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# Long-term maintenance of reducing conditions in a spent nuclear fuel repository

## A re-examination of critical factors

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*Keywords:* redox, nuclear waste, glaciation, hydrogeology

# Abstract


Penetration of oxidising groundwaters to depths of ~500 m in a permeable bedrock, over a glacial/interglacial cycle, may compromise the stability of a spent nuclear fuel repository and cause the release and migration towards the surface of actinides and associated fission products. This report examines the potential for the penetration of oxygen ($O_2$) to depths of ~500 m in a fractured crystalline rock environment, typical of the Fennoscandian Shield.

Previous studies performed for the Swedish program of nuclear waste disposal (principally the SITE-94 safety assessment) have indicated that $O_2$ might reach repository depths during a deglaciation when melt-water from the base of an ice sheet could enter the bedrock, driven by strong hydraulic gradients. This report re-examines aspects of this scenario and finds that:

1. The capacity of flow-path minerals to scavenge $O_2$ from recharging groundwater may be lower than expected due to a previously unrecognised depletion of $Fe^{II}$-bearing minerals in the active flow-paths in a fractured crystalline rock.

2. Assumptions in the SITE-94 assessment, such as the use of a continental-scale flow model, the lack of structural controls on groundwater flow, a preferred horizontal permeability, and the use of permeabilities to depths of 10 km that are up to two orders of magnitude greater than comparable environments, are disproportionately simplistic and represent an extremely conservative case.

3. Assumptions of a thin, discontinuous permafrost, a warm-based ice sheet, and high-$O_2$ content melt-water at the repository site are unrealistic and overly conservative.

A more realistic scenario, which includes a greater influence of permafrost, a cold-based ice sheet, lower bedrock permeabilities and a more-limited, regional-scale flow-path, is recommended as being more appropriate for use in the safety assessment. Under this revised scenario, it is believed that $O_2$ will not penetrate to repository depths over the next 100,000 years, thereby providing greater assurance of the stability of the repository and demonstrating the ability of the geosphere to act as a barrier to migration of nuclear waste.

# Executive Summary


The Swedish Nuclear Fuel and Waste Management Company (SKB) has the responsibility for disposing of spent nuclear fuel at depth in rocks of the Fennoscandian Shield and for demonstrating the safety of this disposal method for a period of at least the next 100,000 years. For the last 10 years, the geosciences program of SKB has centred on the Äspö Hard Rock Laboratory in southeastern Sweden and consists mainly of engineering, rock-mechanical, hydrogeological and hydrogeochemical testing programs.

In 1992, the regulatory agency SKI (the Swedish Nuclear Power Inspectorate) performed a safety assessment study, known as SITE-94, to review the SKB research and development programs. In the report of this study (SITE-94, 1996), the possibility was identified that oxidising meltwaters, driven by high hydraulic heads from a continental ice sheet at the end of the next glaciation, might penetrate to repository depths and so compromise the integrity and stability of the spent fuel.

Several international studies have subsequently been performed to examine this issue in more detail. The study by Guimera et al. (1998) considered a scenario in which oxygen in recharging groundwater was consumed mainly by $Fe^{II}$-bearing chlorite in fractures and by biotite and pyrite in the rock matrix. They used the flow model developed in SITE-94 and its assumptions of glacial and hydrogeological conditions. These included recharge from a warm-based ice sheet, a bedrock permeability of $10^{-16}$ m/s, the dominance of fracture-flow for recharge to repository depths, the absence of permafrost and the use of chlorite as the main scavenger of $O_2$ in fracture flow.

The results of mass transport modelling indicated that in most situations $O_2$ would be scavenged and not penetrate to repository depths. However, Guimera et al. (1998) pointed out that in the situation when fast flow paths become the main transport pathway, oxidising conditions could reach the repository.

The report presented here examines some of the scenarios and critical assumptions underlying the models including:

1. redox conditions and disruptive scenarios,
2. climate change, including the dynamics and characteristics of a continental ice sheet during retreat and deglaciation, and possible scenarios for recharge during a cycle of glaciation,
3. basic assumptions of the groundwater flow models used in Guimera et al. (1998), the SITE-94 assessment and related reports,
4. oxygen content of meltwaters,
5. sensitivity of the model results to variations in input parameters and
6. supporting/contradictory geochemical evidence.

As indicated by Guimera et al. (1998), the penetration of $O_2$ to repository depths is found to be controlled largely by reactions with $Fe^{II}$-bearing minerals (principally

chlorite) in fracture pathways. However, the abundance of chlorite assumed to be present in fracture pathways (35%) may be too high because this value appears to have been determined from data for both low- and high-permeability fractures in the rock at Äspö. In the major permeable zones, chlorite content is found to be significantly lower. Thus, $O_2$ mobility may be greater in the bedrock than previously estimated.

The SITE-94 assessment assumes that permafrost is discontinuous, thin, and melts soon after ice coverage. Therefore, permafrost used in the model does not form an effective barrier to recharge and groundwater flow continues largely without restriction for most of the glacial cycle. Several researchers, however, have predicted that permafrost will be extensive and deeply penetrating (to at least 300 m) and this is believed to be a more realistic scenario.

A warm-based ice sheet has been assumed for the SITE-94 study and related modelling, for much of the period when ice covers the Äspö area. The assumption of rapid attainment of warm-based ice conditions after glaciation of an area effectively allows groundwater recharge to continue throughout most of the glaciation. This is despite abundant evidence (both previously and recently published) which indicates that past ice sheets have been largely cold-based and only melted after considerable time had passed and a substantial thickness (> 2 km) had been accumulated to attain the pressure melting point of ice (~ -2 °C).

The flow model and the central scenario used in SITE-94 and related models is found to be disproportionately simplistic, overly conservative and unrealistic in several of its assumptions. The use of a high and constant permeability for the bedrock, the choice of an immensely long potential flow-path (~1500 km), the lack of structural boundaries on groundwater flow, and the use of a permeability anisotropy that allows preferential horizontal instead of vertical flow, all combine to force the model to predict that groundwater will circulate deep into the bedrock, on a continental scale of distance, under almost any climatic regime, for long periods of time.

Evidence from recent literature indicates that groundwater flow distances much greater than ~ 10 km are unlikely to occur in fractured crystalline rock in a low topographic environment (comparable to that at Äspö) because fracture zones tend to act as major conduits and flow boundaries. Any regional flow that does exist will not be significant over the time period considered for a nuclear fuel waste repository. In addition, the permeability of fractured rock is found to decrease in studies in Canada, Sweden and Finland to less than $10^{-17}$ $m^2$ at repository depths (~ 500 m). The SITE-94 central scenario assumes permeabilities that are up to two orders of magnitude greater than this and that these persist without reduction to 10 km depth.

Hydrostatic heads have been assumed to be high in the SITE-94 study and related modelling due to the combined assumptions of the availability of basal melt-water and the load of ice. Recent literature suggests that heads are high in a melting glacier but they do not exceed the height of the ice surface (ie. they are not driven by distant upstream pressures). The more important parameter for determining if heads are causing groundwater recharge is hydraulic gradient, and this is only likely to be significant in areas where the ice sheet surface is steeply sloping (such as near the margin of an ice sheet).

Recent studies have indicated that $O_2$ may be strongly depleted in dissolved gases in basal ice and, provided that contact with the atmosphere does not occur, much lower $O_2$ concentrations than the value (45 ppm) assumed by Guimera et al. (1998) might be expected in melt-water recharging the bedrock.

Combining the large-scale SITE-94 flow model with the assumptions of thin or non-existent permafrost, a warm-based ice sheet, high hydraulic heads and recharge containing abundant $O_2$, inevitably predicts that, over much of the next 100,000 years, oxygenated water will reach repository depths and thus jeopardise the stability of the spent fuel.

The investigations performed here present an opposing view to that of SITE-94. It is believed that a lower permeability geosphere with discontinuous flow-paths is a more appropriate model. Glacial cycles over the next 100,000 years will not significantly influence the safety of the repository because permafrost will restrict groundwater recharge for much of the time and flow will probably only increase for a short duration during deglaciation, as the ice margin retreats over the repository site. In addition, the $O_2$ content of the recharging melt-water is expected to be lower than used in the transport model. Reduced minerals in the fracture flow-path will scavenge any $O_2$ that enters the system despite the fact that the capacity for removal will likely be lower because the abundance of these minerals is believed to be less than previously estimated in SITE-94.

Under this revised scenario, it is believed that $O_2$ will not penetrate to repository depths over the next 100,000 years, thereby providing greater assurance of the stability of the repository and demonstrating the ability of the geosphere to act as a barrier to migration of nuclear waste.

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# 1 Introduction

The Swedish Nuclear Fuel and Waste Management Company (SKB) has the responsibility for disposing of spent nuclear fuel at depth in rocks of the Fennoscandian Shield and for demonstrating the safety of this disposal method for a period of at least the next 100,000 years. The geosciences program of SKB began in 1977 and several sites in Sweden have been investigated to determine geological properties of rocks of the Fennoscandian Shield and provide data for safety assessment models. This work culminated in the 1990's with the development of the Äspö Hard Rock Laboratory (HRL), located ~300 km south of Stockholm (Fig. 1-1), and the numerous engineering, rock-mechanical, hydrogeological and hydrogeochemical testing programs that were initiated in the HRL.

In 1992, the Swedish regulatory agency SKI (the Swedish Nuclear Power Inspectorate) sponsored an independent performance assessment (PA) study, known as SITE-94, by several international organisations and contractors, in order to provide the SKI with the understanding necessary to review the SKB research and development programs and to be able to review site licence applications, as required by Swedish law. In particular, the SITE-94 project examined data and models for site evaluation, PA methodology, and canister integrity, and reported on these aspects in a comprehensive, two-volume document (SITE-94, 1996). A number of areas of concern were identified that require further investigation. Chief among these, in the area of geochemistry, was the possibility that oxidising meltwaters, driven by high hydraulic heads, from a continental ice sheet at the end of the next glaciation, might penetrate to repository depths and so compromise the integrity and stability of the spent fuel.

Since the SITE-94 review was published, several studies have commenced which aim to provide information useful in addressing this issue, including : 1) the EQUIP (Evidence from Quaternary Infils for Palaeohydrogeology) project, which uses the chemical and isotopic contents of fracture-filling minerals to understand the paleohydrology of a site, 2) the PAGEPA (Palaeohydrogeology and Geoforecasting for Performance Assessment) project and 3) a detailed examination of the scenario of redox front migration by QuantiSci, Barcelona (Guimera et al. 1998).

The study by Guimera et al. (1998) is most relevant for the assessment of the importance of the SITE-94 finding that oxidising conditions may penetrate to repository depth. Guimera et al. (1998) consider a scenario in which oxygen in recharging groundwater is consumed mainly by the $Fe^{II}$-bearing chlorite in fractures (for fracture flow) and biotite and pyrite in the rock matrix (for matrix flow). Both equilibrium and kinetic reactions between groundwater and minerals are examined to determine the rate of migration of the redox front which is largely governed by the concentration of $O_2$ in the recharge. The kinetic reactions were found to be more realistic and represented the more conservative case because recharge under high flow

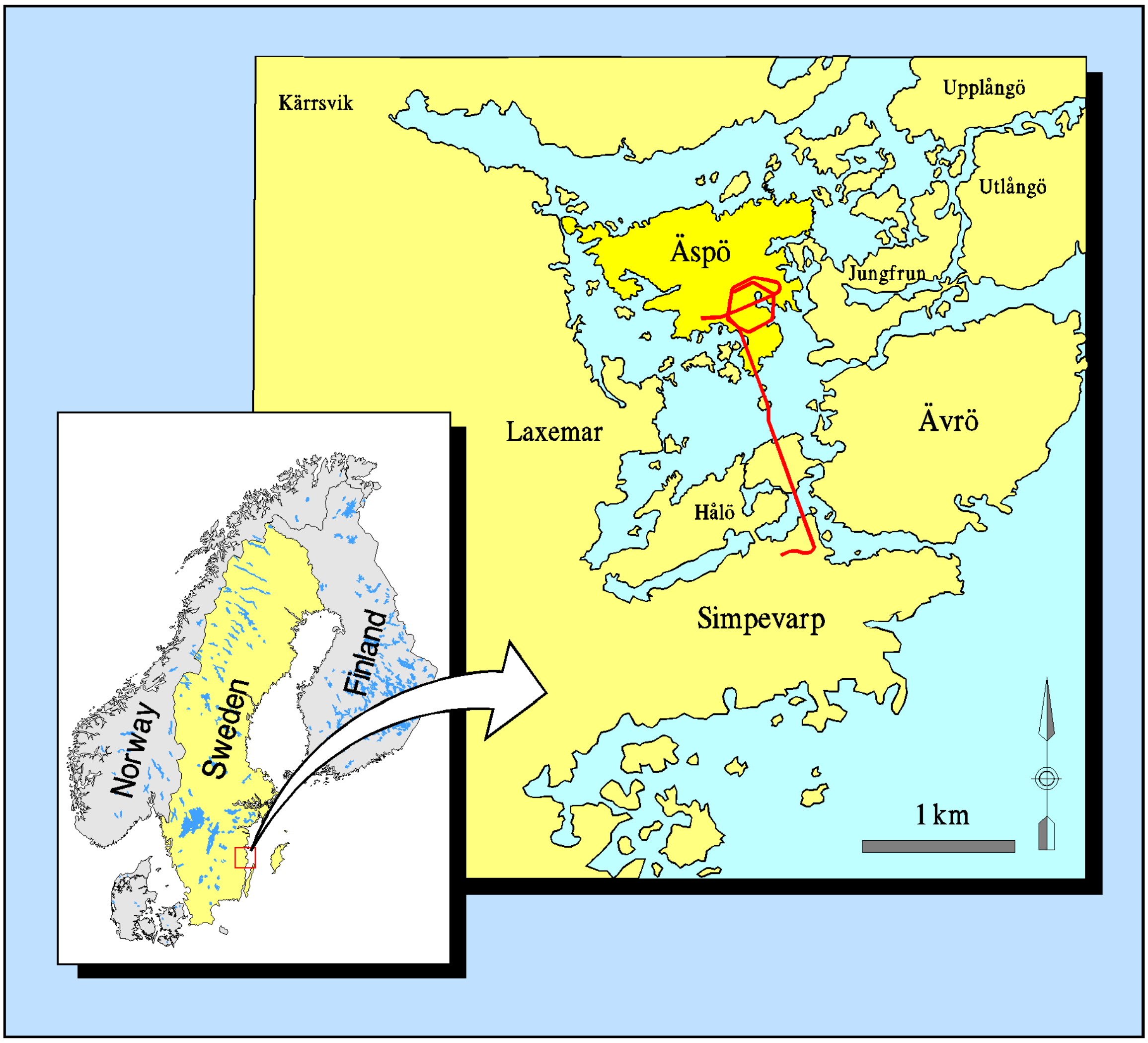



Figure 1-1 *Location map showing the island of Äspö in southeastern Sweden and the plan outline of the Äspö Hard Rock laboratory.*

conditions was believed to be rapid and $O_2$ would have the opportunity to penetrate most deeply before being consumed. Guimera et al. (1998) then applied a mass transport model to the kinetic reaction model assuming certain conditions at recharge and in the bedrock as defined in SITE-94 (1996). These include 1) recharge from a warm-based ice sheet, 2) rock permeability of $10^{-16}$ m/s, 3) dominance of fracture-flow for recharge to repository depths, 4) absence of permafrost and 5) chlorite as the main scavenger of $O_2$ in fracture flow.

