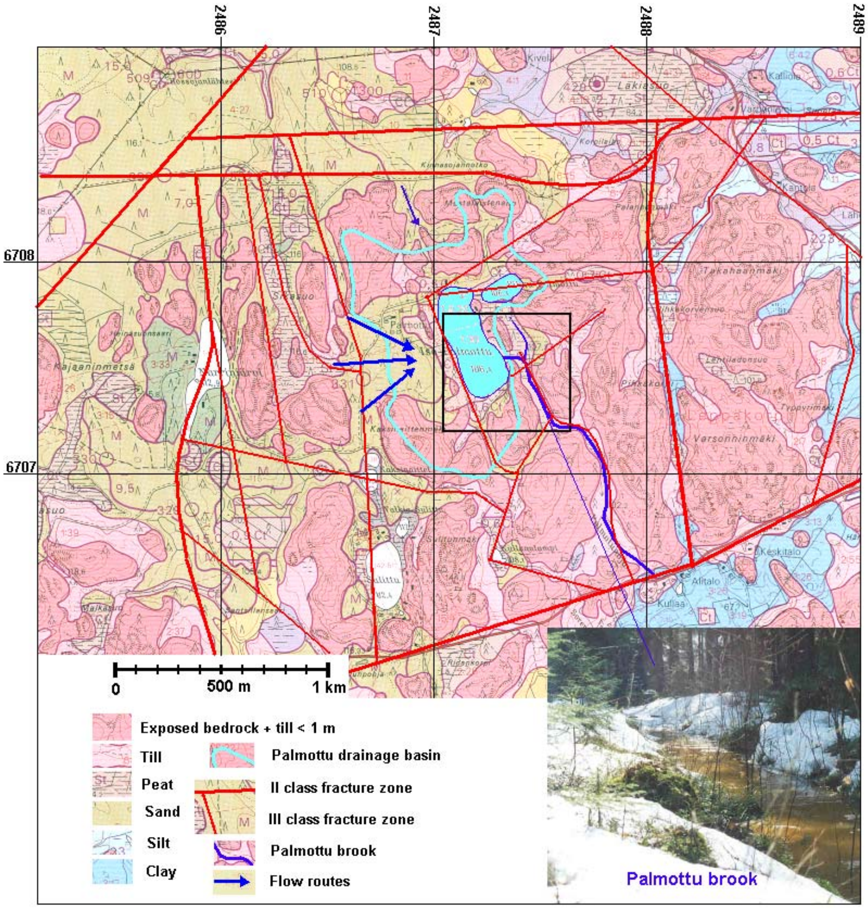

Figure 5. Hydrology and Quaternary formations of Palmottu. The square marks off the area of Figure 2.

3 SITE HYDROGEOLOGY

3.1 Drillhole hydrogeology

Table 1 gives a comprehensive summary of drillhole hydrogeological studies at Palmottu during 13 years. During the earliest stage of Palmottu project, drillhole studies were concentrated on tube sampling and chemical/radiochemical analyses of the Palmottu groundwaters (hydrogeochemistry is treated separately in this report). Tube sampling gives a continuous column of the water composition in a drillhole, and electrical conductivity of water was also measured as a function of depth. Drastic changes in electrical conductivity (EC) may indicate presence of water-conducting fractures. Consequently, tube sampling (with EC data) is also included in table.

Understanding and modelling of groundwater flow requires information on hydraulic head distribution. In the first phase, head could only be measured by logging water tables in open boreholes. As a rule, water tables in different boreholes follow smoothly the topography, as well as the seasonal variation of precipitation (Figure 6).

Table 1. A summary of hydrogeological drillhole studies.

	1987	1988	1989	1990	1991	1992	1993	1994	1995	1996	1997	1998	1999
EARLY PHASE	■	■	■	■	■	■	■	■	■				
Tube sampling (EC-data)	■	■		■									
Temperature		■		■									
Head monitoring (open				■	■	■	■	■	■	■	■	■	■
Fracture logging				■	■	■	■	■					
Galvanic measurements				■						■			
Mise-à-la-masse						■	■						
Light weight packer			■	■									
Borehole radar							■		■		■		
Borehole video									■	■	■	■	
Packers to old drillholes									■	■			
Spinner (+crosshole)								■	■				
Head monit. in plugged									■	■	■	■	■
EU-PALMOTTU, PHASE I										■	■		
Drilling of new research										■			
Crosshole tests									■	■			
BIP borehole-TV										■			
Difference flow										■		■	
Tracer tests (+crosshole)											■		
EU-PALMOTTU, PHASE II												■	■

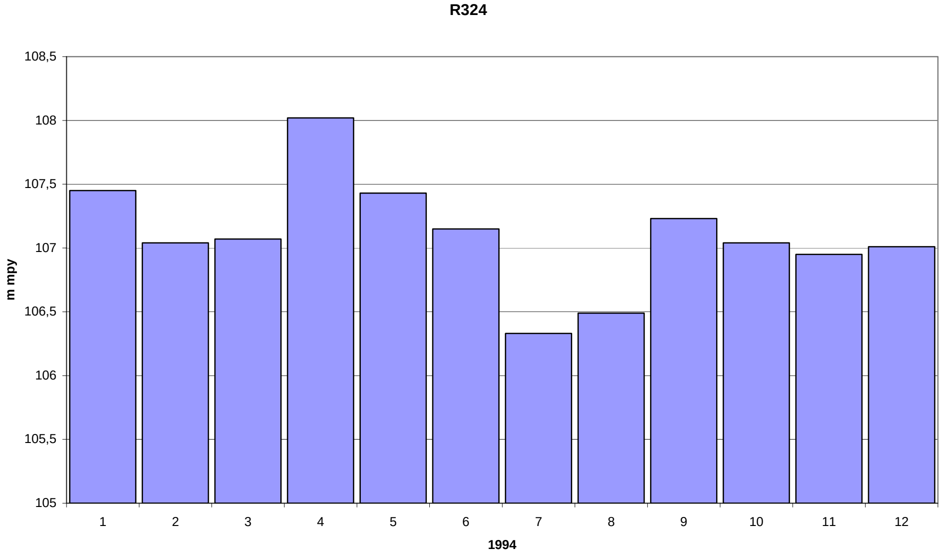

Figure 6. Monthly variation of water table in borehole 324 during the year 1994.

Hydrogeological conditions in bedrock are crucially dependent on the distribution and character of water-conducting fractures. To be able to sample waters from individual (or from only few fractures at same time), a packer sampler was developed at GTK (Laaksoharju et al. 1991). In addition to water sampling, also simple slug-test could be carried out, and the hydraulic conductivity of plugged sections could be estimated. Three boreholes were studied in details by this method at Palmottu (Ahonen and Paananen 1991), but thorough study of hydraulic conductivity variations in all boreholes proved to be too time consuming. Later the equipment was mainly used in groundwater sampling and redox-measurements.

A systematic examination of the drillcore samples, concerning 53 existing old boreholes, was carried out during 1990 – 1992 (Kuivamäki et al. 1991, Paananen & Kuivamäki 1993). The work comprised mapping of fractures and rock types. In addition, a more detailed fracture characterization of 30 boreholes was done in 1993 – 1994 (Lindberg 1994). In order to complement the understanding of the character of in situ fracturing, TV-loggings have been done in 26 boreholes during 1995 – 1998 (Lindberg 1996, Stråhle 1996, Paananen 1997, Paananen et al. 1998). In Figure 7, the results of TV-logs from some boreholes are presented as an example.

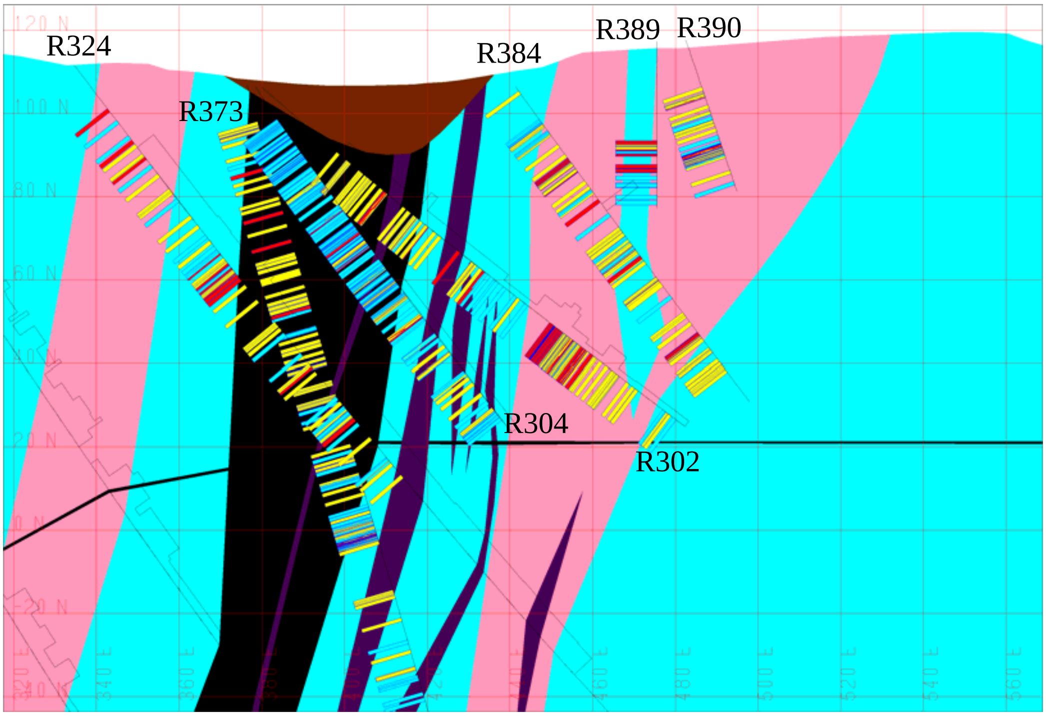

Figure 7. Fractures observed by a video survey in the boreholes. Red = open, hydraulically significant fracture, yellow = open fracture, blue = tight or filled fracture. Lithology is also shown.

Thermal logs in boreholes can be utilized in estimating groundwater flow in the bedrock. When the heat flow is in steady state and thermal conductivity is constant, temperature increases linearly with depth. Under these conditions, gradient changes and anomalous temperature values may reflect groundwater flow. The first thermal loggings in 7 boreholes of Palmottu were done in 1985 and 1988, followed by the loggings of 3 additional holes in 1990.

