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PADAMOT

Palaeohydrogeological Data Analysis and Model Testing

The Long-Term Stability of Groundwater Conditions at Repository Sites: Proceedings of the PADAMOT Workshop, Brussels 2002

PADAMOT Project – EU FP5 Contract No FIKW-CT2001-20129

Technical Report WP1

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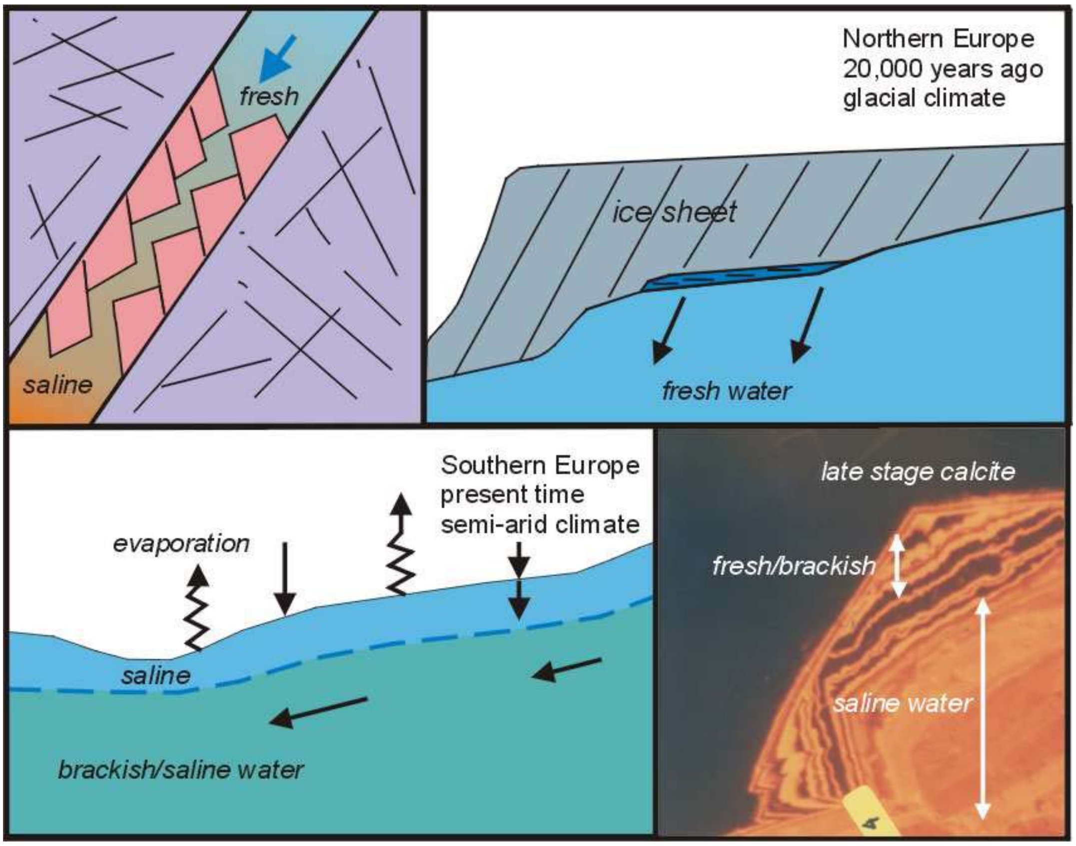

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PADAMOT PROJECT

TECHNICAL REPORT WP1

The Long-Term Stability of Groundwater Conditions at Repository Sites: Proceedings of the PADAMOT Workshop, Brussels 2002

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Foreword

PADAMOT, 'Palaeohydrogeological Data Analysis and Model Testing', is a project within the European Union's 5th Framework RTD programme in nuclear fission safety (Contract Number FIKW-CT-2001-00129). It aims to improve the understanding of past groundwater conditions that supports assessments of future long-term safety of repositories for radioactive wastes. The project began in December 2001 with duration of 36 months. The consortium of organisations involved in the PADAMOT project comprises:

United Kingdom Nirex Limited (UK)

Svensk Kärnbränslehantering AB (Sweden)

Terralogica AB (Sweden)

Empresa Nacional de Residuos Radioactivos S.A. (Spain)

Centro de Investigaciones Energéticas, Medioambientales y Tecnológicas (Spain)

Intellisci Ltd (UK)

British Geological Survey (UK)

Charles University (Czech Republic)

University of Edinburgh (UK)

Universidad Politecnica de Madrid - School of Mines (Spain)

Safety assessments of proposed repositories for the long-term storage or disposal of radioactive wastes must take into account scenarios for environmental change over the long period of time during which the waste will be a hazard, typically up to one million years into the future. The scientific consensus in a number of countries is that disposing of long-lived and/or higher activity radioactive wastes and spent nuclear fuel deep underground in a 'geological repository' is the preferred option for long-term radioactive waste management. The reasons for preferring this option are that the host rock for a deep repository should provide stable conditions for performance of the engineered barrier system and that the rock mass separating a repository from the surface environment is a further barrier to radionuclide migration.

During the last two million years (the Quaternary Period), global climate has fluctuated between extremes of ice ages and warmer conditions than at present. Over various intervals in the past, large areas of northern Europe were covered by ice sheets and experienced extensive permafrost, whilst southern Europe was sometimes more pluvial (wetter). Consequently, the present-day climate is not representative of the climate that has existed for much of the Quaternary. This natural pattern of climatic fluctuation is expected to continue into the future, albeit modified by the impacts of anthropogenic greenhouse gas emissions. Variations in climate and in other environmental factors may affect future movements and compositions of groundwaters in the vicinity of a repository and thus affect the mobility of radionuclides and the rate of their migration back to the surface. It could be argued, therefore, that present-day groundwater conditions may not be an adequate basis for assessing long-term repository safety. However, if it can be demonstrated that, despite significant environmental change at the surface, groundwater flows and compositions at depth remain stable or change in a way that does not impact significantly on safety, then confidence in repository concepts for disposal will be increased.

PADAMOT has sought to address the following questions. How can such groundwater stability be assessed, with respect to climate-driven environmental change? In particular, what evidence is there that a deep geological repository will eliminate or reduce the effects of extreme changes in environmental conditions in the long term? In seeking to answer these questions, PADAMOT has investigated geosphere systems at various European sites, using analytical methods and numerical modelling.

PADAMOT comprises five work packages (WPs) with the following tasks:

- WP1. Convening a preliminary workshop of PA specialists, PADAMOT researchers and other geoscientists on the use of palaeohydrogeology in PA.
- WP2. Making palaeohydrogeological data measurements on mineral samples and groundwaters from sites in Spain, Czech Republic, Sweden and UK, using high resolution and high precision analytical methods, e.g. ion probe and laser ablation.
- WP3. Constructing a relational database and a public domain website to store data from EQUIP and PADAMOT, accessible to project partners and to external researchers via the internet.
- WP4. Developing numerical models to test palaeohydrogeological information interpreted from proxy geochemical, mineralogical and isotopic data, based on understanding of the processes that link the proxy data with climate-driven groundwater phenomena.
- WP5. Synthesising project outcomes and disseminating an improved approach to the use of palaeohydrogeological information in the description of FEPs and hydrogeological scenarios for PA.

There are final reports from each of the five WPs plus a Summary report:

- Technical Report WP1. The Long-Term Stability of Groundwater Conditions at Repository Sites: Proceedings of the PADAMOT Workshop, Brussels 2002.
- Technical Report WP2. Application of Mineralogical, Petrological and Geochemical Tools for Evaluating the Palaeohydrogeological Evolution of the PADAMOT Study Sites.
- Technical Report WP3. Design and Compilation of Database: Final Report.
- Technical Report WP4. Interpretative Modelling of Palaeohydrogeological Data: Final Report.
- Technical Report WP5. Dissemination and Use of Palaeohydrogeological Results for Safety Assessment.
- Summary Report. PADAMOT: Palaeohydrogeological Data Analysis and Model Testing – Overview.

Conditions of Publication

Further Information

Further information on the PADAMOT programme can be obtained from the project website www.bgs.ac.uk/padamot

Abstract

A workshop was held at the Hotel Carrefour de la Europe in Brussels on 2nd & 3rd October 2002 as part of the project 'Palaeohydrogeological Data Analysis and Model Testing' (PADAMOT), being undertaken in the EC/Euratom 5th Framework RTD programme in nuclear fission safety. PADAMOT continues the research directions that were started in the 4th Framework EQUIP project ('Evidence from Quaternary Infills for Palaeohydrogeology'). In addition, it will develop improved methods and recommendations for linking these data with the requirements for long-term safety assessments.

This volume of Proceedings records the workshop presentations and the questions, answers and comments raised in subsequent discussion. The first set of presentations documented here provide overviews of past EC projects on palaeohydrogeology, namely the EQUIP, PHYMOL and PAGEPA projects (the ‘PALHY’ cluster in the 4th Framework Programme) and a related 4th Framework Programme investigation of the Palmottu site in Finland. In addition to these summary papers an introduction to the PADAMOT project itself is provided as is a brief synopsis of the potential use of novel analytical techniques for future palaeohydrogeological studies.

.A key part of the workshop was the involvement of independent expert ‘facilitators’ to synthesise issues. Two papers are included here relating to the value and use of palaeohydrogeological information in past repository safety assessments. Finally, this report documents the summaries of the working group and plenary discussions that were led by the independent experts, addressing the questions:

- How do hydrochemical & mineralogical data support confidence in deep repository stability?
- How can that knowledge be applied more effectively in future safety cases?

Acknowledgements

The PADAMOT consortium acknowledges the active participation by representatives of many organisations, and especially the contributions by the expert ‘facilitators’ Tim McEwen, Andrew Bowden and John Smellie.

The support and involvement of Henning von Maravic and Michel Raynal of EC-DG Research is gratefully acknowledged. The workshop was carried out as Work Package 1 of the PADAMOT project in the European Union’s 5th Framework Programme of RTD in Nuclear Fission Safety, Contract No. FIKW-CT-2001-00129. It was also supported by funds from UK Nirex Limited, ENRESA and SKB.

Contents

1 Introduction and Scope of Workshop

Paul Degnan and Adrian Bath

1.1 THE ISSUES

Many countries generate nuclear power or make extensive use of radioisotopes in health and industrial applications. The safe management of the radioactive wastes that arise from these activities are the responsibility of the producers, national regulatory bodies and radioactive waste management agencies. One option being considered for the long-term management and eventual disposal of such radioactive waste is the construction of repositories deep underground in geological environments, selected to ensure the safety of radioactive waste containment for very long periods of time. Deep emplacement would put a repository beyond the reach of direct impacts from climate change and other processes that might alter the surface and near-surface environment. It would also reduce the potential for human intrusion. In a robust strategy for deep 'geological disposal' there are three steps that should be considered by waste management agencies to support this option:

- Firstly, scientific evidence for the long-term stability of rock, groundwater movement and chemical conditions at depth needs to be collected and examined critically.
- Secondly, predictions of safety should take into account the potential effects of changes in climate and other external impacts, and should represent those changes in terms of a scientific understanding of the processes and of their probabilities.
- Thirdly, but not least, is the need to explain and justify in easily understandable terms the basis for both the scientific understanding and the safety calculations. This will provide increased confidence in the geological disposal option for long-term radioactive waste management.

1.2 SAFETY CASES AND SCENARIOS

Over the past decade there have been several detailed calculations of long-term repository performance that support the concept of deep geological disposal. Most of these exercises have been 'dry runs' in which a generic site concept or a specific 'example' site has formed the basis of performance assessment (PA) calculations of long-term safety, either by a repository proponent or by a regulatory authority. In a few cases, there have been preliminary PA calculations for one or more specific sites as an early stage in the progress through site selection, full site investigation and application for a licence to construct a repository (e.g. SKB-SR97 in Sweden, TILA-99 in Finland, Nirex 97 in the United Kingdom). None of the European national deep repository programmes has yet progressed to the final stage of licensing for construction on the basis of a comprehensive PA and regulatory/public/political consent.

Performance assessments generally contain a detailed calculation for a 'base case' situation that assumes a stable geosphere around the repository, i.e. that the present state of groundwater chemistry and movement at depth will remain essentially unaffected by external changes. The potential impact of future changes to the geological or surface environment is taken into account through the analysis of 'scenarios' or 'base case variants', whereby additional performance calculations investigate system sensitivity to alternative future conditions. To handle uncertainties in the magnitude of processes that link climate changes with impacts on a deep repository, worst case scenarios have been assessed in a conservative manner. The way that these scenarios are handled is central to the demonstration of the long-term safety of deep geological disposal and to its acceptance as a strategy to deal with long-lived radioactive waste.

1.3 THE MULTI-BARRIER CONCEPT AND GEOSPHERE STABILITY

The multi-barrier design concept of a deep repository assumes the stability of the deep geosphere to ensure a continuity of known physical and chemical conditions in which the engineered containment of waste packages and backfill materials can operate as designed for long periods (Figure 1). In addition to tectonic and rock mechanical stability, other geosphere factors, including the rate of groundwater movement and the chemical composition of groundwater, affect the performance of the waste containers and of the buffer and backfill material surrounding the containers, and also affect the dissolution and migration of radionuclides. In some assessments, the predicted performance of the various design concepts for engineered containment is generally so good that less emphasis tends to be placed on the performance of groundwater pathways in the far field in attenuating releases to the biosphere. Nevertheless, the stability of deep groundwater, in comparison with transient effects that are evident in near-surface groundwaters, is part of the safety case for the reasons given above.

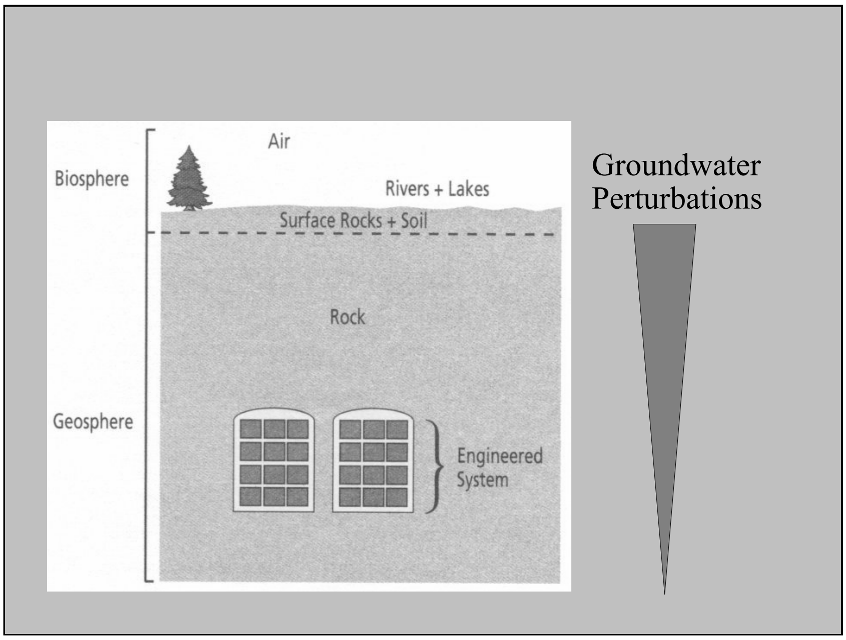

Figure 1. Isolation of radioactive wastes in a deep geological repository to take advantage of a multi-barrier system. The repository (engineered system) would be constructed at a depth and location where the direct effects of any climatic or environmental change in the biosphere would be negligible and where groundwater stability could be assured.

1.4 OUTSTANDING QUESTIONS

Many questions have been identified concerning the use of scenarios and variant models, and whether they are realistic, pessimistic or optimistic for deep repository locations. Some of these questions were: Are there particular characteristics of the deep geosphere that make some locations preferable to others in terms of long-term stability? What evidence is there from studies of past groundwater conditions and its evolution to support the scenarios and variants, and to support the inferred links between climate and groundwater conditions? Are there alternative conceptual models or scenarios that present different safety outcomes, either optimistic or pessimistic? How important are the transitions from variant climate states (concerning subsequent impacts), as opposed to interpretations based simply on the assumption of new equilibrium states alone?

Without evidence to demonstrate the inherent stability of the deep geosphere to long-term climate change, many scenarios will be unconstrained. In such a situation confidence in the long-term safety of geological disposal will be vulnerable to extreme prognoses – the very scenarios that deep disposal should protect against.

1.5 THE WORKSHOP: OBJECTIVES AND PARTICIPATION

The objectives for the workshop were to:

- Consider evidence that is relevant to concepts of long-term stability or instability in the hydrogeological and hydrochemical environment at typical repository depths (>100m), from the EQUIP project and other investigations;
- Identify generic and site-specific data and interpretations that improve understanding of long-term groundwater conditions at repository depth;
- Review how palaeohydrogeological evidence for the evolution of groundwater conditions has been used in safety assessments for deep geological disposal;

Propose new and improved methods for incorporating palaeohydrogeological knowledge into safety assessments for deep disposal.

To achieve the objectives, participation in the workshop by safety assessment experts, geoscientists and site characterisation planners who are involved in assessing groundwater stability was invited. The workshop was planned to present up-to-date views of investigation techniques, the types and accuracy of data that can be obtained, the uncertainties in these data, and the methods for integrated interpretations that can be effectively used in PA.

1.6 ORGANISATION AND BACKGROUND

The workshop was organised as part of the project 'Palaeohydrogeological Data Analysis and Model Testing' (PADAMOT) in the EC/Euratom 5th Framework RTD programme in nuclear fission safety. PADAMOT continues the research directions that were started in the 4th Framework EQUIP project ('Evidence from Quaternary Infills for Palaeohydrogeology'), and in addition will develop ways of linking these data with PA and safety assessments.

A motivating reason for PADAMOT hosting the workshop was to ensure that the compilation of palaeohydrogeological evidence and supporting models remained focussed on assisting safety assessments of repositories for the geological disposal of radioactive wastes. Although much of the PA experience and research to date has been concerned with potential repositories in fractured crystalline rocks, the issues raised in the workshop are of relevance also to proposed repository developments in argillaceous rocks and other geological environments. Members of the PADAMOT consortium represent nuclear waste management agencies and research organisations from across Europe (Table 1).

The first day of the workshop consisted of invited presentations. The starting points were reviews of (i) recent research into the geoscientific parameters and processes that link groundwater conditions with climate and other time-varying influences and (ii) how the question of groundwater stability has been handled in previous PA exercises. To address the first of these, results from previous studies in the EC 4th Framework 'PALHY' cluster - EQUIP ('Evidence from Quaternary Infills for Palaeohydrogeology'), PAGEPA ('Development and Testing of Models for Climate Impacts on Groundwaters') and PHYMOL ('Palaeohydrogeology in a Regional Sedimentary System Containing Clay Layers') – were presented (A. Bath, I. Wemaere & J. Marivoet, and G. Boulton). Additionally, information from the Finnish research site at Palmottu was summarised (J. Smellie) - see Sections 2-5 in this report. P. Degnan described how the PADAMOT project is extending palaeohydrogeological knowledge to support PA (Section 6). To recap on the previous use of palaeohydrogeology in PA, invited speakers (T. McEwen and A. Bowden) provided a succinct and comprehensive summary (Sections 7-8).

Table 1. Organisations involved in PADAMOT

Partner number	Organisation	Type of Organisation	Country
1	United Kingdom Nirex Limited	Rad-Waste Management	UK
2	Svensk Kärnbränslehantering AB	Rad-Waste Management	Sweden
3	Terralogica AB	Consultant	Sweden
4	Empresa Nacional de Residuos Radioactivos S.A.	Rad-Waste Management	Spain
5	Centro de Investigaciones Energeticas, Medioambientales y Tecnologicas	Research	Spain
6	Intellisci Ltd	Consultant	UK
7	British Geological Survey	Research	UK
8	Charles University	University	Czech Republic
9	Edinburgh University	University	UK
10	Universidad Politecnica de Madrid - School of Mines	University	Spain

The second day of the workshop was taken-up with breakout group discussions and reporting. The questions addressed in the breakout groups were (a) How do hydrochemical/mineralogical data support confidence in deep repository stability; and (b) How can that knowledge be applied more effectively in future performance assessments and safety cases. A post-workshop paper by A. Milodowski is presented in this report to provide information on some of the developing technologies that could be used in palaeohydrogeological studies (Section 9). These technologies were discussed in outline during the concluding plenary session. Summaries of the full deliberations are included in Section 10.

2 EQUIP: Field Evidence to Support Chemical and Hydrogeological Stability

Adrian Bath, Intellisci, UK

2.1 INTRODUCTION

Performance Assessments (PA) of proposed repositories for radioactive wastes evaluate how engineering design and geological siting can best contain radionuclides and prevent them from reaching the biosphere for a long time into the future. Climate changes will affect the upper boundary of groundwater systems within that period. The capacity of repository sites deep underground in stable rock masses to mitigate potential impacts of future climate change on groundwater conditions needs to be tested and demonstrated. The possibility of such changes in the future can only be assessed by studying groundwater evolution in the past. Stability of groundwater conditions in the past is an indication of future stability, and is therefore a criterion for suitability of a repository site.

EQUIP: Evidence from Quaternary Infills for Palaeohydrogeology

EC FP4 project FI4W-CT96-0031

Research contractors:	*Co-funding agencies:*
British Geological Survey	SKB, Sweden
Golder Associates, UK	Posiva, Finland
Fintact Oy, Finland	NIREX, UK
Terralogica, Sweden	ENRESA, Spain
Universidad Politecnica de Madrid, Spain	ANDRA, France
BRGM, France	Environment Agency, UK
Bristol University, UK	

Figure 2. Organisations involved in EQUIP, in conjunction with EC DG-Research.

EQUIP ('Evidence from Quaternary Infills for Palaeohydrogeology') was a 3-year project in the EC's 4th Framework Programme, reporting in 2000 (Bath et al., 2000). The project was coordinated with other EC 4th Framework projects (PAGEPA and PHYMOL) in the 'PALHY' cluster. A summary of the PALHY projects was given at the Euradwaste '99 conference in Luxembourg (Bath et al., 1999).

EQUIP Partners from Finland, France, Spain, Sweden and UK (see Figure 2) studied the information retained by secondary minerals about past groundwaters from which they formed, supplementing what can be learnt from groundwater chemical and isotopic compositions and from modelling. Evidence for groundwater conditions during the Quaternary (i.e. up to a million years ago) were studied by EQUIP because this timescale and its climate variability are comparable with those of interest in a repository safety case.

Study of groundwater conditions in the past, or 'palaeohydrogeology', has three basic approaches: numerical modelling of how groundwater flow would have responded to changing boundary conditions, extraction of evidence for past flows and compositions from pressure and chemical data for present-day groundwaters, and studies of rock properties and mineral compositions which record various properties of groundwaters at the times when they were formed or altered (Figure 3).

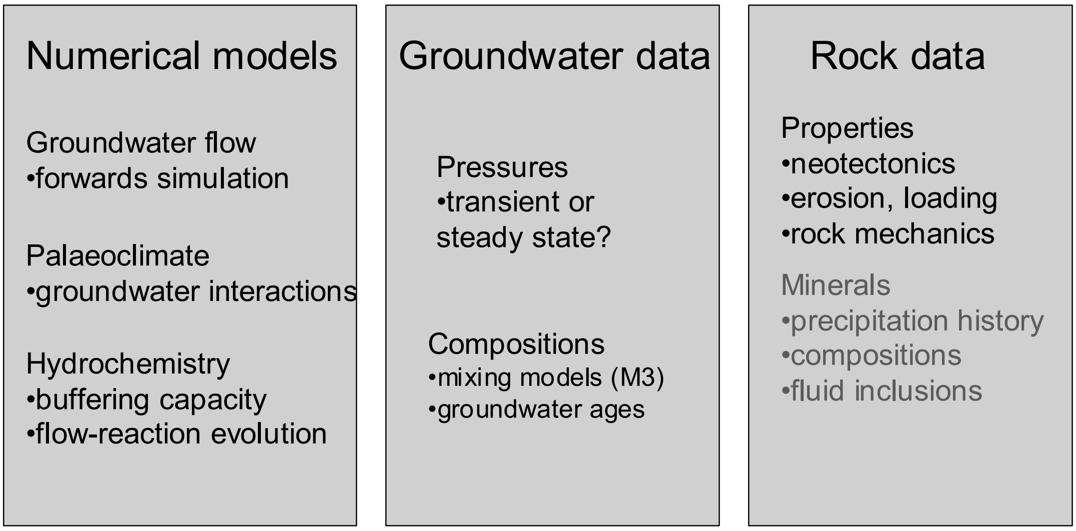

Figure 3. Scientific methods for palaeohydrogeology, i.e. study of groundwater conditions in the past.

EQUIP focused on the third of these methods, and specifically on the use of geochemical data for secondary minerals (predominantly calcite) to gain knowledge about past groundwater conditions.

An overview of the approach used in the project is shown in Figure 4, starting with the identification by petrography and mineralogy of late-stage minerals in fracture rock samples, and concluding with an interpretation of the new data in terms of long-term groundwater evolution.

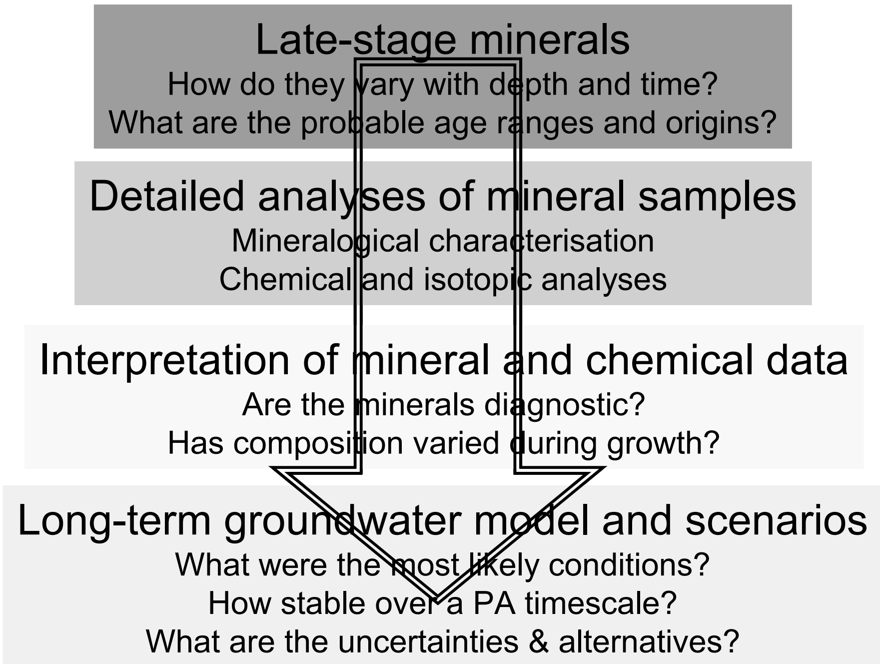

Figure 4. Summary of approach used in EQUIP.

2.2 LOCATIONS OF STUDY

Late-stage fracture minerals (mainly calcite) were sampled from drillcores in low permeability rocks in UK (Sellafield), Sweden (Äspö), Finland (Olkiluoto) and France (Vienne) (Sites 1 to 4, Figure 4). They were petrographically characterised and analysed for compositional variations and fluid inclusion contents. In addition, two sequences of Quaternary sediments in southern Spain at Padul and Cúllar-Baza (Site 5, Figure 5) were studied. They are located in a basin filled with Pleistocene-Holocene sediments for which a continual record of the local Quaternary hydrological environment was sought by studies of carbonate microfauna (ostracods) and organic geochemistry.

Various microanalytical methods were developed and optimised for analyses of trace elements, isotopic ratios and fluid inclusions in fracture minerals. Late stage minerals are generally very much less abundant than older minerals and in many cases this was the main analytical limitation, especially for isotopic age data.

Sites 1-4 were locations of site investigations or research programmes in fractured crystalline rock from which drillcore samples were available (investigations at Sellafield and Vienne have since stopped). Site 5 was rather different, being located in a basin filled with Pleistocene-Holocene sediments in which a continual record of the Quaternary hydrological environment in southern Spain was sought by studies of carbonate microfauna (ostracods) and organic geochemistry.

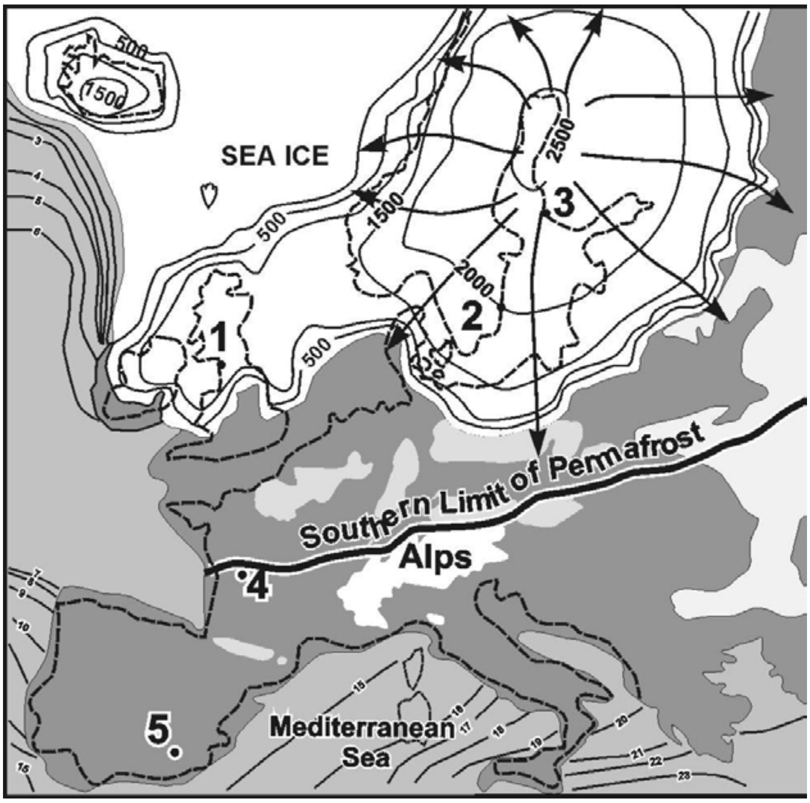

Figure 5. Locations of EQUIP study sites, located in a palaeogeographical map showing the extent of ice sheet advance in the late Weichselian glaciation.

Some of the processes that influence the long-term stability of a typical groundwater system that extends to more than 500 m depth are illustrated in Figure 6. The processes that might be related to climate are shown, including those directly related, such as recharge and sea level change for a coastal location, and also those that may be more indirectly influenced by climate such as mixing between different water masses.