The results of mass transport modelling indicated that in most situations $O_2$ would be scavenged and not penetrate to repository depths. The thorough analysis of Guimera et al. (1998) removes much of the concern raised in the SITE-94 study about the stability of the repository. However, Guimera et al. (1998) pointed out that in the situation when fast flow paths become the main transport pathway, oxidising conditions could reach the repository.

This report summarizes the findings of Guimera et al. (1998) and examines some of the critical assumptions underlying the models used in assessing redox front migration. The assumptions used in hydrogeological and hydrogeochemical models used in SITE-94 are also examined and the following aspects are considered in detail:

1. redox conditions and disruptive scenarios,
2. climate change, including the dynamics and characteristics of a continental ice sheet during retreat and deglaciation, and possible scenarios for recharge during a cycle of glaciation,
3. basic assumptions of the groundwater flow models used in Guimera et al. (1998), the SITE-94 assessment and related reports,
4. the oxygen content of meltwaters,
5. sensitivity of the model results to variations in input parameters and
6. supporting/contradictory geochemical evidence.

Use is made of recent publications in glaciology and evidence from field studies in shield environments both in Sweden and Canada. The importance of the oxygenated recharge issue is then determined and recommendations are made.

# 2 Redox Controls

## 2.1 Redox Potential in Groundwater

Spent nuclear fuel consists largely of $UO_2$ which is very insoluble under a reducing environment. The oxidation potential of fluids interacting with repository materials and the spent fuel, therefore, is undoubtedly the most important geochemical criterion for determining whether the contents of a repository will remain isolated from the human environment for $10^4$ to $10^6$ years into the future.

Groundwaters deep in crystalline rocks are expected to have relatively low redox potentials because of the presence of reduced Fe, S and Mn species in the groundwater and surrounding rock which scavenge dissolved $O_2$ that enters in recharge from the surface. Studies indicate typical redox potentials (Eh) of -100 to -300 mV for the range pH 7 to 9 (Nordstrom 1986, Wikberg 1986, 1988) although somewhat higher values (+100 to -200 mV) have been found in the granitic rock of the Lac du Bonnet Batholith (Gascoyne 1997).

It is well-known that significant uncertainties exist regarding the measurement and interpretation of redox potentials in natural systems. They arise because of operational difficulties of making Eh measurements at remote locations in deep boreholes, the probability that the Eh is a mixed potential arising from the presence of more than one redox couple in the groundwater, and the lack of redox equilibrium at the low ambient temperatures of the groundwater. Some confidence in the measured Eh data can be gained, however, when used in conjunction with measurements of S and Fe species and if dissolved $O_2$ is found to be absent.

## 2.2 Redox Control by Fracture Minerals and Rock Matrix

Intimately related to the importance of redox conditions in preventing spent fuel dissolution and radionuclide migration, is the importance of fracture-filling and host rock minerals in maintaining reducing conditions and in providing a suitable surface for sorption of radionuclides once released from a repository. Maintenance of reducing conditions is best ensured by fracture flow paths (and host rock if diffusion through the matrix is to be significant) that are rich in $Fe^{II}$-containing minerals, such as pyrite, magnetite, chlorite, hornblende and biotite. The ability of these minerals to consume oxygen is indicated by the following reactions:

$$\underset{\text{(pyrite)}}{FeS_2} + 6\,O_2 + 5\,H^+ \rightarrow Fe(OH)_3 + H_2O + 2\,SO_4^{2-} \qquad 2\text{-}1$$

$$\underset{\text{(magnetite)}}{Fe_3O_4} + 5/2\,O_2 + 9\,H^+ \rightarrow 3\,Fe(OH)_3 \qquad 2\text{-}2$$

$$KFe^{II}_3AlSi_3O_{10}(OH)_2 + H^+ + \tfrac{3}{4} O_2 \rightarrow Al_2Si_2O_5(OH)_4 + 2 H_4SiO_4 + K^+ + 3 Fe(OH)_3 \quad \text{2-3}$$
(Fe -biotite)

$$(Fe^{II})_4 (Fe^{III}) Al_2 Si_3 O_{11} (OH)_7 + O_2 + 14 H_2O \rightarrow Al_2 Si_2 O_5 (OH)_4 + 5 Fe(OH)_3 + H_4 Si O_4 \quad \text{2-4}$$
(Fe - chlorite)

In a granite, biotite in the host rock matrix will probably be the most abundant $Fe^{II}$-containing mineral. Dissolution of biotite has been shown to be fairly rapid (from $10^{-16.9}$ to $10^{-15.5}$ mol/cm$^2$/s, Velbel 1985, Nesbitt and Young 1984) and, for reaction 3 above, the equilibrium constant K for the reaction is:

$$\log K = 33.73 = \log [K^+] + 2 \log [H_4SiO_4] + pH - \tfrac{3}{4} \log [O_2] \quad \text{2-5}$$

For a groundwater with $[K^+]$ = 10 mg/L, [Si] = 5 mg/L and pH = 7.8 (Gascoyne 1996), $\log [O_2]$ = -49.5 bar which corresponds to a groundwater Eh of + 40 mV.

Similarly for chlorite dissolution:

$$\log K = 49.86 = \log [H_4SiO_4] - \log [O_2] \quad \text{2-6}$$

For the same groundwater composition, $\log [O_2]$ = -53.4 bar for this reaction which corresponds to a groundwater Eh of - 18 mV. These thermodynamic estimates are close to the average Eh actually measured in groundwaters at the ~500 m depth level in the Lac du Bonnet granite and this suggests that the oxidation of biotite and chlorite to $Fe(OH)_3$ is the dominant redox control for these groundwaters (Gascoyne 1996).

The calculations of Eh for groundwaters in granitic rocks give further support to the conclusions of Malmstrom et al. (1995) and Guimera et al. (1998) that biotite and chlorite are the dominant controls on Eh in groundwater at the Äspö site in Sweden. Guimera et al. (1998) observed that kinetic control of Eh (rather than equilibrium control) by these minerals was the more realistic and conservative situation.

## 2.3 Abundance of Reducing Fracture Minerals

In the SITE-94 assessment and subsequent reports (SITE-94 1996, Glynn et al. 1997, Guimera et al. 1998), mass transport calculations have required an estimate of the abundances of oxidisable minerals in the fractures so that calculations of the extent of consumption of $O_2$ in deeply penetrating oxidising recharge could be made. In the calculations by Guimera et al (1998), the data of Banwart et al. (1992) were used in determining the relative abundance of $Fe^{II}$-bearing chlorite, biotite and pyrite. Because groundwater recharge takes place mainly through fractures rather than the rock mass, chlorite was identified as the dominant control on $O_2$ penetration. An abundance of 35% chlorite with FeO content of 20% reported by Banwart et al. (1992) was used in the transport calculations. These results were based on a detailed analysis of cores from boreholes KR0012B/13B/15B drilled from a depth of 70m in the Äspö entrance tunnel as part of the Large Scale Redox Experiment (Banwart et al. 1992).

In the detailed study of these drill cores, Banwart et al. (1992) noted that the core from borehole KR0012B was dominated by chlorite infillings and Fe-oxides/hydroxides were largely absent. However, the discharge rate of groundwater from the packer-isolated fracture zone intersected in the borehole was the lowest of the three boreholes (0.3 L/min). In contrast, borehole KR0013B, which also intersected the same fracture zone, was dominated by CFC (calcite, Fe-oxides/hydroxides and clay) minerals and had the highest discharge rate (10 L/min). Borehole KR0015B contained both chlorite and the CFC assemblage but a discharge rate was not given.

These observations suggest that the most permeable pathways contain relatively little chlorite and are largely dominated by CFC minerals. The predominance of the CFC assemblage in the major fracture zones is clearly seen in the joint-frequency patterns (Fig. 2-1) in the geological core logs for numerous boreholes up to 700 m deep in the Äspö region (Sehlstedt and Strahle 1989, Sehlstedt et al. 1990). Figure 2-1 shows that although chlorite is an abundant infilling in fractures in all boreholes, its abundance appears to be quite low in the major fracture zones, which are likely to be the main pathways for groundwater flow and, hence, recharge.

In an investigation of the water-conductive fractures in the rock mass (ie. as distinct from the major fracture zones), Sehlstedt and Strahle (1991) identified many permeable fractures that have chlorite as the principal infilling. They note that hematite and 'rust' (iron oxyhydroxides) are seldom found. Unfortunately, no details of the actual permeability of these fractures were given so that their capacity to transmit flow relative to that of the major fracture zones cannot be determined. However, it would appear that abundant chlorite does exist in the permeable fractures in the rock matrix and could scavenge $O_2$ if recharge was directed through them.

A more quantitative estimate could be made of the real abundance of chlorite in the highly conductive fractures using raw borehole logging data which has details of mineral abundances, fracture type, etc. However, because these data were not readily available to the author, it was decided to investigate the relative occurrence of chlorite in various fracture types at another shield location for which data were available, namely a borehole on the Canadian Shield. The core log was examined from a 1-km deep borehole (WA-1) in the granite of the Lac du Bonnet Batholith near Canada's Underground Research Laboratory in southeastern Manitoba. The fracture characteristics and infilling minerals for this borehole had been previously reported by Sikorsky (1991) but details of depth, fracture type and infilling abundances had not been published and were made available through Atomic Energy of Canada Limited by R. Sikorsky (pers. comm., 1999).

The relative abundances of the four most common infilling minerals (chlorite, calcite, hematite and clays) and the type of fracture in which they were observed (open, possibly open and closed fractures) are shown in Fig. 2-2. The dominant minerals can

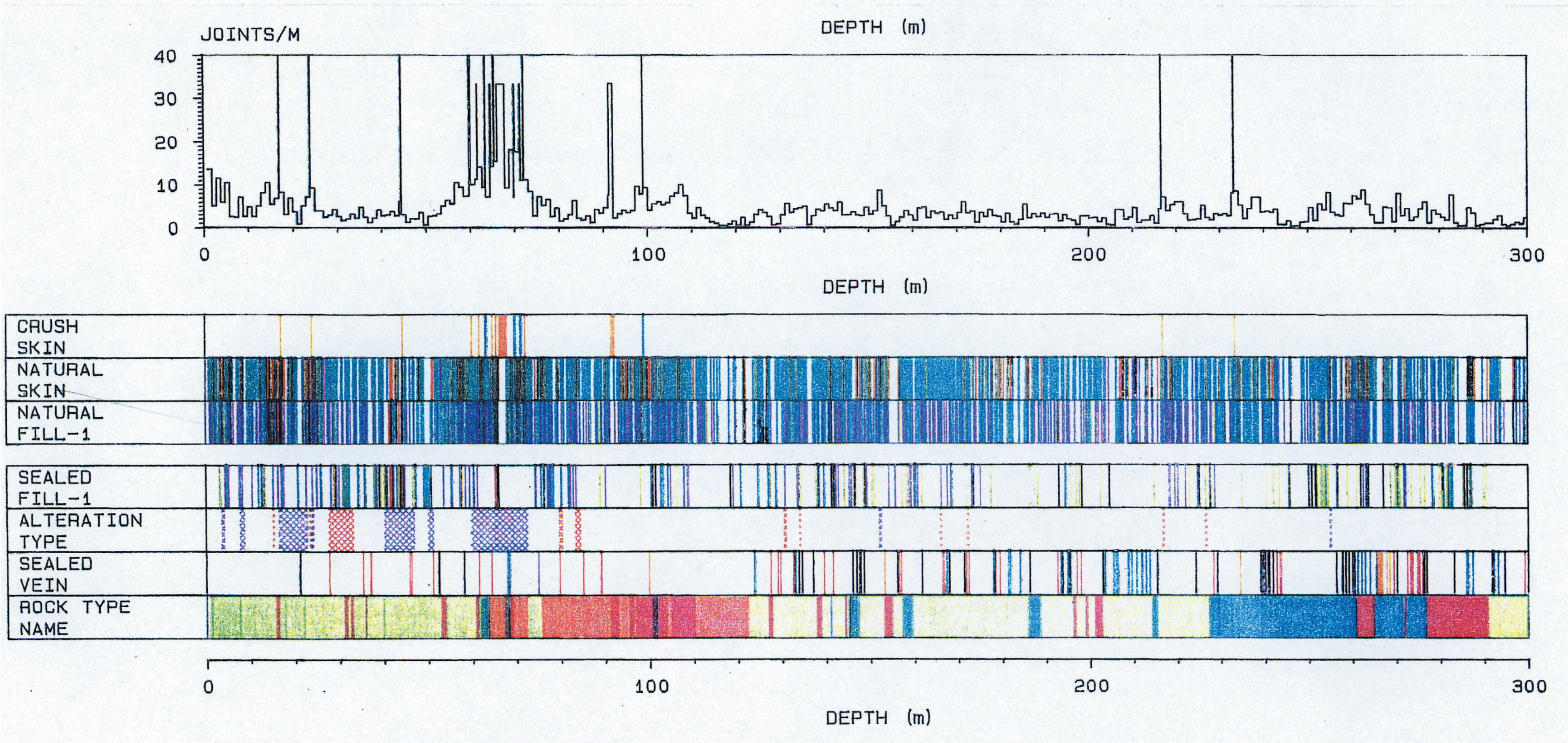



Figure 2-1 *Fracture log of core from borehole KAS06 showing frequency of chlorite (blue), calcite (purple), iron oxide (orange) infillings in the crush and natural skin bars (from Sehlstedt and Strahle 1989).*