Logging of electric resistivity can be used in locating water-bearing fractures and fracture zones, since they are better electric conductors that the surrounding intact bedrock. In Palmottu, the short normal array was applied as the electrode configuration. Altogether 13 boreholes were logged in 1990 (Kuivamäki et al. 1991), added by four holes in 1996 (Paananen 1997). The bedrock in Palmottu is usually highly resistive (several tens of thousands of ohmmeters, Fig. 8), and numerous resistivity minima, related to zones of intense fracturing, can be detected. The strongest resistivity anomalies, however, represent sulphide-rich sections.

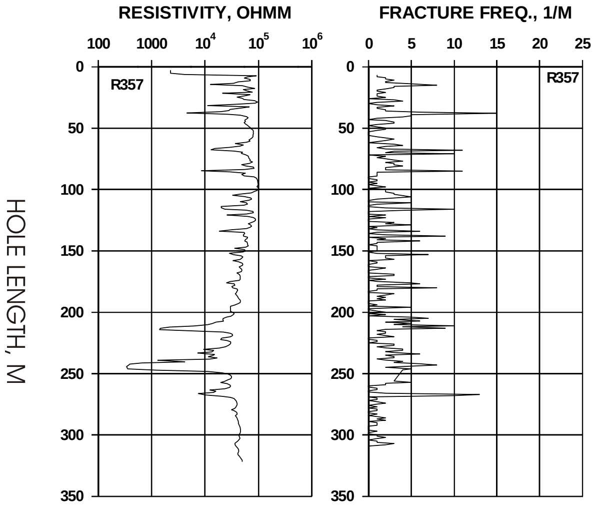

Figure 8. Electric resistivity and fracture frequency in borehole 357 as an example. The strong resistivity minima at ca. 215 m and 240 m are related to sulphide minerals. Smaller anomalies often reflect open fracturing.

Since the drill cores of Palmottu were unorientated, there was a need to gather additional information on the geometry of fractures and fracture zones between the boreholes. For that purpose, galvanic mise-à-la-masse and borehole radar measurements were done.

The mise-à-la-masse method is widely used in ore prospecting, but it has also been used in fracture zone characterization. A current source is positioned at an interesting, electrically conductive section, and the electric potential is mapped either on the ground surface or in a borehole. A galvanic and possibly a hydraulic connection may be observed as a potential maximum, depending on the resistivity contrast and survey geometry.

The mise-à-la-masse survey was done as two separate campaigns in 1992 (Paananen 1993) and 1993 (Paananen et al. 1994). The measurements were done between 13 boreholes, using 32 separate grounding points. The most promising hydraulically conductive sections were chosen as current grounding positions. The results of the mise-à-la-masse survey indicate several galvanic connections between the boreholes. The dips of the conducting structures are usually steep and they also often coincide highly fractured sections in the boreholes.

The borehole radar survey was done in three separate phases in Palmottu. In 1993, the measurements were done with the conventional omni-directional antenna in five boreholes (R357, R346, R324, R348 and R343) at the central study site (Carlsten 1993). In 1996, the survey was complemented by the measurements with the directional antenna in two recently drilled holes R373 and R384 (Carlsten 1996). The third phase of the survey included measurements in the deep geochemical borehole R385 and its neighbouring borehole R386 in 1997 (Carlsten 1997).

The borehole radar survey revealed numerous reflections, indicating open and/or intense fracturing in the bedrock. Some of the most distinct reflectors could also be correlated between the boreholes, resulting unambiguous orientations for the reflectors. The primary concept was a set of 6 - 8 subvertical, concordant fracture zones and a subhorizontal one.

The borehole radar results have been in a major role in compiling the site-scale fracture zone model of Palmottu. They have been correlated with all the existing data in the boreholes, e.g. fracturing, resistivity, hydraulic conductivity, and mise-à-la-masse results.

Spinner is a flow meter designed to detect water-conducting fractures in drillholes. The test probe is a mechanical propel with pulse counter, measuring thus the vertical flow rate in drillhole. Temperature and electrical conductivity sensors are also included in the probe. The flow in drillhole is produced by pumping (air lift was used at Palmottu), and the probe is lowered stepwise (e.g. 4 meters). Water-producing fractures are detected as decreasing of the flow rate. Water coming from a specific fracture zone can also be reflected in changing temperature and electrical conductivity of water (Fig. 9). Due to the simple and 'robustic' principle, the observations are very reliably. However, it must be note that strong water-conductors 'mask' those coming deeper down. When the measurement is completed and pumping stopped, recovery of water table is monitored. Drillhole transmissivity can thus be calculated using conventional methods.

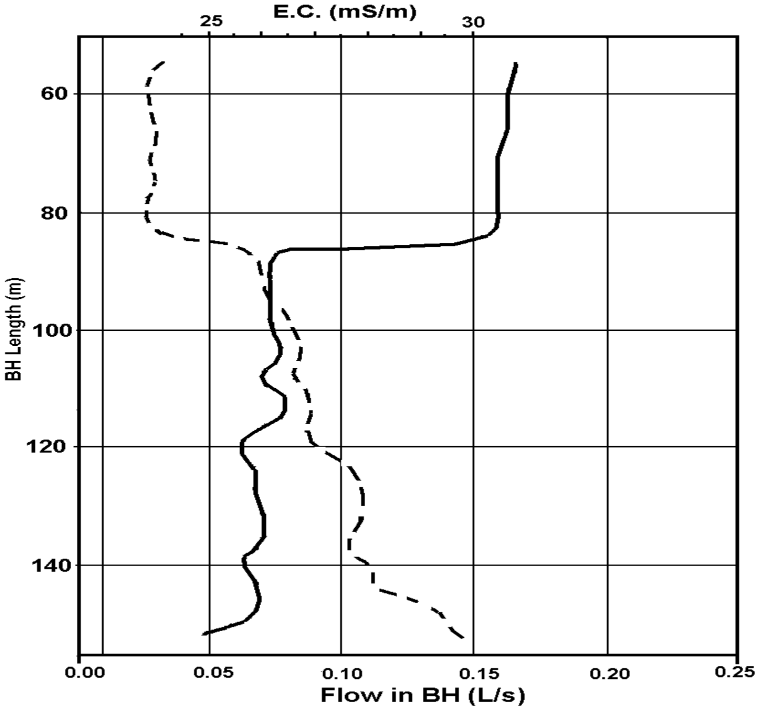

Figure 9. An example of spinner test results, drillhole 357 (compiled from Jönsson et al. 1995). Solid line = flow, broken line = electrical conductivity (E.C.).

It was understood that the large number of open boreholes in a small area has an adverse effect for the monitoring of natural groundwater flow pattern. Consequently, during 1995 practically all open drillholes in the central area were sealed using one to three packers. The packers were pressurized using water, and they were connected to a pressurized nitrogen gas line to ascertain their tightness.

In each drillhole, packers were located so that they would isolate the most important fractures or fracture zones. The locations of the fractures were decided on the basis of available mineralogical, geophysical and hydraulic data.

Each packed-off section was connected to a monitoring tube 15/12 mm for head measurement and water sampling. Continuous monitoring of hydraulic heads in the test sections started soon after the installation, and continued on weekly basis during several years time.

Despite of the large number of exploration boreholes drilled at Palmottu, the Natural Analogue Project considered necessary to drill some additional boreholes to satisfy some specified needs of groundwater research. Additional motivations were that the old boreholes were too narrow (46 mm) to allow the application of some modern down-hole instruments and the recovery of the core was not good enough. The targets were to gain information on fracture systems not revealed by the exploration boreholes (R373, R387), to study the deep part (below 200 m) of the system (R385) or to study selected rock units or flow systems (R384, R388, R389 and R390). Altogether, about 1500 meters were drilled during the years 1994– 1998. Many of these holes were targets for the full-scale hydrogeological and hydrogeochemical research programmes.

Crosshole test campaign (interference test) was one of the major efforts in characterizing hydrogeological conditions of the site. In the method one borehole is pumped and the responses in surrounding boreholes are monitored. Palmottu site provided a unique possibility to do detailed and representative crosshole tests, because of the availability of 23 drillholes sealed by packers in the very central area of about 4 hectares. In those drillholes, there are 50 packed of sections (+23 free water tables). Altogether 17 packed-off sections (9 drillholes) were pumped, and – in every case – practically all (about 70) packer-isolated sections in surrounding drillholes were monitored.

Primary results of the tests are reported by Lampinen et al. 1997 and Ludvigsson 1997. A main conclusion was that the fracturing of, at least, upper 100 – 150 m of bedrock behaves rather as fractured continuum than as idealized planar structures.

Difference flow measurements were done in four of the new Palmottu research boreholes (Rouhiainen 1996, Rouhiainen and Heikkinen 1998). The technique provides a rapid method to determine hydraulic conductivity and hydraulic head in drillholes. In the equipment, the section to be studied is isolated by rubber disks and the flow either out from the section or into the section is measured by thermal pulse method. Hydraulic head and hydraulic conductivity of a test section can be calculated, if measurement is carried out using two different head levels of the borehole (one of them may be natural head). An example of results is shown in Figure 10.

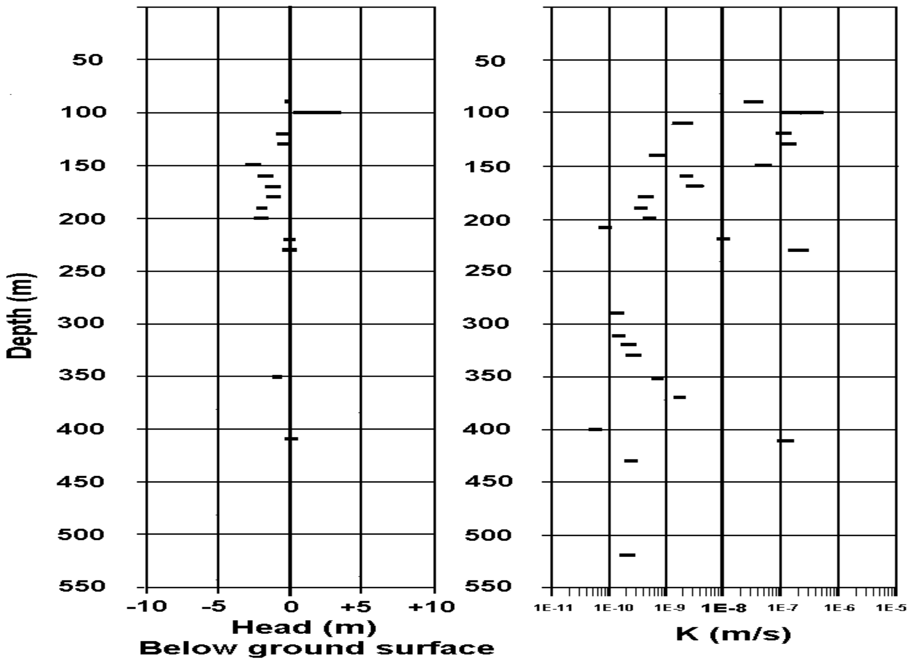

Figure 10. An example output of difference flow measurement. Palmottu, drillhole 385.