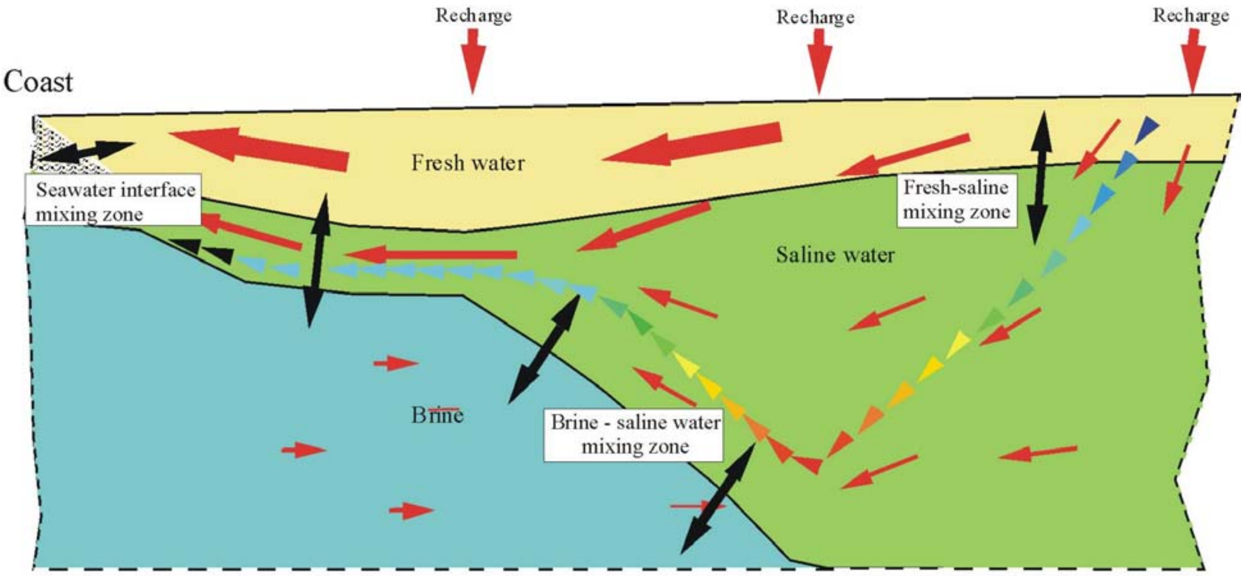

Figure 6. Conceptual model of processes affecting groundwater stability.

The questions pertaining to groundwater stability in northern European regions, i.e. where the dominant climate impacts in the Quaternary have been glaciation and permafrost, are:

- To what depth is groundwater perturbed by the effects of changing climate and topography on recharge?
  - Do glaciation and permafrost have the greatest impacts?
  - Do the observed mineral/geochemical changes correlate with those events?
- Have recharge 'events' (e.g. meltwater) changed redox conditions significantly?
- Have tectonics and sea level changes in last 106 y moved the fresh-saline groundwater distributions?

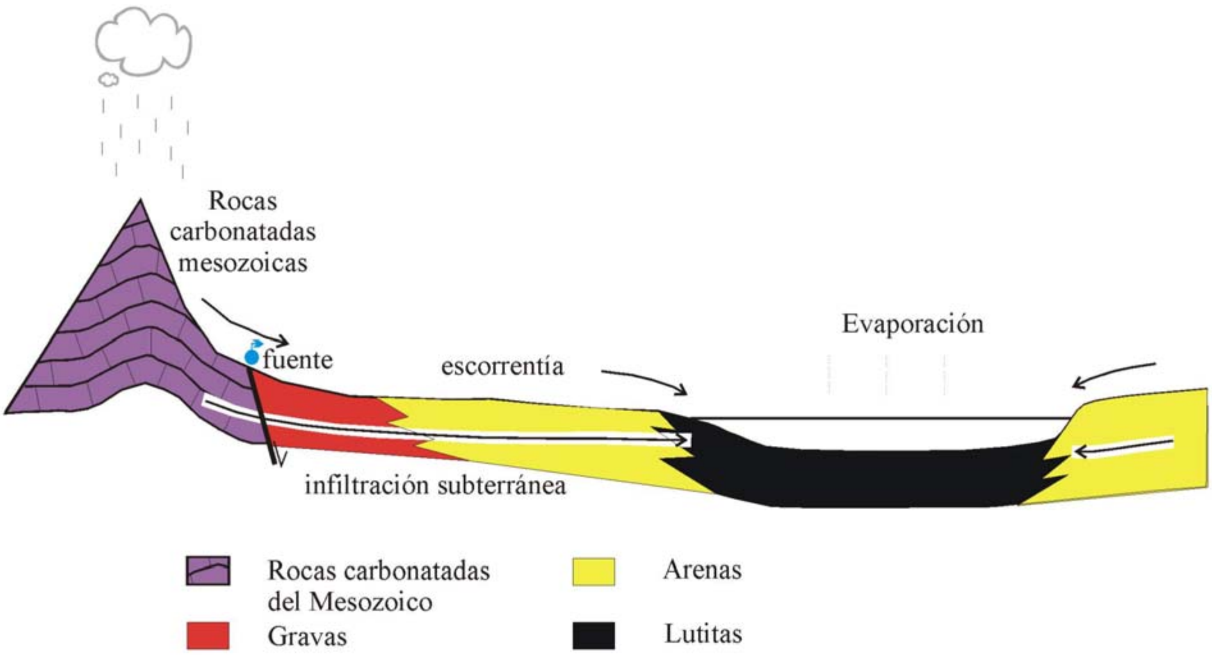

Figure 7. Geological and hydrological system in the Cúllar-Baza basin, Spain, illustrating the interplay between surface water run off, groundwater recharge and discharge, and basinal evaporation.

In southern and central Europe, beyond the reach of glaciation and permafrost (see Figure 4), the palaeohydrogeological issues are rather different:

- To what depth is groundwater perturbed by the effects of changing climate on groundwater recharge and water table?
    - Does evaporation cause significant salinity changes?
    - Do low topography areas switch between discharge and recharge in climate cycles?

For example, these questions are among those posed in the study of the Cúllar-Baza basin by the Spanish partners in EQUIP (Figure 7). Studies of sediment cores from the basin have applied organic geochemistry, trace elements and stable isotopes as sources of evidence for the Quaternary palaeohydrology of the basin.

2.3 SAMPLING AND ANALYTICAL METHODS

Samples of fracture-filling minerals were obtained from drillcores for the crystalline rock study sites. The quality of core recovery varied, and the preservation of secondary minerals was similarly variable, especially since the amounts present were generally very low except in Sellafield core samples. Procedures for drilling and handling cores to optimise the recovery of representative fracture minerals have been recommended for guidance of future site investigations. The numbers of mineral samples from each of the study sites, including soft sediment cores from the Spanish site, were:

- Sellafield: 130 samples from 18 boreholes to 1500m depth
- Äspö/Laxemar: 26 samples from 10 boreholes to 1600m
- Olkiluoto: 35 samples from 7 boreholes to 560m
- Vienne: 65 samples from 7 boreholes to 970m
- Cúllar-Baza & Padul: ~130 ostracod samples & ~100 organic sediments

A fairly comprehensive suite of analytical methods for mineralogical and geochemical analyses was deployed on samples, although not all of the techniques were available in all partners' laboratories:

- optical petrography
- scanning electron microscopy
- backscattered SEM imaging
- cathodoluminescence (CL; cold cathode)
- BSEM/EDXA chemical analysis
- electron probe microanalysis (point analysis & mapping)
- laser ablation ICP-mass spec
- microsampling for stable isotopes & trace elements
- microthermometry & LA-ICP-MS analyses of fluid inclusions
- GC-MS analysis of organics
- stable isotopes & trace elements in ostracod microfossils.

The main limitations arose from the absence of a method for U-Th isotope analyses at sufficient sensitivity for the U content of these calcite samples, and from the uncertainties in the stable O and C isotope micro-analyses of calcite zones at the micrometre scale by laser ablation isotope ratio mass spectrometry. Stable isotope analyses were carried out on some bulk fracture samples (e.g. from Äspö and Olkiluoto), and also on the ostracods samples from Cúllar-Baza.

2.4 PALAEOHYDROGEOLOGICAL INDICATORS IN FRACTURE CALCITES

Following findings from preliminary work by BGS prior to the project, EQUIP focused on a few key indicators of past chemical conditions in groundwaters from which secondary calcite has precipitated in fractured rocks: calcite crystal morphologies, zoning of trace element contents of calcites, compositions of fluid inclusions in calcites, and stable isotope compositions of calcites.

2.4.1 Morphology of secondary calcite

It had been established in earlier work on calcites from the deep boreholes at Sellafield that the morphology of calcite crystals correlated approximately with the location of the transition of groundwater compositions from fresh/brackish to saline, i.e. at ca. 3000 mg/l chloride (Figure 8).

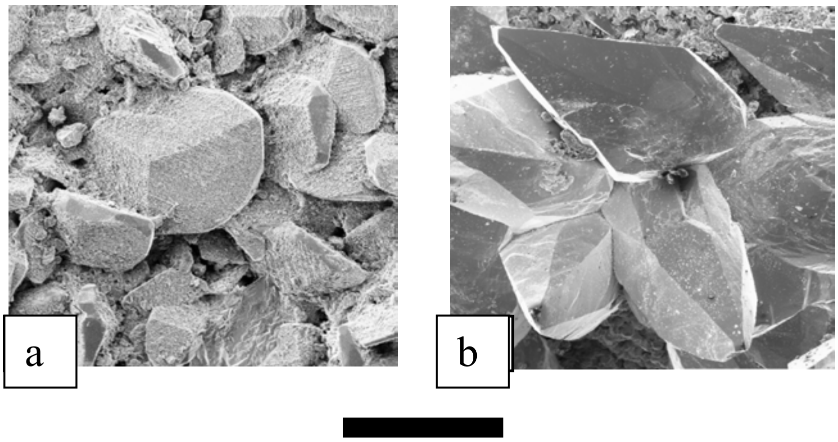

Figure 8. Scanning electron micrographs of fracture-filling calcites in drillcores from boreholes at Sellafield, UK (scale bar = 2 µm), coexisting with (a) fresh/ brackish groundwater and (b) saline groundwater >3000 mg/l Cl^-.

Figure 9 shows how this morphological relationship with salinity provides evidence of changing salinity. The sample shown comes from just above the present-day location of the fresh/saline transition. The change in morphology during calcite growth indicates that the salinity of coexisting groundwater was higher in the past than at the present.

The implications of these observations of calcite morphology can be summarised as follows:

- Calcite morphology correlates with fresh/saline transition (~3000 mg/l Cl^-) in all boreholes at Sellafield and in samples from Äspö.
- Overgrowths indicate that the fresh/saline transition zone at Sellafield has moved vertically by 10's of metres so that it was higher at some time during the growth period of late-stage calcite (duration uncertain but probably 10^2-10^4 y).
- Calcite morphology also correlates with stable isotope ratios at Äspö, suggesting that mixing of cold climate water with saline water persisted through growth period.

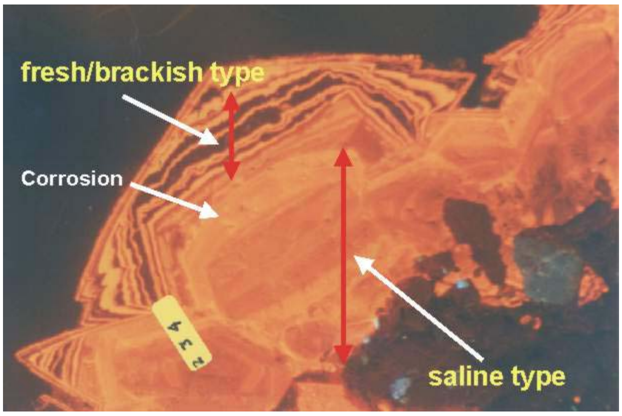

Figure 9. Cathodoluminescence image of secondary calcite from 630 m depth in Borehole 10 at Sellafield, showing how the crystal morphology of growth zoning has changed, suggesting that the salinity of coexisting groundwater has changed from saline to fresh/brackish.

2.4.2 Zoning of trace element contents of secondary calcite

Zones of varying degrees of cathodoluminescent (CL) intensity indicate that the trace element contents of the calcite zones vary. This variation is significant because it involves principally the contents of Fe and Mn which are redox-sensitive. Their variability can therefore be related to variability of redox in coexisting groundwaters. For example, the calcite in Figure 10 comes from the fresh water zone at Sellafield and shows CL banding that reflects varying Fe:Mn ratios. In this sample, the interpretation of Fe:Mn ratios suggests that redox has varied within a fairly small range, maintaining reducing conditions (i.e. <0 mV).

The spatial variations of secondary calcite compositions in samples from Sellafield can be summarised as follows:

1. Late stage calcite with fine luminescent (Mn-rich) bands is typical of fresh groundwater – the pattern does not correlate between boreholes. This indicates local fluctuations between oxidising (Fe-ox + Mn-carb equilibrium = luminescence) and slightly reducing (Fe-carb + Mn-carb = non-luminescent).
2. Late stage calcite with more or less dull luminescence is typical of saline groundwater – the pattern correlates between boreholes locally with fewer zones as salinity increases. This indicates episodes of Fe-sulphide equilibrium (higher Mn/Fe ratio = more luminescent): possibly periods of higher SO_4 ± stronger reducing redox.

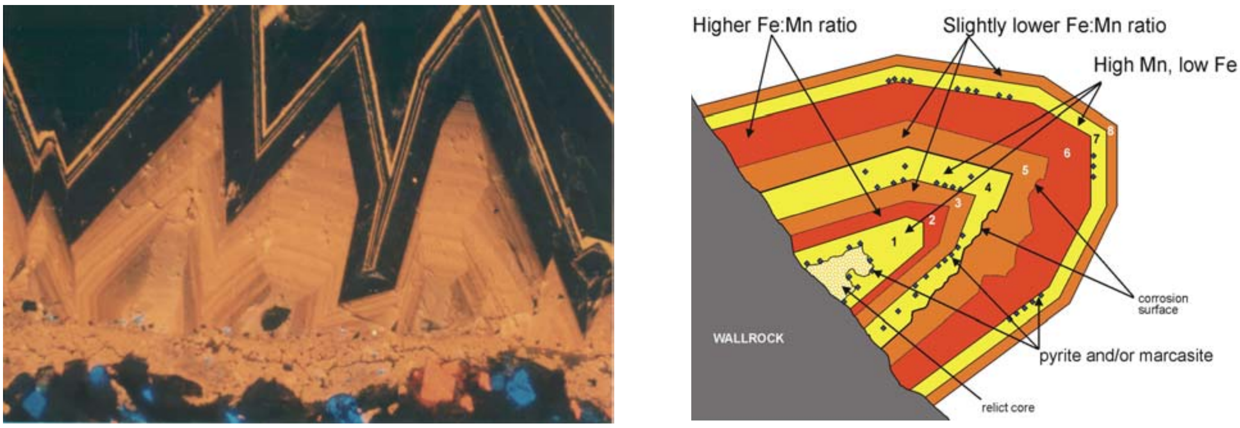

Figure 10. Cathodoluminescence banding of secondary calcite from fresh water zone at Sellafield and schematic sketch showing the compositional variation that accounts for it.

A similar dependence of CL activity on Mn concentration is found in secondary calcite from Äspö for which electron microprobe elemental mapping shows that there is less variation in Fe (Figure 11).

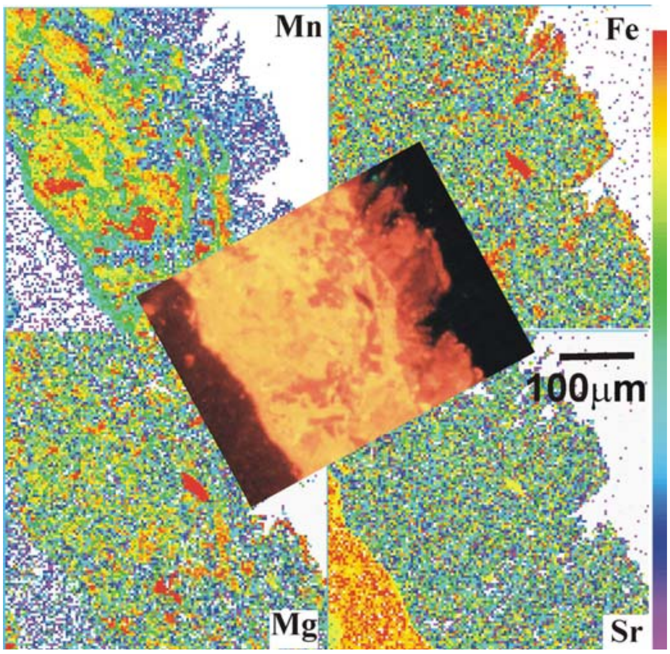

Figure 11. CL and microprobe X-ray maps show that variation in secondary calcite at Äspö (375 m depth) is dominantly in Mn; high Mn^{2+} may characterise periods of dominance of brackish Baltic Sea water.

The implications of trace element zoning in secondary calcites for past groundwater conditions is summarised as follows:

- Similar Mg-Fe-Mn variations are seen in late-stage scaly calcite at Olkiluoto: high Mn is characteristic (but not at Vienne); other trace elements e.g. rare earth elements may also indicate redox variability.
- Zoning suggests that fresh-brackish groundwaters down to 100's of metres have had some fluctuation of redox-related properties during period of calcite growth.
- Variations in Mn, Fe, etc and episodic pyrite probably indicate persistence of reducing conditions with limited range of fluctuation, possibly involving SO_4^{2-} inputs and microbial activity?

2.4.3 Fluid inclusions in secondary calcites

Fluid inclusions trapped in secondary calcite are effectively traces of fossil groundwater, and are perhaps the most direct evidence for past groundwater compositions. They are usually sparsely distributed in calcites that have precipitated from normal groundwater circulation at low temperatures, and therefore there are practical difficulties in extracting and analysing discrete inclusions. The development of laser ablation ICP mass spectrometry has opened a new method for high-sensitivity analyses of inclusions with high spatial resolution of extraction directly from the solid wafer sample of calcite. This new method was used by BGS but was not available at the other partners' laboratories. Because of the difficulties in reliably calibrating such analyses, the elemental abundances can be reported only as ratios to other ions, and not as absolute concentrations.

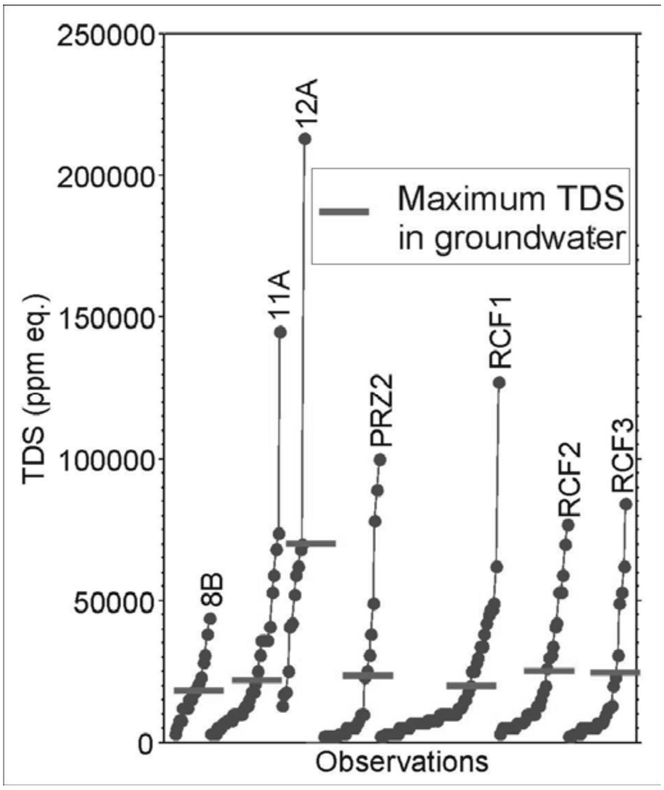

Figure 12. Salinity of fluid inclusions in secondary calcites from the deep boreholes at Sellafield, as determined by microthermometry. Comparison with the maximum salinity of present-day groundwaters in these boreholes suggests that some of the calcites contain evidence of past groundwaters that are more saline than present-day conditions.

Work in this area is summarised as follows:

- FIs were measurable in late-stage calcite in Sellafield samples but not at Olkiluoto, Äspö or Vienne.
- Potentially large uncertainties in metastable monophase FIs.
- Microthermometry analysis of salinity has detection limit at ~3000 ppm.
- Single inclusions can be analysed for element ratios by LA-ICP-MS.
- A few values for salinity in fluid inclusions from each borehole at Sellafield are higher than max salinity in present-day groundwaters (see Figure 12).
- FI data indicate that groundwater was more saline when inclusions were trapped by late-stage calcite.
- Reliability of interpretation must consider uncertainty of MT estimations of TDS.

2.4.4 Stable oxygen and carbon isotope ratios in secondary calcites

Most of the stable O and C isotope analyses were carried out on bulk samples of calcite, i.e. samples that are likely to have contained several growth zones, so that they are not necessarily representative of specific growth episodes. Only a few analyses for Äspö samples were carried out by laser ablation (LA) of single 'points' in the calcite. There is evidence that laser ablation of calcite may cause $^{18}O/^{16}O$ isotopic fractionations associated with solid deposits on the rim of the ablation pit, so data must be interpreted with caution.

The stable isotopic data are summarised as:

'multi-stage' calcites	$^{18}O/^{16}O$	$^{13}C/^{12}C$
Olkiluoto	-20 to -8 ‰	-10 to +10 ‰
Äspö (some LA data)	-20 to -5 ‰	-40 to 0 ‰
Vienne	-15 to 0 ‰	-13 to -5 ‰

The implications of these data are:

- Analyses of 'multi-stage' calcite samples show the large ranges of ratios, but laser ablation or ion probe analyses are required for specific zones.
- There is a specific problem to distinguish early hydrothermal and late stage calcites: (e.g. in Äspö data).
- Very low $^{18}O/^{16}O$ suggests that some calcite (or specific growth zones) have formed from cold climate waters (cf. min -22‰ in preliminary LA data by BGS).
- Low $^{13}C/^{12}C$ (esp. <-20‰) indicates biogenic carbon down to ?500 m at Äspö.

2.4.5 Palaeohydrogeological indicators in Quaternary sediments

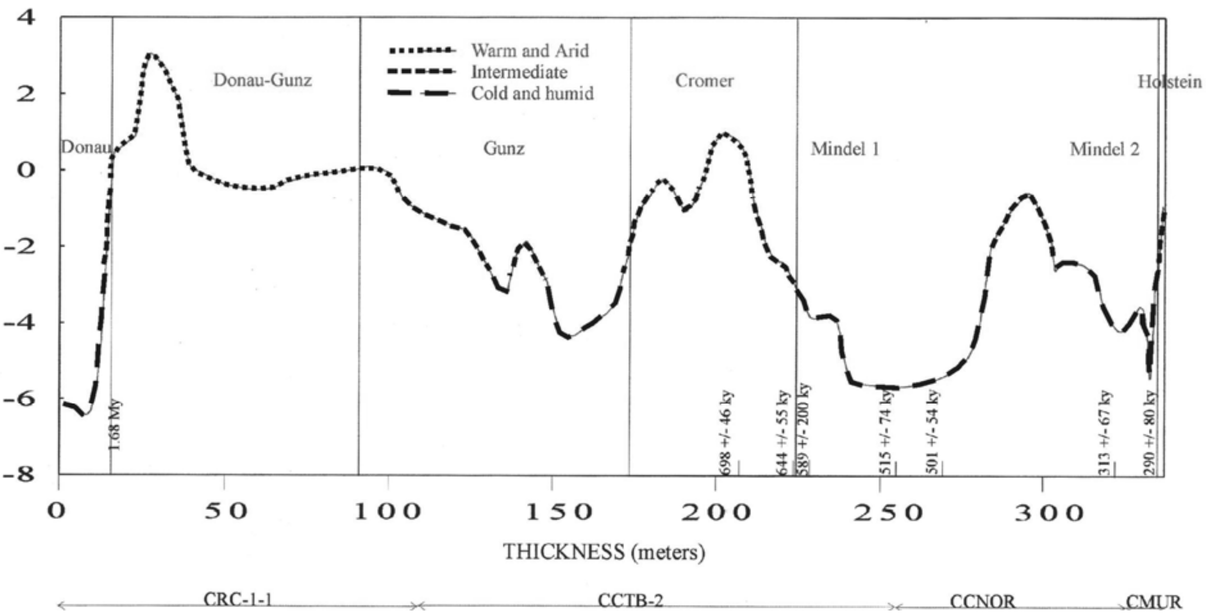

Figure 13. δ^{18}O (‰PDB) of ostracods from lower and middle Pleistocene sediments in the Cúllar-Baza basin in semi-arid Spain plotted against position in the lithostratigraphic column as a proxy for time, to provide a summary of palaeohydrology and climate evolution.

Amongst the palaeohydrological and palaeoenvironmental indicators analysed in ostracod samples from Pleistocene sediments from the Cúllar-Baza basin, Spain (see Figure 7) were $^{18}O/^{16}O$, $^{13}C/^{12}C$, Mg/Ca and Sr/Ca. The results of stable isotope analyses are shown in Figure 13. The correlation of δ^{18}O with palaeoclimate is evident, reflecting the dual effects of climate on direct evaporation from the lake and on the isotopic composition of groundwater recharge that has flowed into the lake. The palaeohydrological interpretation involves using the isotopic variations to aid in interpreting past variations of the balance between input to the lake from run-off and groundwater inflow and losses from the lake by direct evaporation.

2.5 CONCLUSIONS

The results of work in EQUIP are summarised as follows:

2.5.1 History of groundwater sources and salinities

- Groundwaters in the northern European study sites that have stable isotopic compositions indicating cold-climate water also tend to be brackish/saline. This indicates that these groundwaters are now mixtures of component waters with different sources and different ages.
- Waters that are relatively depleted in ^{18}O ('cold-climate water') occur to 100's of metres depth. This suggests that water that recharged during cold stages of the Quaternary has reached those depths by flow and dispersive mixing with saline waters.
- It is uncertain whether fresh meteoric water reached greater depths in the past than the present locations of fresh/brackish-saline transitions. In favour of this hypothesis is the presence at Äspö of secondary calcites that have lower $\delta^{18}O$ than would be given by equilibrium with present-day coexisting water. Against it is the absence of fluid inclusions that are more dilute than coexisting groundwater (but Fluid Inclusions could be characterised at Sellafield only).
- Calcite morphologies at Sellafield suggest that saline water was present at shallower depths in the past than the present-day location of the fresh/brackish-saline transition. The glacial and post-glacial palaeohydrogeological processes that might account for this are isostatic depression by glacial loading, sea level rise or fractionation by permafrost.

2.5.2 Stability of groundwater flows

- At each of the study sites, the upper part of the groundwater system (i.e. to a few 100's of metres depth) has responded to neotectonic impacts and hydrological changes (including glaciation and sea level fluctuation).
- In contrast, evidence from groundwater compositions and secondary mineralisation indicates that deeper, more saline, parts of the groundwater systems have been relatively stable, retaining older (pre-Quaternary?) groundwaters.
- The transition between these more- and less-stable parts of a groundwater system may have shifted in the past, but mineral evidence (e.g. at Sellafield) suggests that the shift has occurred over a rather narrow vertical distance.

2.5.3 History of groundwater redox conditions.

- Oxidation in the shallow parts of groundwater systems is recorded by iron oxide minerals down to ~100m depth;
- Compositional zoning of Sellafield calcites shows that redox has fluctuated in the past, and that the variability decreases with increasing depth and also decreases laterally into more saline groundwaters.
- Variations of Fe and Mn concentrations in calcite zones indicate that Fe and Mn have also varied over time in groundwaters. The range of Fe and Mn variation in Sellafield calcites indicates that redox conditions in deep groundwaters have remained reducing (e.g. max Eh range 0 to -300 mV).
- Trace element and C isotopic variations in calcites at Äspö suggest that redox conditions have been influenced by episodic recharge of organic C, SO_4, and microbes.

2.5.4 Stability of redox conditions

- Shallow systems are susceptible to variations of redox conditions and controls over time: variable equilibria involving O2, SO4, organic C and Fe/Mn-ox minerals as well as microbial activity.
- Deep groundwater systems have a limited range of redox fluctuations which are controlled by Fe-oxide, Fe-S, Fe/Mn-carbonate minerals and SO4/S, with some uncertainty over the effect of microbial activity.

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2.7 DISCUSSION

Q Is there an assumption that carbonate precipitation (in Sellafield fractures) is continuous, or could it be episodic?

A Corrosion surfaces occur in calcite crystals, so episodes of dissolution do occur. However, there is no coherent evidence for mass calcite dissolution in the Quaternary; the mineralisation record appears to be more or less continuous.

Q Are there other areas in the world where morphological changes with salinity are recorded?

A There are lots of accounts in the diagenetic literature, for example in the Bahamas. A large body of literature describes the close relationship between calcite morphology and groundwater Mg content. Fracture-hosted calcites in Japanese Mesozoic granites studied by BGS show similar morphological changes with depth. Peter Fritz (previously with AECL) has some observations of morphology changes from Canadian sites that were investigated as part of the radioactive waste disposal programme. BGS has also recorded similar features from borehole cores at Dounreay.

Q Can tectonic or climatic effects be interpreted from mineral paragenesis studies?

A EQUIP didn't tackle such questions. Proving a mineralogical relationship with climate changes or with subtle tectonic processes such as uplift/erosion requires good age constraints. There is the potential that at some future date radiometric dating of e.g. individual calcite zones might be possible. However, suitable samples would be required (e.g. high in U) and the uncertainties in dating may well continue to be larger than the timescales over which significant global climate change operates. Consequently a two-stage approach is required to infer any climatic impacts on mineral paragenesis: (i) understand shallow patterns, because they are closer to surface processes and more influenced by them; (ii) try to understand the deeper patterns and investigate whether any correlations with shallow patterns are possible (morphological, elemental or isotopic). This approach is being investigated in PADAMOT.

Q From the experience of EQUIP, what is the most important aspect of palaeohydrogeology to consider?

A Individual methods/techniques should not be investigated in isolation; data from different sources need to be considered together, in a holistic manner.

Q If the compositional zonations (in Sellafield calcite) record only subtle changes in redox, what (if any) are the implications for interpretation and Safety Assessment?

A The degree (of change in groundwater redox) inferred from EQUIP is probably of no great significance; the redox fluctuations recorded in calcite zoning patterns are probably very small. However, the approach used in EQUIP is very interpretative, and significant uncertainty remains.

3 PHYMOL: Palaeohydrogeological Study of the Mol Site, Belgium

Isabelle Wemaere and Jan Marivoet , SCK-CEN, Mol, Belgium

3.1 INTRODUCTION

The PHYMOL project (Marivoet et al., 2000), a palaeohydrogeological study of the Mol site, provides an analysis of a methodology, taking climatic changes into account, applicable in the performance assessment of an argillaceous repository system.