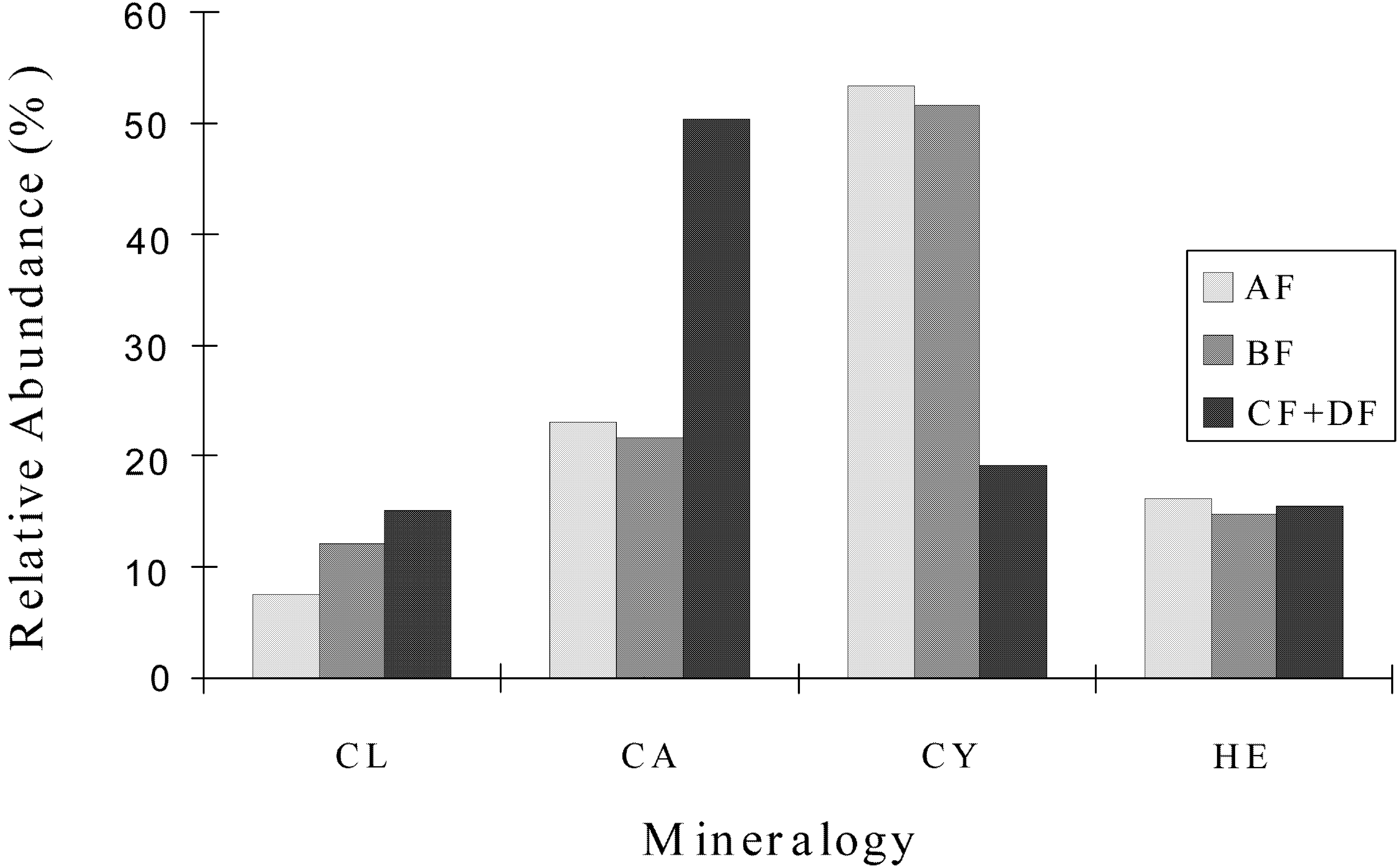



Figure 2-2 *Relative abundance of fracture filling minerals from the core log record of borehole WA-1, near URL, Canada.*

be seen to be the CFC assemblage as observed at Äspö. In this case though, open fractures tend to be characterised more by clay minerals whereas closed fractures are dominated by calcite. Chlorite occurs in all three types of fracture but clearly is less abundant in open fractures than closed fractures (by a factor of about 0.5).The situation at Äspö appears to be similar if not more pronounced; i.e., that chlorite is more abundant in closed or low permeability fractures and less abundant in the large (presumably more permeable) fracture zones. This has important implications for the study by Guimera et al. (1998) as well as related studies (Banwart et al. 1992, Glynn et al. 1997) and the SITE-94 assessment. Chlorite, which is proposed to be the principal scavenger of $O_2$ in a permeable fracture-flow system may only be available at low abundances (<< 35%) in the major groundwater recharge pathways at Äspö. Hence, the capability of flow-path minerals to scavenge $O_2$ from recharging groundwater may be considerably limited and the potential for oxidising groundwaters to penetrate to repository level is greater. Further work in relating fractures and fracture zone permeability to abundance of reduced mineral infillings is needed to resolve this important issue.

## 2.4 Stability of Repository Materials

The stability of materials introduced into the repository (such as the metal container, buffer, backfill and cementitious materials) to degradation caused by reaction with groundwater is important in maintaining the integrity of the repository. The use of Cu or Ti, instead of Fe, as a container material, will help to reduce corrosion of the container, while the use of expanding (smectite) clays in the buffer material surrounding the container, and in backfill sealing the repository, will restrict flow path apertures due to swelling when wet, and will provide an excellent sorbing medium for released radionuclides. The stability of these introduced materials, however, may also depend on the redox potential of the groundwater if the materials can be oxidised or reduced under the ambient conditions of the repository (temperature is ~20 - 100$^{o}$C, pressure is up to 10 Mpa).

Johnson et al. (1994) have calculated the time taken to consume residual $O_2$ in a Canadian repository following closure and sealing. They assume that biotite in the backfill (25% clay containing 0.7 wt. % $Fe^{II}$ and 75% crushed rock containing ~2% biotite with 15-20 wt.% $Fe^{II}$) is responsible for $O_2$ consumption and calculate that it would take about 320 years to remove all $O_2$ at ambient temperatures (~25$^{o}$C) or ~8 years at 80$^{o}$C (the maximum repository temperature in the Canadian concept). They conclude that < 1% of the $Fe^{II}$ in the backfill is consumed in this process and that the bulk of the backfill and buffer remain available for removing $O_2$ from incoming groundwater. As Guimera et al. (1998) point out, these materials represent an additional reducing barrier to the penetration of oxidising conditions to the spent fuel. If the finding of lower amounts of $Fe^{II}$-bearing minerals in fracture pathways, as described above, is realistic, it may be worth considering increasing the content of $Fe^{II}$-minerals in the buffer and backfill to strengthen this barrier. This could be done by selecting higher $Fe^{II}$ clays and possibly adding $Fe^{0}$ to the backfill, as had been proposed in early studies in nuclear waste management programs.

## 2.5 Groundwater Composition

The composition of groundwater at repository level is less critical to the long-term performance and stability of a repository than the composition of the surrounding rock and materials used in the repository. These materials are generally stable in both fresh and saline groundwaters in a mildly alkaline to neutral environment, although salinity, pH and the concentration of certain reactive species (eg. $HCO_3$, F, $H_2S$) are potentially the more reactive and controlling parameters. Saline groundwaters may induce container corrosion at a greater rate than fresh groundwaters, whereas the typical lack of $HCO_3$ in saline groundwaters would prevent the mobilisation of U, once it were oxidised, as a soluble U-carbonate complex, a process that can readily occur in $HCO_3$-rich, fresh groundwaters.

The selection of a relatively fresh groundwater component in SITE-94 and Guimera et al. (1998) that has equilibrated with the shallow rock environment at Äspö is a reasonable choice because the groundwaters predicted to reach repository depths will likely be very dilute (as they are derived from glacial melt-water) and will have interacted to some extent with the upper ~500 m of rock in the flow-path.

## 2.6 Solubility of Spent Fuel

Because uranium (U) in the spent fuel matrix ($UO_2$) is soluble (> $10^{-7}$ M/L) under oxidising conditions (as $U^{VI}$) and almost completely insoluble (<$10^{-9}$ M/L) under reducing conditions (as $U^{IV}$),the release of fission products from the $UO_2$ matrix to groundwaters at repository level and, ultimately, to the surface environment, is largely a function of whether reducing conditions can be maintained indefinitely in the repository.

The penetration of oxidising groundwaters to repository level may result in oxidative dissolution of the fuel once the container has failed. This has been determined to occur for Eh/pH conditions that are above the $U_4O_9/U_3O_7$ line in the $UO_2$ stability diagram (Fig. 2-3). Fortunately the redox characteristics of Fe are opposite to those of U because Fe is largely insoluble in oxidising conditions (as $Fe^{III}$) and soluble when reducing (as $Fe^{II}$). This contrast and the fact that the $Fe^{II} \rightarrow Fe^{III}$ transformation usually takes place at a somewhat lower redox potential than the $U^{IV} \rightarrow U^{VI}$ reaction, help to ensure that $Fe^{II}$ is consumed from the flow path and host rock before $U^{IV}$ in the repository can be oxidised.

Even if U is mobilised by the onset of oxidising conditions, both it and many of the congruently released transuranic elements and fission products are likely to be retarded by sorption on the buffer clays or direct precipitation onto fracture-filling minerals (especially clays) in the flow path. Again, some of these radionuclides are more soluble in the oxidised form (Np, Pu, Cm, Tc) and so maintenance of reducing conditions in the geosphere, away from the repository, is essential to limit contamination from reaching the surface.

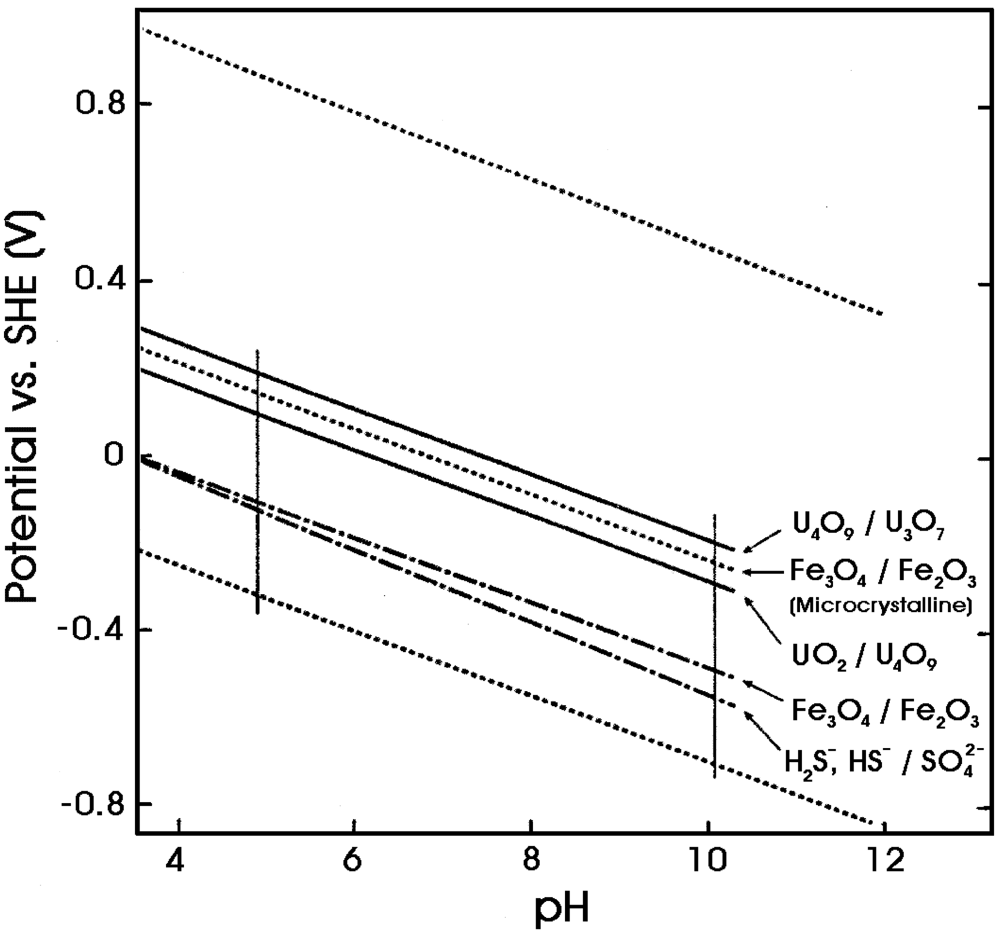



Figure 2-3 *Eh - pH diagram showing U oxide stabilities and region of buffering by Fe redox couples (from Johnson et al. 1994).*

## 2.7 Maintenance of Reducing Conditions

The SKB scenario for spent nuclear fuel disposal relies on maintenance of reducing conditions in the repository. These conditions arise by a combination of several processes:

1. Consumption of dissolved oxygen in recharging groundwater by reaction with organic C in soils and overburden followed by continued interaction at depth with dissolved organic C by the following microbially catalysed reaction:

   $$CH_2O \quad + \quad O_2 \quad \rightarrow \quad CO_2 \quad + \quad H_2O \qquad\qquad 2\text{-}7$$

   In this respiration process, approximately 2.7 mg $O_2$ are removed by 1 mg C. Surface waters typically contain 10 - 20 mg/L DOC (Thurman 1985) and, therefore, possess the capacity to remove most dissolved $O_2$. At depth, DOC concentrations reduce to < 2 mg/L (Vilks and Bachinski 1995) and it is likely that this fraction will be refractory and more resistant to oxidation.

2. Further removal of $O_2$ by interaction with reduced species such as $NH_4^+$, $NO_2^-$, $HS^-$, $Fe^{2+}$ and $Mn^{2+}$ if they are present in groundwater, according to the reactions and sequence shown in Table 2-1.

3. Direct interaction of dissolved $O_2$ with reduced mineral species lining the fracture walls or present in the host rock adjacent to the fracture path-way. These minerals commonly include chlorite, biotite, hornblende, pyrite, epidote and Fe-clays.

4. Interaction with less-common reduced species that may be present in the rock (eg. graphite, magnetite, natural U minerals and organic-rich shales and coal (if sedimentary layers are present in the area).

5. Biological activity catalysing the oxidation of hydrocarbon gases and $H_2S$.

6. Interaction with vault materials. These are likely to include steel, reduced species present in the buffer and backfill, residual organics and any reduced additives that are mixed into the buffer and backfill specifically to maintain reducing conditions.

In the natural recharge situation, oxygen dissolved in groundwater is rapidly depleted (often to <10% of its initial concentration) by interaction with soil and organics in the overburden. At greater depths, the oxygen content decreases to levels that are difficult to measure (< 2 µg/L) and measurement of redox potential using Eh electrodes is the preferred approach. The redox potential of the groundwaters decreases as $O_2$ is consumed and a 'redox' front is typically formed at a certain depth due to the control exerted by a redox couple (eg. $Fe^{II}$ - $Fe^{III}$). It is generally seen either as a depth at which oxidised species in recharging groundwater become reduced and are precipitated in the flow path or the point at which reduced minerals lining the flow-path walls are no longer being oxidised and dissolved by incoming recharge waters.

**Table 2-1 Redox reactions that control Eh and concentrations of dissolved species in groundwaters (from Gascoyne 1996).**

| PROCESS | MECHANISM | REACTION |
|---|---|---|
| $O_2$ consumption | respiration | $CH_2O + O_2 \rightarrow CO_2 + H_2O$ |
| | nitrification | $NH_4^+ + 2O_2 \rightarrow NO_3^- + 2H^+ + H_2O$ |
| | sulphide oxidation | $HS^- + 2O_2 \rightarrow SO_4^{2-} + H^+$ |
| | Fe oxidation | $4Fe^{2+} + O_2 + 10H_2O \rightarrow 4Fe(OH)_3 + 8H^+$ |
| | Mn oxidation | $2Mn^{2+} + O_2 + 2H_2O \rightarrow 2MnO_2 + 4H^+$ |
| Organics* consumption | respiration | $CH_2O + O_2 \rightarrow CO_2 + H_2O$ |
| | denitrification | $5CH_2O + 4NO_3^- + 4H^+ \rightarrow 5CO_2 + 2N_2 + 7H_2O$ |
| | Mn reduction | $CH_2O + 2MnO_2 + 4H^+ \rightarrow 2Mn^{2+} + 3H_2O + CO_2$ |
| | Fe reduction | $CH_2O + 4Fe(OH)_3 + 8H^+ \rightarrow 4Fe^{2+} + 11H_2O + CO_2$ |
| | $SO_4$ reduction | $2CH_2O + SO_4^{2-} + H^+ \rightarrow HS^- + 2H_2O + 2CO_2$ |
| | $CH_4$ production | $2CH_2O + CO_2 \rightarrow CH_4 + 2CO_2$ |
| | $N_2$ fixation | $3CH_2O + 3H_2O + 2N_2 + 4H^+ \rightarrow 4NH_4^+ + 3CO_2$ |

* organic compounds are represented here by the simple carbohydrate $CH_2O$, but may be phenols, esters, etc.