At the end of the Phase I of the EU-Palmottu project, when sufficient data on hydrogeology and –geochemistry of site was achieved, eastern part of the site was selected for a tracer test. The objective of the combined tracer – hydraulic interference test was to verify proposed hydraulic connections, and study their transport properties. The test was made between three drillholes: pumping hole and two injection holes, of which one had two separate injection sections separated by packers (Fig. 11). Thus, three different tracers were injected and the breakthrough was monitored in pumping hole. Distance between boreholes was less than 50 meters. Due to the pumping, hydraulic gradient between injection holes and pumping hole was about one during the test. Hydraulic heads were monitored in all surrounding boreholes.

The experiment lasted about 15 days. During that time about 40 – 50 % of the tracers were received from two test sections, while one of the sections evidently didn’t have hydraulic connection to the pumping hole. Integration of the breakthrough curves indicated that continued pumping would have given total recoveries of about 70 – 80 % after about two months.

Analysis of the results shows that dispersion is large. This indicates that the transport routes behave as interconnected network of discrete fractures approaching fractured porous medium. Tracer test was reported in details by Gustafsson et al. 1998.

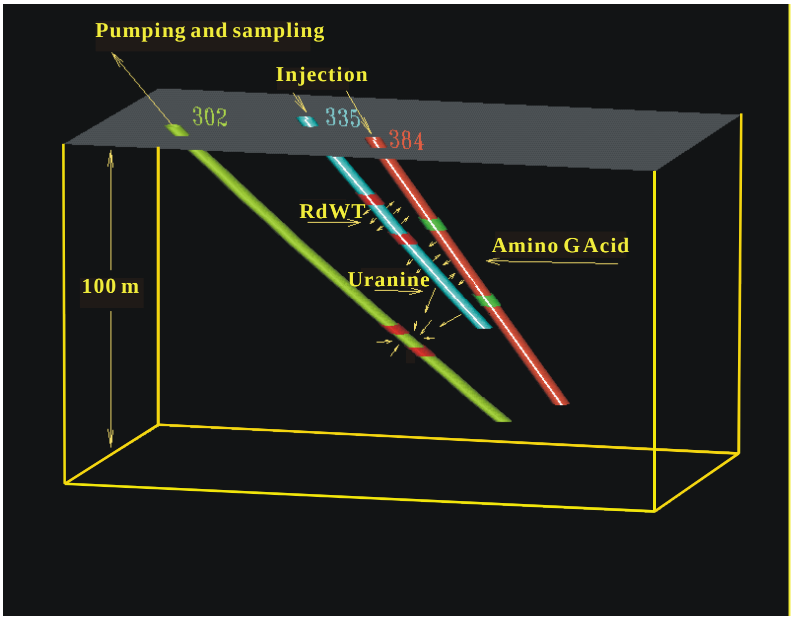

Figure 11. Tracer test arrangement

3.2 Site-scale structures and hydrogeology

Fracture zones and their properties has been a mainline subject in the investigations of Palmottu, since they can provide rapid flow routes for groundwater and dissolved radionuclides even in the deep bedrock. They have been studied using numerous different approaches from regional to site scale.

At the site scale, the fracture zone model is based mainly on geological and geophysical observations from the boreholes, considering also survey results from the ground surface. The 3D interpretation of the diverse data revealed six subvertical and one subhorizontal fracture zones intersected by numerous boreholes. The subvertical fracture zones (V1 – V6, Fig. 12) are roughly NW – SE trending narrow features (typically 1 – 3 meters), observed as intense fracturing and, occasionally, high hydraulic conductivity. Their geometry is mainly determined by combining the borehole radar results with fracturing data in the boreholes. These features are rather steeply dipping, ca. 75°/225° – 240° (dip/dip direction). Two of the interpreted fracture zones (V1 and V2) are closely related to the U-Th mineralisation and the Palmottu Brooklet Valley and three of them (V4, V5 and V6) are associated with the main granitic units. The subhorizontal fracture zone H1 was primarily detected by borehole radar, supported by single hole hydraulic tests. Subsequently, the geometry of H1 was revised by the results of hydraulic cross-hole tests, suggesting a rather thick (c. 100 m) hydraulic zone, dipping gently to the ENE (dip/dip direction 15°/60°). Accordingly, H1 presents the zone of rapid meteoric groundwater flow within the uppermost part of the bedrock. The subhorizontal feature in Fig. 13 depicts the lower boundary of this hydraulic zone.

Extensive hydraulic cross-hole tests and an integrated examination of hydraulic heads and hydrogeochemistry indicate that groundwater flow in the Palmottu bedrock is predominantly limited to the uppermost 100 – 150 m. Within this zone, four separate hydrogeological units have been defined: the Western Mica Gneiss, the Western Granite (bounded by fracture zones V4 and V5), the Palmottu Brook Valley (V1 and V2) and the Eastern Granite (V6). In addition to these units, hydraulic heads and geochemistry suggest a NE-SW trending hydraulic barrier, V7 (Fig. 13). This probable fracture zone appears to be hydrogeologicallly significant, acting as a water divide across the Palmottu Brook Valley.

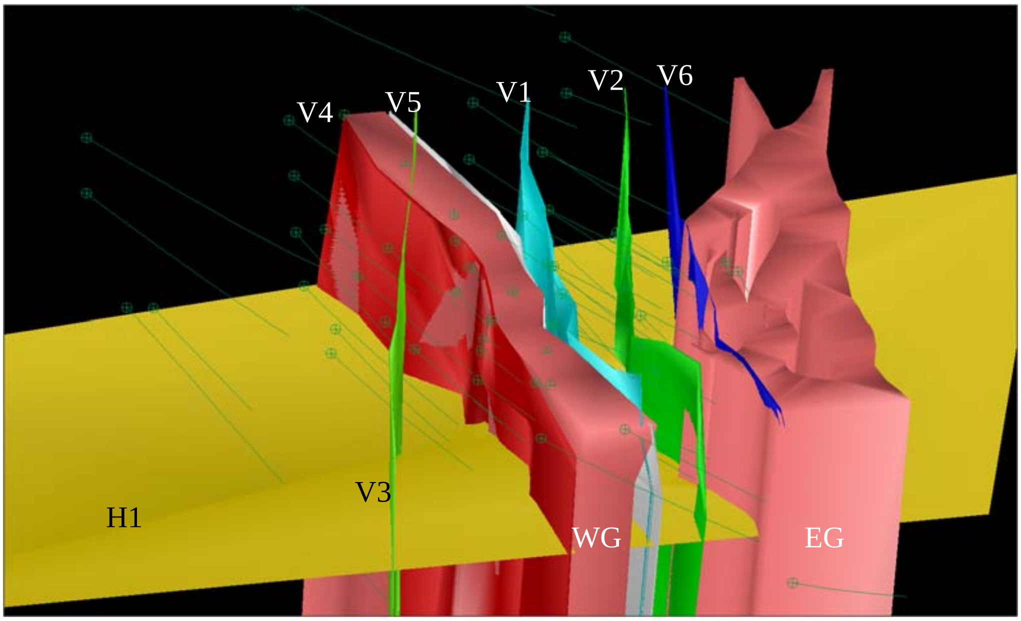

Figure 12. The fracture zones interpreted from the borehole data together with the main granitic units, a 3D view from the SE. WG = Western Granite, EG = Eastern Granite.

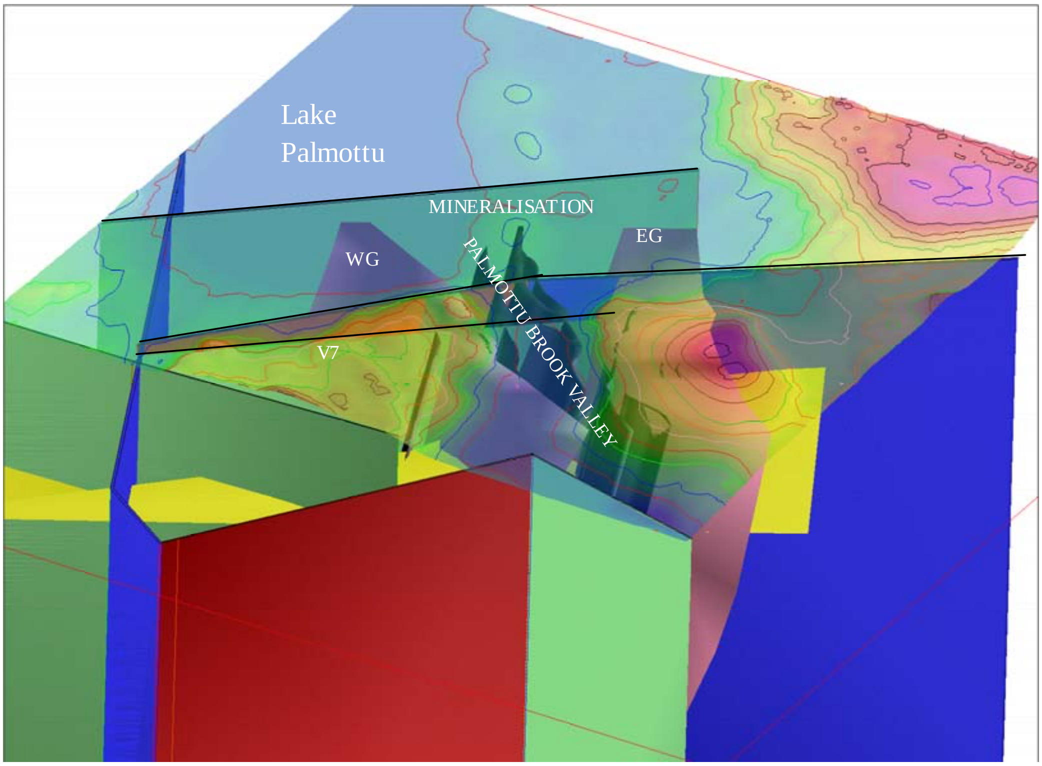

Figure 13. The site-scale hydrogeological units of Palmottu with the interpreted local fracture zones, 3D view from the SE. Topography is also shown. WG = Western Granite, EG = Eastern Granite.