The Boom Clay, considered as the potential host formation, has a very low permeability. It is situated at about 190 metres deep with a thickness of about 100 metres at the Mol site (See Figure 14). This formation is of Oligocene age, situated in the middle of a thick Tertiary sedimentary series related to the North Sea Basin. Above the Boom Clay, the thick sandy Neogene aquifer has a particularly high transmissivity and can be subdivided in to three main sub-aquifers. Below the Boom Clay, the Lower-Rupelian and the Lede-Brussel aquifers, represented by sands and clay layers, are less permeable and are confined in North-East Belgium.

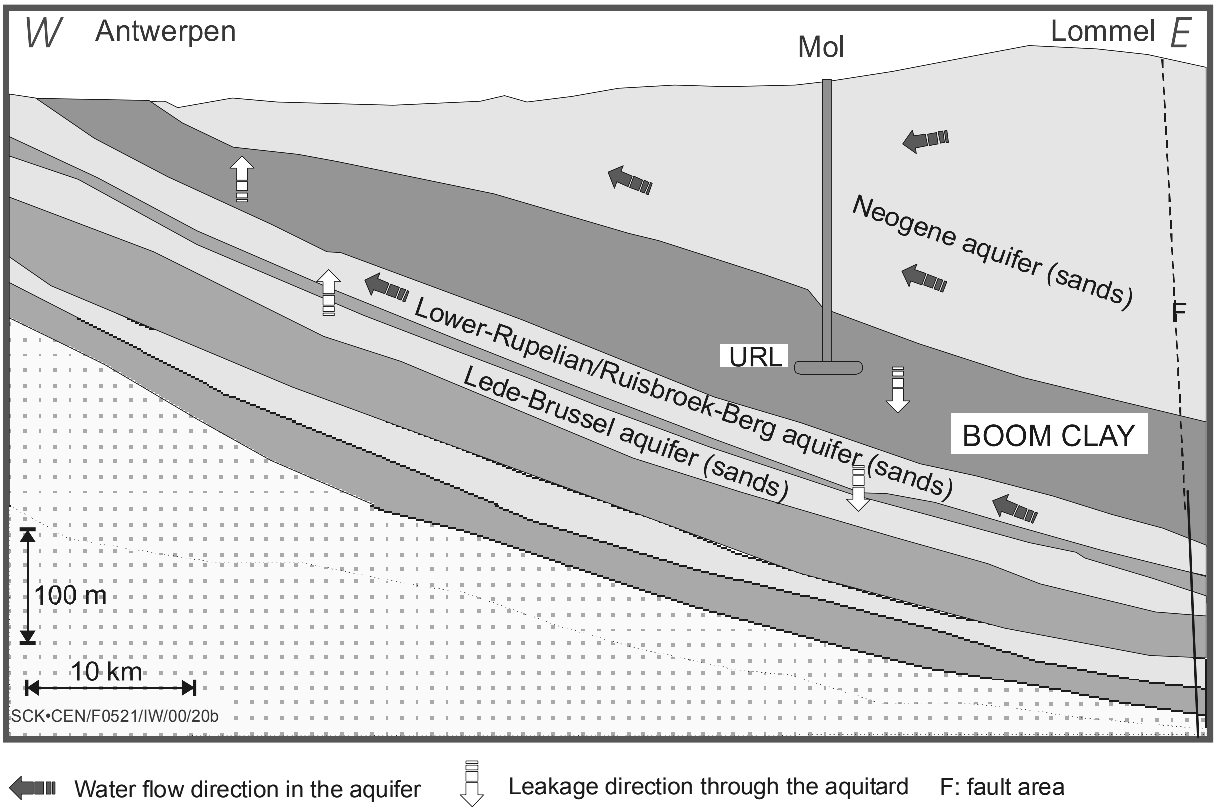

Figure 14. Hydrogeological cross-section at the Mol site

3.2 HYDROGEOCHEMISTRY

A hydrogeochemical campaign was performed on several wells of the SCK regional piezometric network presented in Figure 15. The objective was to examine whether a palaeoclimatic signal could be found in the Lower-Rupelian and Lede-Brussel aquifers and if one could date it. The analyses of major and trace elements have been used to indicate whether the fluids are in equilibrium or not with the host rock. They helped in the interpretation of the isotope and noble gas results. They confirmed the mixing of the deep groundwaters with a marine component, essentially in the north-west of the studied area (Pitsch and Beaucaire, 2000). The geochemical composition of the sampled waters helped researchers to select reliable measurement points (19 of the 27 initial points). This showed the importance of having a well-designed sampling strategy.

The ^{2}H and ^{18}O content and dissolved noble gas contents (He, Ar, Kr and Ne) were measured. A probable palaeo-signal can be found in the deep aquifers based on ^{18}O and ^{2}H distributions indicating that the groundwaters infiltrated during colder climates in the central part compared to in the northern or southern part of the studied area. To date the groundwaters and this signal ^{14}C was measured on total dissolved inorganic carbon (TDIC) and on dissolved fulvic acids (FA). Ages based on FA are systematically younger than TDIC ages. They range from 0 to more than 30 ka from south to north. The deeper Lede-Brussel groundwaters appear to be "younger" than the Lower-Rupelian groundwaters (Philippot et al., 2000).

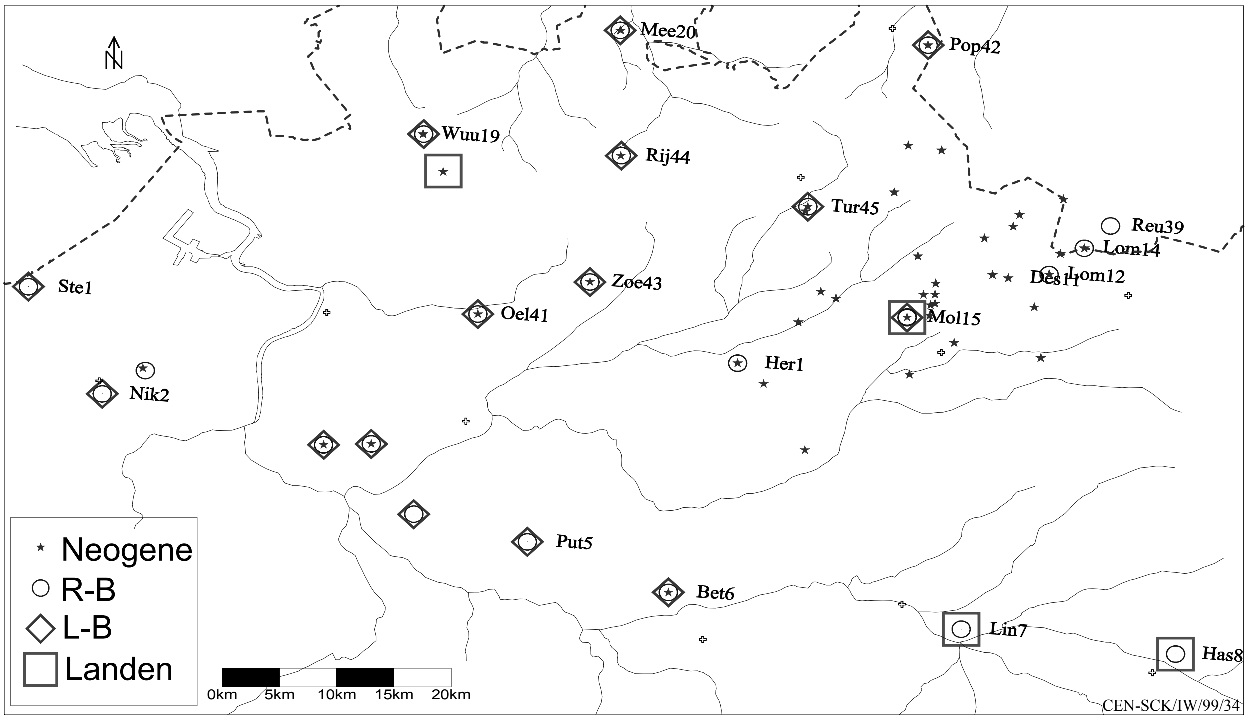

Figure 15. Location of sampling for hydrogeochemistry

3.3 PALAEOHYDROGEOLOGICAL MODELLING

The past climatic evolution, from Eemian times (-126 ka) to the present, was translated in a sequence of climate periods with climatic factors and palaeogeographical conditions for NW Europe and for the specific site of Mol as illustrated in Table 2.

A supra-regional model of NW Europe (part of North Sea Basin) was used to evaluate the large-scale climatic effect on the groundwater flow. The results showed that glacial inflow into the regional area of Mol is negligible because of the presence of large rivers between the ice cap and the Mol area (van Weert and Hassanizadeh, 2000). A more precise simulation of flow was obtained by introducing the palaeo-conditions directly in the regional model used for the Mol site (Wemaere et al., 2000). Both model domains are presented in Figure 16.

Table 2. Sequence of climatic scenarios from Eemian until present adapted for the regional modelling of the Mol site.

Epoch	Time (kyears BP)	Mean annual temperature (°C)	Climate state	Relative sea Level (m)	Infiltration (mm year^{-1})	Permafrost thickness (m) disc/cont
Eemian	116 – 125	11	t	0	220	0
Early Weichselian	116 – 105 105 – 94 94 – 82 82 – 75	-3 0 -2.5 1	dp dp - b dp b	-75 -35 -75 -35	2.6 97 3 155	10 0 15 0
Middle Weichselian	75 – 60 60 – 25 25 – 13	-12.5 -4.5 -12.5	cp dp - b cp	-75 -75 -100	0.14/0 0.36 0	20/50 20 60
Late Weichselian	13 – 10	-2	dp	-75	0.14	20
Holocene	10 – 8 8 – 0	5.5 9.5	b b - t	-30 0	140 220	0 0

t: temperate; **cp**: tundra, cont.permafrost conditions; **dp**: tundra, disc.permafrost conditions; **b**: boreal
disc/cont: discontinu/continu; **BP**: before present

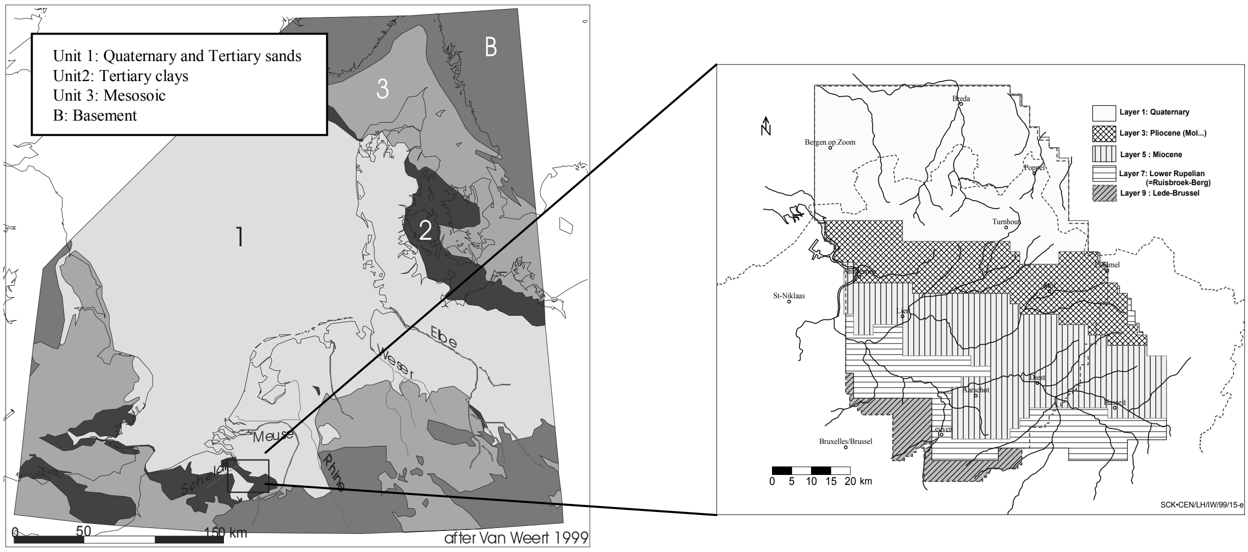

Figure 16. Supra-regional and regional models used for the palaeo-reconstruction.

Flow simulations were performed for each of the climate periods using the finite difference MODFLOW code (US Geological Survey). The permafrost effect was included in the hydrodynamic parameters through a reduction of the upper layer thickness. Rivers were modelled as very conductive drains with elevations adapted for the fall in sea level. The meteoric infiltration rate was adapted based on the climate information and permafrost occurrence. The bottom and side boundary conditions were taken as no-flow for all climate scenarios except for the north-west boundary. In this direction, the aquifers are deepening below the North Sea and a general head condition was used to take account for equilibrium with the sea level and its changes during the glacial cycle. Flow simulations indicate that very low velocities and water pressures are expected in the Mol area during cold periods with permafrost occurrence. An example is given in Figure 17. Critical influential parameters are the infiltration rate and the river elevation.

Transport simulations of ^{14}C and ^{18}O isotopes were realised for the continuous period from Eemian (126 ka BP) to present-day to end in a distribution that could be compared with the observed ^{14}C and ^{18}O content in the deep groundwaters. The simulations were performed with an implicit finite difference algorithm available in the MT3DMS code (Univ. of Alabama).

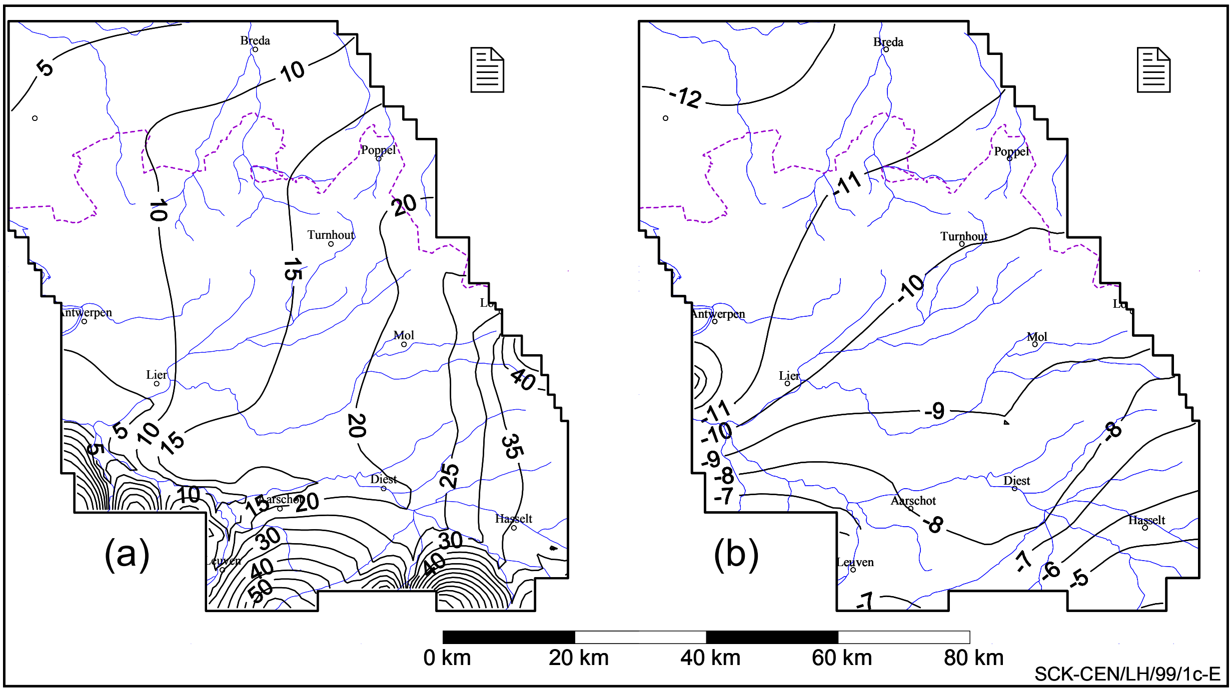

Figure 17. Isolines of calculated potentiometric head in the Lower-Rupelian aquifer (a) present-day condition, (b) Lower Weichselian (116 to 105 ka BP)

The ^{14}C concentration of meteoric water was assumed to have been constant and equal to 100 pmC for 125,000 years. The initial concentration is arbitrarily set to zero. The simulations illustrate that the present day distribution of ^{14}C is poorly influenced by past climates. It is similar to the steady state distribution obtained with the present flow condition. It satisfactorily fits measurements for the Lower-Rupelian aquifer and suggests an additional input of recent water into the Lede-Brussel aquifer at the north-eastern border, which might explain the observed high ^{14}C concentration (see Figure 18). We expect more accurate results by improving the representation of the outcrop zone.

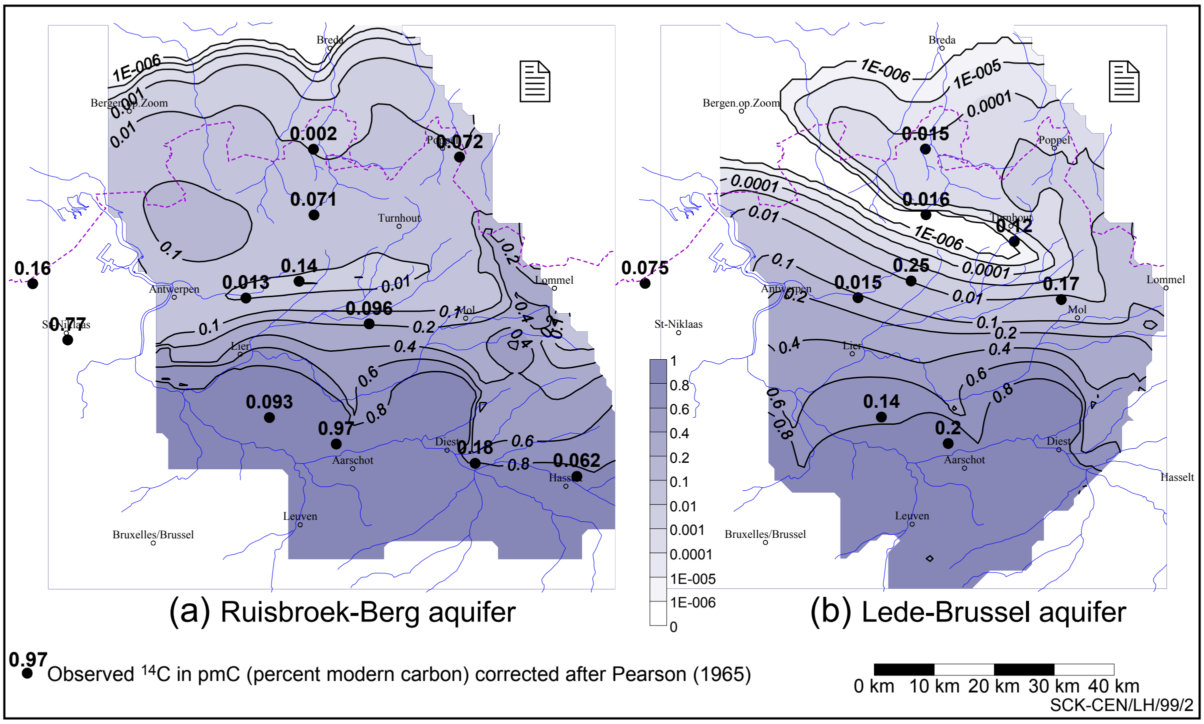

Figure 18. Calculated (isolines) and observed (points) ^{14}C concentrations in the aquifers.

For the ^{18}O simulations a specific concentration of meteoric water was used for each climate scenario since it depends on temperature. The simulations show that the calculated present day distribution of ^{18}O concentration is still influenced by the imposed initial condition. There is a clear spatial shape of the ^{18}O simulated distribution in the deeper aquifers with higher values in the north (older water) and the south (recent water) and lower values in the centre due to the glacial phase. This can be related to the temporal variation of ^{18}O input concentration with a dominance of the wet and moderate climate periods. This shape can also be clearly recognised in ^{18}O measurements of the Lede-Brussel groundwaters (see Figure 19) while those of the Lower-Rupelian aquifer do not allow one to draw clear conclusions. Comparison with observations is more qualitative than quantitative.

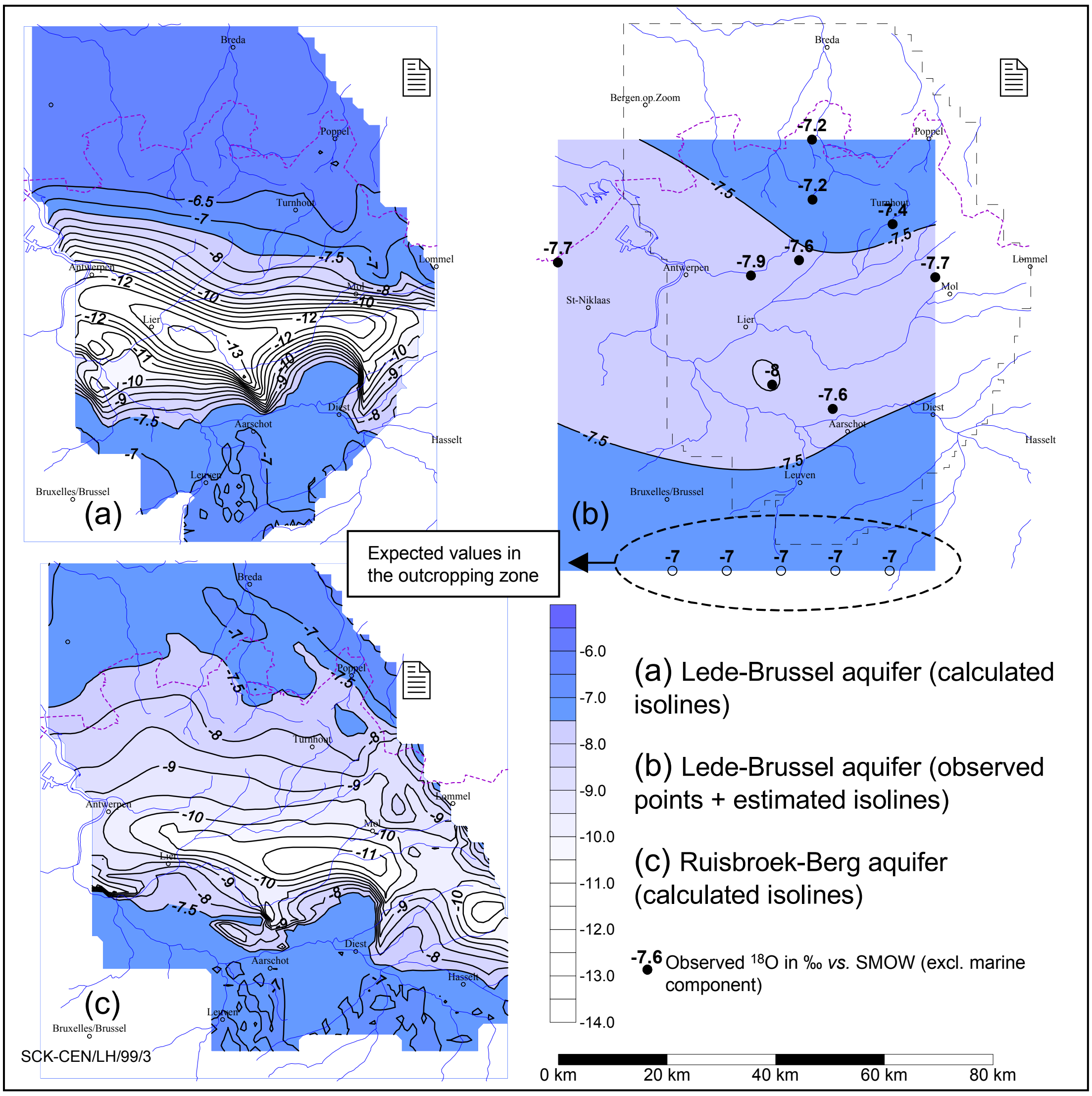

Figure 19. Calculated and observed $\delta^{18}O$ values in the aquifers.

3.4 APPLICATION FOR PERFORMANCE ASSESSMENT

A simulation approach was proposed to evaluate the radiological consequences of radionuclides released from a radioactive waste repository situated in a clay layer for the next 125 ka. The expected climate evolution (Berger et al., 1991) is translated in a sequence of climate periods similar to past climate periods and a continuous transport simulation of radionuclides is performed for this period of 125 ka. The calculated evolution of the radionuclide flux released from the host clay layer is used as input into the aquifer system. The climate effect on this radionuclide flux is strongly limited since the transport in the clay layer is essentially diffusive. The concentrations in the aquifer calculated with and without considering climate changes are given in Figure 20. Drastically higher concentrations are obtained when the climate changes are considered.

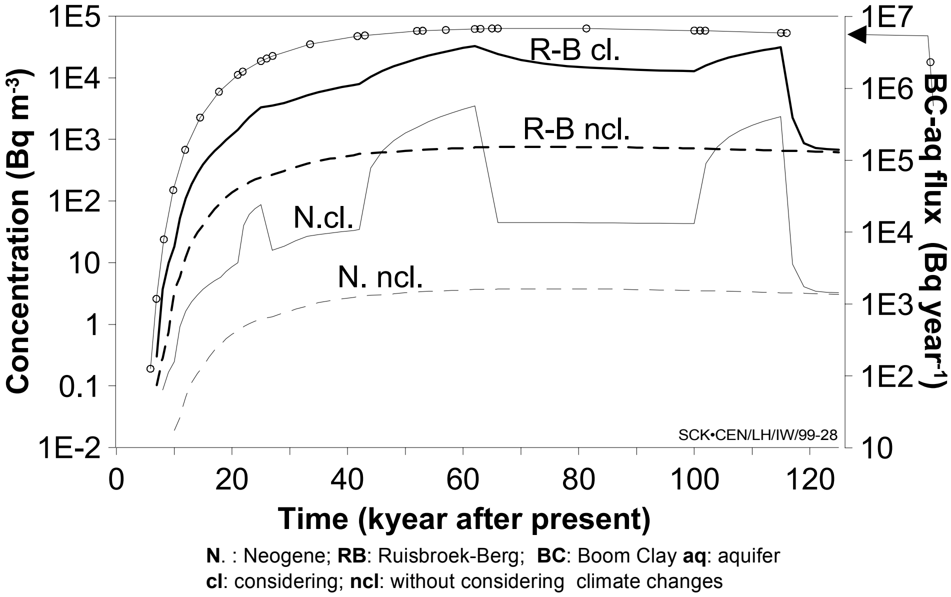

Figure 20. Evolution of the radionuclide concentration and in the aquifers.

3.5 CONCLUSION

Measurements of environmental isotopes provided indications of the water flow during the last glacial cycle. Although enormous uncertainties occur in the data used, a partial reconstruction of the observed distributions was possible by making transport simulations that consider a sequence of climate states giving confidence in the applied methodology.

A similar approach was therefore applied in an assessment of the doses resulting from the disposal of radioactive waste. It appears that in the present moderate climate conditions a strong dilution of the radionuclides occurs in the aquifers, but this will decrease considerably during colder climates. However, the huge uncertainties occurring in these simulations mean that the calculated doses cannot be considered as forecasts of the future dose, but only as an estimated outcome of one of many possible evolution scenarios.

3.6 ACKNOWLEDGEMENTS

The authors like to thank the EC and the partners of this project: Technische Universiteit Delft (the Netherlands) for evaluating palaeoclimatic effects on the North Sea Basin scale, Commissariat à l'Energie Atomique and Université de Paris-Sud (France) for sampling and analysing the aquifer waters and providing the geochemical information.

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3.8 DISCUSSION

Q Was the permafrost thickness (mentioned in the presentation) based on geological evaluation of field evidence or on modelling interpretations?

A It was based on literature data. If we did the study again, we would make the calculation site-specific.

o In the past, permafrost thicknesses in the UK were based on direct geological evidence. More recent numerical modelling suggests that permafrost thickness based solely on geological evidence may be underestimated.

Q Why was a 125 ka span chosen?

A The span coincides with the Weichselian period.

Q Have results been integrated with PA studies?

A Not yet; they may not be.

Q What method(s) were used to measure ^{14}C in inorganic and organic C? Are the presented data based on inorganic or organic C?

A The data presented are for organic C only, spanning 3-30 ka. Information concerning the methodology is provided elsewhere.

Q Is there any difference in the data for inorganic and organic C?

A Ages are lower for organic C.

o One of the problems of not having a good permafrost model is that heads can be miscalculated. The presence of permafrost causes overpressures, while permafrost melting can cause underpressures. New scoping models/calculations would be desirable.

o All observations suggest there is a linear hydraulic gradient top to bottom (at the Mol site) at the present day; i.e. no stored overpressure from glaciations.

Q What length of time is required to reduce overpressures generated beneath permafrost?

A It depends on the materials. It could be quite quick, in the order of several thousands of years.

o Hysteresis, in the form of repeated glaciations, is a problem.

Q Did permafrost reach the Boom Clay at the Mol site?

A No, assuming our calculations are correct.

4 The Palmottu Natural Analogue, Finland: Its Use in Development of Glacial Scenarios for PA

John Smellie, Runar Blomqvist, Shaun Frape, Juha Kaija and Timo Ruskeeniemi (Conterra, Sweden; Geological Survey of Finland; University of Waterloo, Canada)

4.1 INTRODUCTION

The uranium deposit at Palmottu in southwest Finland has been investigated in order to improve the scientific basis for assessing the long-term performance of a disposal site for high-level nuclear waste in fractured crystalline bedrock (Blomqvist et al., 1995, 1998, 1999). The study entailed assessing the present-day flow situation, and identifying and quantifying the processes related to radionuclide mobilisation and migration. This area was affected by glaciation and post-glacial uplift and isostatic effects with respect to sea level. It is therefore valuable for developing palaeohydrogeological scenarios for glaciated terrain.