In stable hydrogeologic situations, the depth of the redox front changes very little and only migrates up or down the flow path in response to gradual shallowing or deepening of weathering effects at the surface. However, over geologic time, changes in climate, recharge rates and surface vegetation coverage may induce larger movements of the front and it is this movement that is of concern here.

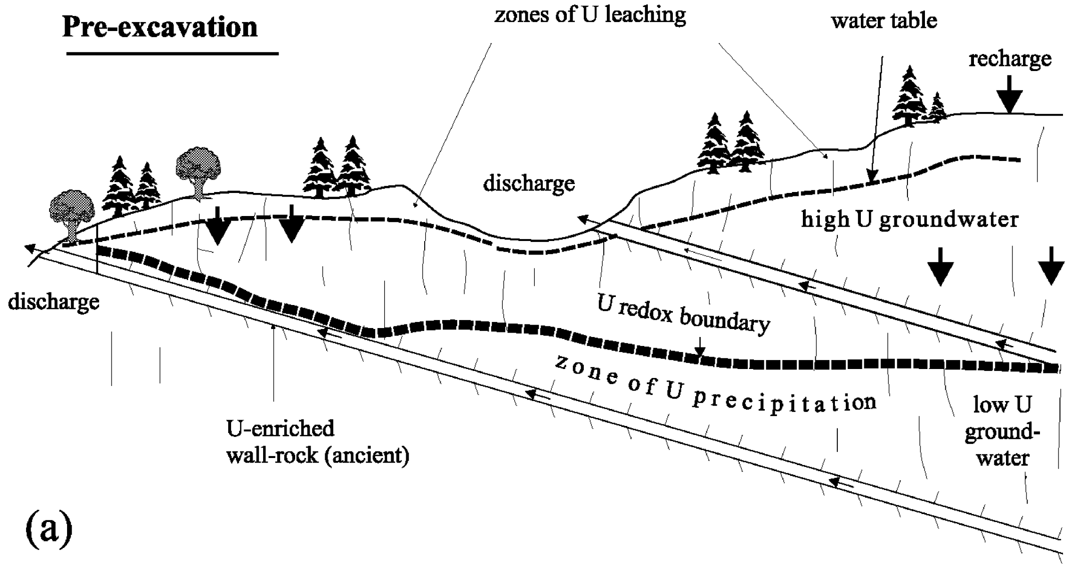
## 2.8 Disruptive Scenarios

Reducing conditions and a stable position of the redox front in the subsurface can be established and maintained almost indefinitely by the mechanisms and natural processes described above, provided that there is no change in initial conditions and controls. There are a number of scenarios, however, in which this situation may be disrupted and oxygenated groundwaters could reach the repository. These include:

1. Surface lowering due to erosion which, in turn, is accelerated by isostatic rebound or tectonic uplift. A good example of this effect is the gradual dissolution of shallow uranium ore bodies at locations such as Alligator Rivers, Austrailia; Key Lake, northern Saskatchewan; and the Oklo and Pozos de Caldos natural analogues.

2. Progressive consumption and depletion of reduced species in the bedrock flow paths causing rapid downward migration of the redox front. This will happen at any site as part of the natural weathering process but is accelerated in areas where the overburden is thin or absent (eg. bedrock outcrops) and where biological activity in the soils is less. In this case, reduction by bedrock minerals takes on a larger role in consuming the initial atmospheric charge of $O_2$ in the recharge water. An example of the potentially rapid consumption of reducing minerals in the flow path is the breakthrough of high concentrations of dissolved U in groundwater draining from fractures intersected during excavation of Canada's Underground Research Laboratory in southeastern Manitoba. Uranium concentrations in the inflow to the facility remained fairly constant for a period of seven years after excavation and stabilisation of the cone of groundwater depression in the fractured granite surrounding the URL (Fig. 2-4a). Then in 1993, U concentrations increased rather suddenly by a factor of ~5 and have remained at or above this level ever since. It is believed (Gascoyne 1997) that the U redox front steadily moved downward over this period as surfaces or minerals that could limit U oxidation were gradually consumed by oxidation, such that after about seven years, the front intersected the URL facility (Fig. 2-4b) and caused rapid and sustained increases of U in the inflow.

3. Increase in recharge rate. This could be brought about in a number of ways, such as change to a wetter climate, increase in fracturing of the host rock, increase in permeability of the groundwater path ways and an increased hydraulic gradient. These aspects are considered in more detail in Sections 4 and 5.



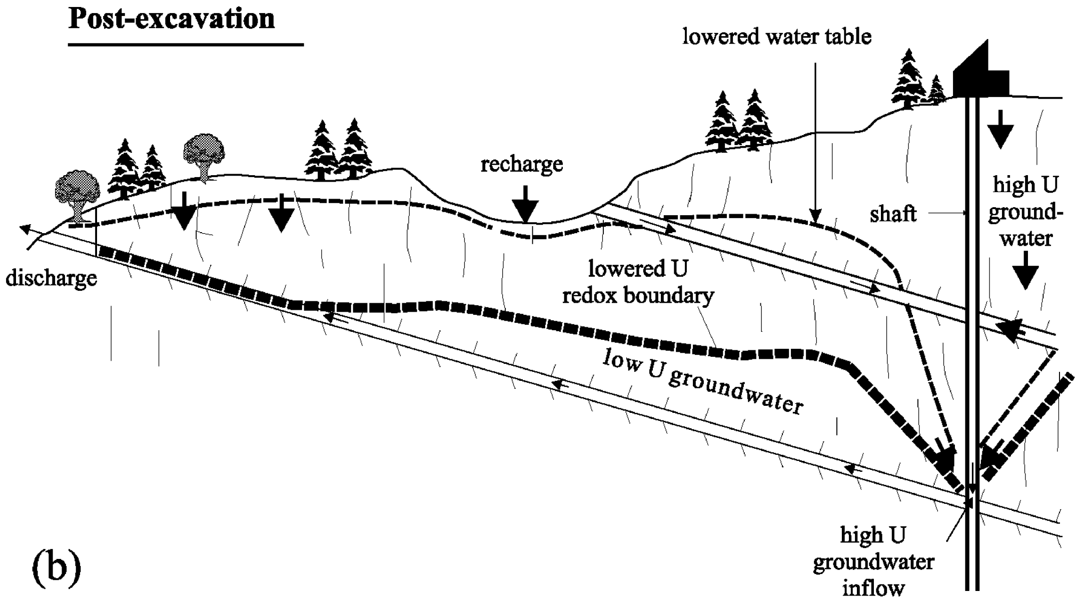




Figure 2-4 *Schematic diagram showing zones of U mobility and retardation in groundwaters at the URL, Canada, a) before excavation and b) following excavation and water table drawdown (from Gascoyne 1996).*

# 3 Oxygen Content of Melt-Waters

Oxygen dissolved in melt-water that is recharged to the bedrock is the principal oxidant that could compromise the geochemical stability of a granitic repository. It is important to know, therefore, the likely concentration of $O_2$ in melt-water beneath an ice sheet and the potential reactions and processes that could consume it.

The $O_2$ content of melt-water underlying an ice sheet depends largely on five factors:

1. initial content of $O_2$ in the melting ice,
2. opportunities for melt-water to re-equilibrate with atmospheric $O_2$,
3. whether partial melting and re-freezing of firn or ice has occurred,
4. diffusion rate of $O_2$ across the gas-water interface and
5. whether $O_2$ is consumed by oxidation of organics in the ice or by reduced minerals in basal sediments.

Cold ice (ie. polar ice, < 0$^o$C) is formed by the sintering of dry snow into solid ice by the elimination of pore space and reduction of surface area (Anderson and Benson 1963). Trapped air in the lattice has approximately the same composition as the atmosphere and is present in ice at a concentration of about 100 cc/kg. In contrast, ice formed by freezing of melt-water (which was in equilibrium with the atmosphere) contains ~ 30 cc/kg of dissolved gas whose composition is more enriched in the soluble gases (e.g., $O_2$). In temperate ice, where melt-water frequently flows through channels in the ice, the more soluble $O_2$ component has been dissolved leaving the ice depleted in $O_2$ (Berner et al.1977). Practically all gas in ice is concentrated in air bubbles and so bubble-free ('blue') ice contains less than 0.1% of the dissolved gases that would be in ice formed in contact with the atmosphere (Scholander et al. 1953).

The processes affecting the $O_2$ concentration in ice have been examined in more detail by Souchez et al. (1995a,b) for the specific case of melting and re-freezing of firn or ice, as may occur at depth in polar ice sheets. Previous work (Stauffer et al. 1982, Craig et al. 1988, Schwander 1989) has shown that the ratio of dissolved $O_2$ to $N_2$ in snow and firn ranges from 0.268 (the atmospheric ratio) to 0.56 (if melting and re-freezing occurs, because of the greater solubility of $O_2$ in water than $N_2$). Ratios slightly lower than the atmospheric value have been observed in basal ice probably because melt-water comes in contact with a gas phase that is already depleted in $O_2$ due to the greater solubility of $O_2$. However, in a study of silty basal ice from the GRIP (Greenland Ice Core Project) core, Souchez et al. (1995a) find strong depletion of $O_2$, to concentrations as low as 0.05%, giving $O_2/N_2$ ratios as low as 0.04. The inverse correlation of $O_2$ depletion with $CO_2$ and $CH_4$ enrichment found by Souchez et al. (1995a) in basal ice is believed to indicate that oxidation of organic matter (from residual peat deposits) is occurring at the base of the ice sheet.

In a second paper on the GRIP core, Souchez et al. (1995b) found considerable evidence to show that several metres of basal ice originated not from the overriding ice sheet but from local ice developed before the main ice sheet arrived. This ice had characteristic properties and strong gradients in several parameters were found beteen the glacier ice and the ice at the bedrock interface, as follows:

1. $CO_2$ and $CH_4$ contents increased from near zero in glacier ice to, respectively, 120,000 and ~6000 ppm by volume in basal ice,
2. Total gas content decreased towards glacier ice, from ~0.1 to 0.05 cc/kg,
3. $\delta^{18}O$ decreased from -25 to -36 ‰ towards the ice.

These data were interpreted as indicating that flow-induced mixing of basal ice was occurring and that the source of the $CO_2$ and $CH_4$ was not oxidation of organics in the ice or entrapped sediments but from the underlying bedrock. This contradicts the previous interpretation by Souchez et al. (1995a) that the high $CO_2$ and $CH_4$ concentrations in basal ice originated from oxidation of residual peaty deposits caught up in the basal ice sediments

Although there have been many studies of the dissolved gas content of glacial ice, there have been relatively few measurements of dissolved $O_2$ in sub-glacial melt-waters. Large variations (from 0.2 to 34 ppm at $0^oC$) in the $O_2$ content of glacial ice have been observed in alpine glaciers and polar ice. Several of the measurements that have been made are summarised in Table 3-1.

One of the more detailed and reliable studies of the $O_2$ content of glacial melt-water has been reported by Brown et al. (1994) who determined dissolved $O_2$ on a diurnal basis for melt-waters draining the Arolla glacier in Switzerland over a 2-month period of melting in 1992. They observed melt-waters with $O_2$ concentrations well below saturation with respect to the atmosphere (which is typically 9.4 to 11 ppm depending on altitude). The lowest values (<40% of saturation) coincided with peak daily discharge while the highest values (>80 %) occurred at lowest discharges. The lowest values were similar to those determined on samples of basal ice (~34%). These results were believed to indicate that the slow kinetics of $O_2$ uptake from atmospheric sources allowed $O_2$ concentrations in melt-waters to remain close to the values in the ice itself. The possibility of $O_2$ loss due to consumption by oxidation of pyrite in the basal rock material was found to be unlikely because low $O_2$ values did not correspond with high dissolved $SO_4$ concentrations, as might be expected if pyrite oxidation were important. In fact, the inverse correlation was observed (high dissolved $SO_4$ coincided with high $O_2$) suggesting that low $SO_4$ concentrations were more likely to be a function of dilution and remained low due to the slow kinetics of pyrite oxidation. However, from Brown et al.'s data, a mass balance of $SO_4$ at maximum and minimum discharge can be calculated and it can be seen that at low discharges, only about half the amount of $SO_4$ is removed compared to that at peak discharge. Thus it is possible that more pyrite is, in fact, being oxidised at peak discharges.

The data in Table 3-1 and the above references indicate that the $O_2$ content of melt-water could vary considerably in water at the base of an ice sheet. Guimera et al. (1998) assumed a concentration of 45 ppm, based largely on a study by Ahonen and

Vieno (1994). This high concentration gives groundwaters an Eh of > +100 mV and is, therefore, a very conservative estimate for the modelling. The data from recent measurements (Table 3-1) suggest that dissolved $O_2$ could be significantly less than 45 ppm, possibly in the range 0 - 5 ppm. Although Eh would be little changed, the ~10-fold reduction in $O_2$ concentration would translate into a similar reduction in the rate of consumption of reducing minerals in the bedrock flow-paths and this is of considerable importance to the safety assessment modelling of a repository.

**Table 3-1 Summary of literature values for $O_2$ concentration in alpine glacial ice (Switzerland and Austria) and polar ice.**

| Location | Type | $O_2$ (ppm) | Comments | Reference |
|---|---|---|---|---|
| Jungfraujoch | glacier | 7.6 | | Weiss et al. (1972) |
| Aletsch | glacier | 0.2 | | Weiss et al. (1972) |
| Arolla | glacier | 3.6 | melt- water | Brown et al. (1994) |
| Arolla | glacier | 3.4 | melted ice | Brown et al. (1994) |
| Arolla | glacier | 21 - 34 | young ice | M.J. Sharp (pers. comm) |
| Arolla | glacier | <0.7 - 28 | old ice | M.J. Sharp (pers. comm) |
| Greenland | ice sheet | 2 - 3 | | Stauffer et al. (1982) |
| Antarctica | lakes under ice | ~ 3 | | Craig et al. (1992) |
| Byrd Stn., Antarctica | basal ice | 0.5 - 5 | | Heron & Langway (1979) |

# 4 Glaciation

Probably the most significant event that might influence the stability of a spent nuclear fuel repository is the growth and subsequent melting of permafrost and a thick ice cover over the disposal site during the next glaciation. It is well recognised that the next 100,000 years will likely see at least one major glaciation in the northern hemisphere. Numerous researchers have examined past ocean core and terrestrial sedimentary records, as well as calculated the earth's orbital variations, to predict the timing of future climatic changes (Hays et al. 1976, Kukla et al. 1981, Shackleton 1987). In particular, several models have been developed (Imbrie and Imbrie 1980, Berger et al. 1989, Boulton and Payne 1993) which attempt to determine the characteristics of ice accumulation, its thickness and flow dynamics, the timing of the glacial maxima over the next 100,000 years, and the corresponding fall in global sea-level.