4 HYDROGEOCHEMICAL STUDIES

4.1 Sampling methods

Groundwater research has been one the key activities of the analogue study. Almost 30 of the original exploration drillholes could be recovered and sampled, first by taking tube samples from the open holes, later by pumping from sealed fracture sections. The tube sampling method was presented by Nurmi and Kukkonen 1986, while details of the packer samplers are given in section 5.1. The drill cores are stored at the central depot of GTK, and they have been carefully studied both for fracturing and lithology. According to the gained knowledge about the location of water-conducting fractures, most of the available boreholes were then permanently packed-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 were isolated by packers.

4.2 Composition of groundwaters

General chemistry and water types

Concentrations of dissolved solids in dilute Ca-Mg-HCO_3- and Ca-Na-HCO_3-types groundwater from overburden range from less than 50 mg/L to over 150 mg/L. Groundwater with low TDS and simultaneous low pH (5.9 to 6.5) are generally found in areas of thin soil layers (< 5 m) and in areas where the hydraulic conductivity of soil is better than in surroundings. In these areas groundwater is in active recharge and normally the groundwater table follows rapidly the changes in precipitation. The composition of groundwater correlates well with the type of soil and according to its hydraulic properties. Based on differences in hydrochemistry, there seems to be more or less isolated local groundwater bodies in the overburden along the Palmottu Brook.

Four general bedrock groundwater types characterize the Palmottu site (Fig. 14). Recently recharged, shallow Ca-HCO_3 type groundwater evolves in the upper part of the bedrock into a Na-HCO_3 type groundwater at increasing depth within a few decades. Brackish Na-SO_4 and Na-Cl type groundwaters (some thousands of years old) prevail below the HCO_3 groundwater bodies; Na-SO_4 is present only around the mineralized zone whilst Na-Cl is found at greater depths in the surrounding area. The content of TDS increases from <100 mg/L of the overburden groundwater to over 1600 mg/L in the brackish Na-SO_4 and Na-Cl type bedrock groundwaters at 300 - 350 m depth. pH increases with depth from slightly acid conditions in the overburden to pH 7 - 8 in the Ca-HCO_3 type groundwater in the upper part of the bedrock and further to 7.5 - 9 in the Na-HCO_3 type groundwater due to dissolution of minerals during its evolution. In the brackish groundwater types pH mainly varies from 8.5 to 9 (Blomqvist *et al.,* 1998, 2000a; Pitkänen *et al.,* 2002).

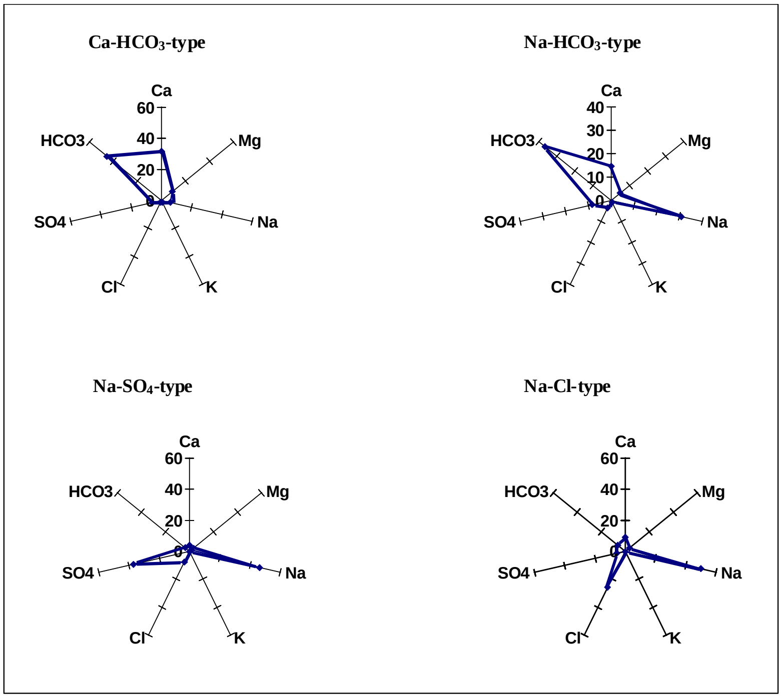

Figure 14. Radial diagram of the four groundwater types that characterise the bedrock groundwater at Palmottu.

As a result of the combined hydrogeological information, a site-scale hydrostructural pattern was established (Fig. 15). Near-surface groundwaters are typically of fresh Ca-HCO3 type, while deeper in more stagnant conditions slightly saline (Total dissolved solids up to 1.7 g/L) sodium sulphate and -chloride bearing water types prevail. Sulphate type seems to be associated with the uranium-rich rocks, while the chloride type represent the typical deeper groundwaters of the area outside the mineralization. Stable isotope data indicates the plausible presence of a glacial water component in the slightly saline waters.

Stable isotopes

The δ^2H and δ^{18}O isotope signatures of the different groundwater types (Fig. 16) indicate a meteoric origin of the water without palaeoevidence of seawater mixing during post-glacial times. In the Na-SO_4, Na-Cl and some of the Na-HCO_3 type groundwaters there is a significant depletion in the δ^{18}O isotope values. The local, present-day meteoric groundwater signature can be observed in the shallow bicarbonate waters. The depleted δ^{18}O values deeper in the bedrock may represent a colder climate recharge, which took place during the retreat of the Weichselian ice sheet about 10 000 B.P (*cf.* Blomqvist *et al.*, 1998). These depleted δ^{18}O values reflect a higher portion of glacial melt water.

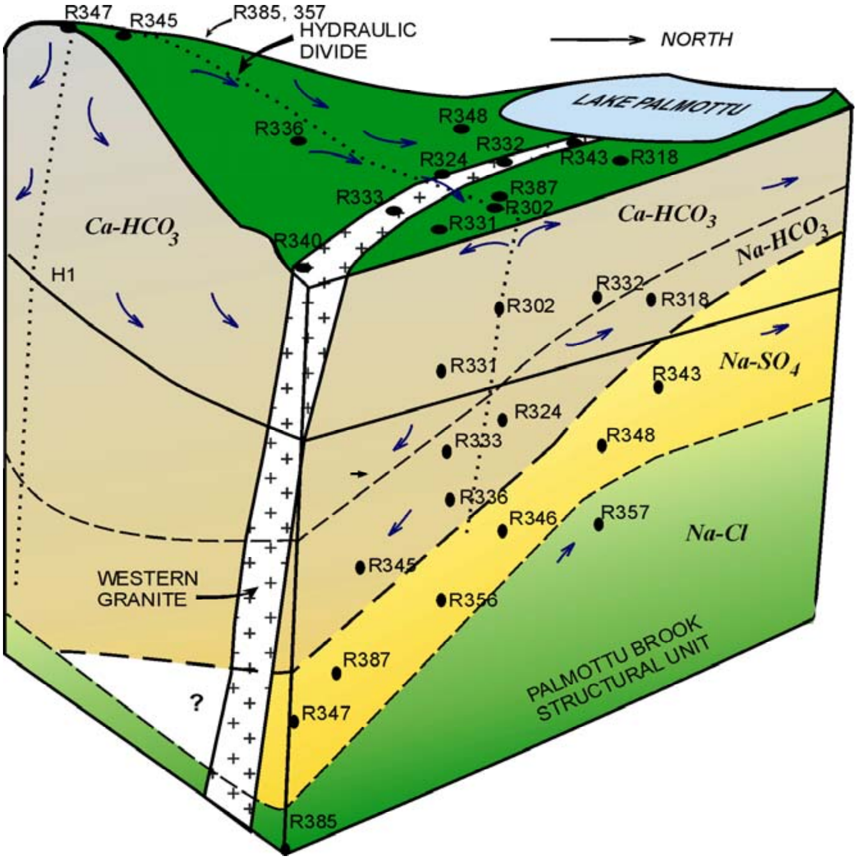

Figure 15. Hydrostructural model of Palmottu

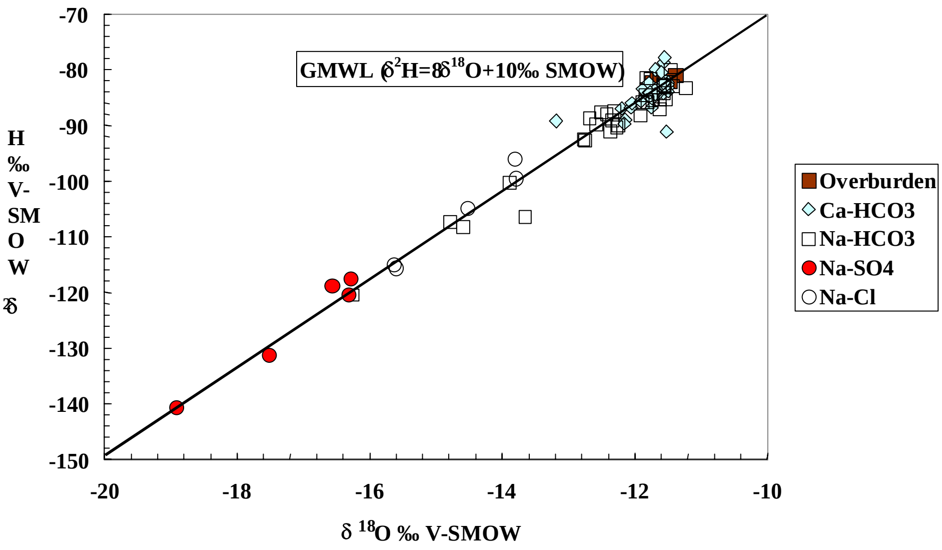

Figure 16. Stable isotope plot (δ2H and δ18O) for the Palmottu groundwater samples compared to the Global Meteoric Water Line ($\delta^2H = 8 \cdot \delta^{18}O + 10$; Craig, 1961).