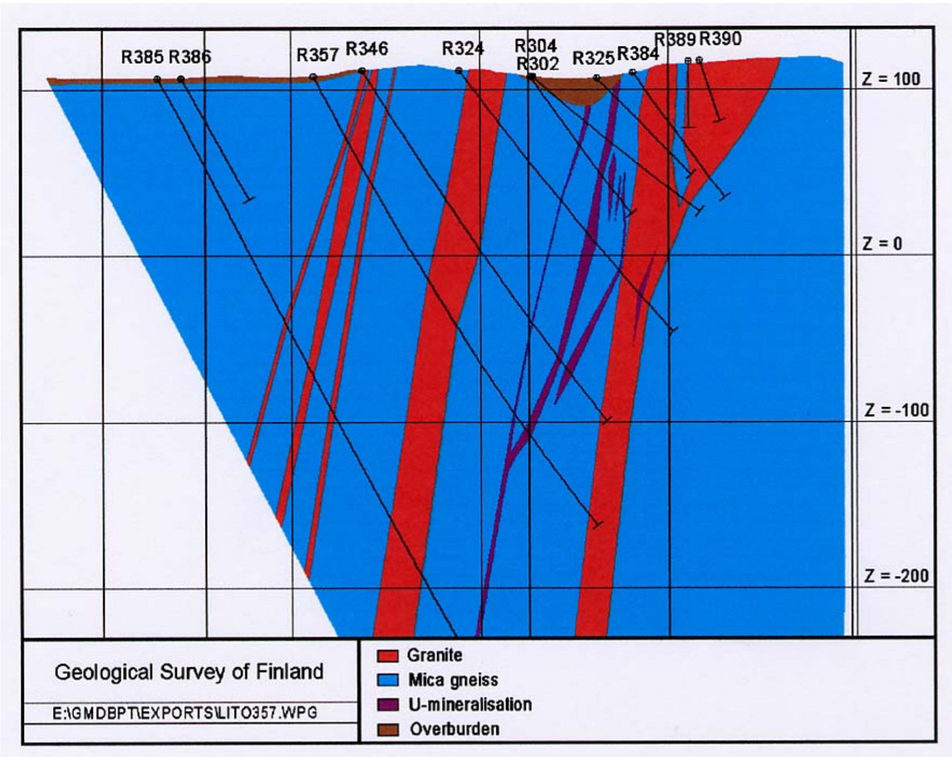

Figure 21. Vertical section through Palmottu, with the uranium mineralisation located between the two granitic bodies. Some of the boreholes are shown, intersecting the uranium ore body.

The Palmottu U-Th mineralisation is hosted by a Proterozoic (1.8 Ga) crystalline bedrock, mainly mica gneiss with granite and granite pegmatite veins. The mineralised part forms a vertical structure that extends to depths of at least 400m (Figure 21). Accordingly, it covers both near-surface oxidative conditions, and low-Eh reduced conditions. Ancient hydrothermal alteration of the uraninite deposit led to substantial re-mobilisation of uranium resulting in the formation of U(IV) silicate (coffinite). Over the last one million years, the site has experienced multiple glaciations during which a proportion of the uranium has been mobilised under oxidising conditions, leading to precipitation of U(VI) silicates (uranophane) at shallow depths (Ruskeeniemi et al., 1999). Volumetrically, significant amounts of uranium, mainly as U(IV), are also found in association with fracture coatings such as impure calcites, clays and iron oxy-hydroxides.

At the end of the last glaciation around 10,000 years BP, Palmottu experienced a continental ice margin palaeoclimate and was subject to permafrost for a long period. However this area was not inundated by seawater and is therefore distinct from the Äspö and Olkiluoto sites in this respect.

A site-specific groundwater flow model was constructed based on an integration of structural, hydrogeological and hydrogeochemical data (Pitkänen et al., 1999). Data are available from a large number of existing prospecting boreholes, in addition to several new boreholes that were purpose-drilled. The deepest boreholes reach depths of 350-400 m (Figure 21).

Two main approaches have been used to obtain evidence of past perturbations of the groundwater system at Palmottu: hydrochemistry and mineralogy.

4.2 HYDROCHEMISTRY AND ISOTOPE GEOCHEMISTRY

Palmottu is characterised by an upper, dynamic groundwater flow system to at least 100 m depth, with typical bicarbonate groundwaters which can be partly explained by mixing processes, but nevertheless show a clear evolutionary trend from Ca-HCO_3 type to deeper Na-HCO_3 type. In the southern part of the site, ^{3}H-containing Na-HCO_3 groundwater penetrates to 200 m depth. At greater depths, where lower permeability conditions prevail, brackish Na-Cl and Na-SO_4 groundwaters are present (Figure 22). A sizeable δ^{18}O-depleted glacial water component is present in the deep Na-Cl and Na-SO_4 groundwaters (and also in some of the deeper Na-HCO_3 water) (Figure 23).

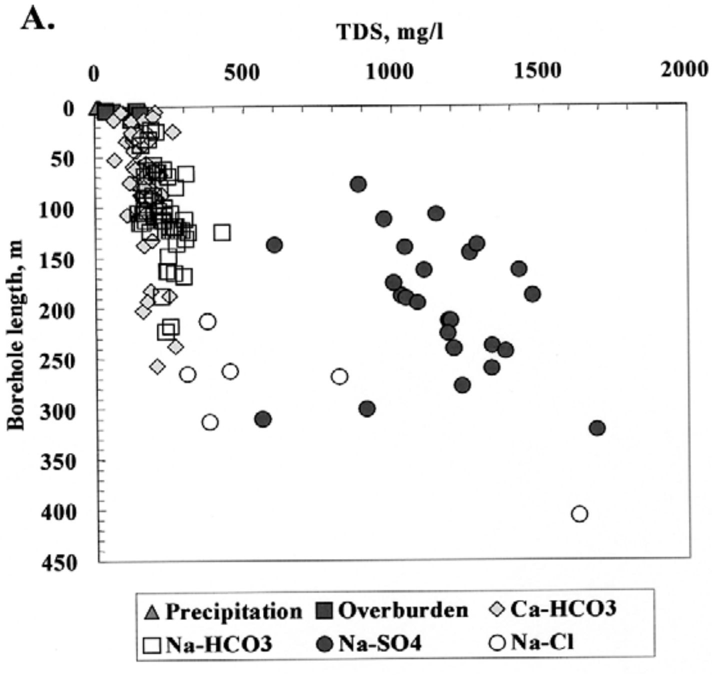

Figure 22. Total Dissolved Solids (TDS) in groundwaters versus borehole length (note: the boreholes are not vertical).

This glacial water component is up to 10,000 years old, and the age suggests that these brackish groundwaters are more or less static.

δ^{18}O values less than -12‰ signify colder climate recharge conditions compared to present ones. The extreme δ^{18}O value of -19‰ is close to the expected isotopic value of glacial melt water, and values lighter than -15‰ indicate a significant glacial water component.

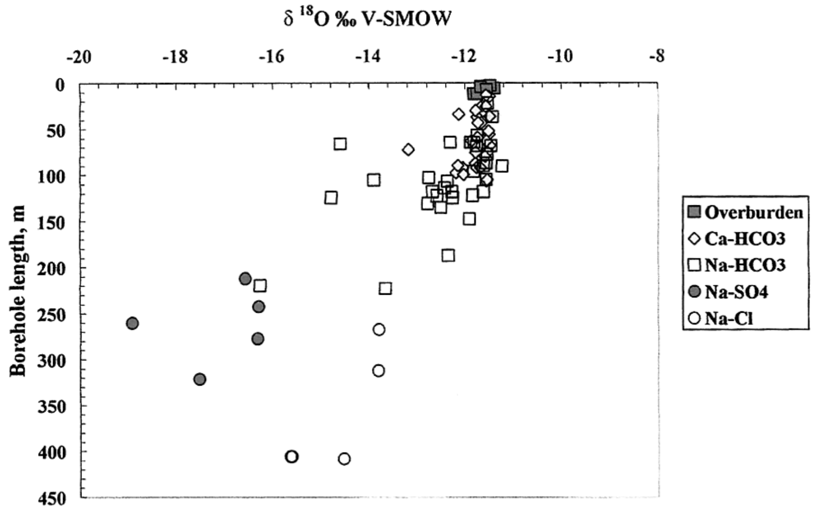

Figure 23. $^{18}O/^{16}O$ ratios in groundwaters versus borehole length.

The presence of glacial water suggests that the system has been open to the incursion of meteoric waters to at least 400 m. It is unlikely, but still uncertain, that the glacial melt waters would have retained their oxidation capacity to any great depth because of the buffering capacity of first the overburden and then the bedrock itself; this is supported by mineralogical evidence. High uranium concentrations (generally from 100-500 ppb) are associated with oxidising HCO_3 groundwaters in the vicinity of the uranium mineralisation down to depths of 130 m. At greater depths, reducing conditions (Eh -300 mV) prevail, with low uranium concentrations (below 10 ppb).

The Na-SO_4 groundwaters at Palmottu are associated with uranium mineralisation and may be explained by sulphide dissolution and oxidation during hydrothermal events. Continuous, long-term oxidation of sulphide minerals during the long-term geological history can also have contributed. During recent glacial melting and unloading, Na-SO_4 groundwaters may have been formed from the breakdown of solid mirabilite ($Na_2SO_4.10H_2O$) formed under permafrost conditions and mixed with glacial melt water as observed today. In addition, the generation of a more saline front under permafrost could have further enriched sulphate concentrations. It is suggested that these cyclic processes have increased gradually the amount of sulphate to presently observed levels. It is possible that a combination of these mechanisms have been involved in creating the observed sulphate concentrations.

Irrespective of the origin of these groundwaters, however, the fact that Na-SO_4 groundwaters, and to a lesser extent Na-Cl groundwaters, are still present at these depths, underlines the stability of the hydrochemical system over long periods of geological time under low permeable conditions, at least since the last glaciation approximately 10,000 years ago.

4.3 MINERALOGY

The major low-temperature fracture minerals at Palmottu that may indicate past changes in groundwater chemistry are calcite and iron oxyhydroxides. Uranophane has a restricted occurrence close to the bedrock surface.

Calcites have been observed at all depths studied; most are hydrothermal or magmatic in origin (>100° C) dating back to at least 1.0 Ga. Younger calcites are also present, but they tend to be fine-grained, characterised by multiprocess compositions, and are generally devoid of fluid inclusions (Ruskeeniemi, 1998). Uranium-decay series model ages from younger calcites selected from the upper 100 m of the bedrock suggest that uranium mobilisation/deposition may have occurred periodically during the last 30-350 ka reflecting palaeoclimatic changes.

Most iron oxyhydroxides occur as poorly crystalline goethite and are restricted to the upper 60-70 m of the bedrock. They appear to be the latest fracture filling phase, forming as coatings on older minerals such as calcite, or directly on the fracture surfaces. Uranium-decay series measurements on these phases suggest uranium mobilisation/deposition over the last ca. 1 Ma.

Small amounts of pure uranophane (U^{VI}-Ca-silicate) occur in sealed, sub-horizontal dilation-type fractures, in the upper 30 m of the eastern granite. Textural fabric patterns suggest slow growth in an open void system that may have resulted from stress-release during glacial unloading. Subsequently, following unloading and destressing, the fractures have become sealed, preserving the uranophane from groundwater contact. Uranium-decay series measurements of two uranophane samples gave geologically young ages of 90-120 and 189-240 ka respectively, which suggests that a ‘uranophane zone’ is still forming at Palmottu. The range of dates corresponds with those of calcite and appears to represent warmer interglacial periods of the past.

4.4 CONCLUSIONS

The presence of glacial water in the deep Na-Cl and $Na\text{-}SO_4$ groundwaters suggests that glacial melt water can be forced down to depths in the order of 350 m, i.e. close to repository depths, possibly under high hydraulic pressures that are thought to have prevailed at the ice margins during glacial melting and retreat (Figure 24). However, it is highly unlikely that these glacial melt waters would have retained their oxidising capacity to any great depth because of the buffering capacity of first the overburden and then the bedrock itself. This conclusion is supported by mineralogical evidence.

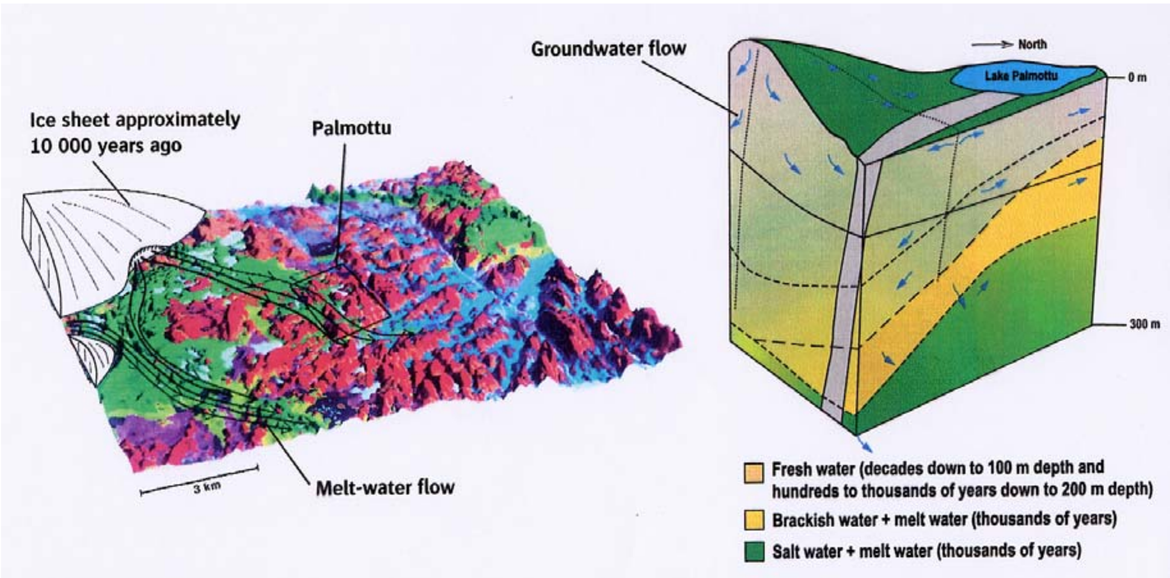

Figure 24. Palaeohydrogeological conceptual model for the influx of melt water at ca. 10,000 years ago, and a model of the present-day groundwater system showing the stratification of fresh, brackish and saline waters with increasing proportions of glacial water.

Labile uranium has been mobilised from the rock matrix around fractures, and this process took place not later than 120,000 years ago, i.e. before the last glaciation. It is likely that this was linked to the incursion of oxidising glacial melt waters, though the distribution of uranophane indicates that oxidising water did not penetrate very deep.

In contrast to other sites at which hydrochemical studies suggest that they have been most influenced by the most recent glaciation, studies of fracture minerals at Palmottu, namely calcite and uranophane, indicate that systematic events related to previous interglacial periods may have affected the site. However, their effect on the long-term stability of the hydrochemical system still needs further study.

4.5 ACKNOWLEDGEMENTS

The financial support of this study by the European Commission (Contract F14W-CT95-0010) is gratefully acknowledged.

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4.7 DISCUSSION

Q Why is there not a hypothesis that the NaSO4 groundwater formed by oxidation of iron sulphide minerals in glacial waters?

A We don’t believe the glacial groundwaters were oxidizing.

Q Groundwaters at Palmottu seem to be less mixed than those at Äspö and Olkiluoto; are they made more stable by the structure of the site?

A The dominant structures are sub-vertical, but there are sub-horizontal discontinuities also, leading to possible compartmentalisation.

Q What is the evidence that oxidizing waters didn’t penetrate to 400 m?

A Oxide/oxyhydroxide minerals are limited to the top 40-60 m at the site. U-series work suggests oxidation extended to a maximum depth of around 200 m. The modelling exercise indicated oxidation at 400 m is not expected.

Q What is the evidence that the glacial groundwaters at depth are from the latest glacial period?

A We don’t know if those waters are the product of several glacial ‘pulses’, or of just the most recent glaciation. We can’t say if the glacial groundwater is from the most recent glaciation or not; there is no evidence.

5 PAGEPA AND BENCHPAR: Groundwater Systems Beneath Glaciers and Ice Sheets and Their Geochemical Impacts

Geoffrey Boulton, University of Edinburgh, UK

5.1 SUMMARY

PAGEPA and BENCHPAR are concerned with the reliability of forecasting the long-term future evolution of geosphere systems through studies of how those systems have changed in the past. The projects have tested whether models can produce reliable predictions of complex systems and therefore whether they are suitable for use in safety assessments. The method adopted has been to identify those episodes in the past, such as glaciation, that had strong impacts on geosphere systems.

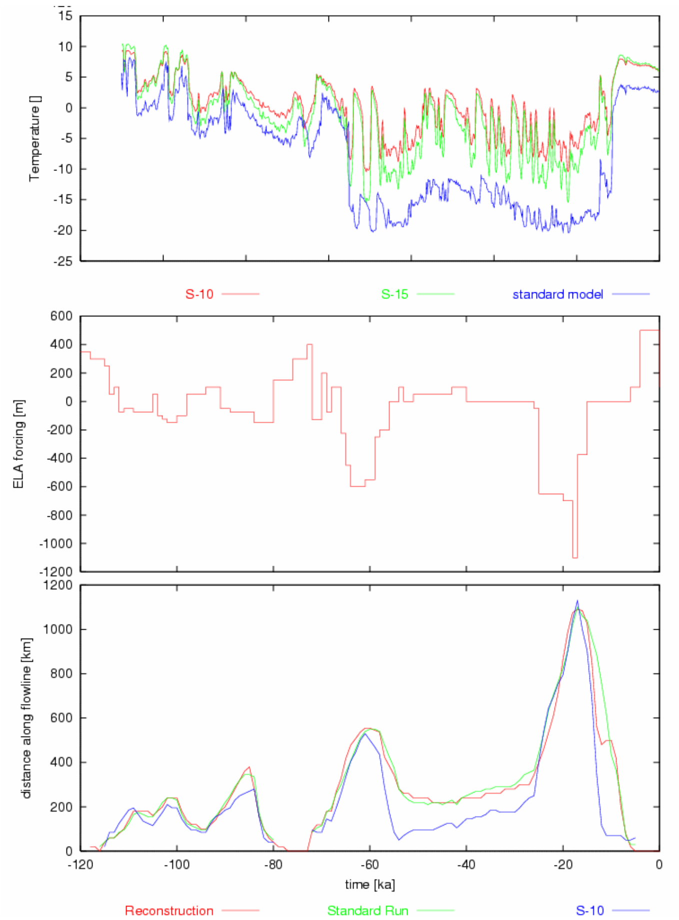

Figure 25. Climate functions used to force ice sheet variation in Europe during the last glacial cycle and the resultant pattern of variation. (a) Three models of temperature change through the last glacial cycle for southern Scandinavia based on the GRIP ice core record calibrated by palaeoclimate records from Europe. (b) The variation of equilibrium line altitude through the last glacial cycle used in conjunction with the temperature model to produce the glacier variation shown in (c). (c) The geologically-inferred pattern of ice sheet margin fluctuation through the last glacial cycle (reconstruction), and the match with the pattern of margin fluctuation created in the model by the forcings shown in (a) and (b).

Climate change is simulated using forcing models calibrated using climate records, and although there are substantial simplifications these models are relatively reliable (Figure 25). For example, the climate model is a statistical model based on the continuous, high-resolution deep core record of climate change from the Greenland ice sheet. Models for the ice sheet-geosphere system are more complex models that represent mathematically our understanding of the physics of the ice sheet system. Extrapolating the former into the future assumes that the future will be like the past. If physics-based models can be effectively tested, they can be used more rigorously in future forecasts, but ultimately depend upon confidence in climate forecasts or scenarios. The present aim is to produce models that predict impacts that can be tested from known geological history.

Reconstructions were made of the evolution of climate and of surface palaeo-environments in Europe during the last glacial cycle. These were used as boundary conditions for modelling the evolution of groundwater through the glacial cycle at the Äspö and Gorleben sites. Three environments had particularly powerful effects on groundwater evolution: organisation of groundwater systems beneath ice sheets; the impact of sub-marginal and proglacial permafrost on proglacial groundwater flow; the impact of dense saline waters in glacio-isostatically high postglacial sea levels on groundwater evolution.

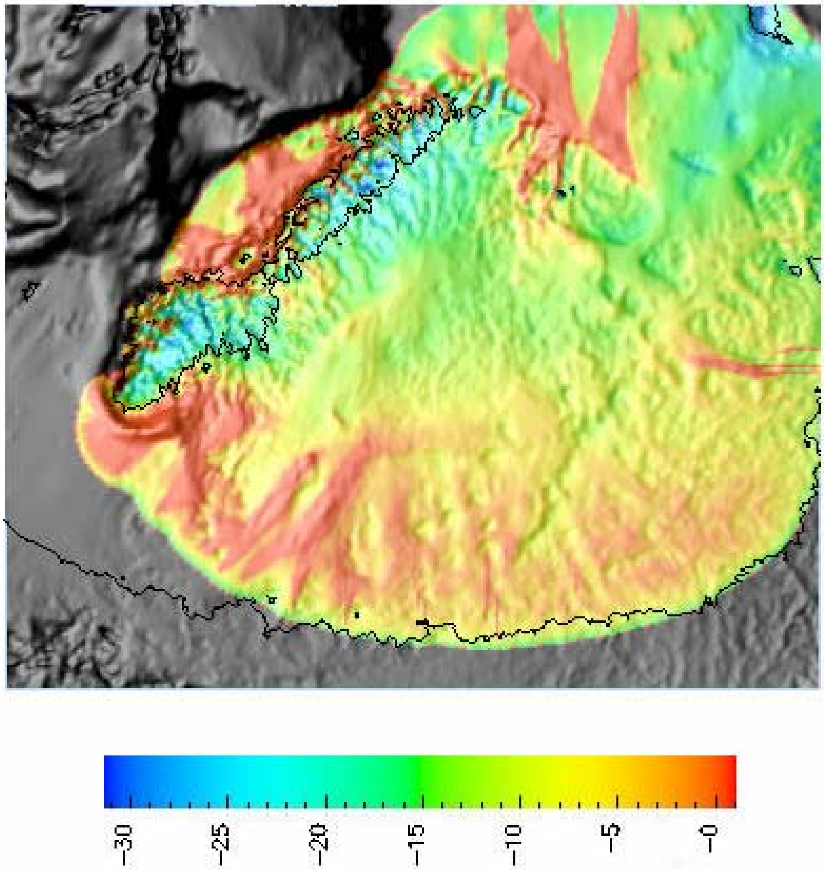

Figure 26. The simulated areal pattern of basal temperature distribution beneath the European ice sheet near to the last glacial maximum extent. Basal freezing conditions occur in the central area of the ice sheet and near to the extreme margin where the ice sheet overrides pre-existing permafrost. The red and yellow areas are areas of basal melting.

The advance of the northern European ice sheet to its maximum extent over Scandinavia has been simulated, driven by a model of climate forcing derived from the Greenland (GRIP) ice core, calibrated by palaeotemperature data from Europe (Figure 26). An Earth model is coupled to the ice sheet model to simulate crustal flexure, and zones of basal melting are computed using a thermo-mechanical model that couples temperature-dependent ice flow, frictional heat generation and the geothermal flux. The model predicts the pattern of relative sea level change through the glacial cycle, which can be tested by comparison with geologically inferred patterns of relative sea level change. The modelled ice sheet extends from central Scandinavia to Germany. A zone of basal melting extends for 100's of kilometres back from the ice sheet margin, generating very large aggregate fluxes of basal meltwater that must escape from beneath the ice sheet (Figure 27). Basal freezing occurs beneath the ice divide zone of the ice sheet, and in the extreme terminal zone during advance when the ice sheet is advancing over proglacial permafrost. There is no such terminal permafrost during final retreat of the ice sheet.

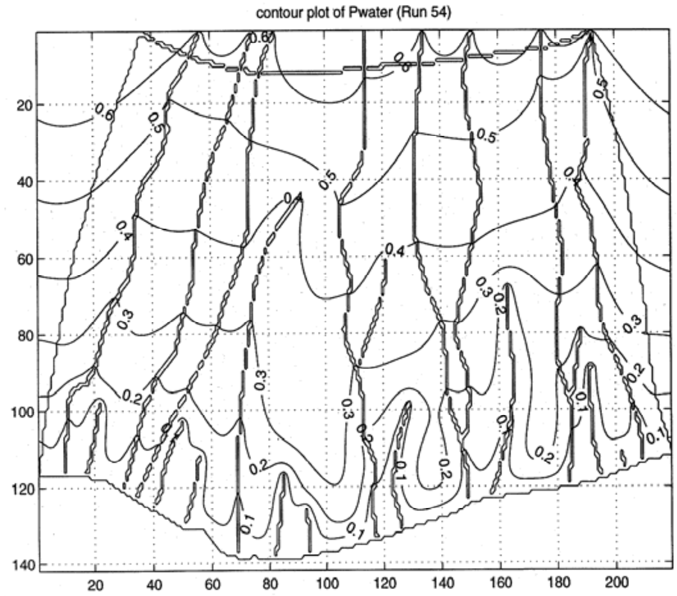

Figure 27. Simulation of groundwater heads in an area of Finland lying north of the Salpausselka moraines (near the lower margin of the diagram). The double lines show the locations of eskers, interpreted as the locations of former subglacial tunnels. The equipotential surfaces are normalised to the ice pressure. Scales are in kilometres.

The thermo-mechanically coupled ice sheet/lithosphere/permafrost model has been used to simulate evolution of the boundary conditions for groundwater flow, and 2- and 3-D groundwater codes have been used to simulate the groundwater response. In general, it indicates that basal meltwater will be injected into the ground with a downwards hydraulic gradient beneath the ice sheet, but with a predominantly upward gradient at the ice sheet margin. However, it was found that at some distance down ice from the beginning of the melting zone, the integrated meltwater flux was so large that even the most transmissive rocks are unable to discharge the meltwater by groundwater flow alone, without the hydraulic head exceeding the glacier head, an unstable and highly transient state. It is suggested therefore that when this happens, tunnels form at the ice/bed interface so as to discharge the excess meltwater flux, and draw down hydraulic heads to stable values. This situation has been modelled using realistic values of bedrock hydraulic conductivity from Scandinavia. It produces the surprising result that the spacing between tunnels should be in the range 5-25 km, and that the frequency of tunnels will increase in the down ice direction. These attributes happen to be characteristic of the esker systems and tunnel valleys that occur over the whole of the area covered by the last European ice sheet.

Using this pattern of eskers as a template, a model has been created, driven by the European ice sheet simulation, that computes the hydraulic gradients along tunnels lying along the lines of eskers, and a groundwater flow model determines the groundwater flow and potential fields associated with them (Figure 28). The simulations suggest that subglacial tunnels will be major groundwater sinks; that beneath the groundwater divides that separate groundwater catchments, there will be downward groundwater flow along downward directed potential gradients, and that tunnels will draw groundwater flow towards them, with the potential to create very large upward potential gradients sufficient in many cases to generate hydrofracturing, supercooling and strong local heating at the base of the glacier (Figure 28). Simulations suggest that overpressures created by these processes could survive at depth long after deglaciation.

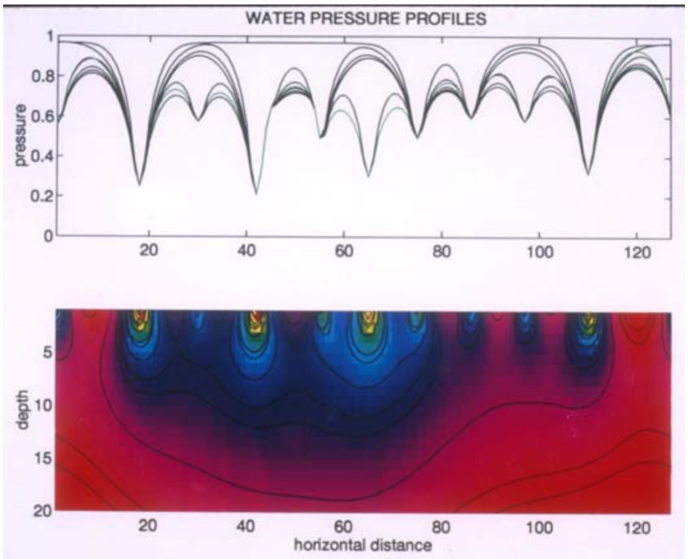

Figure 28. Section transverse to ice flow showing the subsurface heads associated with esker tunnels. The upper diagram shows the heads at the ice/bed interface. Heads are drawn down very strongly along the lines of tunnels, and are high between them. The lower diagram shows heads in the subsurface. There are very low heads below the areas of tunnels and very high potential gradients associated with strong groundwater flow towards tunnels. Normalised scales.

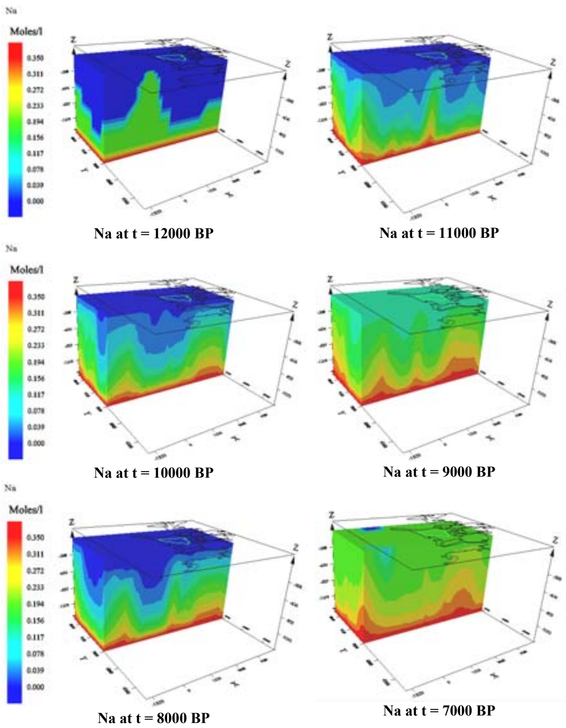

Figure 29. Modelled molar concentrations of sodium in groundwaters in the rock mass below the Äspö site. Early postglacial groundwaters are glacial waters with low sodium concentrations. They are partially replaced by saline, sodium rich waters during a phase of marine inundation at about 9000 y BP, are freshened by waters of the Baltic Ice Lake at about 8000 y BP, and finally replaced by marine waters after about 7000 y BP.

The strong depressurisation of carbonated groundwaters as they move along steep pressure gradients can theoretically be enough to produce contemporary carbonate precipitates that represent an archive of contemporary groundwater geochemistry. A field observation that might confirm this phenomenon is that of carbonate-cemented sand pillars, surrounded by uncemented sands, in an esker in northern Estonia. Geochemical analyses of the carbonate cements shows $^{18}O/^{16}O$ ratios that are consistent with precipitation from isotopically depleted waters typical of those forming from precipitation at high elevation on the summit dome of an ice sheet, which is the ultimate source of meltwater produced at the base of an ice sheet. As glacier ice contains large concentrations of air trapped in bubbles, meltwater also has a high partial pressure of oxygen. As a consequence, meltwater recharged to the ground is highly oxygenated and corrosive, and is not stripped of its oxygen by a soil layer as happens under non glacial conditions, because pre-existing soils will have been removed by the strong erosion typical of melting bed conditions. Under these conditions, solution and re-precipitation will be more important than under non-glacial conditions, although the depth to which they penetrate will depend on the extent of buffering by fluid-rock interactions.