The onset of glacial conditions will directly influence recharge, groundwater flow paths and the permeability of fractures in the bedrock. An approaching ice sheet will not likely compromise the stability of the repository but would reduce rather than increase groundwater flow rates, and fracture permeabilities will initially decrease due to permafrost and ice-loading at the surface.

For the case of the Fennoscandian Shield, a scenario describing expected climate change over the next 100,000 years has been prepared by Ahlbom et al. (1991). Peak glaciations are predicted at 20,000, 60,000 and 100,000 years from now with the 60,000 year event been the most severe. These predictions are summarised in Table 4-1. Some deglaciation is likely after ~ 23,000 years although interstadial (cold) conditions are likely to persist until the onset of the next glaciation. A second deglaciation is expected at about 75,000 years and this is likely to lead to warm climate conditions comparable to the present. A third deglaciation may occur at about 125,000 years.

The effects of these climatic cycles on a repository site at Äspö have been considered by Ahlbom et al. (1991), McEwen and de Marsily (1991) and King-Clayton et al. (1995), and have been subsequently modelled in the SITE-94 study (SITE-94 1996) and reported in detail by Provost et al. (1998). Several aspects of this work are examined below and some of them raise concern regarding the validity of the modelling in light of recent field observations and previous work.

## 4.1 Permafrost

The flow of groundwater in a repository location will be strongly influenced by the presence of permafrost (perenially frozen ground). From the above description of climatic conditions for the next 100,000 years, it is likely that more than one period of extensive permafrost will develop at the repository site. To some extent, the intensity

**Table 4-1. Summary of predicted climatic events in the Fennoscandian Shield over the next 125,000 years (based on Ahlbom et al. 1991).**

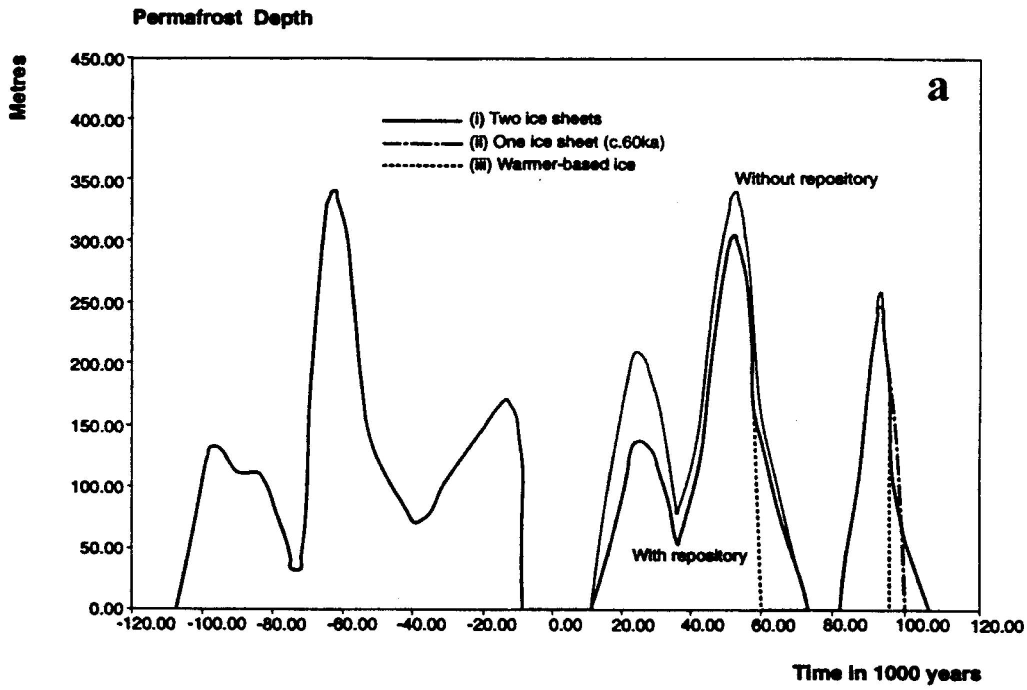
| Future Date (years) | Event | Ice Centre | Ice Thickness (m) | | Crustal Depression (m) | | Sea-level Depression (m) |
|---|---|---|---|---|---|---|---|
| | | | Maximum | S. Sweden | Maximum | S. Sweden | |
| 5,000 | minor glaciation | Caledonian Mntns | 1000 | 0 | 300 | 0 | 5-50 |
| | interstadial | | | | | | |
| 20,000 | major glaciation | Gulf of Bothnia | 1500 | 800 | 500 | 60 | 85 |
| | interstadial | | | | | | |
| 60,000 | major glaciation | Gulf of Bothnia | 3000 | 2000 | 700 | 600 | 150 |
| 75,000 | deglaciation | - | | | | | |
| 100,000 | major glaciation | | 1500 | 1000 | 400 | | |
| 125,000 | interglacial | - | | | | | |

and depth of influence of permafrost will be offset by the insulating effects of an ice cover, the ambient geothermal gradient and the initial thermal effect of the repository. McEwen and de Marsily (1991) have calculated that permafrost in Sweden may develop to depths of between 500 and 700 m for prolonged cold climatic cycles (up to 120,000 years long). However, if a repository is present and there is an overlying ice sheet, the base of the permafrost may only extend to 400 m depth, thus leaving the repository is surrounded by unfrozen ground.

The formation of deep permafrost requires a long period (~ 10,000 years) of continuously cold climate. Because permafrost retreat takes place from the base upwards, loss of permafrost conditions will similarly take considerable time and so it is unlikely that it will be absent by the time deglaciation begins. This is especially true if the first significant deglaciation in Sweden is not until ~ 75,000 years in the future.

The calculations of McEwen and de Marsily (1991) have been revised by King-Clayton et al. (1995), taking into account the latent heat capacity of freezing, other modes of heat transfer than conduction, and a more realistic inclusion of heat from the waste. The revised model examined three scenarios: the central scenario in which cold-based ice forms over the Äspö site at about 50,000 and 90,000 years; the same scenario but with warm-based ice; and a scenario where a cold-based ice sheet forms at 50,000 years only. These calculations revised the depth of penetration of permafrost at Äspö over the next 100,000 years (Fig. 4-1a) showing that a maximum depth of 300 m was attained after 50,000 years if a repository was present. They also derived profiles of the change in temperature with time (Fig. 4-1b). The results of the warm-based ice model only deviated from the those of the other scenarios after ~60,000 years and indicated faster melting of the ice at the time of deglaciations.

The SITE-94 central scenario for the Äspö location is summarized in Fig. 4-2 and includes predictions of sea level, ice thickness, permafrost thickness and the presence or absence of saline water in the Baltic Sea for the next 120,000 years. Despite the revised modelling predictions above, that still show extensive, deep permafrost at Äspö lasting from ~ 10,000 to at least 60,000 years, King-Clayton et al. (1995) argue that permafrost is not likely to be spatially continuous but would be broken by 'taliks' (unfrozen ground in a permafrost environment) which underlie deep lakes and rivers and through which groundwater can recharge or discharge. In the Äspö area, one of the three large fracture zones (NE-1) is assumed to be a talik throughout the entire permafrost period. King-Clayton et al. (1995) also assume that the ice sheet is cold-based only towards the advancing front and under its central portion. Basal melting, due to pressure and ice movement is believed to occur soon after ice advances over the area and remains until deglaciation except for the source area in the centre of the ice sheet. In addition, it is also assumed that permafrost that developed before ice overran an area is discontinuous and represents a minimal barrier to groundwater recharge and lateral or vertical flow soon after the ice cover develops. These assumptions are based principally on previous models of glaciation by Boulton and Payne (1992), Boulton et al. (1984, 1993). Only during the initial advance of the ice is it assumed that a cold-based ice sheet and a permafrost barrier will exist at the site.



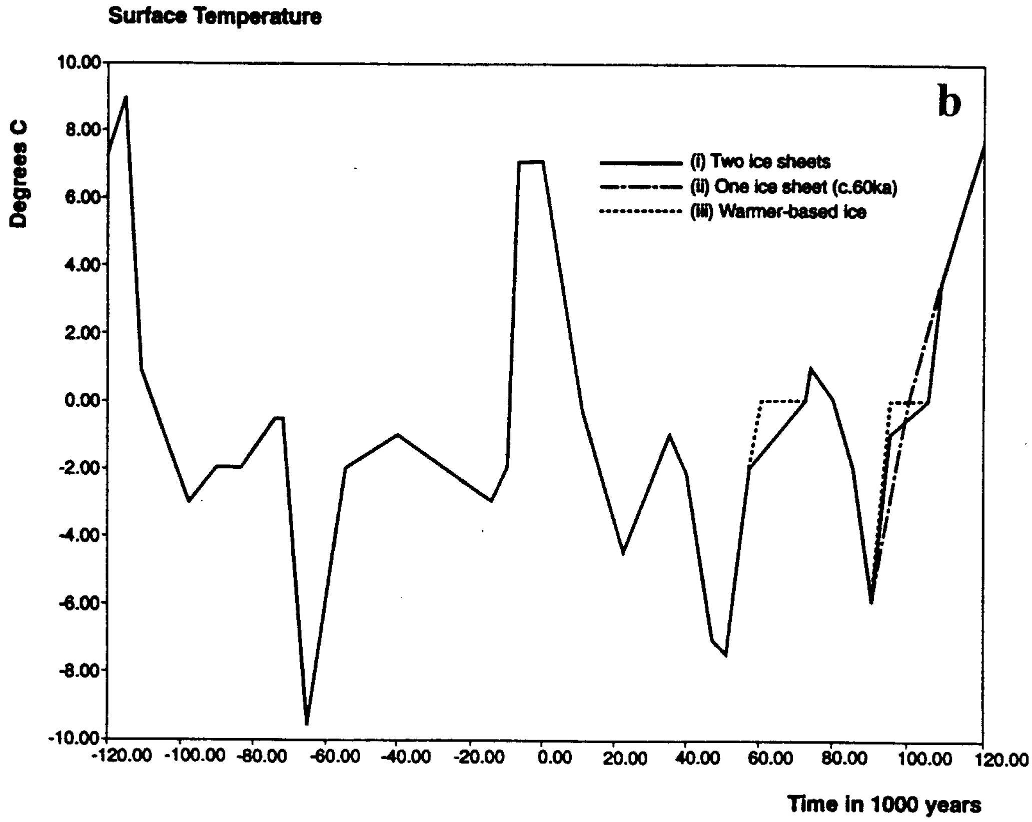




Figure 4-1 *Predicted variation in time, from -120,000 to + 120,000 years, for three different glaciation scenarios (see text), for a) permafrost depth and b) rock surface temperature at the Äspö site (from King-Clayton et al. 1995).*

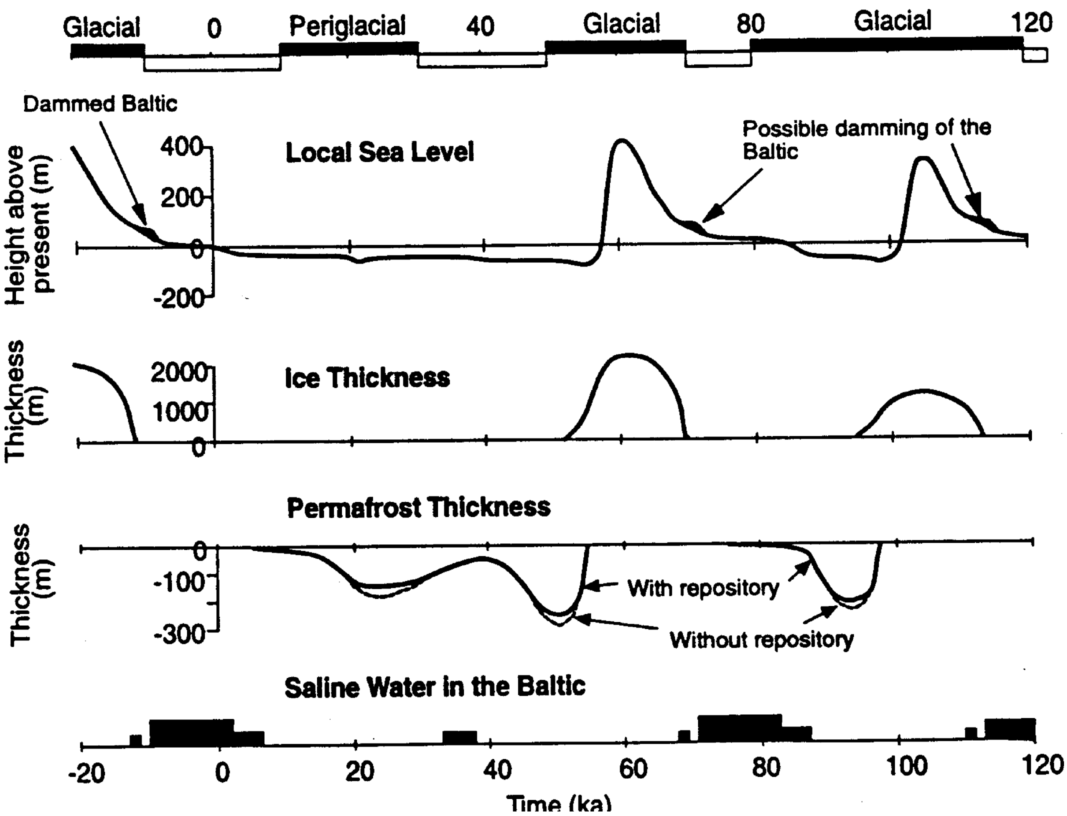



Figure 4-2 *Results of the central climate change scenario in SITE-94 showing sea levels, ice and permafrost thicknesses at Äspö over the next 120,000 years (from King-Clayton et al. 1995).*

The modelling done for SITE-94 goes further in this direction and assumes permafrost at Äspö is very thin (3 m) and disappears completely soon after being overrun by an ice sheet (Provost et al. 1998, Appendix D). This is clearly illustrated in Fig. 4-3. Therefore, groundwater flow for the bulk of the modelled period is performed with no permafrost barrier to surface flow, in contrast to the arguments presented above, by McEwen and de Marsily (1991) and the revised model of King-Clayton et al (1995), for an extensive influence of the permafrost barrier. King-Clayton et al. (1995) identified a number of alternative scenarios to address uncertainties that they perceived in some areas of the model, such as the continuity of permafrost, cold- versus warm-based ice, variable responses of fractures to ice loading and changes in sea level due to ice damming. The likelyhood and effects of these alternatives were discussed qualitatively but were not quantified as in the central scenario.

Several questions remain unanswered in the determination of the extent and duration of permafrost at the repository site over the next 100,000 years: will permafrost be spatially continuous and deep for the initial ~ 50,000 years or will it be thin and punctuated regularly by taliks and, therefore, be an ineffective barrier to groundwater recharge? Recent evidence (described in the next section) seems to indicate that permafrost will be continuous and the ice sheet will be cold-based for most of its life. This will clearly affect the groundwater flow rates at the repository site and, hence, challenge the SITE-94 finding that oxidising groundwater may penetrate to repository depth.