Dissolved uranium

The median concentrations of uranium in groundwater in the overburden are very low, over 300 times lower than in the oxidative bedrock groundwater of Ca-HCO_3-type. In fact, the concentrations are close to uranium concentrations of the deep bedrock groundwater, which are in a strongly reducing environment. The groundwater observation tubes in overburden are installed in fine-grained silt layers where the hydraulic conductivity is low. Due to poor conductivities, the groundwater flow is limited and reducing conditions prevail causing low uranium concentrations. Groundwater samples taken from tubes, which have been installed in sand-till layers, near the main uranium mineralisation show slightly higher uranium concentrations. The hydraulic conductivities of sand-till layers are higher and, thus more oxidative conditions prevail allowing higher uranium concentrations.

With respect to the present-day mobilisation of uranium in bedrock groundwaters, a continuous alteration and oxidation process is proceeding at Palmottu. This is seen in the high uranium contents usually from 100 to 500 µg/l, clearly associated with the oxidative HCO_3 type groundwaters of dynamic flow system down to depths of 150 m in the immediate proximity of the uranium mineralisation, *i.e.* the Eastern Granite and the Palmottu Brook structural unit.

The upper uranium-rich groundwater plume forms a horizontal, tube-like system with a diameter of 100 m and a horizontal extension of 300 m (Fig. 17). Below this hydraulically active zone (*i.e.* depths > 150 m) reducing conditions prevail and groundwaters are characterised by low uranium concentrations. The south-east extent of the uranium plume is presently not known because there are no suitable boreholes for groundwater sampling.

Uranium concentrations in Ca-HCO_3 and Na-HCO_3 type bedrock groundwaters varied from 2.2 to 765 µg/L and 0.6 to 363 µg/L, respectively. The groundwaters in the Western Granite and Western Mica Gneiss have much lower uranium concentrations, generally less than 10 µg/l. At greater depths, below 200 m, in Na-SO_4 and Na-Cl type stagnant waters where reducing conditions prevail, uranium concentrations very seldom exceeded 10 µg/L (Kaija, 1998).

With increasing depth, uranium concentrations are thought to be limited according to the solubility versus pH relationships. At greater depths the system approximates steady state with both uranium concentrations and isotopic ratios likely to be controlled by equilibrium with uranium rich fracture minerals.

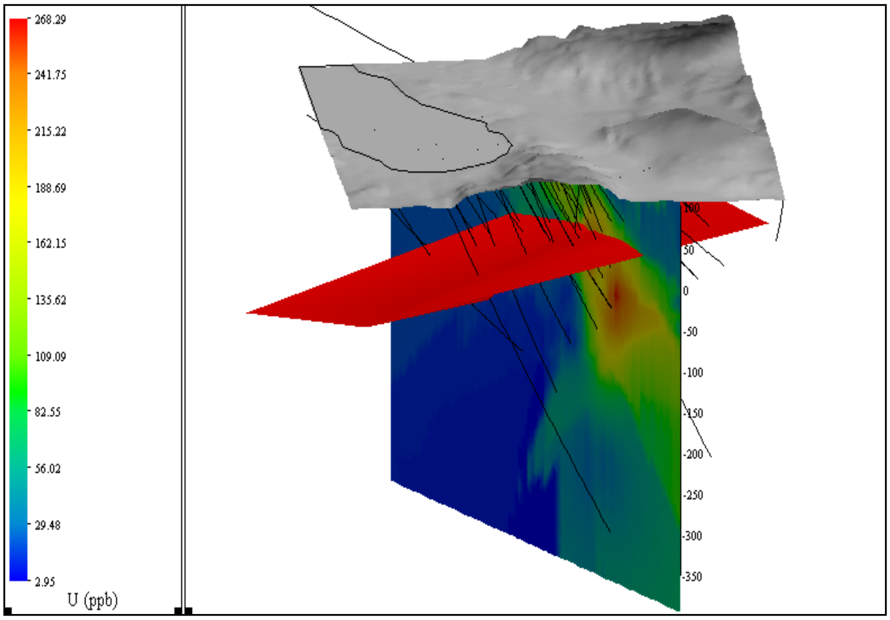

Figure 17. Uranium concentrations in the Palmottu Brook structural unit with the location of boreholes. Elevation contour data © National Land Survey of Finland, permission number 13/MYY/03.

4.3 Uranium inventories at Palmottu

Uranium inventories at Palmottu have been estimated for a 300 m × 500 m × 250 bedrock block (Kaija et al. 2003). This work was performed in the framework of the IAEA coordinated natural safety indicator research project. The results are summarized in Figure 18 and Table 2. The U-inventory and fluxes in the Palmottu case are well justified. As also concluded in the report, past and present processes in the Palmottu bedrock are well studied and understood, demonstrating the improving understanding of the scientific basis of nuclear waste performance assessment.

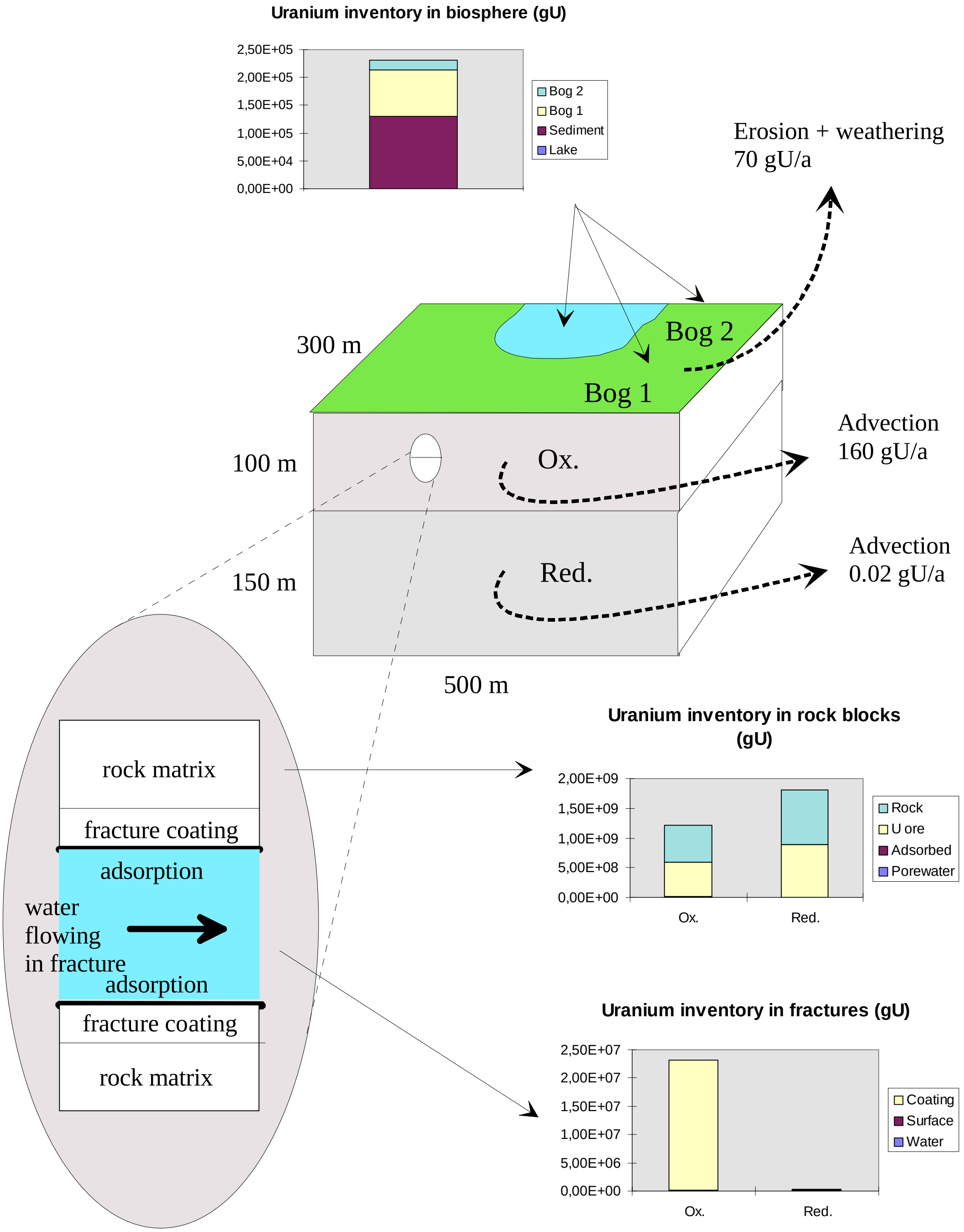

Figure 18. The most important uranium sinks at Palmottu based on the uranium balance calculations (Blomqvist et al., 2000).

Table 2. A summary of uranium concentrations from the Palmottu study area.

	URANIUM CONCENTRATION
Precipitation	0.01-0.2 µg/l
Surface waters	
Bedrock hollows of Eastern Granite	0.01-4.35 µg/l
Lake Palmottu	0.11-0.25 µg/l
Palmottu Brook	0.11-0.22 µg/l
Groundwater	
Springs (background)	0.06-0.55 µg/l
Overburden	0.02-11 µg/l
Bedrock groundwater	
Ca-HCO3-type	2.2-880 µg/l
Na-HCO3-type	0.6-363 µg/l
Na-SO4-type	1.2-19 µg/l
Na-Cl-type	0.3-8.2 µg/l
Lake sediments	
Organic	4.2-20 ppm
Minerogenic	< 2 ppm
Quaternary deposits	
Peat	0.1-180 ppm
Silt	~2 ppm
Till	4-106 ppm
Bedrock	
Eastern Granite	60 ppm
Western Granite	30 ppm
Mica Gneiss	9 ppm
Main uranium mineralization	1600 ppm
Main uranium minerals	
uraninite	UO_2 77-82 %
coffinite	UO_2 50-74 %
monazite	UO_2 1.4 %
β–uranophane	UO_2 67-75 %
Dispersed uranium phases	UO_2 > 2000 ppm
In fracture infillings	High U associated with calcites, kaolinites, iron oxyhydroxides in the upper 150 m of the site

4.4 Colloids and microbes in groundwater

The undisturbed inorganic colloid concentrations in Palmottu groundwaters are low: about 40 – 70 mg/L, consisting mainly of silica and ironhydroxide (Laaksoharju and Degueldre 1999). Measured concentrations of humic and fulvic colloids are about 5 mg/L and 3 mg/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 in U(VI) reduction and immobilization (Pedersen and Havemann 1999). Microbial biomass is not included in the colloid determinations.