The geochemistry of source waters has been characterised and both simple advective and complex coupled geochemical models has been used in PAGEPA to simulate time dependent geochemical responses at the Äspö and Gorleben sites (Figure 29). At the glaciated coastal site of Äspö, PAGEPA modelled the groundwater flow systems during the whole of the last glacial cycle. Äspö is known to have a complex distribution of mixed groundwaters from various sources and ages. The programme attempted to test the model by comparing the range of modelled variation with the observed pattern of variation. A block model was constructed, ca. 8 km square and 2 km deep. Modelling of this evolution using a computer programme that couples flow with geochemical reaction, it was possible to simulate groundwater mixtures that could be compared with measured present-day groundwater samples. The modelling also suggested that the impact of glaciation on sub-surface water structure and composition was particularly powerful, with fresh glacial waters having entirely flushed the sub-surface to depths of the order of a kilometre. However, models and site data suggest that high postglacial sea levels led to rapid displacement of these glacial groundwaters by denser saline waters that descended from the surface (Figure 29).

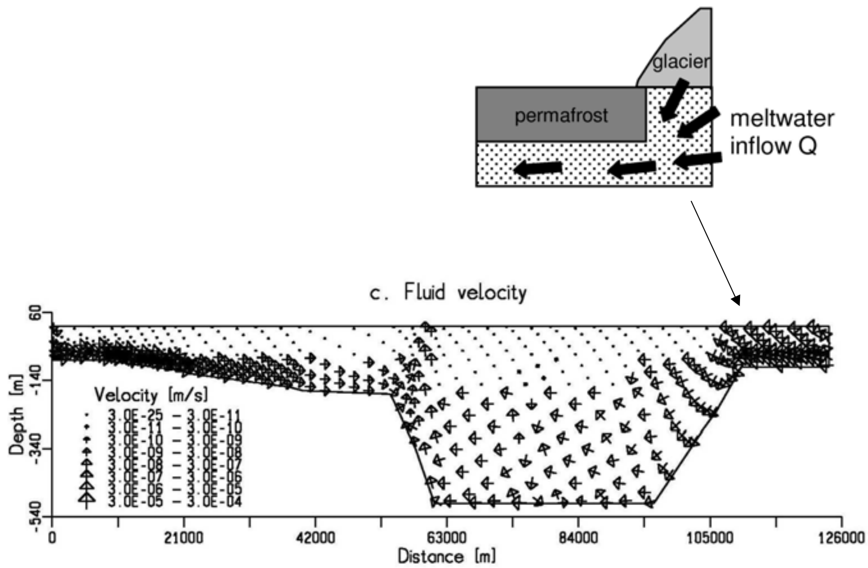

Figure 30. Model of glaciation and permafrost in the Gorleben area at the last glacial maximum. In this scenario, permafrost extends up to the glacier margin. Subglacial meltwater is confined beneath the proglacial permafrost and maintains high groundwater velocities until waters are discharged from the talik that exists beneath the Elbe River, in the middle of the valley. The surface is underlain by permafrost to a depth of about 140 m, except at the talik, where water is shown rising to the surface.

The Gorleben site lay some 60 km outside the maximum extent of the last glaciation ice sheet, but modelling and geochemical data suggest that the extension of permafrost beyond the ice sheet prevented subglacial groundwaters from emerging from beneath the glacier at its margin. A permafrost cap beyond the glacier confined groundwater flow beneath it, sustaining high heads and stimulated a strong circulation of glacially-driven groundwater flow as far as a talik underlying the Elbe River, some 60 km beyond the glacial maximum (Figure 30).

PAGEPA also studied the large-scale interaction between glaciers and permafrost. The depth of permafrost was calculated along a line from Sweden to France. Modelling the advance of the ice sheet over this permafrost suggested that even very thick permafrost could be degraded after about 1-2000 years.

Data from the AECL Whiteshell site in Canada have been used in BENCHPAR. The groundwater system in the low permeability crystalline rocks of the Shield is noteworthy for the persistence of overpressures, i.e. groundwater heads are higher than hydrostatic. At 11.5 ka ago, this area was covered by an ice sheet about 2000 m thick. It is suggested that the overpressuring is relict from ice loading, its decay being inhibited by the very low permeabilities.

General conclusions drawn from the PAGEPA and BENCHPAR studies include:

- Glaciation has the capacity dramatically to reorganise the physical and geochemical structure of groundwater systems and the distribution of stress within the Earth;
- Modelling of discrete processes, properties and geometries provides insights about site-specific phenomena, such as overpressuring at Whiteshell, complex groundwater mixing at Äspö and the coupling between permafrost and glaciation at Gorleben;
- Forward models designed to assess future scenarios can best be tested by using them to simulate evolution from the past to the present.

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5.3 DISCUSSION

Q What is the effect of sea-level on periglacial conditions?

A Permafrost is not able to penetrate far below sea level because of the large thermal capacity of either saline or fresh water bodies. We should also bear in mind that although global sea levels at the last glacial maximum were about 120 m lower than present on average, the isostatic effect of ice sheet loading is generally large enough to create locally higher than modern sea levels in the vicinity of ice sheet margins. As a consequence many shallow sea areas, such as the North Sea fringes of the ice sheets, are unlikely to have suffered permafrost conditions during the glacial maximum.

Q Could cemented esker sediments reflect the different sedimentology of eskers (compared to materials around/beneath them)?

A That is unlikely, but there is a need to establish more firmly the observational base.

6 PADAMOT: Palaeohydrogeological Data Analysis and Model Testing

Paul Degnan, Nirex, UK

6.1 SUMMARY

PADAMOT (Palaeohydrogeological Data Analysis and Model Testing) is a three year-long programme of research organised under the EC/Euratom 5th Framework RTD programme in nuclear fission safety. It began in January 2002 and is the successor to the EC 4th Framework project EQUIP (Evidence from Quaternary Infills for Palaeohydrogeology). The work in PADAMOT is directed towards providing a permanent solution for the disposal of radioactive waste, specifically by improving confidence in the concept of radioactive waste emplacement in deep geological repositories.

During the Quaternary period global climate has alternated between cold climate states, characterised by extensive permafrost/ice sheets/glaciers, and climate states warmer than the present. In northerly latitudes, the potential for cold region processes to affect groundwater pathways, fluxes, residence times and hydrochemistry is significant, whilst for southern European localities the alternation between pluvial and arid conditions is equally important. Because the long-term safety of an underground repository depends on the continued performance of the repository environment, in sufficiently isolating the wastes and attenuating any releases, demonstration that climatic impacts do not degrade containment properties is important. Scenarios for groundwater evolution relating to climate change are currently weakly constrained by data and process understanding. There is criticism by some opponents of deep disposal that current hydrogeological models are over-simplified and not addressing the full variability inherent in natural systems over time. PADAMOT is using mineralogical and hydrochemical data to investigate changes in groundwater conditions over time, and attempting to link these to changing global climate. From detailed analysis of high quality quantitative data, project scientists aim to develop improved process understanding, conceptual models and process models.

The four main work packages in the PADAMOT project are described. The Brussels Workshop comprises the first work package (WP1). In WP2, project members are investigating minerals and groundwater at sites in Spain, the Czech Republic, Sweden and the UK to relate their evolution to the environmental record. Isotopic signatures of fracture minerals from near-surface rocks are being compared with deep fracture minerals to find out what are the differences between deep and shallow impacts of past environmental conditions. New samples and some that have already been characterised are being used to evaluate analytical techniques that were not previously available in EQUIP, e.g. ion probe and laser ablation. These new techniques are analysing at spatial resolutions that distinguish compositional zones relating to specific groundwater episodes. Data are being investigated to see if it is possible to calibrate zones in calcite with the chronology of past climatic events. Analysis at the resolution of individual zones has not previously been practicable.

A relational database and a public domain website has been created as part of WP3. The database will store data from EQUIP and PADAMOT in a consistent manner. It will be accessible to all contributing partners via the internet and at the conclusion of the project the data will be made more widely available.

At least two types of geochemical process model are being used as part of WP4, to test the hypotheses erected under WP2 to explain generic and site specific aspects of groundwater

evolution. The geochemical models being developed and applied will relate variations in the compositions of calcite and redox-sensitive minerals (e.g. pyrite, iron oxide) with groundwater chemistry to the evolution of groundwater systems in typical repository formations. This task includes innovative modelling of reactions coupled with transport.

In WP5, valid conceptual models will be identified to describe hydrogeological scenarios linked to long-term climate and other types of environmental change, for use in safety assessments. Results from PADAMOT will provide guidance on the use of analytical techniques, process models and data requirements on varying geosphere conditions in response to changing climate. Knowledge acquired throughout the project will be interpreted and synthesised in order that it can have direct application to safety assessments. The enhanced understanding will have generic importance of value to all radioactive waste management agencies and regulators, as well as site-specific significance.

6.2 WORK PLAN

The work plan for PADAMOT builds on the outcomes of the preceding projects in FP4: EQUIP ('Evidence from Quaternary Infills for Palaeohydrogeology'), PHYMOL ('A Palaeohydrogeological Study of the Mol Site') and PAGEPA ('Palaeohydrogeology and Geoforecasting for Performance Assessment in Geosphere Repositories for Radioactive Waste Disposal'). The PADAMOT work packages and their broad objectives are:

1. Workshop on the use of palaeohydrogeology in PA: Direct interaction with PA specialists and the wider scientific community. Views concerning possible future research directions will be considered in developing the on-going PADAMOT programme.
2. Palaeohydrogeological data measurements: Application of analytical methods to mineral and water samples from new areas, in addition to detailed investigation of samples from well characterised sites. As well as applying tried and trusted techniques, new analytical methods will be used to get improved data at finer resolution (spatial and temporal) for mineral compositions and dating.
3. Design and compilation of database: Design of a database to ensure that all interpreted summary information is 'captured' and put into a format that is useable by PA models and for building confidence. Database will be accessible over the World Wide Web.
4. Development of conceptual and numerical models for process understanding and testing: Building a number of interpretative models for 'process-understanding' that will be calibrated using palaeohydrogeological information and will be the interface between the database, FEPs and PA.
5. Dissemination and recommendations concerning use of results for PA: PA teams will be updated throughout the project as results are obtained to evaluate how they contribute to selecting sites and demonstrating safety; a final report will be prepared and information placed on a web site.

The EQUIP project developed and evaluated methodologies for obtaining palaeohydrogeological information from minerals formed under past groundwater conditions (see paper by A. Bath, Section 2 in this report). It investigated calcite mineralisation that formed in fractures in crystalline rocks relatively recently in geological history, i.e. the last 2 million years or so. Calcite was chosen as the principle mineral for study because its reactions (both precipitation and dissolution) in low temperature groundwater environments are fairly well understood, and it is also fairly ubiquitous in both crystalline and sedimentary rocks. Geochemical modelling suggested that plausible timescales for growth of individual calcite crystals are such that they may accumulate a record of groundwater evolution over periods of significant climate change. A number of mineralogical, geochemical and isotopic methods were employed to unravel the evidence of past groundwater conditions. Uncertainties in the results arose mostly from the limitations of analytical techniques then available to the project and from the interpretative models of processes.

Work on Sellafield samples confirmed that calcite morphology might provide a record of changes in salinity. Modelling shows that these changes in salinity are related to changes in the boundary conditions of a groundwater system. The need for further study of the factors that control the precipitation rates and geochemistry of calcite precipitated in rock fractures was identified, so that interpretation could take account of generic and site specific factors. Compositional zones in calcite were inferred to reflect changes in co-existing water chemistry, especially redox conditions, over the period of crystal growth. Such changes in the past, and the recognition of factors that mitigate such fluctuations, are of potential significance for predictions of long-term stability. Techniques for analyses of uranium series isotopes and stable isotopes that would indicate mineral ages and sources of coeval groundwaters were not generally applied in EQUIP, and it is a primary aim of PADAMOT to exploit recent developments in these techniques where possible.

A number of recommendations were put forward by EQUIP for the continuation of this approach. PADAMOT addresses these issues. They include (a) further developments of analytical techniques that exploit the rapid advances in instrumental capabilities especially for quantitative microanalysis for trace elements and isotopes for dating, (b) developing modelling tools to interpret such data quantitatively and to relate this both to water-rock reactions at the scale of mineral crystals and also to evolution of the groundwater system at larger scales, and (c) the focusing of further research to investigate specific processes that might link climate and groundwater in low permeability rocks. It was also appreciated that more could be done to integrate palaeohydrogeological research results into Performance Assessments (PA) for repository siting studies, both generic and site specific.

Acquisition of data for palaeohydrogeological interpretation will be guided by what is quantitatively or qualitatively useful in limiting the ranges of PA scenarios, especially with respect to boundary conditions and responses at depth. Thus the necessary data sets will be identified by working back from the inputs in a time-dependent PA model to the categories of data with which those inputs are quantified or constrained. The types of data are (these might be regional or site-specific):

- Palaeoclimate and future climate data set containing estimates for climate variables at time steps with appropriate resolution;
- Hydrological data set containing estimates for surface water conditions;
- Topography data set containing land elevation data and water table elevation data for representative locations at different times;
- Rock properties data set: estimates of time-variant changes (if any) in physical properties of hydrogeological units;
- Borehole and other sample location information;
- Hydrochemical data set for depth profiles through groundwater systems;
- Secondary mineral data set from drillcore profiles including sample locations and structural setting information;
- Mineralogical data including morphology, compositions and relative ages of zones;
- Fluid inclusion data including salinity and chemical analysis.

6.3 WORK PACKAGES

6.3.1 WP1: Workshop on palaeohydrogeology in PA

The PADAMOT workshop in Brussels (October 2002) was held to achieve a wider understanding of what has been achieved in EQUIP and related research, and what are the priorities for the use of palaeohydrogeology in PA. The workshop was intended for participants who have expertise in PA and in repository site selection and characterisation, but who are not necessarily directly otherwise involved in PADAMOT. Other interested geoscientists e.g. palaeoclimate experts and aquifer hydrogeochemists were also invited. The intention was that PA practitioners, experimentalists and field scientists could interact freely and feed back to each other their expectations concerning information from future work programmes. For example, what are the types and quality of basic data necessary for developing conceptual models? What are the requirements for populating numerical models with appropriate parameters and what is the feasibility of providing such information (at the required scales) given realistic acquisition techniques? In addition, addressing issues such as how scientists could further contribute to the specification of FEP ('features, events and processes') descriptions, and other confidence-building information, was an expected outcome.

6.3.2 WP2: Palaeohydrogeological data measurements

This section of the work programme describes where samples will come from, what measurements will be made and what techniques will be available. The selected techniques will be capable of getting data that are of adequate quality for the proposed interpretations and that will stand up to broader scientific scrutiny. PADAMOT will exploit state-of-the-art technology to achieve this.

Specific questions to be addressed are:

- Is the occurrence and distribution of fracture calcite sufficiently consistent and well enough understood that it could reliably be used for characterisation of some or all site types?
- Under what geological, hydrogeological and climatic conditions does the growth of calcite and other minerals provide a useful record of groundwater change/stability in the recent past?
- Can a 'calcline' (i.e. the depth at which there is a transition from calcite dissolution to calcite precipitation) be identified, and how does this depend on palaeoclimate and geological conditions (both in terms of degree and rates)? Are there similar transitions for other key minerals such as pyrite and Mn/Fe oxy-hydroxides?
- What are the ages of recently-formed calcites (bulk calcite and compositionally-distinct zones), and does age information with other isotopic and chemical indicators correlate reasonably with the timing and environmental impacts due to climate change during the recent past?
- Are there distinctive changes in the mineral properties (zoning, morphology, etc) or isotopic composition of calcite and other minerals in different parts of the same groundwater flow path? Do these changes indicate that minerals formed in recharge areas are more sensitive to surface environmental changes than those in deep flow paths and discharge areas?

Samples of recently-formed (?Quaternary) calcite are available from generic research sites in different palaeoclimatic regions of Europe at Sellafield and Dounreay (UK), Äspö (Sweden) and Los Ratones (Spain). Near-surface samples will also be obtained because they are expected to have different fracture mineral characteristics due to the different degrees of climatic impact between deep and shallow groundwaters. Comparable samples will be obtained from locations in eastern Europe – these will be in fractured crystalline or sedimentary rock at shallow depths. They will allow a comparison to be made of the impacts on shallow groundwater and mineral compositions in the central European palaeoclimate regime relative to the northern and southern European regimes. Co-existing groundwater samples will be available for these sites so that the geochemical and isotopic partitioning between minerals and water can be studied. Additional fracture mineral samples will be obtained from a new site in fractured rock that will be drilled in Year 2.

The EQUIP procedure for sampling and characterising fracture minerals will be used and modified to incorporate the new techniques. This work topic will be led by BGS. Sampling will be carried out in the first and second years of the project. Basic petrographic analysis will be done locally, and a set of samples will be compiled that are representative of the various sites. It is anticipated that this 'priority' set will comprise about 30-40 samples which will be analysed as follows:

- Cold-cathode cathodoluminescence (CL), back-scattered scanning electron microscopy (BSEM) and elemental mapping by electron microprobe;
- Ion microprobe analysis of trace elements and isotopes;
- Accelerator mass spectrometry (AMS) analyses of carbon-14 in calcite (subject to availability);
- Stable O and C isotope analyses of bulk calcite;
- Sr isotope analysis by laser ablation microprobe – plasma ionisation multi-collector mass spectrometry (PIMMS);
- Chemical and isotopic analyses of groundwaters;
- Microthermometric (MTA) and laser ablation (LAMP-ICPMS) analyses of fluid inclusions;

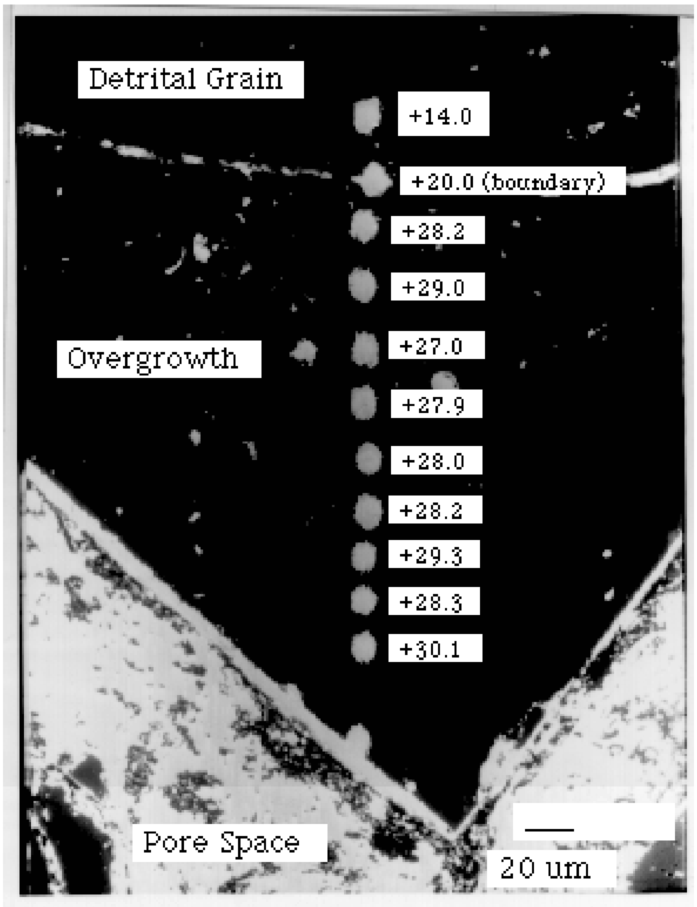

Figure 31. Ion probe microsampling of individual growth zones in minerals permits isotopic and trace element (ppb) analysis at high resolution (1-20µm for trace elements, ~50 µm for isotopes).

Samples will be prepared as polished sections and examined initially by optical and BSEM petrography techniques. Samples containing calcite mineralisation will subsequently be examined by cold-cathode cathodoluminescence petrography (CL) to examine the details of growth zoning fabrics. Chemical characterisation of the mineralisation will be carried on the same polished sections using electron probe microanalysis. Appropriate, well-characterised samples will be selected, on the basis of these petrographic analyses, for further investigation of isotopic and chemical composition (ion probe, PIMMS and laser ablation microsampling-mass spectrometry – Figures 31 and 32). The methodology will ensure consistency of data from different techniques in the same laboratory, and will provide a basis for cross-comparison of data from alternative techniques or different laboratories.

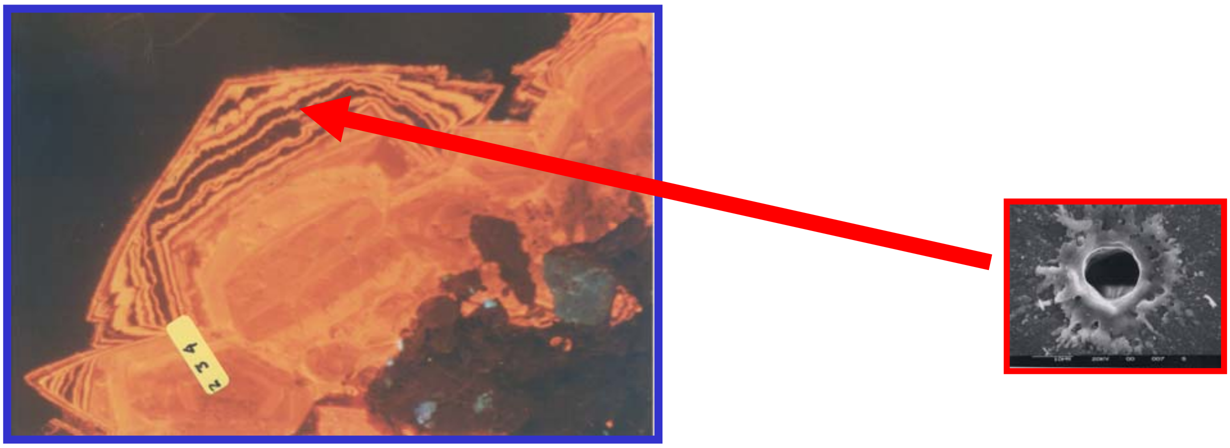

Figure 32. Laser microsampling of individual growth zones in calcite (C, O) provides indirect indication of age/groundwater processes. Resolution 80-100µm

The ways that these analyses will be used to address the above questions are described below:

6.3.2.1 Comparison of fracture minerals from deep and shallow locations

Calcite and iron oxide from shallow locations in the groundwater systems will be compared with minerals in deep samples. Mineral samples at shallow depths will be more likely to show the impacts of present and past climates on their compositions than those from greater depths. The aim will be to ‘calibrate’ the morphology and variations of trace elements and stable isotopes in calcites against known climatic impacts.

Trace elements analyses can be considered in two parts: minor element components of calcite (e.g. Mg, Fe, Sr) presumed to be present at ppm levels, and trace elements (e.g. REE) presumed to be present at ppb levels. Conventional electron microprobe analyses were used in EQUIP to investigate Fe and Mn variations as potential indicators of redox changes during calcite growth. EQUIP also showed that other minor and trace elements show variations through zoned calcites (e.g. Sr, Ba, Mg, Ce, and Eu) and that these may be significant as indicators of residence time, redox, etc. Calcites will be analysed using a combination of ion microprobe (for low level trace elements: e.g. REEs) and electron microprobe analysis (for minor elements: Fe, Mn, Mg).

6.3.2.2 Estimation of palaeoenvironment and age with stable carbon and oxygen isotopes

Correlating oxygen isotopic zoning patterns with known climatic or environmental events will provide an indirect means of constraining the age of the calcite. Analysis and interpretation of stable isotope compositions of recent calcites may be the only feasible approach to dating recent minerals and may therefore be important for site characterisation.

The ion microprobe (University of Edinburgh) will analyse up to twenty samples of calcite for ^{13}C and ^{18}O isotopic zoning. The selected fracture calcites will be prepared for ion microprobe analysis using the methods developed in EQUIP. The sections will first be examined using CL and BSEM petrography and electron microprobe elemental mapping to characterise the main chemical zonation characteristics. Zonation of stable isotopes will be compared with the identified crystal growth fabrics.

BGS will analyse the $\delta^{13}C$ and $\delta^{18}O$ isotopic composition, and $^{87}S/^{86}Sr$ isotopic ratio of coarser growth zones in some calcite samples using the laser ablation micro-sampling - mass spectrometry technique, as a comparison between the two techniques for high-resolution isotopic analyses. Laser microsampling and isotopic analysis will undertaken using polished mineral grain mounts, after detailed characterisation and 'mapping' of the mineral growth zoning by BSEM, CL and electron microprobe analysis.

Analyses of the isotopic composition of corresponding 'conventional' samples will be carried out by Terralogica. This information will be important alongside the micro-scale analyses of individual zones because it will identify where the use of conventional 'bulk' carbonate samples might or might not be adequate for site characterisation.

6.3.2.3 EVALUATION OF NEW ANALYTICAL TECHNIQUES FOR ANALYSING AND DATING FRACTURE CALCITES

The feasibility of analysing ^{14}C by accelerator mass spectrometry for studying the ages of a sub-set of fracture calcites from shallow locations will be evaluated (University of Edinburgh). Only shallow material has any likelihood of detectable ^{14}C, and detecting its presence would provide a strong constraint on how recently the flow and chemical conditions for calcite precipitation have prevailed. Sample requirements dictate that analysis could not discriminate between growth zones, so samples will be selected on the basis of consistency between stable isotope compositions of calcite from bulk calcite and individual zones.

For most samples, U-Th dating of individual zones of calcite is unlikely to be feasible with existing methods because of the low abundance of U. However, developments in ion probe instrumentation that will become available in Year 2/3 may enable that analysis to be carried out (University of Edinburgh). This would be a very significant development if successful, allowing the potential for relative or even direct correlation of calcite zone development (and associated geochemistry/hydrochemistry) to climatic events. U concentrations in fracture calcite samples from the new sites may be sufficient to allow isotope analyses. Therefore the strategy with respect to U-Th dating will be to carry out analyses only when there is a reasonable chance of success in obtaining dates for 'calibration' of the stable C and O isotope compositions.

6.3.2.4 DIRECT ANALYSIS OF PAST GROUNDWATER COMPOSITIONS IN TRAPPED FLUID INCLUSIONS

Fluid inclusions, the microscopic droplets of water trapped in minerals during crystal growth, are a unique means of measuring the compositions of past groundwaters. Chemical compositions of inclusions representing past groundwaters were analysed in EQUIP by microthermometric analysis (MTA) and laser ablation ICP mass spectrometry (LAMP-ICP-MS) in Quaternary calcites, though recently-formed fluid inclusions were not sufficiently abundant for analysis for all of the studied sites. The aim in PADAMOT is to extend the database and to improve the interpretation of fluid inclusion data.

Fluid inclusions will be analysed in 30 to 40 samples of calcite from new sample locations, using MTA and LAMP-ICP-MS (BGS). The mineral samples will be screened and characterised using BSEM and CL petrography prior to analysing the fluid inclusion. Additional fluid inclusion analyses will be carried out by CIEMAT and BGS. MTA will provide information on salinity (total dissolved solids, TDS) estimates for 5-20 µm diameter aqueous inclusions in calcite to ±3000 ppm over the salinity range 3000-250,000 ppm. LAMP-ICPMS and novel cold plasma techniques will measure relative concentrations for Li, Na, K, Rb, Mg, Ca, Sr, Ba, B, Fe and Mn in 20-40 µm diameter fluid inclusions. Fluid inclusion data will be interpreted to give a relative chronology of changes in water chemistry with time.

6.3.3 WP3: Design and compilation of database

Establishing a purpose-designed database from the outset will ensure that all interpreted data from the proposed work, as well as from previous work (e.g. EQUIP) and literature review, are stored in a quantitative and recoverable form. Characterisation tasks and analytical specifications in the previous work topic will be more focused if these data requirements are established at the outset. The central role of the database is shown in Figure 33. This work package will construct a relational database to handle the different types of data and to retrieve them for interpretation and visual presentation. Raw data will be available as web links to PADAMOT partner organisation websites. Given the quantity of data to be included, the database will be developed using Oracle. The principal deliverable from the database will be:

- Retrieval of quantified and interpreted information, derived from the palaeohydrogeological studies, that can be used to constrain FEPs and scenarios for PA.

Other potential uses of the database will be:

- Presentational illustration of data, information and estimated trends in palaeoclimate (and future climate) and hydrological parameters versus time;
- GIS-based 3D visualisation of distributions of samples, mineral data, hydrochemical data, etc;
- GIS-based 2D visualisation of time-variant patterns in climate features such as ice sheet growth, permafrost development, water table fluctuation, saline interface movement, etc;
- Presentational illustration of depth patterns in mineral, hydrochemical and isotopic data for single boreholes;
- Calibration and input data for interpretative models that estimate palaeohydrogeological information from mineral and geochemical data;
- Graphical or statistical comparisons between similar data sets from different regions or sites;

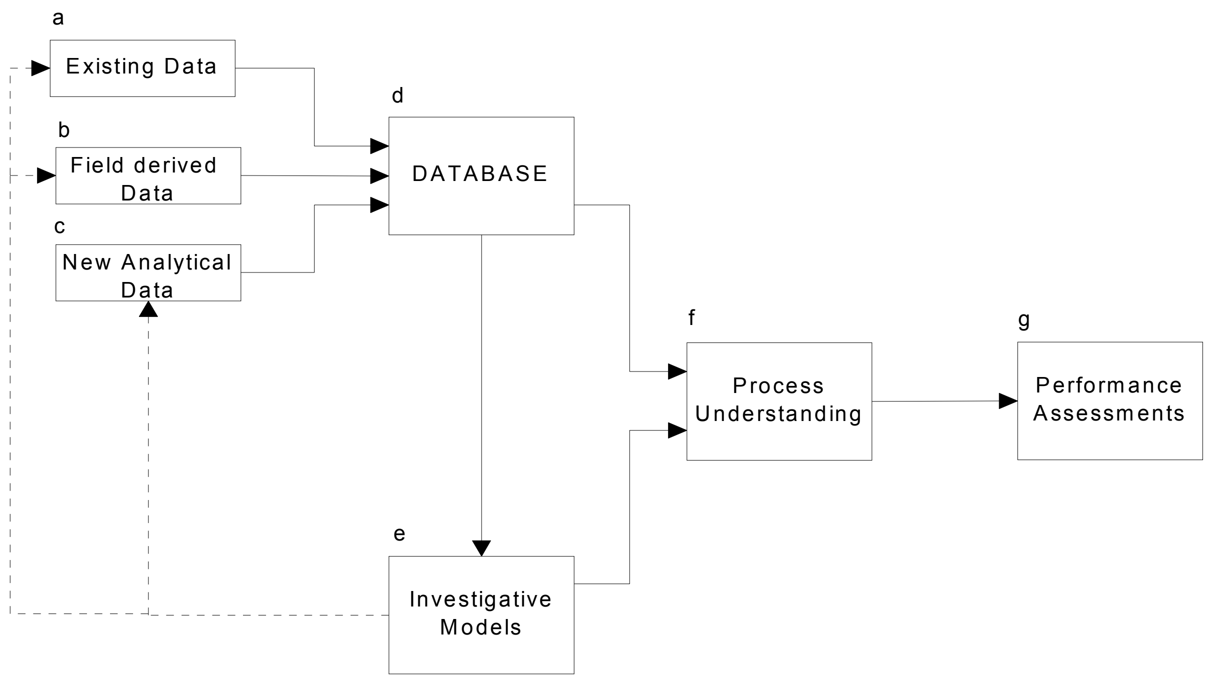

Figure 33. Schematic diagram showing the central role of the database for assembling data that can be used by process models and by PA.