## 4.2 Basal Ice Conditions

The base of an ice sheet may remain frozen to the bedrock for some time during a glaciation and, because it moves only by internal deformation, it will cause little erosion of the bedrock surface. However, once a thick ice cover has been established, freezing conditions at the bedrock - ice sheet interface may disappear due to geothermal heat flux and internal heating by viscous flow of the ice, which could raise the temperature to the pressure melting point. This transition has an important influence on the properties of the ice sheet and its development.

The characteristics of an ice sheet, such as its formation, thickness, flow velocity and temperature distribution, have been modelled for the last glaciation in a detailed study by Boulton and Payne (1993). They derive a coupled thermo-mechanical, 3-D model driven by changes in the equilibrium line altitude (ELA), the boundary between net ablation and net accumulation of ice. The ELA is, in turn, a function of precipitation input and air temperature and, therefore, a reflection of sea-surface temperature. This ice sheet model satisfactorily simulated the behaviour of the last ice sheet in Europe and was then used to predict future glaciation conditions in the Fennoscandian Shield.

In this application, the model predicted that the ice sheet would be cold-based during its initial expansion and until it had extended approximately 100 km from its source, at which point basal melting began. Surface melt-water, although generated largely during the summer season, did not penetrate to the basal ice except within a few

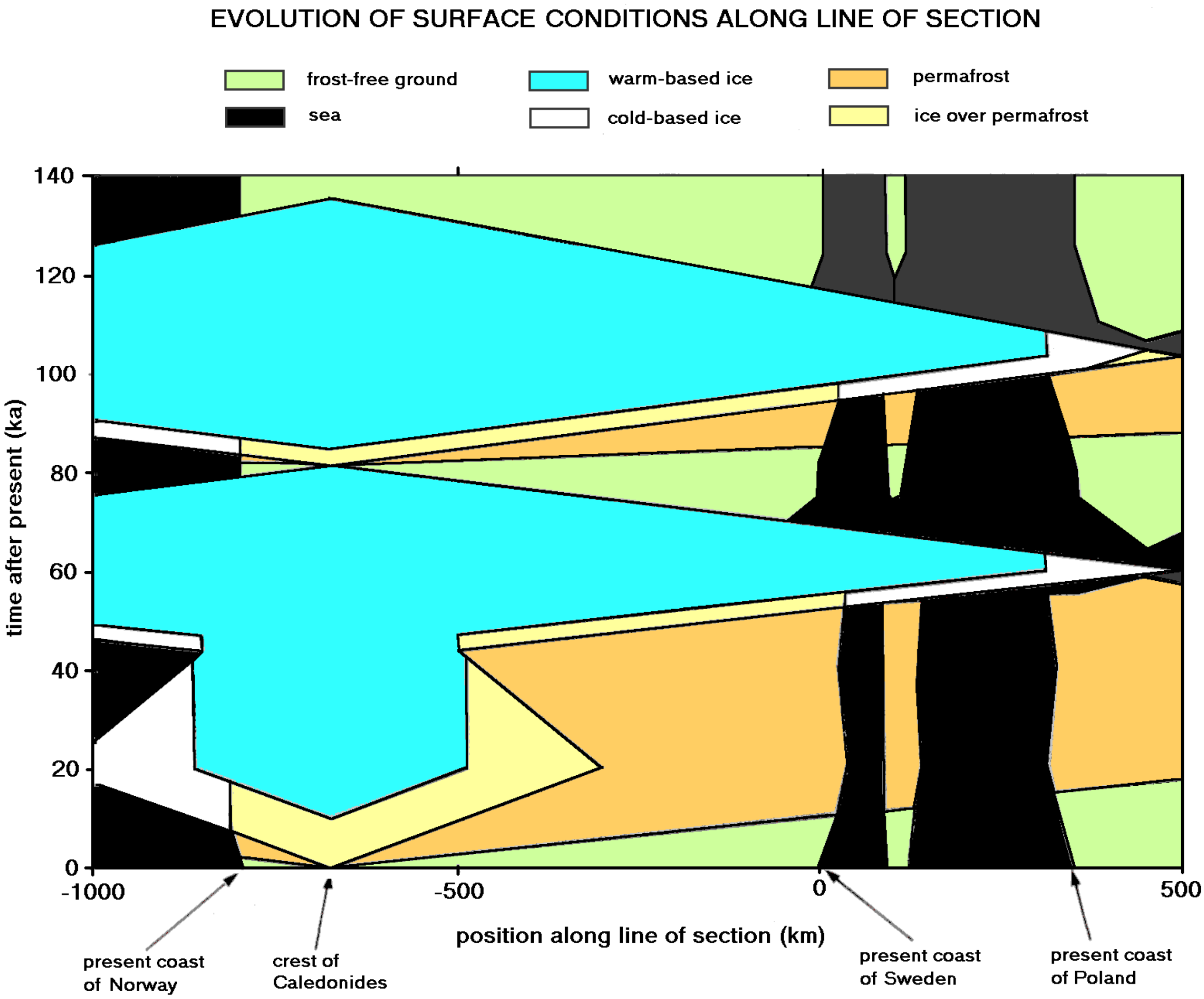



Figure 4-3 *Variation in climatic conditions at the surface for the regional cross-section of the flow model (from SITE-94, 1996).*

hundred metres of the terminus because of the rapid closure with depth of moulins and crevasses that might otherwise have conducted water to the base. This has important implications for the characteristics of basal melt-water in that 1) the basal melt-water is likely to be isolated from $O_2$ uptake from the atmosphere, 2) it may have high hydraulic heads and 3) the lithostatic load (the weight of ice) might be the main control on the hydraulic head of groundwater that enters the bedrock. As described in section 4.1 above, the modelling done for SITE-94 assumes that basal ice conditions change over the period of glaciation, from cold-based during the early advancing stage, to warm-based, caused by a combination of geothermal heat, frictional heating in the ice itself, and lowering of the freezing point by up to 2$^{o}$C due to pressure of overlying ice. The SITE-94 model and supporting models of Boulton and Payne (1993) and Boulton et al. (1995), propose that warm-based ice would persist at the Äspö location for most of the period of ice cover. However, in the last few years, a number of papers have been published in glaciological journals which contradict the assumptions in the SITE-94 model for basal ice conditions.

Heine and McTigue (1996) have modelled the evolution of the thermal profile in a growing ice sheet using realistic values for geothermal heat flux, snow accumulation rate and thermal conductivities and heat capacities of ice and bedrock. They find that the thermal profile is influenced strongly by the surface temperature at the start of ice sheet growth: the colder the temperature the longer it takes for the pressure melting point to be attained (Fig. 4-4a). Even with a relatively warm initial surface temperature (~ -10$^{o}$C), basal melting does not occur until 35,000 years. Ice-sheet thickness also affects the time to basal melting. For an initial surface temperature of -12.5 $^{o}$C, a thick ice sheet of 3 km reaches basal melting conditions (~ -2$^{o}$C) in about 60,000 years (Fig. 4-5b). However, if the ice is thinner than 2 km, basal melting may never occur. Also, the rate of downward advection of cold surface ice greatly affects the basal ice temperature such that at high rates of snow accumulation (5 cm/year), basal melting is not attained until after 100,000 years.

The model of Heine and McTigue (1996) indicates that basal temperature is strongly time-dependent and is largely determined by the initial surface temperature and the rate of advection of surface ice to depth. Basal melting will not occur due to geothermal heat flux alone, especially if the initial ground temperature is colder than -10$^{o}$C. The central part of an ice sheet, therefore, takes tens of thousands of years to reach pressure melting point at its base. For these reasons, therefore, Heine and McTigue believe that central parts of Pleistocene ice sheets were cold-based throughout most of a glacial cycle. This argument is supported by observation of preserved periglacial features on the Fennoscandian Shield (Kleman and Borgstrom 1994, Kleman et al. 1997). The change to warm-based ice only occurred near the end of the glaciation and towards the margins of the ice sheet. Thus, the duration of melt-water flow at any particular location would have been short and would have resulted in only slight surface erosion and limited recharge to the bedrock. It has been argued that a change in the basal conditions of the Laurentide ice sheet, from cold- to warm-based, could have resulted in a dramatic thinning of the North American ice sheet (MacAyeal 1993). This would account for evidence indicating massive discharge of icebergs into the North Atlantic Ocean (so-called 'Heinrich' events). Heine and McTigue (1996) believe that these events do not contradict their model but they are

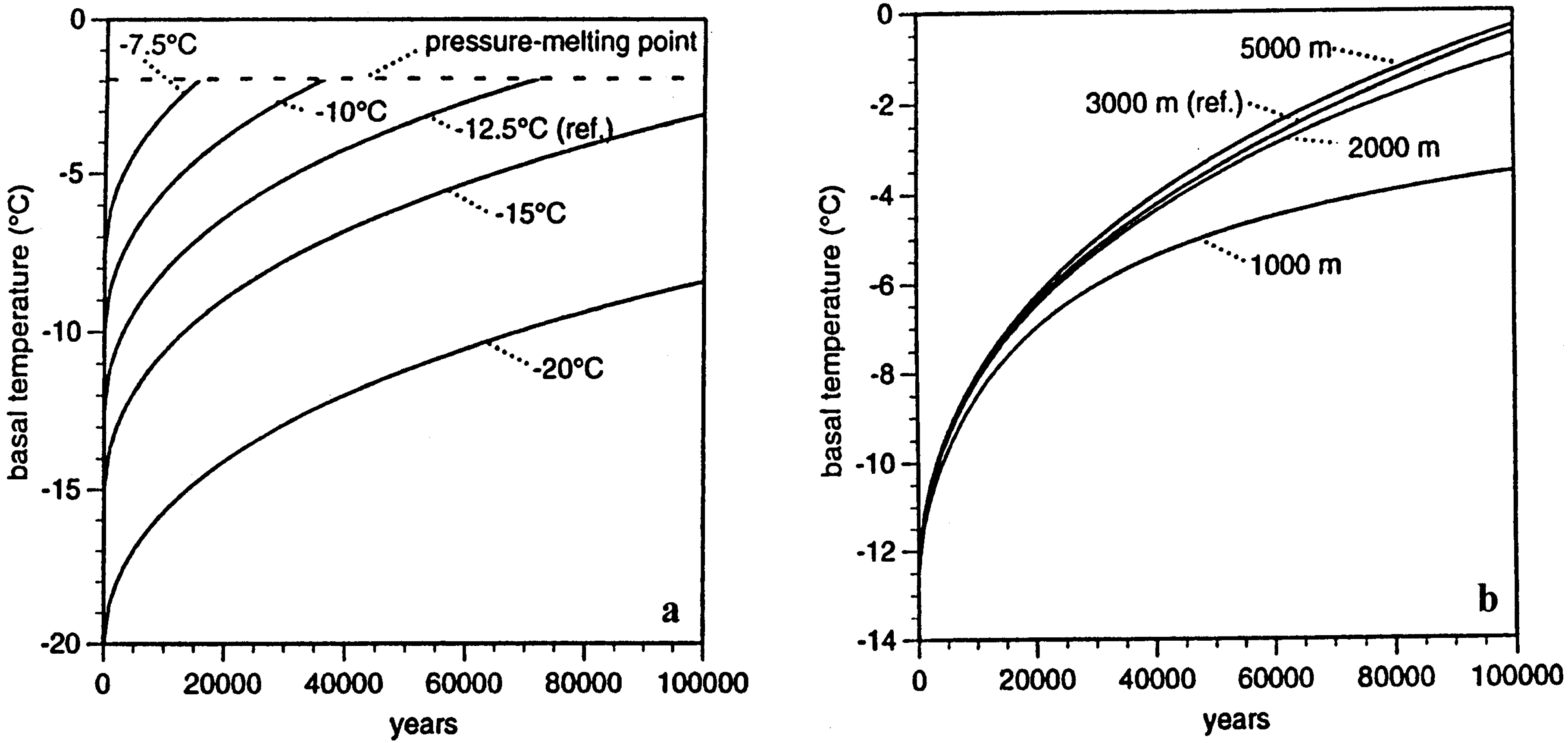



Figure 4-4 *Influence over time of a) initial surface temperature on basal ice temperatures and b) final thickness of the ice sheet on basal ice temperatures (from Heine and McTigue 1996).*

probably confined to the margins of the ice sheet, while the main part remains cold-based. If an ice sheet becomes warm-based, groundwater circulation could return, although on a limited basis as normal discharge path-ways would probably be blocked by ice. During a deglaciation event, groundwater flows will return in the bedrock and likely increase due to the potential for greater hydraulic heads as the ice melts and the base of the ice becomes an important conduit for melt-water.

Detailed studies of the last (Weichsel) glaciation in Europe (maximum ~18,000 years ago) have shown that it was characterised by the development of an extensive permafrost in front of the ice sheet which caused loss of forested cover and increase in steppe and tundra terrain. Studies of modern glaciers and ice sheets (e.g., Boulton and Spring 1986, Hindmarsh et al. 1989) have indicated that, as an ice sheet advances over frozen terrain, basal ice temperatures would initially be below the pressure melting point of ice. As the ice thickens, geothermal heat may cause melting at the base and thinning of the permafrost to allow limited recharge to bedrock. These studies suggest, however, that cold-based ice sheets are most likely to occur during major northern-hemisphere glaciations, rather than under modern glaciers, although some parts of the ice sheet may be warm-based. In Scandinavia, preserved landforms have been shown to be pre-Weichselian and indicate that the Fennoscandian ice sheet had a frozen bed and that any melting occurred at its outer limits (Kleman et al. 1997)

The Antarctic ice sheets provide a limited amount of data that addresses the question of basal ice conditions in polar ice. In a 1000-km section of the West Antarctic ice sheet (Fig. 4-5a), Boulton and Spring (1986) found zones where basal conditions range from a fully frozen bed (Fig. 4-5b, mainly in the upland source regions) to one of melting and refreezing (in the lower regions). Deep-drilling at the Byrd Station showed that refreezing of the basal ~5m of ice had occurred, suggesting a zone of basal melting in a bedrock trough, up-glacier of the Byrd borehole. The ice sheet modelling of Boulton and Spring predicted sub-glacial water flow occurring under large sections of the ice sheet (Fig. 4-5c).

Use of ice-penetrating radar (Oswald and Robin 1973) and, recently, radio-echo sounding (Kapitsa et al. 1996, Siegert and Ridley 1998) has also shown that large areas of the East Antarctic ice sheet are underlain by lakes of melt-water. In some cases, individual lakes that do not appear to overlap spatially from the limited sounding data may be shown to be a single lake by the use of satellite radar altimetry (Kapitsa et al. 1996). Other data that do not show the existence of a single flat surface nevertheless show that a reasonably flat interface exists with a strong radio-echo signal. These interfaces are believed to indicate the presence of basal water and water-saturated basal sediments (Siegert and Ridley 1998).