5 REDOX PROCESSES

Characterization of the groundwater redox-conditions was considered to be one of the crucial tasks from the point of view of uranium behaviour. First redox measurements using packer equipment were carried out during early 90's, and the results were published by Ahonen et al. 1993, 1994. However, the most important results were obtained during the EU-financed phase from the new research boreholes, because their larger diameter (56 mm) allowed the use of the SKB mobile field laboratory. In this equipment redox potential is measured both in the hole 'in situ' and on the surface 'on line'. After comprehensive hydrogeological characterization of the Palmottu site the potential water-conductive fracture zones were packed off in most of the boreholes. A new slim sampling device was developed to fit the narrow monitoring tubes connected to the different isolated borehole sections.

5.1 Field measurements

First redox-measurements at Palmottu were done using the portable packer equipment (Figure 19) developed under the auspices of GTK. For practical reasons, those measuments could not be prolonged overnight. Measuring times were from about five hours to about eight hours, and about 300 - 600 litres of water was pumped during one measurement. Redox potentials were measured in a flow-through cell equipped with a combined Pt-AgCl electode. The portable packer device was also used in normal operation mode in the overflowing borehole (DH 302). Redox potential obtained from one-day pumping correlated well with those obtained in long-term monitoring of overflowing borehole.

SKB mobile field laboratory (Figure 20) constitutes a comprehensive equipment for complete hydrochemical characterization of deep groundwaters. The equipment has been developed and used in the Swedish nuclear waste disposal studies (Almén et al. 1986). In the SKB mobile laboratory, *in situ* electrodes receive the water sample immediately after passing the downhole pump in underground pressure and temperature conditions. Later, when water enters the ground surface, redox potentials are measured again in a gas-tight flow-through cell (*on line*). Both configurations (*in situ* and *on line*) are equipped with three different inert redox-sensitive electrodes (Pt, Au, carbon) and a reference electrode. Continuous monitoring times of single fracture systems were measured in weeks.

During the long-term isolation of the most important fracture zones in drillholes, new pump for water sampling and continuous redox measurements was developed at GTK (Figure 21). As in the older sampler (Fig. 19), pumping of water was based on pressurized nitrogen pulses. Single redox measurements in permanently packed drillhole sections lasted from a couple of days to more than a month. Stable values were normally attained within 3 to 30 hours, but prolonged stabilization times were occasionally required because of technical malfunctions. Stable readings were recorded in about 20 to 300 hours time before terminating the measurement. Measurement in overflowing (OF) borehole (302) was repeated several times, the longest continuous monitoring time being more than a month. All measured values coincide well, grouping themselves within some tens of millivolts.

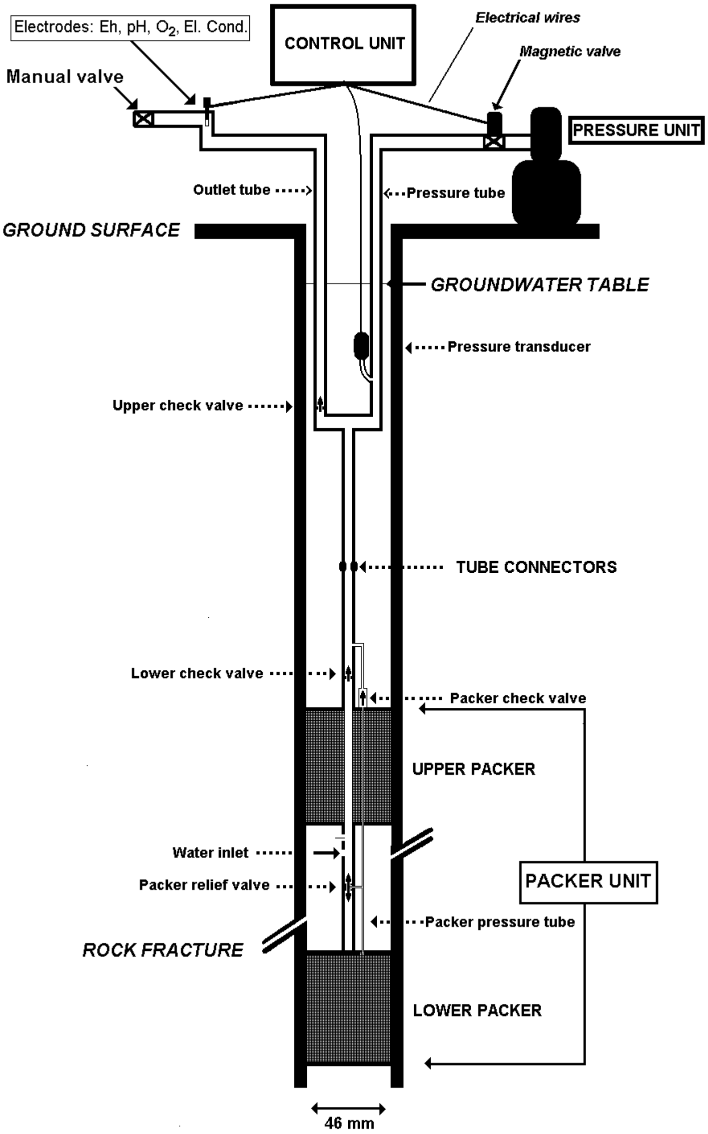

Figure 19. Schematic depiction of the light-weight double packer equipment (redrawn from Laaksoharju et al. 1995).

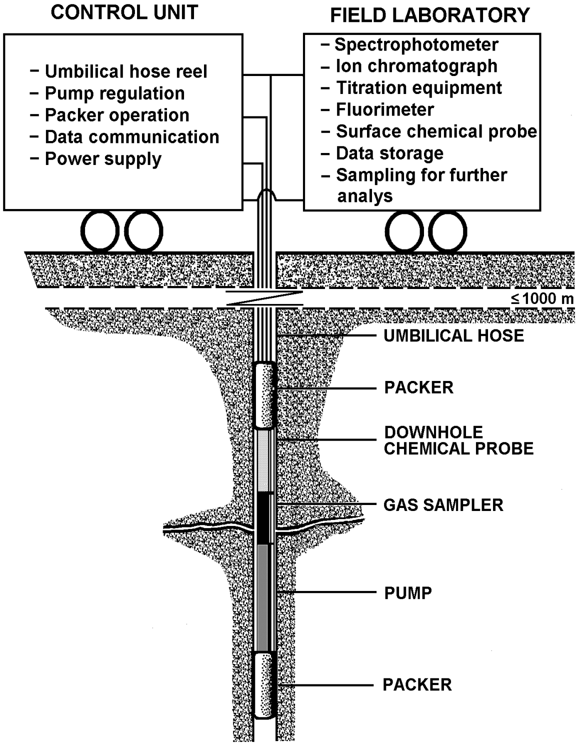

Figure 20. SKB mobile field laboratory for deep groundwater characterization (Almén et al. 1986).

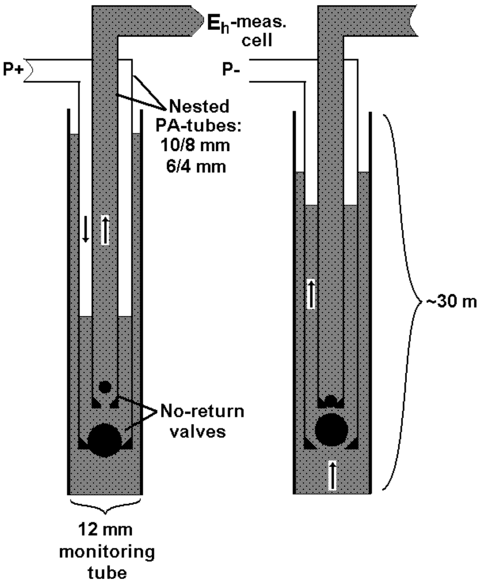

Figure 21. Schematic depiction of the slim tube sampler.

The obtained redox data were evaluated. Values probably erroneous due to technical problems were rejected. In case of SKB-laboratory measurements, values obtained by different electrodes were compared. Only values supported by more than two electrodes were chosen, and clearly discrepant values were rejected. The redox modelling was mainly based on the data obtained by *on line*-platinum electrodes. A summary of the representative redox data from Palmottu is given in Table 3.

Table 3. Selected redox data of Palmottu. Sampling equipments are described in methods section. SKB mobile laboratory pH- and redox (platinum, gold, carbon) electrodes are denoted as follows: (s) = electrode in the on-line cell on the surface, (b) = electrode in situ in the borehole chemical probe. GTK sampling systems are denoted as follows: GTK1 = light-weight double packer system; GTK2 = slim sampler for observation tubes; OF = overflowing borehole.

Drill H. ID	DH sec. (m)	Sampling system	pH		Eh (mV)					
			pH (s)	pH (b)	Pt (s)	Pt (b)	Au (b)	Au (s)	C (b)	C (s)
385	94-100	SKB	8.5	8.4	-480	-470	-50	-425	-291	-66
385	94-100	SKB	8.1		-105	-202	-182	-91	-270	-260
385	112-118	SKB	9.4	8.8	-459	-485	-375	-405	-400	-380
385	112-118	SKB		7.9		-435	-329		-363	
385	112-118	SKB	8.7	8.5	-336	-309	-311	-333	-316	-327
385	217-222	SKB	9.2		-420	-470	-290	-370	-324	-380
385	217-222	SKB	9.4		-420	-410	-320	-370	-325	-380
385	403-408	SKB	8.4		-370			-325		-320
386	72-84	SKB	8.5		-145	-270	-220	-170	-290	-300
387	32-38	SKB	8.4		480			270		380
387	101-106	SKB	8.5	8.6	-308	-290	-287	-308	-293	-308
387	101-106	SKB	8.4		-317			-316		-310
387	119-127	SKB	8.5		-300			-300		-300
387	257-264	SKB	8.5		-245			-210		-330
387	257-264	SKB	8.6		-250			-140		-200
387	304-308	SKB	8.5	8.6	-317	-300	-245	-305	-316	-302
302	90-95	OF	8.4		-55					
302	80-95	OF	8		-34					
318	50-80	GTK2	7.66		51.5					
324	95-101	GTK1	6.87		5					
325	38-81	GTK2	7.6		345					
335	45-81.5	GTK2	8.72		141					
337	80-100	GTK2	7.7		22					
346	65-71	GTK1	8.05		-11					
346	122-128	GTK1	8.4		-75					
346	240-246	GTK1	9.05		-92					
348	200-225	GTK1	8.82		-70					
384	29.5-56.5	GTK2	7.7		413					
384	56.5-94	GTK2	6.8		454					

5.2 Interpretation of redox processes

Even though the redox-potential measurement as such is a simple and straightforward process, the meaning and interpretation of the result has been a matter of debate for decades (e.g., Bruno et al. 1986). Schematic presentation of the redox measurement process is given in Figure 22.