The database will be interfaced with a GIS (e.g. ArcInfo, ArcView) for display and manipulation of spatial information.

Two additional factors must be considered in the design of the database. Firstly, uncertainty ranges will be stored with many types of data. These can be stored as simple statistical distributions (e.g. uniform or triangular) between upper and lower limits to reduce the complexity. Secondly, time series will be stored for some data sets, e.g. climate or hydrological properties over the timescale of interest (e.g. 10^6 y).

Design construction and implementation of the database will be undertaken by BGS, with input from Partners Nirex, SKB and ENRESA. Nirex has provided a suitable high specification NT server (with Oracle 8, ColdFusion and Webserver software.) to hold the database. The server will initially reside with BGS on a temporary basis while the database is developed and implemented. It will then be returned to Nirex who will arrange for it to be sent to a suitably identified server farm or ISP, where it will be co-located for permanent internet access by other Partners.

6.3.4 WP4: Development of models for process understanding and testing

Palaeohydrogeological information will be obtained from 'proxy' data (geochemical, mineralogical, isotopic, etc) by using interpretative models. Additional information, e.g. climate data from independent sources, geophysical data, analogue data from present-day 'extreme climate' environments, will be used as validity checks on the models. Interpretative models are based on understanding of the processes that link the proxy data with climate-driven groundwater phenomena, the 'cause-effect' relationship.

This work package will develop and apply geochemical models that relate the compositions of calcite and redox-sensitive minerals (e.g. pyrite, iron oxide) with groundwater chemistry to the evolution of groundwater systems in typical repository formations. Two types of interpretative models will be produced: (i) geochemical equilibrium and mass balance calculations, and (ii) a mixing model that simulates the spatial and temporal evolution of groundwater compositions, and of co-genetic minerals.

6.3.4.1 GEOCHEMICAL EQUILIBRIUM AND MASS BALANCE CALCULATIONS

Geochemical equilibrium modelling will be used to understand the past conditions under which calcite is likely to have precipitated or dissolved. Geochemical modelling carried out in EQUIP showed that uncertainties in the in-situ pore-water chemistry (in particular pH and P_{CO2}) have a major influence on the modelling. Mass balance calculations will initially make estimates of the amounts of fracture calcite in the system, both as proportions of total rock mass and fracture void space, and also in relation to estimated present-day closed-system or open-system fluxes of component solutes in groundwaters.

The rate of calcite precipitation can be related to groundwater compositions and flow by a model where water equilibrated with the atmosphere travels to depth and mixes with water that is equilibrated with rock at depth. The geochemical computer codes EQ3/6 and PHREEQC will be used to simulate the water compositions taking into account the importance of reactions involving the dissolved gases O_2 and CO_2.

The interaction between the two fluids will be modelled by BGS using a modified version of the code PRECIP to give an indication of the quantities of calcite precipitating. PRECIP is a 1D coupled reaction-transport code where changing porosity directly influences flow rate. Precipitation rate will be described as a function of variables such as starting temperature, fluid flow rate, and degree of fluid mixing.

Separately, CIEMAT will use a hydrological model (VISUAL-BALAN) with data from WP2 to calculate water mass balance and characteristics of past surface water conditions that are necessary inputs for groundwater modelling as described in the following section.

6.3.4.2 Hydrodynamic/Geochemical Mixing Model for Spatial and Temporal Evolution of Groundwater Systems

A numerical 2D model will simulate the hydrodynamics of a site with time-varying boundary conditions. This will be coupled with calculations of transport and mixing of non-reactive and reactive species to forward-model the time variant geochemical evolution of groundwater mixtures. The model codes $CORE^{2D}$ and RETRASO have already been developed and demonstrated (ENRESA and CIEMAT). The simulated chemical and isotopic compositions of groundwaters and co-existing minerals will be compared with what is observed. Initially the model would simulate the conservative palaeohydrogeology/palaeoclimate indicators (stable isotope compositions, salinity, solute residence/transit time indicators). The model would be used (a) to constrain the impacts of various palaeoclimatic conditions on the groundwater system and thus to guide scenario development for PA, and (b) to illustrate and communicate how alternative conceptual models can be considered against the geochemical evidence.

The mesh for $CORE^{2D}$ is being constructed with boundaries that will be able to represent discrete time-variant changes in water pressure and/or composition (e.g. water table, sea-water interface, lateral inflow). The model mesh will be more detailed for the zones where geochemical and mineral data are actually available, allowing an adequate simulation of the processes that have produced the present minerals with the observed chemical and isotopic zoning. Simulations of the time-variant compositions of groundwaters and co-existing mineral precipitates will be compared with observed data for macro-scale distribution of calcite, Fe-oxide and pyrite in the present-day system, Fe-Mn compositions and inferred redox regime for individual zones in calcites and for inclusions of Fe-oxide and pyrite, salinity distribution and oxygen isotope composition of present-day groundwaters, and oxygen and carbon isotope compositions of individual zones in calcites.

This work package will deliver a demonstration and assessment of the modelling approach as a tool for interpreting geochemical measurements and significantly, for providing a visualisation to communicate the interpretation. The model will be generated for one site initially, but the development, methodology and the learning derived from the model will be applicable at any site specified at a later date.

6.3.5 WP5: Dissemination and use of results for PA

The PA teams in the radioactive waste management organisations will be regularly updated on progress and their continued involvement will ensure that the palaeohydrogeological results from PADAMOT are relevant to their needs. The review of PA uses and needs will be updated yearly, in which the outputs from PADAMOT will be evaluated in terms of their significance to FEPs and scenarios.

A final report will be published at the end of the three-year period to disseminate the results obtained to a wide international audience. The choice and the approaches made by the collaborative experts involved in the project will be presented and justified and the main results will be made available. The advantages and the limits of the approaches and the results will be addressed as well as their possible uses for site specific and generic performance assessments. All partners involved in the proposal will assist in the preparation of this publication.

6.4 DISCUSSION

Q How will human activity have influenced the mineralogical and groundwater record being studied in PADAMOT?

A In developing any future scenarios it may be important to ensure that we do not ignore past and on-going human influences on natural systems. For example, there have been major anthropogenic effects on hydrology over the last few thousand years due to deforestation and urbanisation, resulting in changes in runoff and local recharge patterns. From a global and long term perspective, however, the effects of past anthropogenic influences on the groundwater system could be considered insignificant before about 10 ka BP. This date corresponds roughly to the rise of agrarian societies and the establishment of large permanent centres of human population. It also marks the end of the last period of major glaciations. Therefore the mineralogical record before this time, which is likely to record the effects of significant 'natural' climate change, is unlikely to record any anthropogenic effects. Whilst it is true that more recent human activity has perturbed the natural system, the mineralogical effects that we observe today would not be extensive when viewed in the context of the longer record.

Q What are the criteria for 'shallow' and 'deep'?

A 'Shallow' often extends to around 100-200 m depth, corresponding to the zone containing evidence for oxidation related to surface processes and evidence of responses to climate change. A depth of 500 m, representing a possibly appropriate target depth for repositories, should be considered to be 'deep'. There is a growing debate in the UK about long-term 'shallow' underground storage. Interactions between the effects of climate and groundwaters tell us something about the inherent differences between 'shallow' and 'deep' components of a groundwater system. Characterisation of shallow and deep zones and an understanding of the differences represents good progress. However, there are no hard and fast criteria to differentiate shallow from deep domains and, depending on the precise questions being posed, it may be difficult to decide upon appropriate measures (permeability, redox, etc.). Even if a reasonable measurement parameter is chosen for a specific problem, the cut-off value between the two domains is usually an arbitrary choice. Using the change in permeability with depth, for example, would pose a problem as it is generally gradational - unless there are abrupt lithological (including diagenetic and weathering effects) or structural changes, as for example observed at Sellafield, in which case a feature based classification could be used. The terms 'shallow' and 'deep' should be considered to be an aid to conceptualisation rather than a strict limit on system boundaries.

o Such definitions prejudge conditions: for example, 'deep' conditions could be oxidizing. Definitions like these tend to prejudice arguments.

o It's not helpful to have generalised definitions; definitions need to be site-specific.

o Groundwater systems should be referred to as 'fast' or 'slow'. For example, at 150 m depth near a Czech installation there exists very old (^{14}C) water ('slow') surrounded by much younger ('fast') waters.

7 Potential Applications of Advanced Analytical Techniques for Geochemical Analysis of Secondary Minerals

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7.1 PALAEOMAGNETIC METHODS

The palaeohydrogeological studies that were undertaken during the EC Framework 4 'EQUIP' Project and that are currently being undertaken within the subsequent EC Framework 5 'PADAMOT' Project, focus on the use of mineralogical and geochemical information recorded in late-stage calcite mineralisation. In sites such as at Sellafield and Dounreay, late-stage calcite is relatively common and forms crystals large enough to be readily examined by routine petrographical methods. At other sites (e.g. Äspö, Laxemar and Olkiluoto), petrographical observations are more limited because the calcite mineralisation is much finer grained and of lower abundance. Even in the Sellafield site, calcite is absent or reduced in abundance at shallower levels. In these situations other minerals such as iron and manganese oxides and oxyhydroxides could potentially furnish useful palaeohydrogeological information. Palaeomagnetic methods are one way of improving understanding.

Iron atoms in iron bearing minerals crystallising and precipitating from groundwater or hydrothermal fluids become polarised by the Earth's magnetic field, and acquire magnetic characteristics that reflect the ambient magnetic field at the time of their formation (chemical remnant magnetism). Thus, they can preserve a record of the past magnetic states and it might be possible to correlate mineralogical palaeomagnetic characteristics with the history of changes of the Earth's magnetic field. This methodology has been utilised in the dating of fault movements, by correlating the magnetic properties of authigenic iron minerals such as authigenic limonite (goethite) (e.g. Hailwood et al., 1992) or iron-bearing minerals such as calcite (e.g. work by Torsvik and Sturt referred to by Maddock, 1992) with past magnetic field states.

Iron and manganese oxides and hydroxides are commonly present as late-stage minerals in the weathering zone and in the shallow oxidising parts of the groundwater systems. Examples of this type of mineralisation are found at Sellafield, Dounreay, Äspö, Laxemar and Olkiluoto. Although, their distribution and formation can be related to the development of the present day groundwater system (e.g. Milodowski et al., 1998; 2002), little use has been made of iron and manganese oxyhydroxides in palaeohydrogeological investigations, often because they are very fine grained and poorly crystalline, and consequently difficult to study. However, this type of mineralisation may lend itself to palaeomagnetic investigations, and may provide a means of constraining ages of rock-water interaction processes if the magnetic properties can be correlated to changes in the Earth's magnetic field during the Quaternary.

Magnetic susceptibility measurements might also provide useful palaeohydrogeological information. The magnetic susceptibility of the rock mass will potentially increase as secondary iron and iron-manganese oxyhydroxides form. Measurement of relative differences in magnetic susceptibility properties in the rock mass may provide a means of identifying the flow paths of oxidising groundwaters.

7.2 MICROBIOLOGICAL METHODS

7.2.1 General

Biomarkers were studied during the EQUIP Project, in order to investigate the palaeohydrogeology of Quaternary sequences of the Padul and Cúllar-Baza Basin in southern Spain (Bath et al., 2000). This study used interpretations based on the analysis for n-alkanes, elementary sulphur and carbon and oxygen isotopic analysis of organic material extracted from the groundwaters. However, microbiological 'biomarkers' might also furnish useful palaeohydrogeological tools.

Micro-organisms have long been known to be important in many groundwater systems, influencing geochemical and mineralogical processes (Erlich, 1990; West and Chilton, 1997; West and McKinley, 2002). Interest in subsurface microbiological activity has grown significantly in the last 20 years, and has focused on (West and Chilton, 1997; West and McKinley, 2002; Powell et al., 2003; Pedley et al., in press):

- The biodegradation of organic contaminants in groundwater;
- Groundwater denitrification processes and nitrate contamination
- The behaviour and migration of pathogens in groundwater including: •the burial of farm animals during the foot and mouth disease outbreak and the potential for pathogen migration from these burials to water supply boreholes; the survival and transport of prions in the environment – particularly the BSE agent; outbreaks of cryptosporosis linked to contamination of groundwater sources with Cryptosporidium oocysts; the disposal of sewage sludge on fields and the potential of pathogens from these sites to migrate to water supply boreholes; the migration of pathogens from faulty septic tanks, sewerage pipes into water supply boreholes; the potential for pathogens from cemeteries to migrate into water supply boreholes; deliberate release of pathogenic agents)
- The role of bacteriological activity in relation to the migration of radionuclides from nuclear waste repositories.

The ability of the sub-surface to support microbiological activity depends on several factors, including:

- Availability of nutrients (principally, carbon, phosphorus, nitrogen, sulphur);
- Energy sources for microbial metabolism (electron donors and acceptors);
- Availability of water;
- Physical characteristics of the aquifer (e.g. groundwater flow, nature and availability of sites for attachment and growth);
- Environmental conditions experienced by the organisms (e.g. salinity, pH, redox, temperature).

Microbes can survive in a wide range of environments, and each micro-organism will have a range of conditions under which its growth is viable. However, they have been found in extreme conditions of acidity, alkalinity, redox, temperature, pressure, radioactivity and salinity (West and Chilton, 1997), and may remain active even under low nutrient supply (as in the Äspö groundwaters). Different groups and assemblages of micro-organisms develop and adapt to the different environmental conditions that may exist in the surface and sub-surface environments.

In the context of Quaternary climate changes, it might be expected that different microbial assemblages will be developed and adapted to cope with the different soil and sub-surface environments produced in response to climate change. Soil-derived micro-organisms and/or their products may be carried into the deeper groundwater system from the recharge zone (Price et al., 1992; Tranter et al., 1997). Climate-driven changes within the groundwater environment (permafrost, redox, salinity, organic carbon) may also give rise to the development of locally specially adapted microbiota. Consequently, it may be possible to utilise data from the microbiological analyses of groundwater, for palaeohydrogeological interpretation, if these assemblages (or biological markers such as DNA) are preserved and can be related to past environmental conditions.

To date, most groundwater microbiological investigations have focussed on identifying populations present and assessing their bio-geochemical impacts at the present-day. Very little is known about the long-term preservation of microbial assemblages that may have been introduced into the groundwater system. They may die and/or be replaced by new biota in response to changes in the groundwater environment. The new microbe populations may destroy any evidence of any earlier assemblage by utilising it as a carbon source. However, in some cases, it is believed that microbes may persist for hundreds or even thousands of years.

7.2.2 Bacteriophage

Bacteriophages ('phages') are virus-like micro-organisms that 'prey on' bacteria. Numerous different phages are known since almost all bacteria (including actinomycetes) are susceptible to infection by specific phages. Bacteriophages are highly specific, and each type is able to infect only one group of closely-related organisms – usually a single species. This characteristic behaviour of phages is utilised to provide a method for 'typing' bacterial isolates in modern epidemiological studies.

There is a growing interest in bacteriophage in groundwater studies. Phages have a particle size between 10^{-5} to 10^{-9} m and behave as colloids in groundwater, and several recent studies have used phages as tracers (Skilton and Wheeler, 1989; Ward et al., 1997, 2000, 2001).

Like other virus, bacteriophages are only active when in the presence of their host organism. When the host is absent, the phage crystallises and remains dormant in this state until the host organism is re-established. It can survive for a very long time in this dormant crystalline state for much longer than the host organism. Therefore, there is significant potential for using groundwater phage in palaeohydrogeological investigations. Different palaeoenvironmental conditions – in either the soil/recharge zone or the sub-surface environment – might be expected to produce specific phage assemblages that reflect the different microbial communities. Consequently, if the phage preserved in the groundwater can be identified and associated with particular microbial assemblages, it may potentially provide information that reflects the past recharge and sub-surface environments.

Bacteriophages might sorb or come to rest on the surfaces of minerals growing in the groundwater. Continued growth of the mineral might cause them to be 'entombed' as minute solid organic inclusions or trapped within fluid inclusions. Because of their potential durability, the trapped phages may remain viable in this state for a significant period of time. Thus, there may be possibilities of extracting and identifying useful phage material from late stage mineral inclusions.

7.3 SYNCHROTRON RADIATION SOURCE ANALYSIS TECHNIQUES

A wide variety of Synchrotron Radiation Source (SRS) analytical methods are now available with spectroscopic, diffraction and imaging techniques using radiation with wavelengths extending from the infrared to hard X-rays. Sophisticated SRS analyses can be used to provide information on mineral surface structure and physical properties, mineral surface interactions, surface chemistry, chemical speciation, chemical coordination and oxidation states, crystal and molecular structure. SRS techniques are finding increasing use in environmental and geoscientific areas. Information obtained from SRS techniques is largely being applied particularly to understanding and modelling of the structure, surface complexation and reactivity of mineral surfaces. However, a detailed review of this is outside the scope of this present paper. Nevertheless, some of these modern analytical methods may find useful applications, alongside other geochemical, mineralogical and petrological techniques used in palaeohydrogeological studies, and a few examples are given below.

X-ray absorption fine spectroscopy (EXAFS - or sometimes abbreviated as XAFS), ex situ X-ray standing wave (XSW) and surface extended X-ray absorption fine structure (SEXAFS) spectroscopy have been used to study the nature of interactions, speciation and uptake of metals at the mineral water interface. It has recently been used to investigate the uptake and coordination chemistry of trace metals on the surface of calcite, to study how this influences the incorporation of trace metals within, and the growth of, the calcite crystals (e.g. Cheng et al., 1998; Elzinga and Reeder, 2002). Developments in this area could potentially impact on how compositional and isotopic zoning in calcite mineralisation is interpreted, in terms of past groundwater conditions. This forms an important aspect of the palaeohydrogeological investigations of the PADAMOT and EQUIP projects.

SRS X-ray microprobe analysis and imaging methods (Integrated Microanalyser for Imaging and X-ray – IMIX; and X-ray capillary microprobe) are being developed to provide highly sensitive and high-resolution trace element chemistry information from laminae within speleothems, for use in palaeoclimatic studies (Kuczumow et al., 2001). Notable information obtained from these techniques included: information on the inclusion of iron and silicon; measurements of calcium/strontium/barium ratios; profiles of anionic species – including the presence of iodine. These data were considered in the potential estimation of the climatic conditions during the deposition of the calcite material.

Potentially SRS techniques could provide valuable trace element chemistry data for zoned fracture calcites from deeper groundwater systems, or possibly could be used to the study the nature and distribution of trace elements within clay minerals or compositionally late-stage iron and manganese oxyhydroxides at shallower levels (e.g. Randall et al., 1998; 2001).

7.4 ACKNOWLEDGEMENTS

The author thanks Dr Julie West and Mr Mike Raines (British Geological Survey) for useful discussions on the potential application of microbiology and palaeomagnetic methods in palaeohydrogeological studies.

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8 Palaeohydrogeology: Lines of Evidence for Understanding the Past Evolution of Groundwater Systems and Its Use in Safety Assessments

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8.1 ABSTRACT

Palaeohydrogeology, the study of the past evolution of groundwater systems, combines evidence from several sources including observations from the depositional rock record, measurements of physical hydrogeological parameters such as groundwater pressure and flow rates, measurement of hydrochemical and isotopic variations in groundwaters, and measurement and analysis of geochemical and morphological data from mineral precipitates either in rock fractures or as diagenetic minerals within the rock matrix. Understanding, presented as a conceptual model, results from interpretation and compilation of the various lines of evidence and may be tested using a variety of numerical simulation and modelling techniques. A broad overview is presented of the various lines of evidence, their usefulness to understanding the past evolution of groundwaters in different geological and geographical settings, and challenges in terms of data acquisition, interpretation, conceptualisation and modelling.

8.2 INTRODUCTION

Palaeohydrogeology, the study of the past evolution of groundwater systems, combines evidence from several sources including observations from the depositional rock record, measurements of physical hydrogeological parameters such as groundwater pressure and flow rates, measurement of hydrochemical and isotopic variations in groundwaters, and measurement and analysis of geochemical and morphological data from mineral precipitates either in rock fractures or as diagenetic minerals within the rock matrix.

Understanding, presented as a conceptual model, results from interpretation and compilation of the various lines of evidence and may be tested using a variety of numerical simulation and modelling techniques.

A broad overview of the various lines of evidence, their usefulness to understanding the past evolution of groundwaters in different geological and geographical settings, and challenges in terms of data acquisition, interpretation, conceptualisation and modelling is presented.

8.3 PALAEOHYDROGEOLOGY – WHAT IS IT?

Although the concept of, or at least the use of the term, palaeohydrogeology is a relatively new one it is argued that the science underpinning the terminology is, or at least should be, a fundamental a part of any site characterisation programme for radioactive waste disposal. However, the use of palaeohydrogeology, both directly in performance assessment and indirectly to support a safety case, has not yet been fully demonstrated despite recognition of its potential almost a decade ago (Chapman and McEwen, 1993).

According to the NEA definition (NEA, 1993) palaeohydrogeology is:

"A combination of observations on hydrochemical and isotopic differences in various groundwater zones or bodies, mineralogical data on the rock formations and the hydraulic properties of the same formations, which are then compiled to allow interpretation of the evolution of the rock-water system over long time periods in the past."

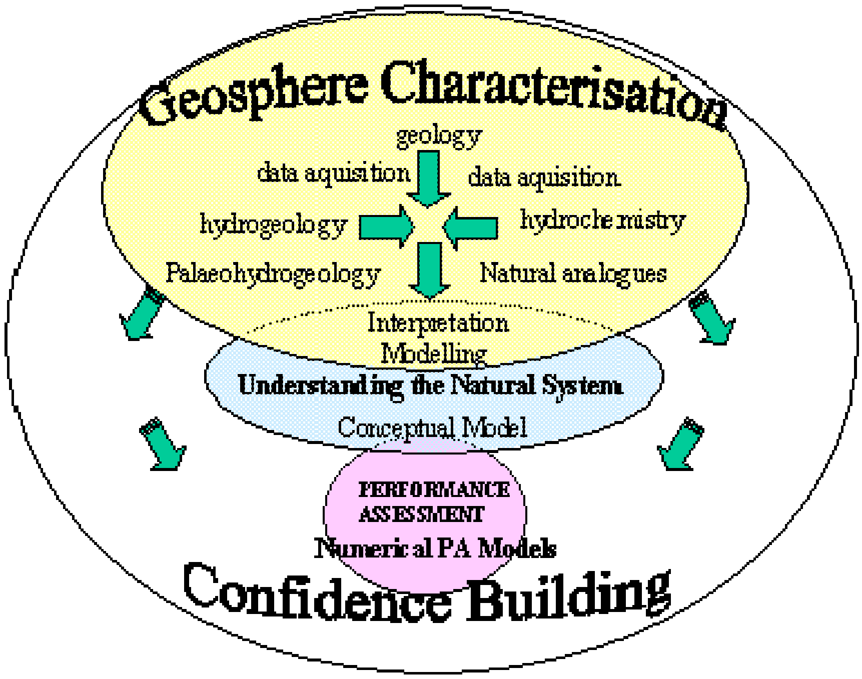

Figure 34. The roles of palaeohydrogeology in Safety Assessment.

This definition shows the linkage between palaeohydrogeology and site characterisation but falls short of demonstrating the application of palaeohydrogeology to performance assessment and the development of a safety case. From this perspective, palaeohydrogeology together with natural analogue studies are most appropriately viewed as ways of integrating knowledge and understanding from a variety of sources in order to build confidence in the past and future evolution of a site (see Figure 34).

The main contribution of palaeohydrogeology, therefore, may be in demonstrating an understanding of the site and its evolution such that that confidence can be placed in the results of the safety assessment rather than in providing direct input into performance assessment calculations.

8.4 LINES OF EVIDENCE

As noted in the introduction, palaeohydrogeology, combines evidence from multiple sources (Figure 35). Broadly, the interpretation of data acquired from these sources contributes to the development of an understanding of the evolution of the groundwater system through application in three main areas:

- The system description
- The boundary conditions and driving forces
- The system response mechanisms

System description – includes, for example a description of the geology and hydrogeology of the site and its principal characteristics in terms of repository host rock type (e.g. crystalline, clay, salt) and the current distribution and chemistry of groundwater bodies present in different regions of the deep geological environment. Palaeohydrogeological studies will contribute to understanding the origins of different groundwaters, the degree to which mixing has occurred and the geochemical mechanisms operating within and at the interfaces between the different water bodies. Evidence of the principal pathways for water flow in the system, at present day and in the past, and the extent to which these have influenced the overall evolution of the groundwater regime may be contained within fracture zone minerals.

Boundary conditions and driving forces - includes the identification of external features, events and processes (FEPs) controlling the driving forces affecting the system and defining the boundary conditions for modelling purposes. One of the main questions to be answered at any repository site is "Will hydrochemical conditions still be suitable in the future or will future external processes (e.g. climate change) induce important modifications in the hydrochemistry of a repository?" Palaeohydrogeology can provide insights, for example, into the possible effects of long-term climate change on groundwater fluxes and groundwater chemistry.

System response mechanisms - includes, for example, the signal transmission properties of the site in terms of say, rates of penetration of oxidising groundwater or the physical and chemical buffering capacity of the rock mass. Palaeohydrogeology studies can contribute to an analysis of the capacity of the geological barrier to provide a chemically and mechanically stable environment in which the functions of the repository engineered barriers remain robust on a long time scale. For example, it may be possible to determine the residence times of the different groundwater bodies and hence comment on their stability with respect to changes in external conditions.

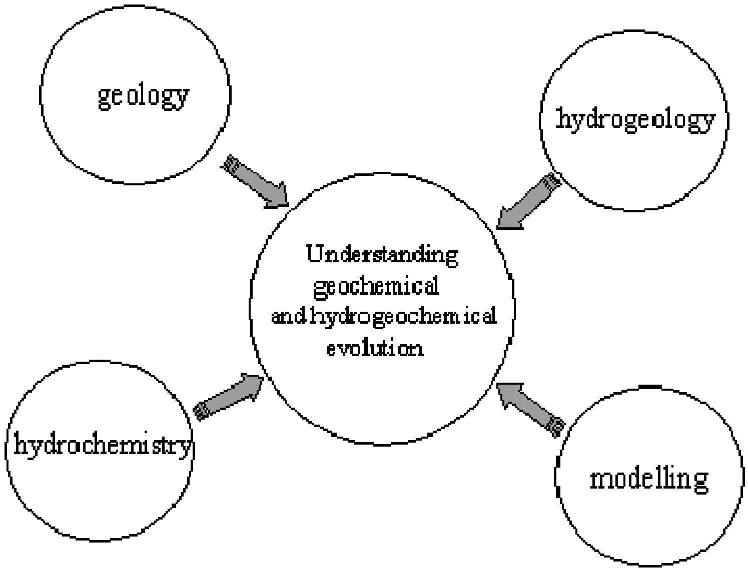

Figure 35. Multiple lines of evidence

The nature of palaeohydrogeological evidence potentially available within the different disciplines illustrated in Figure 35 and the main uncertainties and challenges associated with data acquisition and interpretation are briefly described below.

8.4.1 Geological Evidence

The general trend in application of palaeohydrogeology, in response to the requirement to carry out performance assessments over a forward period of 1 million years, has been in understanding groundwater evolution over the past 1 million years i.e. over the latter part of the Quaternary epoch. However, the rock record may contain useful information with respect to past groundwaters over much longer time periods. Variations in the depositional environments of sedimentary rocks, for example, may relate to the uplift and subsidence history of the area.

Rock facies types provide markers to depositional environments e.g. sandstones (shallow coastal waters, mudstones (deeper water continental shelf); evaporites (evidence of dry arid climate), coals (warm humid climate), phosphates and cherts (ocean upwelling); reef limestones (warm surface ocean). Analysis of sedimentation rates, grain morphology and chemical composition all provide information on climatic conditions prevailing at the time of parent rock weathering and the type, morphology and distribution of fossils and microfossils may be indicative of warm and cold water regimes. Oxygen isotope analysis of nanofossils from deep-sea cores was a breakthrough in Pleistocene climate reconstruction.

8.4.2 Mineralogical Evidence

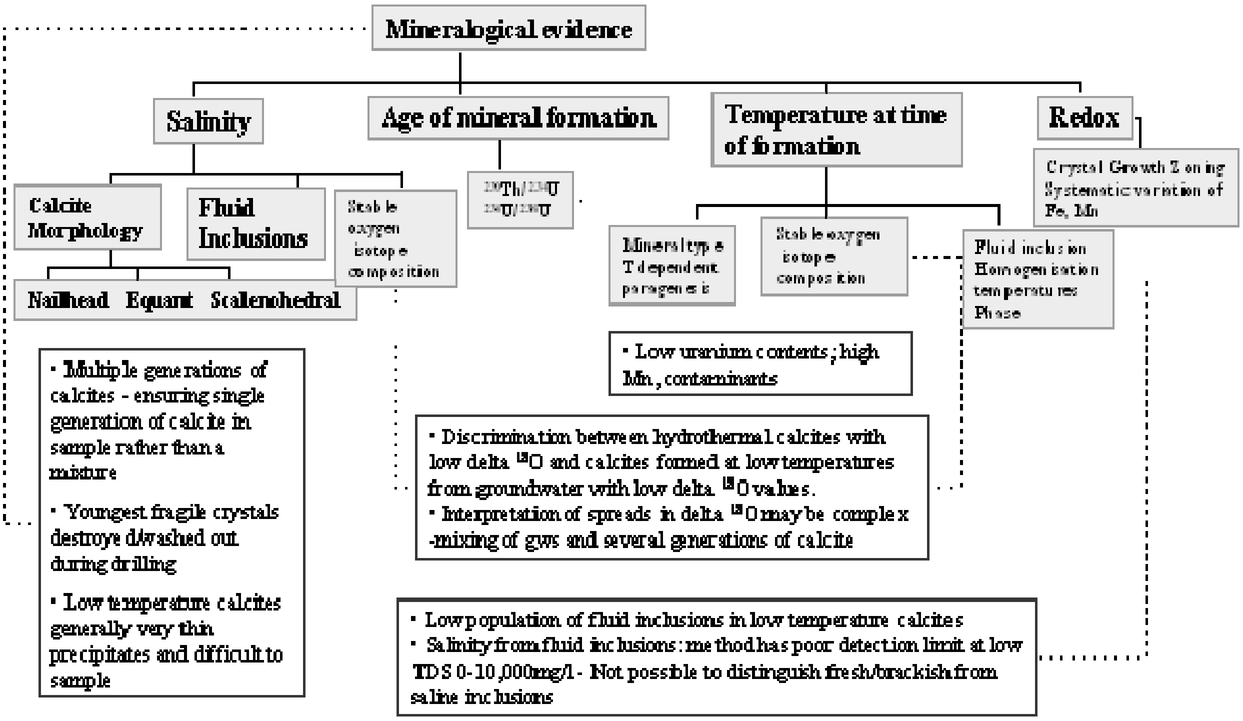

Figure 36. Summary of data types available from mineralogical studies and associated challenges and uncertainties

Arguably the most fruitful source of palaeohydrogeological evidence comes from the study of the mineralogy of fracture fillings. Vein minerals are an 'archive' of groundwater information with an associated timescale. Figure 36 summarises the information and data types arising from mineralogical studies and the processes to which they contribute palaeohydrogeological evidence.