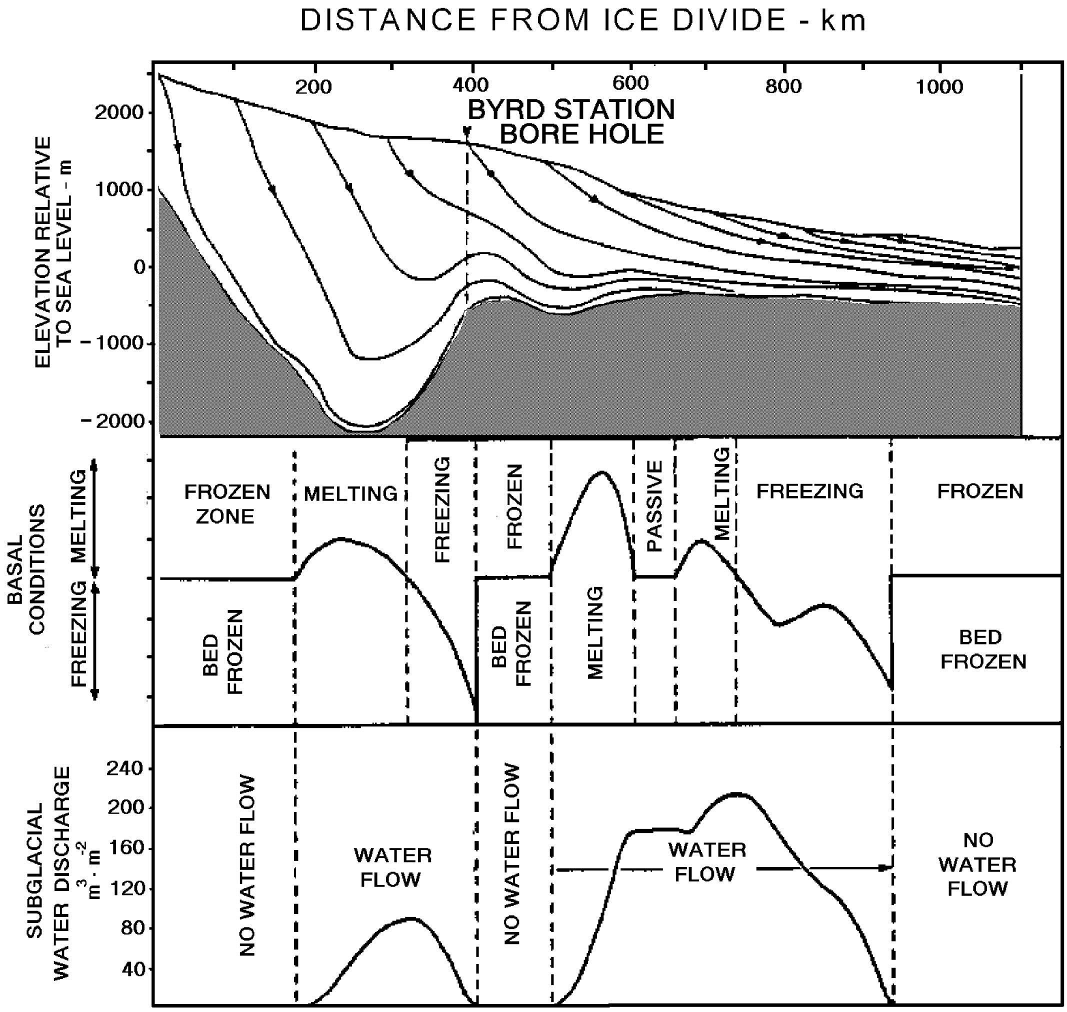



Figure 4-5 *Vertical section along a transect through the Byrd Station, West Antarctica showing patterns of basal melting, water flow and regelation (from Boulton and Spring 1986).*

The recent research described above generally indicates that a polar ice sheet is cold-based (ie. frozen) in both its margins and central portions during the expansion phase. Only after it has been established for a considerable time could basal melting occur due to geothermal heat and the energy of viscous flow. This is likely the case for the Antarctic ice sheets which have been established for most of the Pleistocene and now have formed extensive basal 'lakes'. Ice sheets formed over a short time-base, say 50,000 - 100,000 years, however, are more likley to be frozen at the base. Melting at the margins can occur, generally during deglaciation, although underlying permafrost may remain for some time, preventing groundwater recharge to the bedrock.

The model used in SITE-94 and detailed in King-Clayton et al. (1995) and Provost et al. (1998) for the Fennoscandian Shield assumes that the ice sheet is only cold-based west of the region of the Caledonides and is warm-based from the Caledonides eastwards over Sweden to the Baltic (Fig. 4-3). This is clearly in contrast to the above research and earlier SKI reports (e.g. McEwen and de Marsily, 1991) which indicate that the next ice sheet will likely be cold-based for much of its duration and permafrost will be extensive and deep and, probably, spatially continuous.

# 5 Groundwater Flow Modelling

Groundwater flow in crystalline rock typical of shield environments occurs almost entirely through the fracture network. The unfractured rock matrix typically has a hydraulic conductivity that is 4 to 5 orders of magnitude lower than that of the permeable fractures and so, for most purposes, can be ignored. Fracture frequency tends to decrease with depth and the major pathways are usually sub-vertical joints in shallow zones. At greater depths, faults or fracture zones are the major flow paths.

## 5.1 The SITE-94 Model

In the SITE-94 model of groundwater flow (Provost et al.1998), a cross-sectional, variable-density, flow-simulation model was developed and tested for various glacial scenarios. At the outset, a regional scale section was chosen to represent groundwater flow across this extensive region, stretching from the coast of north-west Norway, across the Caledonides, south through the Swedish Highlands and across the Baltic Sea into Poland (a distance of ~ 1500 km). The model included the effect of brines, changes in density and viscosity, ice sheet thickness and large-scale topographic gradients.

Provost et al. (1998) made several assumptions in their model that should be considered when assessing the validity of the model's predictions. These include:

1. Hydraulic head is equal to the local elevation.

2. The Fennoscandian Shield is hydraulically conductive to a depth of 10 km. This is justified on the grounds that some permeability was seen in the Kola super-deep borehole to this depth and, because the areal extent of the model is very large (1500 km), as a first approximation the depth dimension is negligible and groundwater flow can be described as two-dimensional in an areal plane (Fig. 5-1a).

3. The flow system is anisotropic. Horizontal permeability ($10^{-15}$ $m^2$) exceeds vertical permeability ($10^{-16}$ $m^2$). These values are used for the entire 10-km depth range of the model.

4. No specific geological structures are included in the model. Therefore, large-scale regional flow-paths are allowed to develop in the crystalline rock (ie. of the order of tens to hundreds of kilometres) and the only boundary conditions for the flow system are those defined by the limits of the maximum extent of the Scandinavian ice sheets.

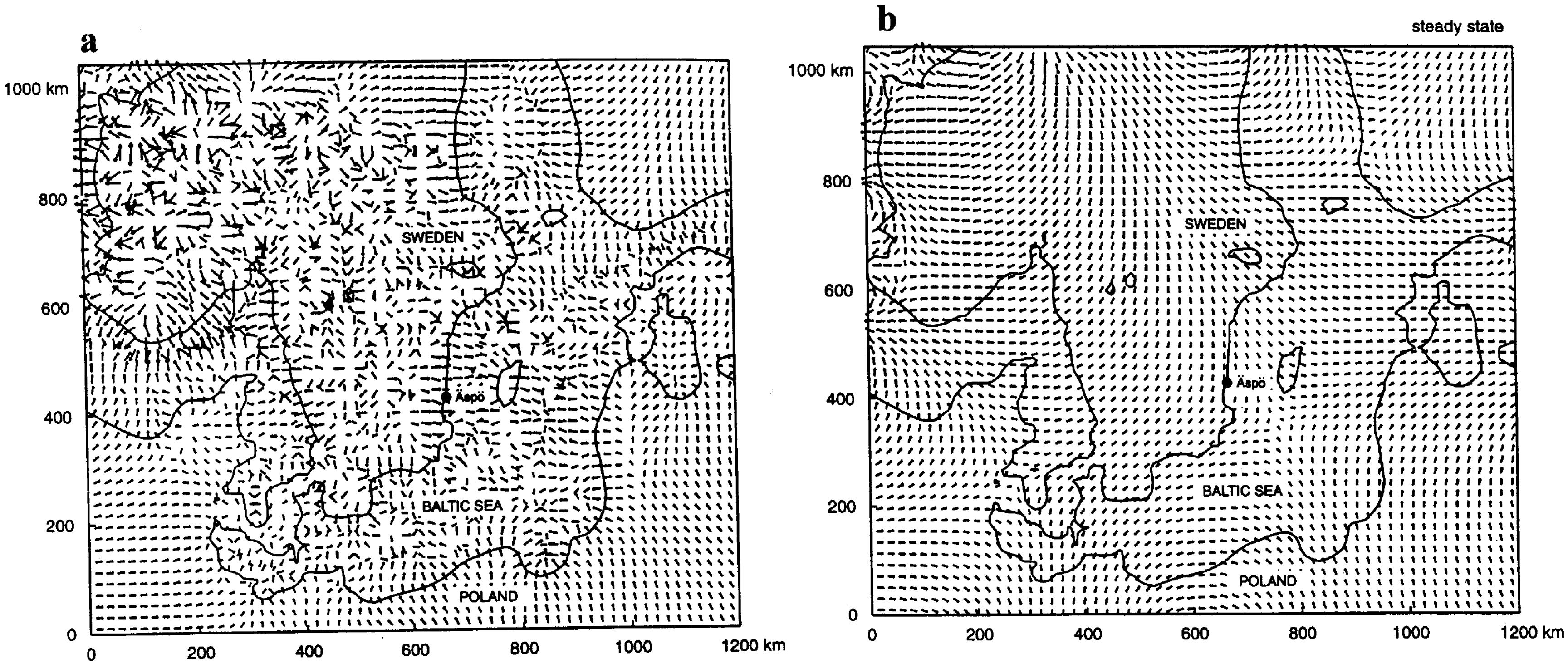



Figure 5-1 *Groundwater velocity field driven by elevation heads a) at the present, b) at steady state (>50,000 years), predicted by the regional flow model of SITE-94 (from Provost et al. 1998).*

5. There is no water in the Baltic Sea. This is justified by the model calculations that steady-state groundwater flows were unaffected by the presence or absence of the Baltic Sea.

6. The line of section through the regional flow field is determined from the modelled velocity field (Fig. 5-1a), when the system is at steady-state with an overlying, cold-based ice sheet (this allows no groundwater recharge or discharge to or from the flow field). Steady-state conditions are obtained in the model after ~50,000 years (Fig 5-1b) and these conditions are used to identify a general northwest-southeast regional flow direction which is close to the expected line of advance of an ice sheet.

7. The varying density of the groundwaters is included in the model. To avoid spurious flows at the outset between zones of different assigned density, the evolution of the salinity of the groundwater (as total dissolved solids, TDS) is controlled by rock-water interaction so that groundwaters attain saturation with increasing contact time. This saturated brine is assumed to be an end-member of the range of groundwater compositions.

Several of these assumptions appear to be overly simplistic and questionable. For instance, the crystalline bedrock is treated as a hydraulic continuum with an anisotropic effective hydraulic conductivity that accounts for all fracture zones. The result of this and the allocation of greater horizontal than vertical permeability is to induce long flow-paths whether they are realistic or not. It is known from detailed studies of the Lac du Bonnet granite in the Whiteshell Research Area, Manitoba, (Ophori et al. 1995, Stevenson et al. 1996) that vertical permeability usually exceeds horizontal permeabilty in the upper ~ 300 m of rock and below this depth there is no evidence for a preferred orientation. Furthermore, fracture zones vary considerably in their properties, both internally and between one another and, rather than inducing regional flows, tend to define the boundaries of a flow system by acting as major conduits to capture and direct groundwater flow.

Although regional groundwater flows over many hundreds of kilometres are believed to exist in fractured sedimentary rocks (Freeze and Witherspoon 1967, Toth 1988), no evidence of such extensive flows has been found in the relatively low-lying topography of fractured crystalline rocks of Precambrian shields, as has been proposed by Toth and Sheng (1994). Domenico et al. (1995) believe that the relatively low-topographic relief of shield areas may preclude the presence of regional flow particularly for the time scale of interest for nuclear waste disposal. The work that has been done on flow systems in crystalline rocks so far indicates that there is no evidence for regional groundwater flows on the scale of tens to hundreds of kilometres let alone the 1500 km flow path of the SITE-94 model.

In addition, the assumption that the bedrock is equally permeable to groundwater flow irrespective of depth within the upper 10 km, is highly questionable. There is abundant evidence (see Fig. 5-2) that permeability decreases with depth in the upper 1 km of fractured crystalline rock and, in all cases studied, decreases to less than $10^{-18}$ m$^2$ at 1 km (Stanchell et al.1996). This value is already two orders of magnitude

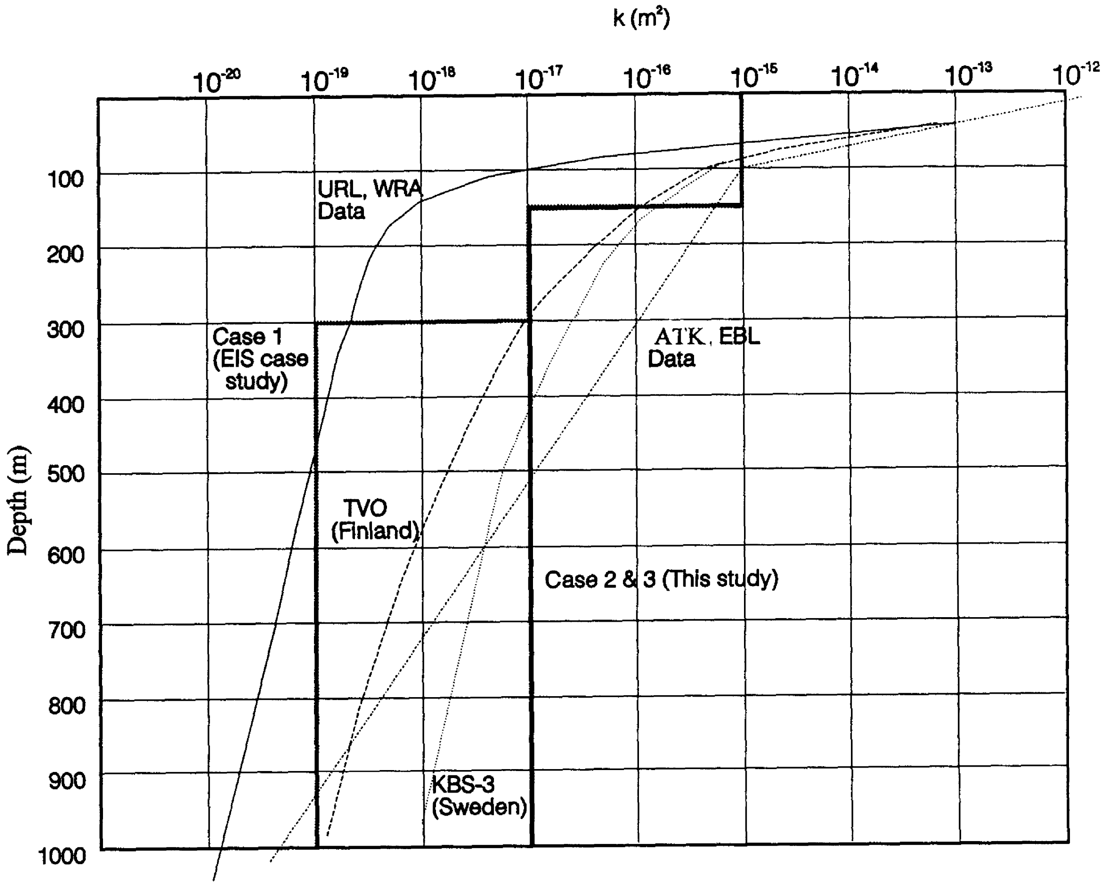



Figure 5-2 *Permeability versus depth data for rocks at AECL's Research Areas (the URL, Whiteshell Research Area, Atikokan and East Bull Lakes) and Swedish and Finnish assessments of Fennoscandian Shield conditions (from Stanchell et al.1996).*

less than that assumed in the SITE-94 model. There are large technical difficulties in measuring permeability at lower values than $10^{-17}$ m$^2$ and so any measurements of this magnitude should be assumed to be maximum values. Provost et al. (1998) examined alternatives to the central scenario in their hydrogeological model and this goes some way to addressing the concerns raised above. They run several variations in the base case, such as changes in basal melting rate, decreasing permeability with depth, different horizontal to vertical anisotropies of permeability, reduction of rock porosity and increase in ice-loading efficiency. These variations are treated rather perfunctorily but show, as might be expected, marked changes in groundwater residence time, salinity, depth of penetration of melt-water and hydraulic head gradients. In particular, the reduction of permeability below 500 m to $10^{-18}$ m$^2$ shows that the flow system has a slower hydraulic response and much less water penetrates to repository depth. These alternatives, which may, in fact, be more realistic than the base case, are not addressed further in the SITE-94 modelling.