The system redox conditions are defined by the intensive parameter redox-potential, and by the associated extensive parameter redox buffering capacity. The first mentioned, usually expressed using normal hydrogen electrode as a reference (Eh), is dependent on the activity of soluble redox-ions and their reactivity (kinetics) with each others and with the inert electrode. Kinetics of U(IV)/U(VI) –pair in U-rich Palmottu-groundwater was demonstrated by Ahonen et al. 1994. Persistency of the redox conditions is defined by the redox buffer capacity, which is largely determined by the redox-sensitive mineral phases (e.g. Fe- and U-minerals). Possible redox-buffering processes were first described by Ahonen et al. 1993 and, more thoroughly within the EU-Palmottu phase (Cera et al. 1999).

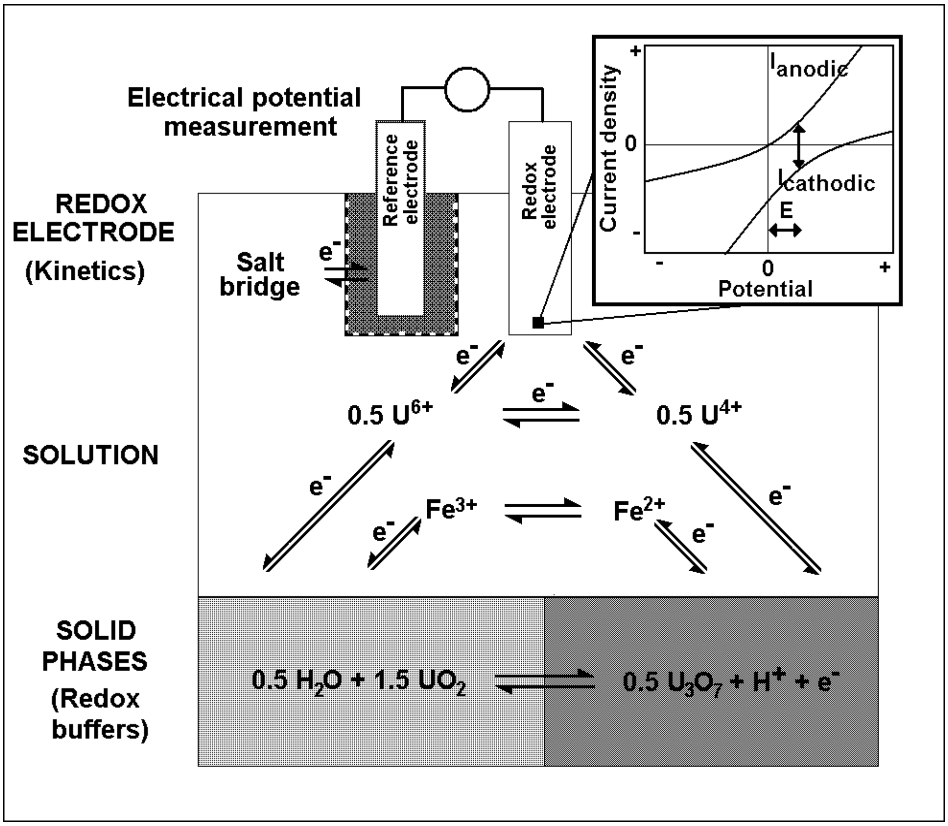

Figure 22. Principles of redox processes and redox measurement.

The results of Cera et al. 1999 indicate the presence of a redox evolution sequence. Near-surface waters are clearly oxic, as indicated by the high redox potentials; oxygen depleted waters show Eh-values matching with iron-dominated redox conditions, while sulphide system seems to control redox potentials of the deepest very reducing waters. There is also strong evidence on the impact of uranium system on the redox potentials of the mineralized zone. The results coincide with the redox-regime classification of Berner 1981, complementing it with respect to the crystalline bedrock conditions (Figure 23). A special feature of the bedrock of Palmottu is the anticipated role of uranium minerals in redox buffering of uraninite-rich environment, which was also verified by studying the redox capacity of Palmottu uranium minerals in laboratory conditions (Cera et al. 1999).

In Figure 24, representative redox data are plotted as a function of depth. Uranium concentrations of groundwaters are strongly dependent on the redox conditions and, thus, on the depth. Mobilization of uranium takes place only in oxic near-surface conditions, where uranium concentrations up to 1000 mg/L were analysed. Contrary to uranium, thorium concentrations remain below 0.05 mg/L in all conditions.

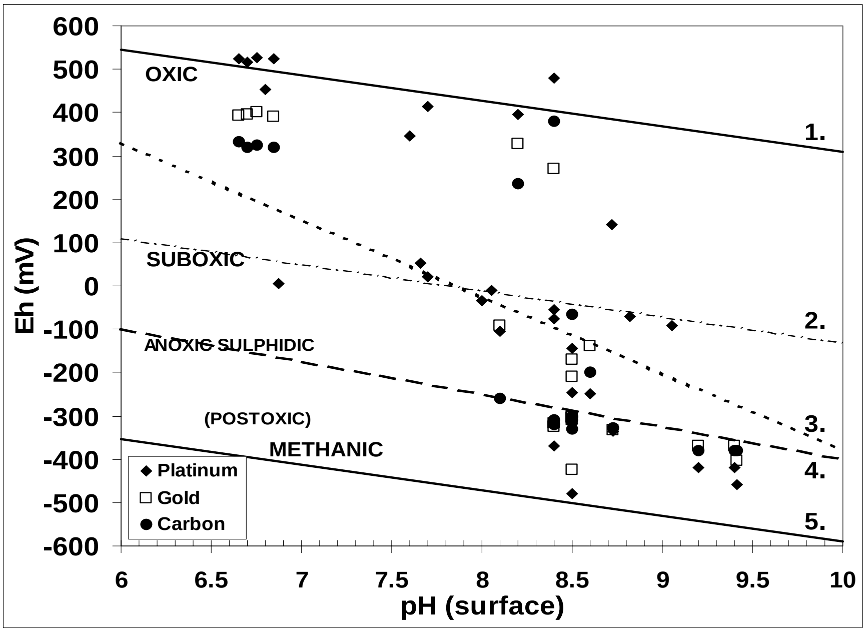

Figure 23. Measured redox values on an Eh - pH diagram. Only surface-electrode data of different electrodes are included. The numbered lines indicate different redox regimes ensuing from the concept of Berner 1981. The redox processes indicated by the numbers are:

1. $\frac{1}{2} O_2 + 2e + 2 H^+ = H_2O$
2. $0.33\ U_3O_7 + 2/3\ e + 2/3\ H+ = UO_2 + 1/3\ H_2O$
3. $Fe(OH)_3 + e + 3H^+ = Fe^{2+} + 3\ H_2O$
4. $SO_4^{2-} + 7\ e + 8\ H^+ + \frac{1}{2} Fe^{2+} = \frac{1}{2} FeS_2 + 4\ H_2O$
5. $2\ H^+ + 2e = H_2$

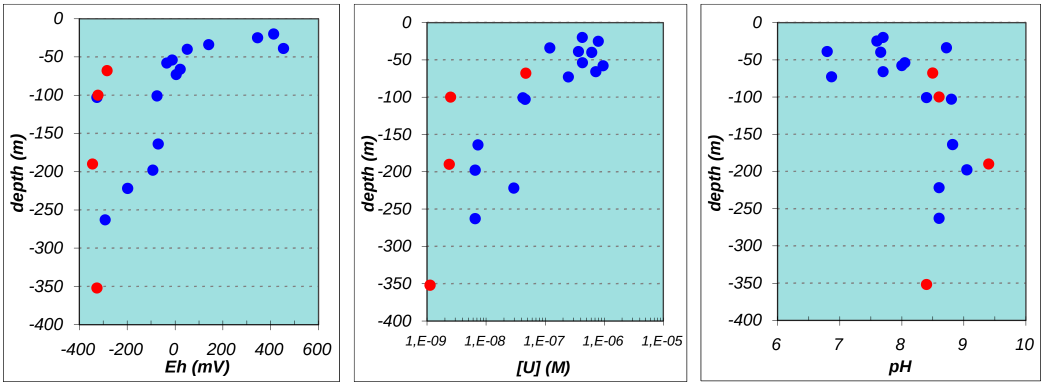

Figure 24. Redox conditions, uranium concentration and pH as a function of depth. Blue dots = data from eastern area, red dots = data from western area.

6 GEOCHEMICAL MODELLING APPROACHES

Geochemical modelling is currently a standard tool in the interpretation of hydrogeochemical results. In Palmottu research the first modelling efforts were published by Pitkänen et al. 1991, concentrating on the equilibrium-disequilibrium concepts. Later modelling efforts also included statistical approach in classification and description of mixing and evolution processes. Groundwater evolution was also modelled using mass balance calculations and reaction-path simulations (Peña et al. 1997, Gimeno & Peña 1999).

6.1 Blind predictive modelling (BPM)

Palmottu analogue study provided a good possibility to test the applicability of the predictive geochemical modelling tools. A blind predictive modelling exercise (BPM) was carried out to test the conceptual understanding on 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 of QuantiSci (Spain), Ciemat and GTK-VTT (VTT Building and Transport).

The modelling task consisted of the speciation calculations performed using different thermodynamic databases and of the associated solubility calculations. The models applied for solubility controls included precipitation/dissolution of pure mineral phases as well as the solubility controls by co-precipitation/co-dissolution process with predominant mineral phases.

The elements chosen for modelling were Rare Earth elements (REE), which may be considered as chemical homologs of actinides, and some of the elements of importance in the performance assessment of nuclear waste disposal (U,Th, Ni, Sr).

An example of performed inter-comparisons is given in Figure 25 for strontium. For uranium, the results indicate that uranophane or soddyite (U(VI) silicates) are the most likely limiting phases controlling uranium concentration in oxic conditions. Carbonate minerals and phosphates are important in controlling compositions of many trace elements and rare earths. For nickel, the results remain ambiguous.