8.4.3 Hydrogeological Evidence

Hydrogeological data are essentially snapshots of present day or relatively recent hydraulic pressure regime – although low permeability rocks may retain relict pressures that are out of equilibrium with present boundary conditions and thus may hold information relating to past hydrogeological conditions. However, this complication may pose problems for interpretation and understanding as there may be several alternative interpretations and modelling solutions capable of explaining the head distribution (e.g. Sellafield).

8.4.4 Hydrogeochemical Evidence

The hydrogeochemical record comprises a large number of variables incorporating information on the history, reactions, mixing and flow paths of groundwaters. These variables include:

- 20-30 major water constituents
- Isotopes
- Gases
- Microbes
- Colloids
- pH, Eh, temperature

Figure 37 summarises the information available from study of groundwater chemistry and identifies the areas to which the palaeohydrogeological evidence applies. Some of the difficulties and challenges in acquiring and interpreting representative chemical data on groundwaters in low permeability rock types are also identified.

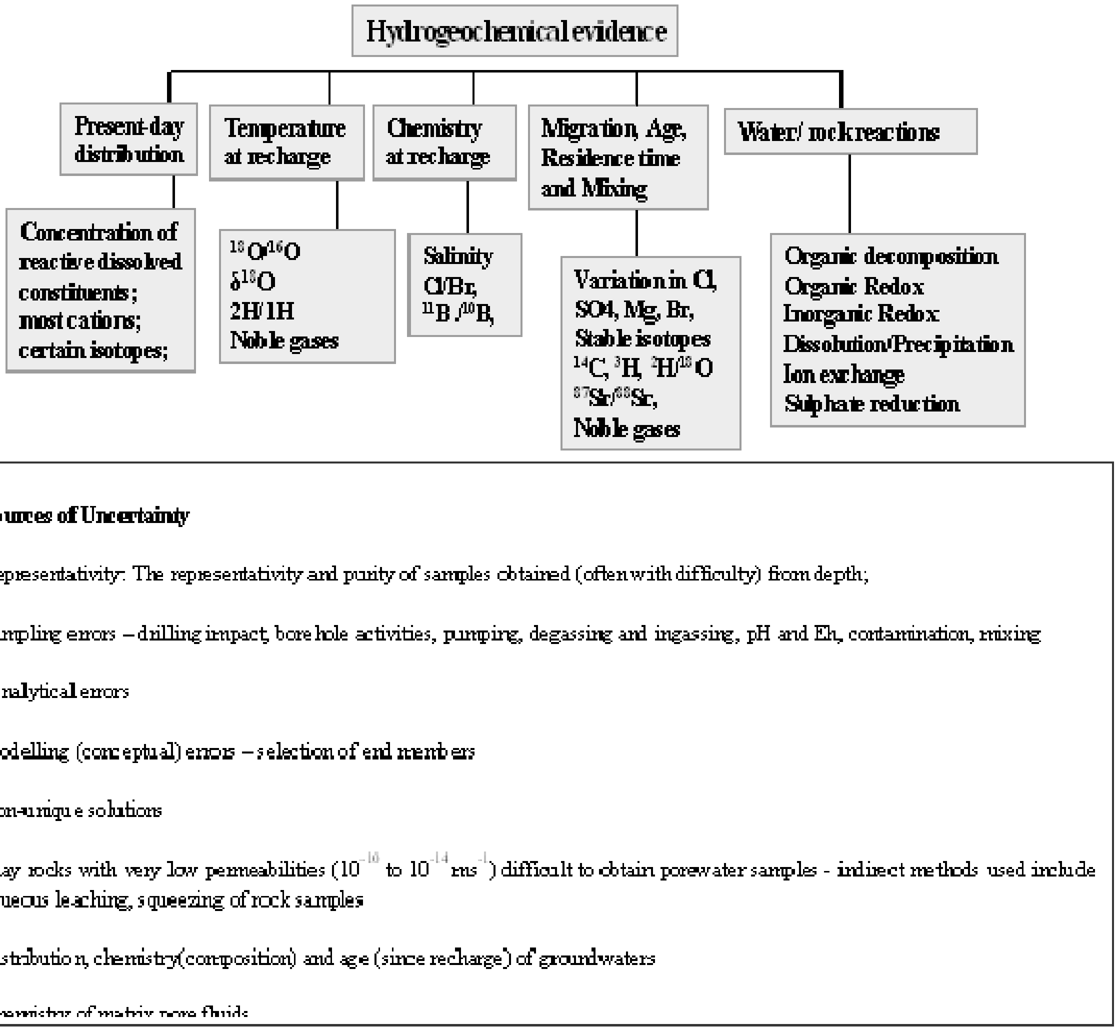

Figure 37. Summary of data types available from hydrogeochemical studies and associated challenges and uncertainties.

8.4.5 Modelling

A range of modelling techniques is available for exploring groundwater flow, mixing and reactions. Ideally, 3-dimensional, time-dependent, coupled chemical-hydrogeological models are needed to untangle the effects of all processes that have occurred at the site. Chapman and McEwen (1993) identified a number of specific difficulties and complications:

- The conversion from apparent isotopic 'dates' for groundwaters to true 'ages' by accounting for dilution and mixing processes between different waters;
- The overlapping nature of many rock-water interaction processes that can provide several mechanisms to explain certain observations;
- The natural in situ production of some of the environmental isotopes used in isotopic dating systems;
- The lack of appropriate kinetic data for some rock-water interactions;

The nature and contribution of what are generally regarded as secondary mechanisms or sources to the evolution of the groundwater system (e.g. tectonic effects on flow, very deep sourced fluids affecting hydrochemistry etc).

In practice a range of separate but interlinked models is used to fully explore the groundwater system behaviour (Figure 38). One modelling approach that has found popular acceptance recently, possibly as a result of its relatively simple approach to penetrating the mass of hydrogeochemical data and its presentational qualities, is the Multivariate Mixing and Mass Balance model (M3).

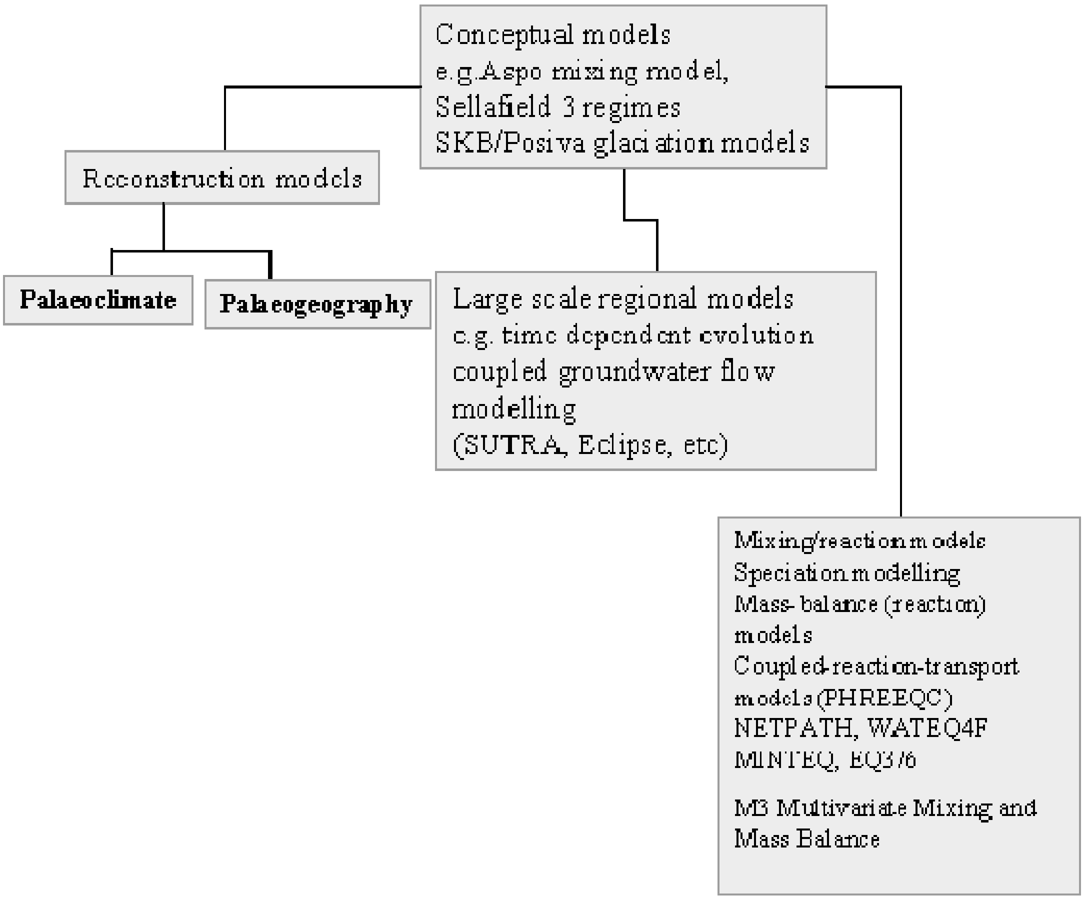

Figure 38. A range of separate but interlinked models is usually required to explore the groundwater system behaviour.

8.5 APPLICATION OF PALAEOHYDROGEOLOGICAL EVIDENCE

Two alternative, but not mutually exclusive, approaches to the application of palaeohydrogeological evidence are (R. Metcalfe, pers.comm.):

- To deduce a 'history' of the groundwater flow system
- To focus on specific issues relating to establishing site stability

There are difficulties in the first approach because of the absence of large parts of the geological record and because of the many intrinsic uncertainties. As with more quantitative assessment approaches, palaeohydrogeology is more powerful if the geological environment is relatively simple. However, a difficulty arises with sites that may apparently be very suitable for hosting a radioactive waste repository due, for example to the low permeability of the rock types and apparent robustness to external influences but for which evidence is difficult to obtain and assemble into a coherent 'history' of the groundwater flow system. The second approach may be more successful in focussing palaeohydrogeological arguments to support specific hypotheses. This approach requires the presentation of a reasoned argument in support of the hypothesis whilst ensuring that uncertainties are made transparent and propagated through the logic flow and that 'confirmation bias' is avoided by explicitly identifying negative evidence. One method of achieving this is through the application of evidence-based reasoning.

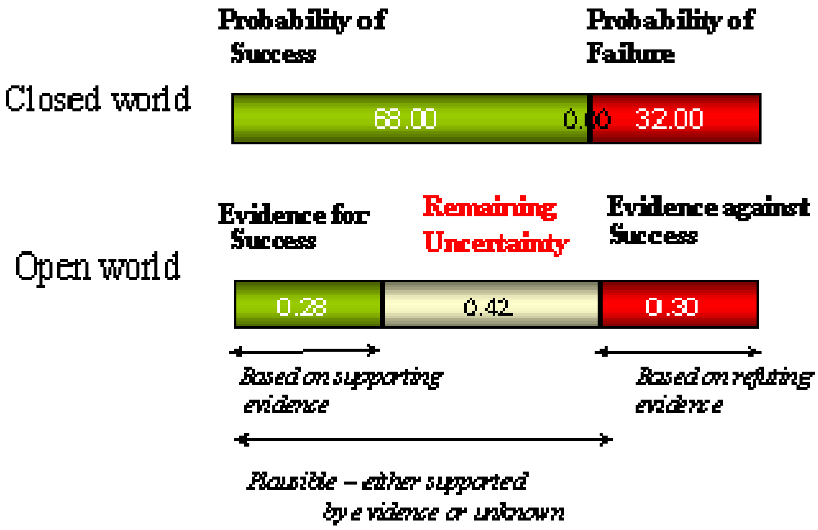

Figure 39. 'Open world' evidence-based reasoning differentiates remaining uncertainty from the evidence for and against a hypothesis.

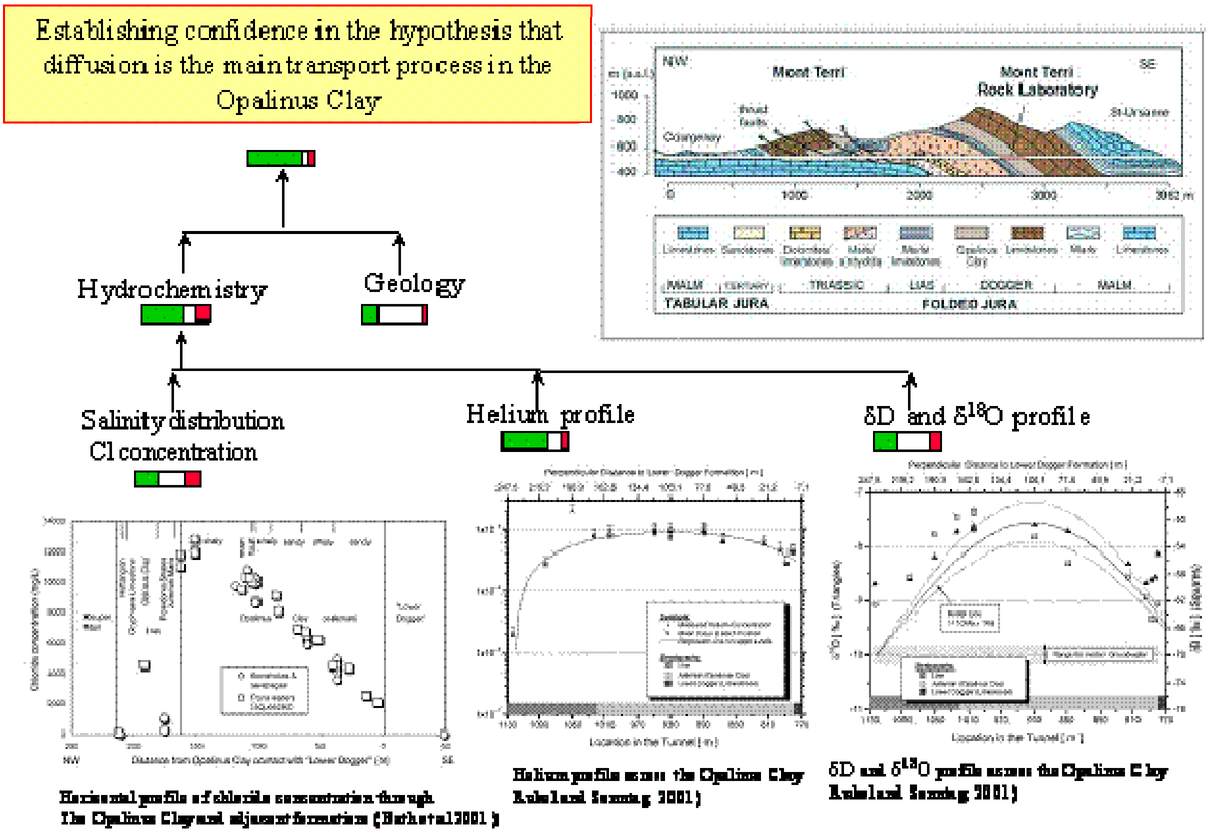

Figure 40. An evidence-based process framework for assembling evidence in support of the hypothesis that 'diffusion is the main transport process in the Opalinus Clay'.

Given the high uncertainties associated with the geosciences generally, and with the acquisition of representative sample data for measurement of groundwater chemistry in particular, it is appropriate to use an evidence-based approach to provide support for hypotheses arising from palaeohydrogeological investigation. Evidence-based reasoning applies expert judgements to the data, and to the quality and meaning of the data in the context of a particular hypothesis (Figure 39). Interval representation of evidence in support of, evidence against and remaining uncertainty in the processes contributing to the top-level hypothesis are accumulated and propagated through a model of the problem. Support for the hypothesis is gauged from the model result in terms of the balance of evidence in support to the evidence against the hypothesis and the remaining uncertainty in the system.

An example (schematic) of the possibilities for using this approach was presented based on experiments undertaken at Mont Terri (Figure 40).

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8.7 DISCUSSION

Q How do you know how big your uncertainty is in ‘Italian Flag’ models?

A You don’t; it is an open-world approach.

9 The Use of Palaeohydrogeology in Performance Assessment and, in Particular, Its Use in Previous Assessments

Tim McEwen, SAM, UK

9.1 INTRODUCTION

The use that has been made of palaeohydrogeology in safety assessments has been reviewed, using the following assessments as examples: TILA-99, SR 97, H-12, Nirex 97, Kristallin-1, AECL 94, SAFIR 2, with input also from other assessments and their parallel research programmes. This review is placed in the context of the PADAMOT workshop regarding the evolution of groundwater conditions at depth, the extent to which it is believed that they have been stable and the implications for their possible lack of stability.

9.2 HOW PALAEOHYDROGEOLOGY HAS BEEN USED IN ASSESSMENTS

The use that has been made of palaeohydrogeology in safety or performance assessments is not as great as had originally been envisaged almost ten years ago (e.g. Chapman and McEwen, 1993). Palaeohydrogeology has, for example, not been used in any predictive capacity in performance assessments and there has been no real quantitative testing of aspects of performance assessment models using a palaeohydrogeological approach. However, it is now accepted that it is necessary to understand the evolution of a disposal site to its present-day conditions, either in order that there can be sufficient confidence in developing ideas as to its future behaviour (i.e. more in a predictive capacity via scenario analysis) or simply in demonstrating that sufficient is understood about the site that confidence can be placed in the results of the safety assessment (and that the disposal concept developed at the site will be sufficiently robust). It is believed that palaeohydrogeology is probably best used in this confidence-building role rather than directly in any performance assessment calculations.

Figure 41 illustrates in conceptual form the types of processes that occur in the geological environment that will influence the stability of the groundwater and affect its age. It is these processes and the effects that they have on the rock mass and groundwater system that need to be understood. Their study forms the core of a palaeohydrogeological study of a site, but the extent to which this evidence is used within a performance or safety assessment has varied considerably.

The majority of performance assessments have used palaeohydrogeology in a variety of ways. The assessments that are considered here are: TILA-99, SR 97, H-12, Kristallin-1, AECL-94, SAFIR 2, Nirex 97, together with the forthcoming Opalinus Clay assessment and information from the assessment work for the WIPP and Yucca Mountain. The use that has been made of palaeohydrogeology can be summarised as:

- It is sometimes not used directly,
- Its use is sometimes not apparent – it has, for example, been used in the development of conceptual models for groundwater flow through fractured rocks, without any specific reference being given to its use,
- It has not generally been used to constrain parameter values for use in assessments,
- It use is helpful in defining scenarios and

- It has been used more in a general sense as support for assessments.

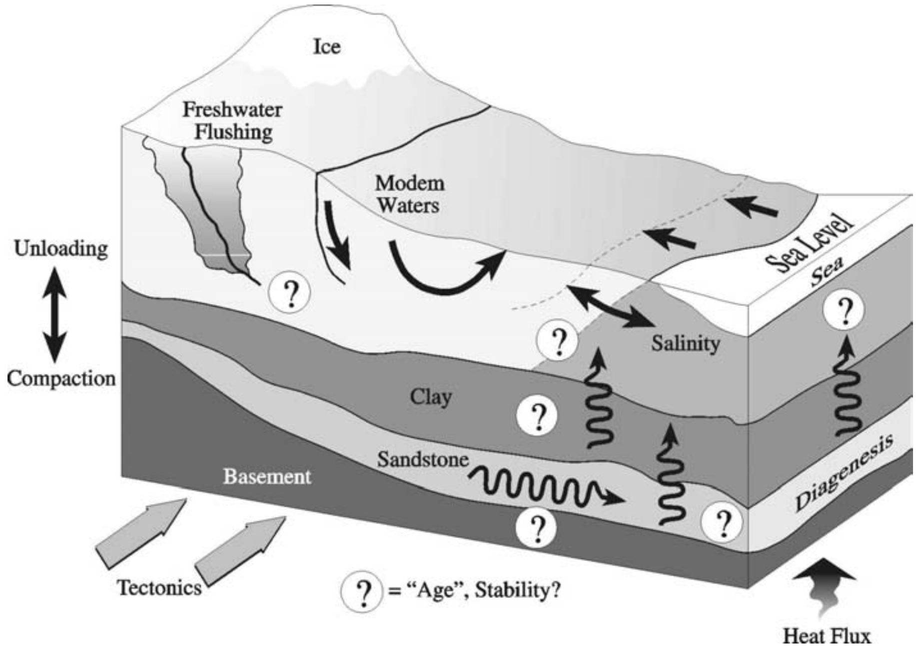

Figure 41. The types of processes that occur in the geological environment that will influence the stability of the groundwater and affect its age. An understanding of these processes is required to unravel the evidence provided by the minerals in the rock mass (in particular, those in the fractures) and the chemistry of the groundwater and understand how a site has evolved to its current state.

9.3 SUMMARY OF ASSESSMENTS STUDIED

A short summary of the use that has been made of palaeohydrogeology is presented below for eight assessments.

TILA-99

- The "normal evolution" (i.e. that considered most likely) of the natural system over a period of 1 million years is considered as the basis for calculations:
- This is based on a review of past changes to the hydrogeological system
- It provides the basis for "what-if" calculations, e.g. the effect of intrusion of glacial meltwater into the repository
- No further use is made of palaeohydrogeology.

SR 97

- Palaeohydrogeology is used to provide insight into the effects of long-term climate change, such as:
  - changes in groundwater fluxes
  - changes in groundwater chemistry
- And, thereby, provides indications of future changes in these conditions over the forthcoming 130,000 years
- The understanding of the changes that have taken place to the groundwater system in the past is used in the definition of calculational cases

- The overall conclusion of the assessment is that even extreme climate changes do not affect the stability of a repository to an unacceptable degree.

H-12

- Similar use is made of palaeohydrogeology to that of Posiva and SKB
- The results of these studies are used to develop scenarios and, thereby, calculational cases
- The main influence of climate change is associated with sea level change (especially for a coastal site), i.e. the effect on the saline interface, etc.
- No specific climate change scenario is developed
- The effects of changes in groundwater chemistry and groundwater fluxes are considered using "what-if" calculations

Kristallin-1

- The use of palaeohydrogeology is seen as one way of building confidence in the assessment
- It use is seen as an adjunct to natural analogues
- The calculational cases developed are based on the understanding developed of the evolution of the groundwater system over the last few hundreds of thousands of years

AECL-94

- Extensive palaeohydrogeological studies were carried out, e.g.
  - Investigations of calcites in fracture zones
  - Investigations of groundwater mixing (related to fracture zones)
- In fact, this was the first assessment when palaeohydrogeological investigations were so extensively applied
- Changes in groundwater salinity were sufficiently great that geosphere retardation factors had to be made a function of the salinity
- A 10,000 year cut-off in assessment calculations, so no "what-if" calculations of climate-related changes, as carried out by Posiva
- The essential elements of the groundwater system typified by the conditions at the Canadian URL site, that was used for the AECL-94 assessment, are shown in Figure 42.

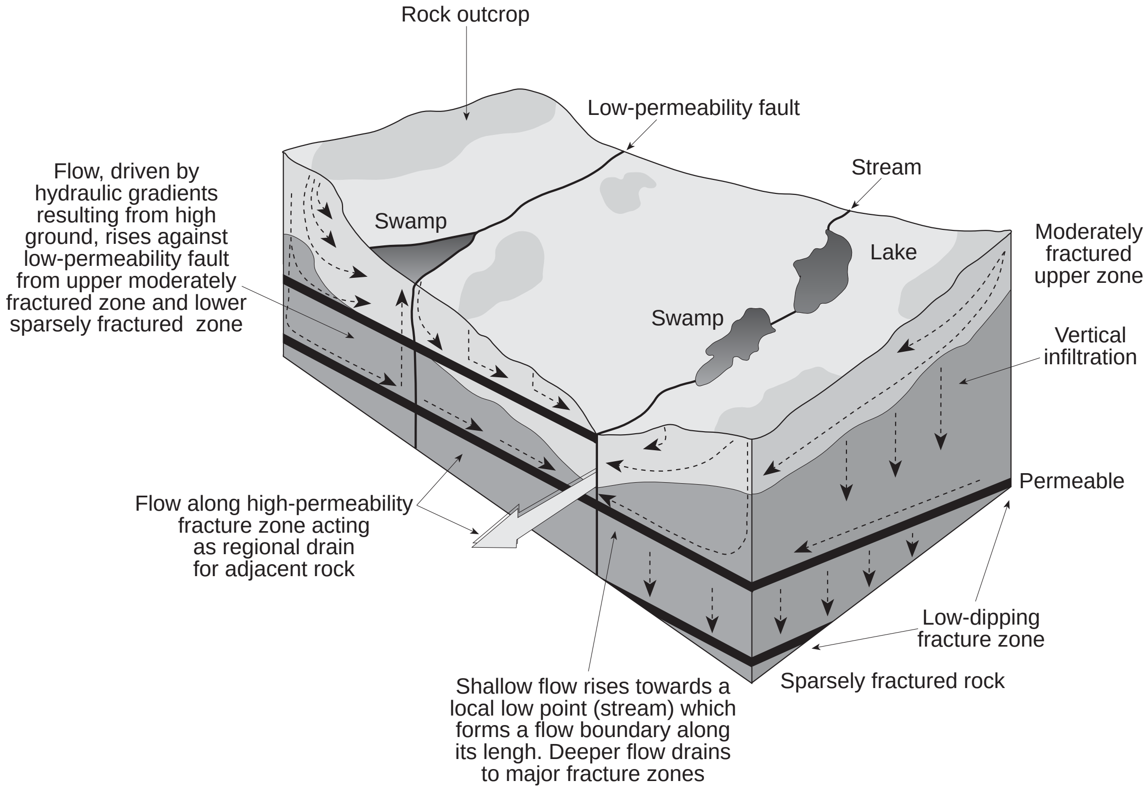

Figure 42. Conceptual model of groundwater flow in the Canadian Shield, showing the influence on groundwater flow of low-angle, transmissive fracture zones (from AECL, 1994).

SAFIR 2

- No direct use of palaeohydrogeology in assessment calculations
- Normal evolution scenario assumes future glaciations similar to last three, however geological barrier remains unchanged
- Severe glaciation scenario assumed to result in damage to geological barrier
- Effects of climate change studied as part of EVEREST, PHYMOL programmes
- Recommendations of review committee for SAFIR 2 related to future geochemical stability of host formation and adjacent aquifers (using a palaeohydrogeological approach)

Nirex-97

- Palaeohydrogeology not used directly in the assessment (a palaeohydrogeological approach was followed in some of the site investigations, but its use is not apparent in the assessment reports)
- Palaeohydrogeology used in developing an understanding of the evolution of the site and in the development of conceptual models related to:
  - The fracture system and its evolution, and the fracture mineralisation and its development over time
  - The saline interfaces and their past movements (Figure 43)
  - Hydraulic head profiles and the reasons for their current values.

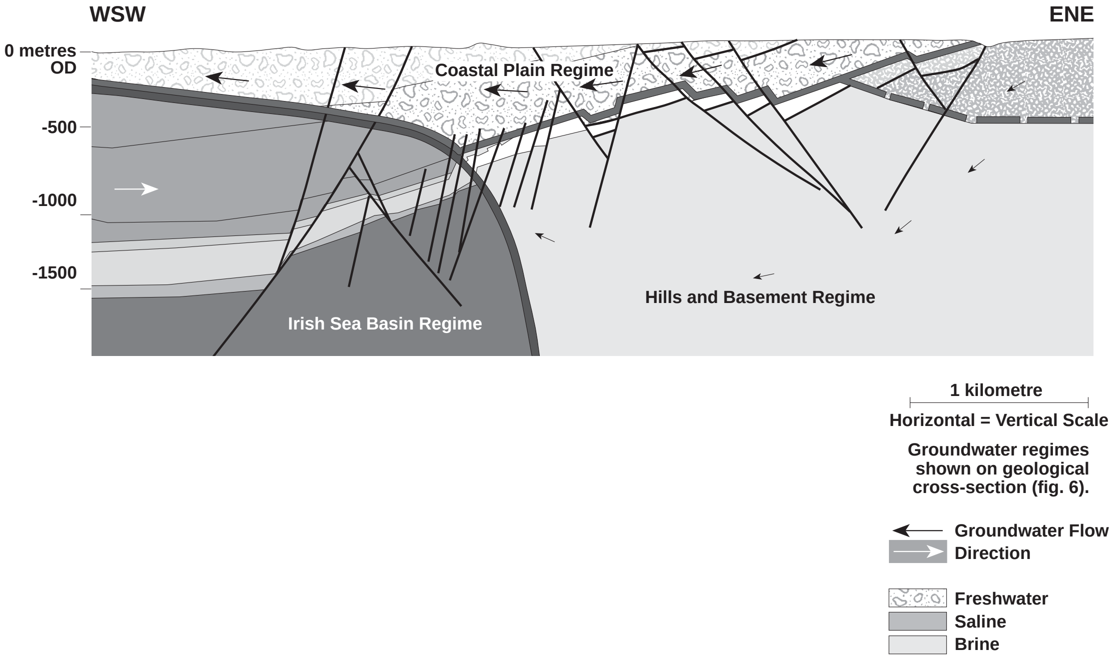

Figure 43. Conceptual model of the current groundwater flow system at Sellafield. Palaeohydrogeological studies at the site have demonstrated, for example, that the boundary between the fresh and saline water has moved only a few tens of metres at most.

Opalinus Clay assessment

- Assessment currently in preparation
- A palaeohydrogeological approach has used:
  - Isotope profiles into the Opalinus Clay from adjacent aquifers
  - Isotope profiles around faults in Benken borehole and in the Mt Terri URL
  - Isotope data to demonstrate consistency of time scales between groundwater movement and groundwater age.

9.4 CONCLUSIONS FROM THE ASSESSMENTS AND THE ASSOCIATED SITE INVESTIGATION AND R&D PROGRAMMES

The conclusions from the assessments and the associated site investigation and R&D programmes referred to above can be summarised as:

- Investigating the palaeohydrogeology of a site is probably the only sound basis on which to build a performance assessment.
- Its use should help in presenting results of both the site investigation and the associated performance or safety assessment in a comprehensive manner.
- Palaeohydrogeology is likely to provide some of the most useful information for building confidence in the results of comprehensive safety assessment.
- Palaeohydrogeology should form a focus around which the investigation of a disposal site is constructed.
- Site investigations need to place as much, and probably greater, emphasis on obtaining high quality hydrochemical and mineralogical data as on hydraulic measurements.
- There are likely to be many ambiguities and problems in data interpretation when considering the palaeohydrogeology of a site, but this is not a good reason for abandoning this approach.