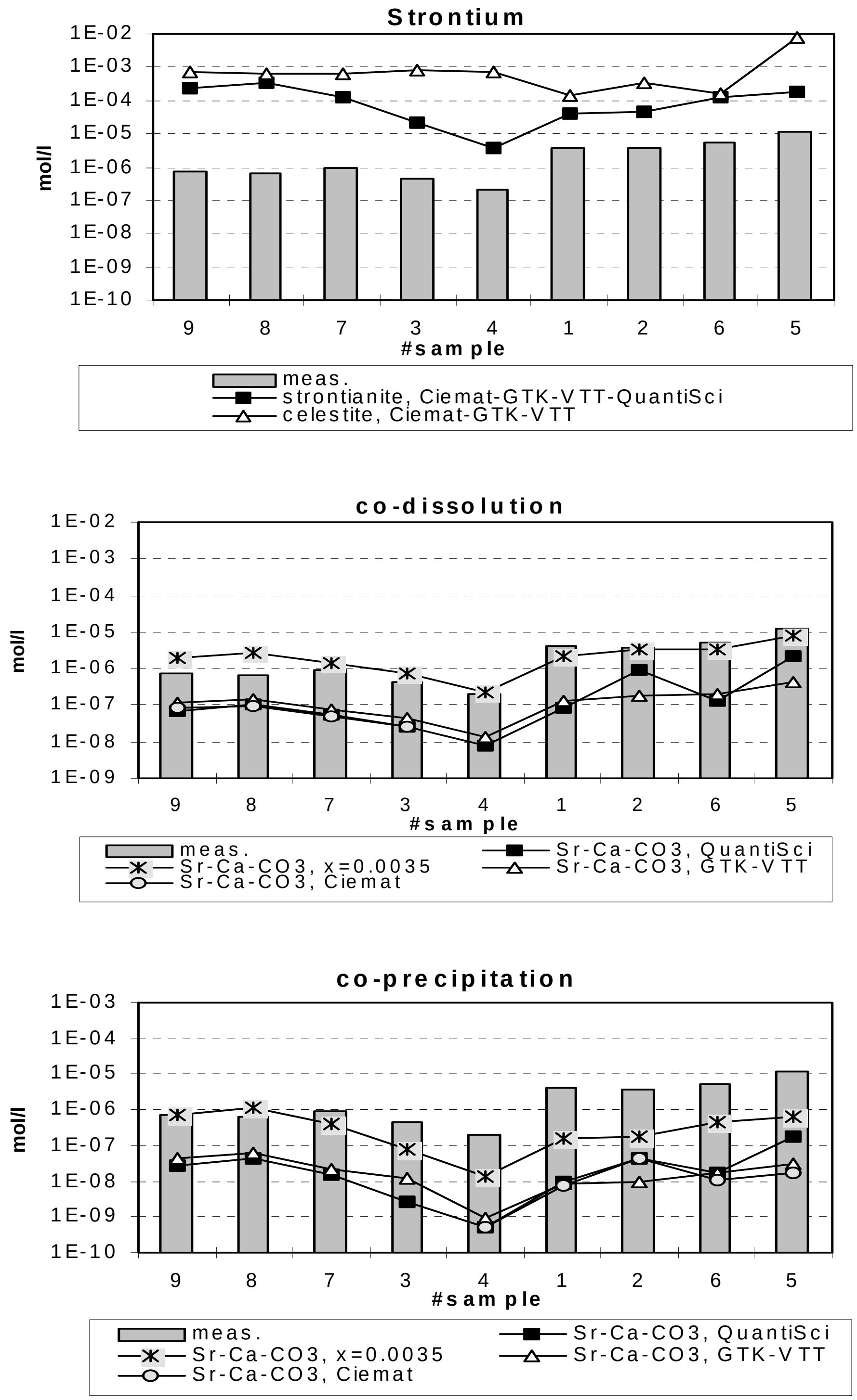

Figure 25. Comparison of actual Sr concentration with results of different modelling approaches.

7 MIGRATION PROCESSES AND MODELLING

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

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 surface (up to 25 mg/m^2) and comparing the value with the natural uranium concentration of corresponding natural groundwater. From weight to volume ratio of 50 g/L, K_D-values up to about 2×10-3 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 in many calcite samples.

Matrix diffusion is considered to be an important retardation process. 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 a-recoil.

Finally a migration modelling exercise was carried at Palmottu. Modelling study was done in steps: a 'blind' predictive phase and a modelling phase allowing calibrations and model refinement. Modelling was done by different teams using different modelling tools: VTT utilized the FTRANS code, which is based on the advection-dispersion-matrix diffusion model; Institut de Ciències de la Terra, CSIC, Barcelona used the RETRASO code (Salas and Ayora 1999), which couples flow, heat transport and geochemical processes; BRGM used the versatile ALLAN-NEPTUNIX software package, which can be used to generate specific chemical simulators. In Palmottu, a compartment type of model emphasizing geochemical processes was used by BRGM. A summary of 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 refer to a well defined hydrogeological system. The migration route starts from unsaturated bedrock of a small hill and passes through a granite or its contact zone. The groundwater evolves from rain water to very uranium rich water type along the flow path to the depth of about 100 meters, being deeper depleted in uranium. Mineralogical observations show that the fractures of the upper bedrock contain easily soluble, well crystallized U(VI) silicate uranophane, which has been considered to be the source of dissolved uranium of this flow system. Uranophane is a geologically young component in the system; the USD-dating of three samples indicates ages of 90-120 ka and 189-240 ka. These ages match with the interglacials of the Fennoscandian Quaternary history. Further study of the uranophane is still going on.

8 LESSONS LEARNED

The scope of this report is partially retrospective, with the particular aim to delineate guidelines for possible future utilization of the Palmottu research site. From the beginning, the Palmottu natural analogue study has been a part of the publicly financed nuclear waste disposal research program. During the years, the objectives and methods applied have evolved to meet the requirements and questions posed by the safety and performance reasoning of the disposal. At the same time, also definition and emphasis of natural analogue studies in general have evolved (e.g. Miller et al. 2000).

Originally, many natural analogue studies were understood rather literally to represent natural systems, which are similar to the planned disposal systems or parts of them. Soon the point of view became more process oriented, as also reflected by the general definition of the Palmottu study: “The behaviour of natural radionuclides in and around uranium deposits”. From the evolving conceptual understanding of processes of disposal systems, many natural analogues grow to a more ambitious target to produce “hard numbers”, i.e., data for mechanistic models of processes. In this respect, natural systems have the advantage and disadvantage of being realistic, i.e., extremely complex. It seems to be evident that most powerful use of natural analogues is in testing and demonstration of natural processes, rather than production of input for models. However, as shown in the Palmottu exercises, ability to model is a powerful tool for the demonstration of proper understanding of natural systems.

First years of Palmottu reconnaissance studies concentrated on the testing and development of sampling and analytical methodologies. Deep bedrock environment was still new for hydrochemical studies. In addition to the tube sampler for deep-borehole groundwaters (Nurmi & Kukkonen 1986), new samplers were developed to meet the new requirements. Analytical methods for low-level concentrations and redox state of actinides were also developed in the course of the Palmottu study (Pilviö 1998, Ervanne 2004). Methodological achievements in sampling and analysis of Palmottu groundwaters are considerable.

Studies of Palmottu bedrock concentrated on the structural geology, with a strong emphasis on the possibly water-conducting structures. Structural model, mainly based on map information, was a primary input for the local geohydrological model of the Palmottu surroundings (Palmottu bedrock block). Structural bedrock modelling performed at Palmottu is well comparable with that done in the nuclear waste site selection studies.

Site scale structural studies utilized exhaustively the boreholes drilled during the earlier ore exploration phase. The large amount of boreholes in the relatively small area of the site is an exceptional feature of Palmottu. This fact, on one hand allows very detailed studies, but on the other hand was also considered as a potentially disturbing factor for natural groundwater conditions. In order to overcome the disturbance, most of the boreholes were sealed by packers after borehole studies. The variety of geophysical and hydrogeological methods used in the borehole studies was probably wider than ever applied in Finnish hydrogeological research. Structural model of the site could thus be based on a well-established dataset. However, the hydrogeological synthesis between the interpreted, well-defined hydro-structures and the observations in the pumping- and tracer tests was ambiguous. Higher hydraulic conductivity of the uppermost hundreds of meters of the bedrock was an obvious feature, but the vertical structures could not be discerned as preferred orientation(s) of drawdown in pumping. In line with many other tracer tests, results of Palmottu-tracer test also showed that unexpected results are common in the hydrogeological fieldwork. As the hydro-structures of the crystalline bedrock are one of the key issues for the Finnish nuclear waste disposal concept, study of this theme is now carried forward within the current public research programme on nuclear waste management (KYT).

Reduction-oxidation processes have a key role in the hydrogeochemical characterization of the nuclear waste disposal systems. However, there are still conceptual uncertainties in the use and the usability of the parameter reduction-oxidation potential (usually expressed as Eh). The special strength of the Palmottu redox-studies was that the interpretations could be based on best possible measuring technology. Comparison and analysis of the results improved substantially the understanding of the redox processes of crystalline bedrock. The value of the Palmottu results for the nuclear waste disposal studies is underlined by the fact the deep-bedrock redox-processes are not very site-specific in a given type of geological environment.

Palmottu-study provided a large amount of data on bedrock groundwater chemistry and isotopic composition. As in many studies, salinity of groundwaters was observed to increase with depth. Stable isotope composition in one of the water types indicated precipitation at temperatures clearly colder than the present average, suggesting a contribution of glacial meltwaters. The obtained paleohydrogeological indications give a clue for the study of possible future cold climate scenarios.

Sorption and matrix diffusion are potentially important processes in dispersion and retention of radionuclide transport. Palmottu, as a geochemically well-defined, U-Th-rich environment is an ideal site also for future studies of these processes. Detailed studies of fracture surfaces, applying the uranium series disequilibrium methods provided important information about geologically young mobilization/immobilization processes and their time scales. Processes related to the Quaternary Ice Age may thus be traced back.

Palmottu data was available in studying the use of natural safety indicators in the safety assessment of nuclear waste. The objective of this IAEA coordinated project was to contribute to the safety assessment by means of additional safety indications derived from natural systems.

Palmottu is one of the high-priority natural analogue studies in the present EU-financed NAnet project with the aim to promote the usability of natural analogues for nuclear waste disposal. Finally, Palmottu as a well-studied site with a research friendly infrastructure provides good opportunities for various kinds of future research activities.

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