9.5 REFERENCES

AECL, (1994) Disposal of Canada's nuclear fuel waste: The geosphere model for post-closure assessment. AECL Report AECL-10709, COG-93-9.

Chapman, N.A. and McEwen, T.J. (1993) The application of palaeohydrogeological information to repository performance assessment. In: Palaeohydrogeological methods and their applications. Proceedings of NEA Workshop, Paris 1992.

9.6 DISCUSSION

Q What do PA teams expect from palaeohydrogeology: numerical information, conceptual models?

A The main use of palaeohydrogeology is in a supportive role, building confidence in PA and safety assessments. PA can be used, for example, to constrain fluxes or to address specific 'worst-case' scenarios, such as having glacial meltwater at repository depth. It depends also on regulation in different countries; in some cases, numerical data are required.

o Too much reliance on numbers leads to extreme conservatism.

Q How do you take account of alternative concepts? Typically, the bulk of effort is put into one conceptual model, when there may be other equally valid ones.

A All possible conceptual models should be addressed seriously.

o By creating worst-case scenarios (the 'what-if' approach) there is a possibility of knock-on/coupled processes that may complicate matters unnecessarily. For example, if you make a worst-case assumption that glacial meltwater gets into your repository, you have to then consider its effects on bentonite backfill and so on. You risk ending up with over-complications and enormous conceptual uncertainty.

o There are three issues for discussion:

- What would an adequate geochemical characterisation be for palaeohydrogeology?

- Is it self-evident that we could explain the outcomes of palaeohydrogeology?

- Are all worst-case scenarios self-evident, or might we miss some?

o We ultimately need truth to develop a working model. How much palaeohydrogeological information is sufficient? When do you judge you have enough? This needs expert judgement, which in turn needs palaeohydrogeological information: a circular situation.

o Every model is based on expert judgement.

o We try to make models dependable by testing them; this doesn't mean that they are true.

o If you use a limited amount of data to produce a forward model, you produce a coherent result. Going the other way leads to many options. We have the consequences of process and we're trying to determine cause, which is extremely difficult.

o There is no way around the fact that we cannot investigate an undisturbed system. We face a paradox that the most easily obtained and best quality data come from shallow environments not relevant to deep disposal.

o Studying samples of actual groundwater (as preserved in fluid inclusions in late-stage minerals) can resolve many palaeohydrogeology problems and contribute significantly to confidence building. For example, good agreement between measured element ratios in fluid inclusions from Sellafield late-stage (ME9) calcite and those of present-day groundwaters suggests strongly that ME9 palaeo-groundwaters were not dramatically different from present-day groundwaters. What is critically required is the ability to date accurately the minerals and/or compositional zones that host the fluid inclusions.

10 Working Groups' Discussions of Issues

Participants formed three working groups, each having a diverse membership in terms of expertise and representation. The discussions of these working groups were chaired by Andrew Bowden, Tim McEwen and John Smellie respectively, who also acted as rapporteurs of the working group discussions to the final plenary session.

All three working groups considered topics associated with two inter-related issues: (i) How do hydrochemical & mineralogical data support confidence in deep repository stability? and (ii) How can that knowledge be applied more effectively in future safety cases?

The following reports include the rapporteurs' summaries of working groups and the points raised in subsequent discussion.

10.1 HOW DO HYDROCHEMICAL AND MINERALOGICAL DATA SUPPORT CONFIDENCE IN DEEP REPOSITORY STABILITY?

Six issues were addressed:

1. What is meant by 'stability' (in terms of groundwater rather than geotechnical or other forms of stability), how to define, describe and test it, and what are the favourable conditions for near field stability?

A distinction was drawn between *pre-repository (or natural) stability* and *post-repository stability*. A definition put forward for *stability* was 'the capacity of a site to respond to external influences with minimal impact to the environment of a deep repository'. This, however, requires further definition of the term 'minimal'.

There was broad agreement that the term 'stability' and the concept of a 'stable' site are not ideal because:

- it is unlikely that any natural system is steady-state
- it is sometimes more difficult to identify and measure parameters characteristic of a 'stable' system than one that is changing – in the sense that it is problematic to prove long-term stability (or guarantee future long-term stability) by short-term measurements.

Site-specific factors, such as geological/hydrogeological character, waste forms, and engineered barrier design lead to many difficulties in creating a specific definition of stability.

Several alternative terms were considered:

- *predictable/predictability* are not favoured because they imply that an unrealistically high degree of confidence can be applied to a particular future outcome; predictability implies that the chain of conditions leading to a particular outcome can be anticipated and that those conditions as well as the outcome can be quantified.
- *normal* is used by POSIVA, but raises the question 'what is normal?'
- *suitable/suitability*, where a suitable site might be one in which 'changes to the geological environment lie within certain limits and are acceptable' requires further definition of terms such as 'certain limits'.

No conclusion or consensus was reached. It was recognised that current disposal concepts in some countries (e.g. Sweden and Finland) seek to demonstrate that the engineered barrier will provide the required degree of isolation necessary to ensure safety. This might imply that it reduces the need for the rigorous definition and testing of geological stability that would be expected if the engineered barrier was not so robust.

SKB defines stability simply as ‘the capacity to maintain favourable conditions’. In SKB’s case, favourable conditions for the HLW repository concept are defined as groundwater conditions with the combination of the following properties:

- a reducing environment
- pH of 6 to 8
- TDS <100 g/L
- DOC <20 mg/L
- colloids <0.5 mg/L
- Ca+Mg >4 mg/L

These are based on safety assessment ‘suitability criteria’ suggesting that within these ranges the bentonite buffer will function optimally with no corrosion of the copper canister. However, these parameters and appropriate values are likely to depend to some extent on other site properties and engineered barrier characteristics and so perhaps they should not be regarded too rigidly.

2. Palaeohydrogeological research should seek to better understand what are the possible hydrodynamic aspects of far-field stability. For example, scenarios should consider the controls and nature of groundwater flows for a range of climatic conditions (glacial, periglacial, temperate, semi-arid). There is scope for investigating this issue by the use of present-day natural systems that represent different climate conditions. Such work should be supported by models that can be independently validated where possible.

Two phenomena were discussed to illustrate the issue: the development and decay of permafrost and the effect of glacial unloading.

The *development and decay of permafrost* may change groundwater salinity and may cause fracture dilation or sealing, thus altering groundwater flow patterns. Permafrost could also impact on bentonite stability by either freezing the clay or by raising groundwater salinity to a point that cation exchange alters the clay particle structure. The evidence for, and significance of, permafrost at depth was discussed.

Literature reports suggest that 50 metres is the maximum depth to which permafrost has extended in the UK. However, in Canada it has recently been shown that 500 metres thickness of permafrost can develop in 3,000 years; the estimate for the UK and elsewhere may therefore be an underestimate. There are no cases reported in the literature where the former presence of permafrost at depth (>100 m) in rock has been identified unambiguously. Contortions and freeze-thaw effects can develop in soft sediments, but no analogous marker phenomena have been identified in crystalline rocks. It was suggested that a record of the saline front moving ahead of a descending permafrost layer might be preserved in fluid inclusions developing in late-stage crystals. Evidence of freezing phenomena discernible in clay morphologies has been reported from Germany. A recommendation was made by one of the working groups to carry out more clay morphology studies to evaluate the effects of the presence of permafrost. The absence of evidence for permafrost at depth may mean that it never happened, or that it leaves no record. One view on permafrost (and other ‘events’ that leave no obvious record) is that if there is no direct evidence or record of an event, it probably did not occur and therefore is probably not relevant to Safety Assessment studies. However, this view is not supported by other scientists currently studying the effects of permafrost on groundwater systems, often because of theoretical or analogue arguments that still have to be evaluated[1].

[1] e.g. some members of the Permafrost project (GTK, POSIVA SKB Nirex, OPG) PERMA

The effect of glacial unloading on the physical integrity of the underlying rock mass was discussed briefly in another working group session. Effects include dilation/sealing of fractures (and consequent potential opening/closing of pathways between groundwater of different chemical characteristics), potential incursion of fresh glacial meltwater deep into bedrock, and upward movement of saline water from greater depth to the repository environment. The timeframe is important; the impact will likely be greater the more rapid the unloading. Several recommendations were made for further studies to investigate these effects:

- drill through an ice sheet to characterise the recharge water chemistry
- obtain more/better mineralogical information
- measure rock stress to investigate the physical integrity of bedrock in response to glacial unloading
- make further attempts to produce integrated hydrodynamic and hydrogeochemical models (forward modelling)
- perform scoping studies of the effects of glacial unloading on bedrock integrity (micro-tectonics), considering time-dependent changes

It was noted that the BENCHPAR project within the DECOVALEX programme has been investigating the effect of glacial loading through coupled hydro-mechanical (HM) modelling.

3. More information needs to be provided to define the relevant timescales for investigating mineralogical evidence and to assess how well are these covered by supporting hydro-geochemical evidence?

The meeting was informed that an NEA workshop in April 2002 focussed on timescales in PA. The report of that workshop[2] contains some very relevant discussions and observations regarding both the 'scientific approach' to handling the wide range of timescales for which safety cases must be adequate (and to dealing with the associated degrees of quantification and uncertainties) and also the 'public acceptance' approach to addressing people's concerns with the right order of priorities.

Research related to long-term safety of radwaste disposal often neglects the 'up to 1,000 years' timescale, but it is the timescale of greatest concern to most members of the public. Can we assume that systems are 'stable' within this timescale? Millennial scale climate variations i.e. events or 'spikes' which could impact on repository performance, are unlikely to be reflected in deep groundwater changes, which may respond only to a prolonged major climate change process. It was agreed generally that considering timescales such as 1,000 and 10,000 years might be a useful way to frame and communicate changes to groundwater systems. PADAMOT outcomes could be presented in a multi-timescale format.

A challenge in the use of information from secondary minerals in palaeohydrogeology is to obtain reliable age constraints for mineral growth and from these to calibrate the processes that are recorded in their geochemistry. Absolute dating is difficult to achieve in many cases with meaningful accuracy and precision. Attempts have been made to constrain the age of late-stage calcite at several of the study sites; however some results may not be sufficiently reliable. Modelling suggests the fastest that calcite crystals of their size and crystalline nature (several mm's) could precipitate is within 10's of years, but it is more likely to have taken 1,000's to 10,000's of years. Calcite is the most common and widespread late-stage mineral in the investigated European sites. Ideally we want to date parts of calcite crystals (e.g. centres and edges, as opposed to whole-crystal analysis) using the U-Th series method. However, very low U contents in late-stage calcites combined with the very young age of the crystals generally

[2] The Handling of Timescales in Assessing Post-closure Safety of Deep Geological Repositories. Proceedings of NEA Workshop, Paris, 16-18 April 2002, OECD-NEA.

prevent a successful outcome. It was suggested that AMS (accelerator mass spectrometry) might help in future U-series dating studies. The single most important constraint would be an age for initiation of late-stage mineralisation. This would provide a timeframe within which all late-stage processes could be set and evaluated. Successful absolute dating of events in past groundwater evolution may open up new questions; for example it may point to long gaps with no constraints on the transition from one groundwater system state to another.

4. It will be important to understand the different approaches that will be required for investigating crystalline rock and clay rock formations? What generic approaches (if any) can be used regardless of the environment? A statement to clarify this question would be a useful PADAMOT outcome.

Although it is more difficult to track and understand hydrogeological and mineralogical changes in low transmissivity bedrock (in which a repository is likely to be placed) than in high transmissivity settings, there is increasing evidence that similar approaches are applicable to a range of low transmissivity environments. Studies of the palaeohydrogeological evidence from the Sellafield area, which has a crystalline host rock at depth and a variable sedimentary cover, showed a direct correspondence between the fracture-filling mineral sequence and the diagenetic mineral sequence, both of which could be related to the groundwater changes. Secondary mineral studies should be equally applicable in fractured crystalline rock and in indurated clay rocks. Methodologies and objectives for groundwater studies have been different for the two types of host formation because the conceptual models for water flow and solute transport are somewhat different.

5. In addressing the question 'Are there new sampling/analytical techniques that may impact the science?' there was little knowledge of new analytical developments beyond those already proposed in PADAMOT.

We need to first define clearly what critical information is required, then to pursue appropriate funding and technological development. For example, an important requirement in secondary mineral studies is for improvements in absolute dating of the sequential stages of deposition of late-stage minerals (see discussion above). That may become possible through developments in TAMS (tandem accelerator mass spectrometry), in plasma source magnetic sector mass spectrometry, or in ion probe analysis. This is an active area of technological development where our research strategies need to take a realistic view of the developments in scientific capabilities and the new uncertainties that might be revealed.

Another area of relevant analytical development is the increasing power of techniques for examining mineral surfaces, i.e. the atomic layers that account for interactions with solutes and therefore relate microscopic mineral properties such as morphology and zoning with present and past groundwater compositions. X-ray absorption fine structure spectroscopy (XAFS) is the most promising of these new methods (but perhaps it should be noted that this is a very sophisticated method requiring a synchrotron light source of which there are very few).

This issue is discussed in Section 7 of this report.

6. The question was raised 'What are the potentially significant areas of uncertainty?' Consensus at the meeting was that all aspects of palaeohydrogeological investigation were subject to varying degrees of uncertainty. The more pressing question was 'How do we deal with uncertainty?'

The key points made in discussions covering this topic were:

- Proxy parameters require interpretation and are therefore inherently uncertain. Parameters derived from mineral attributes and groundwater compositions all have associated uncertainties in measurements and therefore in the models that they inform; perhaps we should be more critical of these data. On the one hand using data to build confidence is appealing, but on the other it is not systematic about demonstrating safety.

- Can we ever have enough information to construct scenarios that will slot into safety cases and improve upon the conservative approach of resorting to worst-case scenarios? If not, then palaeohydrogeological information can be used only for 'building confidence' by improving and illustrating the general understanding.
- It would be helpful to have a workshop to test alternative interpretations for generally accepted models. However, the question of how to compare equally valid alternative models presents a significant difficulty. For example, the relative sensitivities of different parameters in PA models will determine how important each dataset is.

During one of the three working group sessions, individuals in the group were asked what they considered to be the main palaeohydrogeology issues of interest to them or the organisation they represented. The main generic issues were:

- How to obtain proof of the past influx and 3-dimensional spatial distribution of groundwaters of differing character (redox, salinity, temperature). One of the main objectives of the PADAMOT project is to correlate stable isotope signatures in zoned late-stage minerals to see how groundwater 'signals' have moved through the system, and from this evaluate drivers/causes and effects.
- How to transfer palaeohydrogeology results into decisions regarding repository siting. Similarly, can palaeohydrogeology be used to improve site selection criteria, and can it be used to improve/inform the decision-making process?
- Erecting site-specific timeframes through absolute dating of different parts of the system.
- Assessing the nature and timing of the response of a system after an 'event' and whether pre-perturbation conditions are recovered.

10.2 HOW CAN THAT KNOWLEDGE BE APPLIED MORE EFFECTIVELY IN FUTURE SAFETY CASES?

There are four areas where the workshop participants thought that knowledge derived from palaeohydrogeological investigations could be more effectively applied to support a safety case for a deep geological repository:

1. Transfer of data and knowledge to Performance Assessment

More effective transfer of data and knowledge from site investigation personnel to performance assessment modellers was required. This means establishing close and effective working relationships. It is a recognised problem for many organisations, but one for which there does not appear to be a universally successful structure.

It was suggested that knowledge from these studies would be applied more effectively if it were being used to answer specific questions that were posed by the safety case. To achieve this, the safety case must develop a number of alternative scenarios. However, it was recognised that such development should be an iterative endeavour that relies on site data at the outset and throughout the process.

Most PA studies have had inadequate communication and understanding between PA modellers and the researchers providing palaeohydrogeology data. This situation can be attributed to the contrast between the parameter requirements of PA and the attributes that are typically measured in palaeohydrogeology. There was general agreement that this situation needs to be improved. Two requirements were identified: (i) having a common set of objectives at the outset is crucial to the two sides working together effectively; (ii) each other's capabilities need to be understood at the outset.

An essential distinction has to be made when evidence is being interpreted for use in a safety case. This arises from the likely situation where evidence remains incomplete and where interpretation contains areas of significant uncertainty. Consistency and rigour have to be exercised in dealing with the absence of evidence for a particular evolutionary condition of a repository site in the past, for example one that might challenge the stability and suitability of a site. When does the absence of evidence positively indicate that the evolutionary condition has never occurred, and when does it indicate a level of uncertainty that affects the safety case?

2. Performance Assessment (PA) and demonstration of general understanding of a site: where should the emphasis lie?

Uncertainty in all its forms needs to be addressed in safety assessments. It was felt by some participants that there would be merit in investigating further the use of 'Evidence-based Reasoning such as proposed by A. Bowden (see Section 8 for further information).

The key points were:

- The question of whether a site investigation programme should include a large palaeohydrogeology component depends on site-specific issues. For example, if dating suggests that all groundwater at a site is old, that will influence all modelling and safety cases, whereas if it is young these will be different. So, it is important that safety assessment includes palaeohydrogeology, but the way in which it is used needs to be site-specific.
- Very little palaeohydrogeology would be done if we were restricted to data for direct input to PA, but if we want to understand site evolution we need a significant body of palaeohydrogeological information to support conceptualisation of the system and the processes that are operating.
- Palaeohydrogeology is inherently valuable for testing our confidence in PA models; the crucial thing we need is goals and focus. It may be better to use palaeohydrogeology to answer specific questions of relevance to PA than to try to develop a complete picture of groundwater evolution. A balanced approach would be to aim for both measured properties to inform PA and conceptual understanding through palaeohydrogeology.
- An alternative (or perhaps complementary?) view is that research should address the issues that are identified as being of greatest concern to non-specialist members of the public (stakeholders).

3. Communication to the wider scientific community and public – is the geochemical evidence for stability a valuable area for communication?

Issues concerning communication to the wider scientific community and public were discussed. Outcomes and issues being investigated as part of the EC RISCOM project should be taken on board and considered as a means to better describe results from palaeohydrogeological research and how they impact on a safety case.

The key points made in discussions covering this topic were:

- Presentation of results in a understandable form to the public should form an important part of palaeohydrogeology.
- A quantitative model is not always required; a qualitative model may be sufficient for public communication. SKB uses 'snapshot' models covering the last and next 100,000 years as an effective way of communicating system change over geological time. A combination of visualisation and animation is effective.
- Information from natural analogue studies should be included. These studies are not used to maximum benefit in PA, but people like to see how processes operate in nature.
- All sides of the argument should be presented, i.e. using palaeohydrogeology to identify difficult scenarios and shortcomings in present approaches to PA, not just 'building confidence' by rubber-stamping the current position.

- Getting across a clear idea of timescales is crucial. The 1,000-year timescale is crucial in painting a word-picture for the general public. In the NEA timescales meeting earlier this year, 1,000 years was considered the maximum that the public considers to be ‘a long time’. Beyond that, they are not interested and/or don’t believe you.
- Two-way communication between scientists and the public is important. We should produce a drill-down/layered information source for the public, so they can see the story ‘evolve’ and engage through discussion at a level with which they feel comfortable. There is a place for illustrating how the environment changes; for example TV presentations of popular science. Palaeohydrogeology has a role to play in this ‘picture-painting’.
- ‘Safety case’ and ‘confidence building’ are not the same thing, they are parallel issues. There is a general mistrust of science and scientists because the public tends to see scientific debate as arguments with no agreement and a great deal of incomprehensible specialist jargon. A long process of building public confidence in scientific methodology and science in general is required. The key is being open, and providing lots of publicly accessible information consistently.
- When the public asks ‘Is it safe?’ we need to be able to demonstrate it. The questions behind that are ‘Is it believable?’, and ‘Are the people presenting the science believable?’ There needs to be a clear, comprehensible argument.
- The long-term future is much less tractable as a scientific concept than the long-term past. We should take events in the past and use them as a measure of the rate at which processes work; then we can say, for example, on a 1,000-year scale this process is very slow/insignificant.
- We need to demonstrate the contrast between degrees of confidence in near-surface and deep stability over the 1,000 year timescale, and thereby illustrate the rationale for deep disposal. For example, the relative sluggishness in the deep environment as opposed to the shallow environment in terms of response to events.
- Outside the geochemical community there is very little understanding of the principles/concepts that underlie groundwater evolution/processes, so any success in getting the message over is worthwhile.

11 Summary and Conclusions

Paul Degnan and Adrian Bath

The presentations of recent and ongoing EC-funded palaeohydrogeology projects are summed up in the following paragraphs:

The EQUIP Project (which was the forerunner to PADAMOT) developed geochemical and mineralogical techniques for palaeohydrogeological study. These techniques involve careful sampling and some 'state of the art' mineralogical analyses that pose some challenges in transforming the proxy data into relevant palaeohydrogeological and geochemical information. Nevertheless, this type of geochemical approach and the complementary studies on water samples are really the only source of primary data for palaeohydrogeology in deep groundwaters. Isotopic and organic geochemical techniques for obtaining information on past surface water and shallow groundwater regimes corresponding to changing climate conditions in semi-arid southern Europe were also demonstrated in EQUIP. Palaeohydrogeology methods are potentially attractive for communicating long-term safety issues and the corresponding state of understanding to both the geoscience community and to the wider public but, just as with natural analogues, the rationale and the scientific uncertainties need to be explained clearly as a foundation for confidence building.

The PHYMOL project has been an exemplary study in the use of groundwater isotopic data for palaeohydrogeology, specifically for calibrating a hydrodynamic model. Independent palaeoclimate information was used to estimate time-dependent boundaries and initial conditions for a hydrodynamic model which then transports the isotopes in a forward model to reconstruct present-day conditions. Comparison of observed with modelled isotope data is a clear test of the model which has then been used for a PA calculation of radionuclide concentrations in aquifers either side of the clay host formation.

The Palmottu site in Finland is a natural analogue (U-mineralisation) site for which intensive study has added substantial knowledge about hydrogeology and palaeohydrogeology of Fennoscandian crystalline rocks. Groundwater chemistry and isotopes have been supplemented by fracture mineral studies from which U-Th dates are correlated with interglacials, i.e. mineral growth during wet periods. As with EQUIP there is a significant amount of interpretative uncertainty, but this is still a landmark study that exemplifies both the conceptual and quantitative importance for groundwater models of incorporating geochemical data and palaeohydrogeological interpretation.

Numerical models were developed in PAGEPA and are being further developed in BENCHPAR to simulate the processes in groundwaters overlain by, or adjacent to, ice sheets. This work is the scientific backbone of palaeohydrogeology for areas subject to glacial processes. Amongst the phenomena modelled are infiltration of glacial meltwaters, discharge through ice tunnels or eskers, permafrost, and overpressuring due to loading. Hydraulic and geochemical processes have been modelled. It is clear that water movement must be modelled in 3-dimensions to represent the geometrical heterogeneity of these phenomena. As modelling gets more complex, so the requirement for modern analogue and interpreted palaeohydrogeological evidence gets more demanding.

There are three strands to the PADAMOT project, which is the successor to EQUIP. The first strand is the continuing development of the analytical techniques which are revealing the geochemical archive of past groundwater conditions, which is contained in secondary minerals. In particular, new instrumentation is being used which offers the possibility of calibrated microanalyses of isotopic compositions in individual growth zones of secondary minerals. The second strand is the construction of a database which will collect together a wide range of data types in appropriate formats for use in interpretative modelling. The latter objective will serve to gather together many disparate datasets, which are emerging from site investigations and natural analogue studies and which will become increasingly valuable for testing otherwise uncalibrated numerical models. The third strand is the use of numerical models to simulate groundwater flow and transport in order to test hypotheses generated on the basis of palaeohydrogeological information being provided by the analytical studies.

Following these 'project-oriented' presentations, our expert rapporteurs presented their commentaries on the general approach to, and value of, palaeohydrogeology as starting points for discussion. Two particular aspects were considered: (i) the data sets from site investigation and the lines of palaeohydrogeological evidence that constitute a potential input to Performance Assessment, and (ii) the extent to which such data and lines of evidence actually have been used in existing PA's.

The conclusion drawn by reviewing the topic from both aspects is that this type of evidence has so far been used very variably in site characterisations and PA's, and has not really achieved the potential value that had been expected and promised. However the breadth of the topic, and the conceptual overlaps with many other scientific aspects of site characterisation, mean that its value is often implicit in the way that models of groundwater systems are constructed and justified, even if it is not explicit and 'up front'. A number of reasons have been put forward for this, but the most prominent ones seem to be the ambiguities and problems of extracting information that is quantitatively usable by PA and especially the often-subjective way that uncertainties are handled in that process. Nevertheless, the view was put forward that palaeohydrogeology provides the only sound scientific basis on which to prioritise data collection and to build a safety case. Accepting this view leads to the recommendation that more emphasis and effort needs to go into improving the ways that we tackle interpretation, uncertainties and communication of results, and also improving the balance between short and long timescales in our scientific strategy.

Discussions among the working groups were focused on how the present states of expertise and knowledge in palaeohydrogeology could be taken forward to improve future value of this approach. Two directions of discussion were tackled: (i) how scientific methods, data and uncertainties relate to the concept of groundwater stability, and (ii) how the knowledge gained from these studies can used effectively in PA. There was apprehension about trying to be prescriptive about a definition for stability when there are so many site-specific factors and also variability in the dependence of multi-barrier containment on geosphere stability for different repository designs, locations and safety cases. Whilst there are many steps that need to be taken (and are already being taken) to improve the interaction and iteration of data and safety implications between site characterisation staff and PA teams, it was also recognised that we should ensure that wider public concerns about safety scenarios and timescales are given adequate attention. Palaeohydrogeology is one of the tools to address those concerns, but a more consistent and rigourous approach to evaluating evidence coupled with more innovative methods of communicating concepts, implications and uncertainties will be required.

A few of the suggestions that arose in discussion include:

- More focus on potential impacts (or positive evidence of negligible impacts) of permafrost in northern and central Europe;
- Field investigations of processes (especially ones that are time-dependent and/or localised) that are associated with present-day ice sheets;
- Further studies to investigate the palaeohydrogeological significance of isotopic and organic geochemical proxy indicators in sedimentary sequences representing the Quaternary pluvial/arid climate cycles of southern Europe;

- Wider use of a range of timescales, e.g. 1,000 and 10,000 years, to focus research aims and to communicate safety-relevant understanding;
- Continuing development and exploitation of analytical techniques to improve primary data in areas such as the chronology and compositional variations in secondary minerals;
- Interactions between PA teams and palaeohydrogeology researchers that are focused on specific issues, for example by a structured process of constructing and testing ranges of alternative models and scenarios;
- Adopting a broad approach to discussing groundwater 'stability', rather than trying to be prescriptive, so that the many relevant aspects of this issue and the associated uncertainties can be considered realistically.

Finally, it is recommended that there could be a more strategic use of palaeohydrogeological research in conjunction with other research initiatives. For example, it is now more widely recognised that palaeohydrogeological studies are only one component in a 'timeline' of understanding (Figure 44).

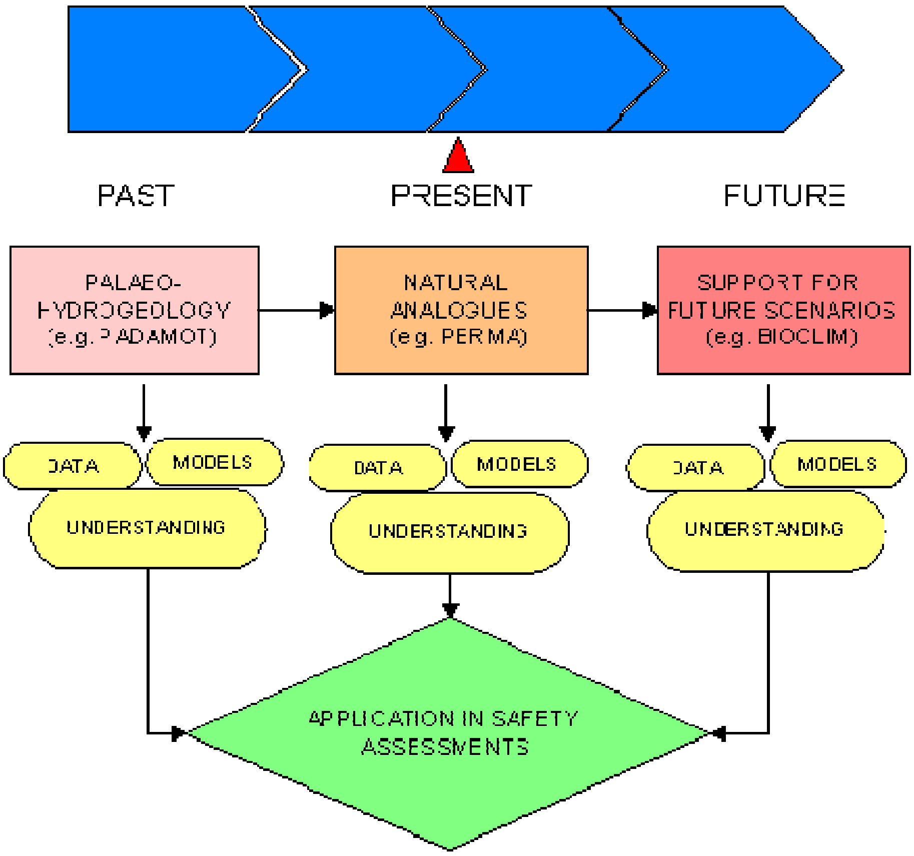

Figure 44. Palaeohydrogeology as part of a timeline of investigations to support safety assessments.

Palaeohydrogeology by its nature concentrates, through geochemical and mineralogical analyses, on understanding past groundwater movements and hydrochemical evolution. Many studies of natural analogue sites, on the other hand, investigate processes that are currently operative. Clearly, even at such sites knowledge of the past conditions is required in order to properly understand present day conditions, but the emphasis in many natural analogue studies may be different from explicit palaeohydrogeological research. With knowledge of past and present conditions derived from palaeohydrogeology and natural analogues, we have the potential to extrapolate our understanding into the future. Because of uncertainty, such projections can never scientifically be called predictions, but they do allow us to construct defensible scenarios. Climate change is likely to be one of the major factors in disrupting future surface and near-surface environments. Consequently a major effort is being made in the EC 5th Framework

BIOCLIM project to construct defensible scenarios of future climate change, over the timescales of interest for repository safety assessments. That project is also investigating the potential radiological impacts of transitions between climate states, as opposed to what radiological impacts might be expected under different, but steady state, climate conditions. Outcomes from BIOCLIM and other studies that are based on palaeohydrogeological understanding and information derived from natural analogues should help constrain scenarios of future environmental change on the surface and in the near surface. As such they will provide appropriate boundary conditions for models of the deep geosphere, where the effects are expected to attenuate with depth.

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