A result of the extremely long flow-paths and high permeability of the SITE-94 hydrogeological model is that groundwater flow in the Äspö region during maximum ice sheet thickness is being driven by the topography in northern Norway. This effectively ignores the potential of the local topography (such as the southern Swedish Highlands) to deflect the regional flow from the higher Caledonides to the northwest. In their modelling of groundwater flow-paths under a cold-based ice sheet, Provost et al. (1998) see an effect of flow deflection by the Highlands that is still strong in the model after 10,000 years. The use of a model that obscures the influence of the Highlands for time periods shorter than 50,000 years is questionable, therefore, particularly since it is eventually applied to dealing with groundwater flow patterns of shorter durations, such as a deglaciation.

The model also assumes that there is no water in the Baltic Sea. This effectively strengthens hydraulic gradients beneath Äspö and forces groundwater to flow laterally, and at a higher velocity than if the Sea were present. Provost et al. (1998) state that presence or absence of the Baltic Sea makes no difference to groundwater flow rates beneath Äspö. However, in something of a contradiction, they go on to state (in Appendix A) that ‘had the presence of the Sea been taken into account, the model would have computed zero flow in the region occupied by the Sea’.

The assumption of continuously increasing salinity (as TDS) to saturation conditions implies that sufficient salts are available in the fractured rock adjacent to the groundwater to allow salinity to increase to saturation. These rocks will likely contain saline fluids in pores, discrete fluid inclusions and on grain boundaries. However, these fluids are unlikely to be saturated as this requires concentrations of ~360 g/L in the case of NaCl or >600 g/L for $CaCl_2$. Pore fluid salinities have seldom been observed to be saturated with these salts and values ranging from 0 to ~200 g/L (Nordstrom et al. 1989) have been typically observed in fracture-filling minerals and host rocks. The assumption of saturated fluids in determining the influence of their density for inducing or retarding flow, therefore, is not valid. Provost et al. (1998) actually set a maximum concentration of 300 g/L but this is still probably much larger than that present in the rock, particularly for the groundwaters in the major flow-paths.

Careful examination of the salinity model they use reveals a contradiction between their emphasis on the importance of including salinity in a model to describe how denser groundwaters are developed and flow in the model, and with the actual salinity increases that are calculated. The equation describing rock-water interaction and the generation of salinity is (Provost et al. 1998):

$$\text{Rate (of brine formation)} = k_{mt} (C^{max} - C) \qquad 5\text{-}1$$

where $k_{mt}$ is a rate constant (or mass transfer coefficient) for rock-water mass transfer, $C^{max}$ is the maximum allowable concentration of dissolved solids in the groundwater and $C$ is the TDS concentration in the groundwater at the start. Using values for $k_{mt}$ ($2.0 \times 10^{-15}$ $s^{-1}$) and $C^{max}$ (0.3 kg/L) from Table 1 of Provost et al. (1998) and using a very low value of $C$ (to represent the case of dilute recharge), the rate is calculated to be about 19 mg/L per 10,000 years. This is negligible for contributing to groundwater salinity even if extrapolated over the full period of climate change being investigated (~ 100,000 years).

The first author (A. Provost, pers. comm., 1999) has subsequently stated that this mechanism, although recognised to be ineffectual in building salinity, was introduced 'as a means of generating realistic looking, mutually consistent, initial, near-steady-state ("present-day") pressure and concentration fields, thereby avoiding the spurious flows that can result from simply imposing an "artificial" initial concentration profile'. The inability to model the effects of the variable densities of groundwaters in southeastern Sweden further undermines the credibility of the SITE-94 flow model.

The SITE-94 report examines data on the isotopic composition of groundwaters in the Äspö area and concludes that the general lack of groundwaters with a glacial signature (i.e. depleted $^{2}H$ and $^{18}O$ content) is evidence for rapid flushing of the bedrock down to at least 1 km depth and glacial waters that have been recharged to these depths have now been displaced by warmer-climate, recent groundwaters from the regional flow system. This argument is somewhat self-serving because it assumes glacial melt-waters were once present and then were flushed out and their absence today provides support for the regional flow system. An equally feasible scenario is that melt-water penetration did not take place to the extent and depths proposed and that is why there is little evidence for glacial waters in the subsurface today.

The above concerns challenge the validity and credibility of the hydrogeologic model used in the SITE-94 performance assessment study. The use of a preferred-horizontal permeability, flow-paths of equal permeability to 10 km depth, the lack of any structural controls on groundwater flow, the assumption of the presence of warm-based ice and the negligible influence of permafrost during a glacial cycle essentially guarantees that the model will predict the recharge of surface water to great depth for long periods of time and over extensive distances during the next glaciation. This flow model is used by Glynn et al. (1997 ) to show that dissolved oxygen may be advected to repository depths during full glacial and deglacial conditions.

## 5.2 Hydraulic Heads

An important assumption in the SITE-94 flow model is that hydraulic heads at a repository location may be dramatically greater during deglaciation than at full- or non-glacial times (Provost et al. 1998). These heads are interpreted by Provost et al. (1998) as being able to drive freshwater down to depths of over 1 km in bedrock for a prolonged period. They may also be capable of hydro-fracturing the rock in extreme situations (King-Clayton et al. 1995).

The conditions of hydraulic head and gradient at the base of an ice sheet have been difficult to define and model because of the coupling of ice-sheet dynamics, groundwater flow, bedrock permeability, presence or absence of permafrost and extent of interconnectivity of bedrock fractures. Boulton et al. (1993) and King-Clayton et al. (1995) have proposed that recharge to the bedrock under an ice sheet will be large if bedrock permeability is high. King-Clayton et al. (1995) propose that regional groundwater flow could be high in the Äspö area during the ice advance once basal melting begins, owing to high pore pressures and high hydraulic gradients exerted by the ice sheet well upstream from the Äspö area. They state that (King-Clayton et al. 1995, p.91):

*Groundwater flows may be many times that of the unglaciated or present-day case owing to the excess head gradient exerted by the ice sheet and potential influx of sub-glacial meltwater. Groundwater discharge is less likely to occur within the Äspö region during this stage since water will be forced out towards the margins of the ice sheet, far to the SE of Äspö.*

These statements again indicate the belief that large-scale (regional) flow-paths exist in the Fennoscandian Shield, that their permeability is high and they are not blocked by permafrost or a frozen ice sheet base.

A number of recent papers have been published which consider the internal hydraulic heads of alpine glaciers and those of the basal ice of polar ice sheets. These are summarized below in an attempt to shed more light on this complex issue.

An important question in understanding hydraulic pressures in glaciers and ice sheets is whether sub-glacial water flows in pressurized conduits or in open channels at the ice-bedrock interface. Hooke (1984) has examined this question and concluded that, for most of the glacier, conduits would be at atmospheric pressure except in the over-deepened areas associated with bedrock ridges. This would indicate, therefore, that while apparent hydraulic gradients in a melting ice sheet (i.e., those between the surface, where meltwater may sink in a crevasse, to the point of discharge at the snout of a glacier) might appear to be large, sub-glacial melt-water channels are comparable to rivers flowing along graded valleys or streams flowing through karst conduits and, therefore, will not enhance the penetration of recharge to the deeper bedrock groundwater system. Support for this argument has been given by Fountain (1994) who measured water-level variations in the South Cascade Glacier, Washington, USA. He found that subglacial water pressures were close to local ice-overburden pressures and drainage occurred via a subglacial debris layer into conduits with near-atmospheric pressures.

In a detailed testing of the lower part of the Storglaciaren, a valley glacier in northern Sweden, however, Kohler (1995) concluded that an appreciable length of the subglacial conduit system existed as a pressurized conduit (to within ~ 300-400 m of the snout). The pressurization may be due to closure of ice tunnels by ice flow and was larger than assumed in models of ice dynamics.

Souchez et al. (1993) have found evidence for elevated hydraulic heads and melting of basal ice towards the margins of the Greenland ice sheet. They identified three types of ice near the base (debris-free, fine-dispersed debris-laden, and stratified, debris-laden ice) each of which had characteristic dissolved gas contents and compositions. In particular, the $CO_2$ content of the ice increased towards the margins and exceeded the concentration expected for equilibrium with the atmosphere close to the margin. This was interpreted as indicating that the water is pressurized and may imply that increased hydraulic heads could occur near the margin of an ice sheet during deglaciation.

Basal-ice water pressures have been determined in Ice Stream B of the West Antarctic ice sheet (Engelhardt and Kamb 1997). Boreholes drilled from the ice-sheet surface to the base of the ice stream were found to have hydraulic heads that were 96-117 m below the ice sheet surface. This water pressure was slightly below that required (~ 100 m) to 'float' the ice stream and was found to vary considerably across the ice stream and over time. Nevertheless, water pressures remained close to those expected from the ice-stream thickness at the measurement locations and did not reflect the higher altitudes of upstream glacier ice.

In a study of an alpine glacier (the Haut Glacier d'Arolla) in Switzerland, Hubbard et al. (1995) also drilled boreholes to the bedrock interface and determined variations in water pressure to understand the subglacial water flow pattern and its response to diurnal cycles of melting and freezing. They observed that diurnal changes were localised and this indicated that most flow occurred in large sub-glacial channels but, at times of peak flow, a more distributed network of smaller channels was used which tended to increase basal sliding. Again, water pressures did not exceed the height of the local ice surface.

While local groundwater pressures may be high due to overlying ice thickness, the evidence for steep hydraulic gradients induced in bedrock fractures as a result of overlying ice is limited. It would appear that the addition of 1 - 2 km of warm-based ice over a gentle-topography, crystalline-rock area such as Äspö, would not significantly increase groundwater recharge and flow to depth unless the ice surface was steeply inclined (e.g. the ice margin was close to the repository site) and there was regional interconnectivity of fractures in the bedrock to allow these flows to be transmitted. The hydraulic gradient over repository-scale distances must therefore be large to induce recharge to depth. Except for locations near the margins of the ice sheet, appreciable flow through bedrock fractures would be unlikely to occur. In addition, in the case of the Äspö area, the present land surface would likely be below sea level and flow gradients would be countered by a sea-water head over the discharge point.

The potential for deeply penetrating recharge rests largely on whether an extensive, interconnected fracture network exists in the crystalline rock of the Fennoscandian Shield. Evidence for this has not been found in other Shield environments ( as discussed in Section 4.1 above) and so its application to the Swedish situation in the SITE-94 and other studies must therefore be questioned.

# 6 Summary and Conclusions

Climate changes for the Fennoscandian Shield over the next 100,000 years, will include a prolonged cold event, with permafrost developing over the period 10,000 to 50,000 years, followed by a full glaciation of the Äspö area of Sweden until ~ 70,000 years and deglaciation at about 75,000 years, followed by a return to cold conditions with glaciation again at about 100,000 years. These conditions might be expected to lead to a reduction of groundwater recharge due to extensive permafrost, the presence of cold-based ice (i.e., frozen to the bedrock) and compression of the sub-horizontal fracture pathways due to ice loading.

To assess the performance and long-term safety of a spent nuclear fuel repository in Sweden, in crystalline rock of the Fennoscandian Shield, the SITE-94 study and related reports (King-Clayton et al. 1995, Glynn et al. 1997, Provost et al. 1998) have developed and applied models of groundwater flow and geochemical interaction under various climatic regimes. The flow model that has been developed extends to 10 km depth and has a 1500 km path length (from northern Norway to Poland). The assumption is made that during a glaciation, permafrost is discontinuous and the ice sheet is warm-based (unfrozen) for most of its duration. Under these circumstances, in the Äspö area, the flow model predicts that oxygenated melt-water from the base of the ice sheet will recharge the bedrock over much of the next 100,000 years and, because of high hydraulic heads due to the thick ice overburden, oxidising groundwater will be advected relatively quickly to repository depths or deeper.

Guimera et al. (1998) have examined the geochemical aspects of this scenario in more detail using kinetic water-rock interaction mechanisms and mass transport modelling. They find that penetration of oxidising groundwater to repository depths will be unlikely because of the ability of reduced minerals in the fracture pathways to scavenge $O_2$. They did concede, however, that oxidising conditions could penetrate to repository depths "in the unlikely case of flow through fast fractures.

The work performed in this report has examined the models of SITE-94 and related reports, and that of Guimera et al. (1998), to determine the validity of assumptions in these models and to compare the results with those of recent field observations of modern glaciers and ice sheets. The following conclusions can be formulated:

1. Control of $O_2$ mobility to repository depths will take place mainly by reactions with $Fe^{II}$-bearing minerals in fracture pathways as indicated by Guimera et al. (1998). However, the reaction model used by Guimera et al. assumes a concentration of chlorite of 35% in fracture pathways which may be too high because this value appears to have been determined for low-permeability fractures in the rock at Äspö as well as the major permeable zones where chlorite content is significantly lower. Because most recharge to depth takes place through the higher permeability fractures, $O_2$ penetration may be greater in the bedrock than previously calculated.

2. Discontinuous permafrost (ie. permafrost that is punctuated by melted areas called taliks) has been assumed for the models and, in SITE-94 specifically, a very thin permafrost layer (3 m thick) was assumed, which melted soon after ice coverage. The net result of these assumptions is that the permafrost used in the model does not form an effective barrier to recharge and that groundwater flow continues largely without restriction for most of the glacial cycle. This is despite the fact that several researchers and ice-sheet models have predicted that permafrost will be extensive and deeply penetrating (to at least 300 m).

3. Warm-based ice has been assumed for the SITE-94 study and related modelling over much of the period when ice covers the Äspö area. This is despite abundant evidence (both previously and recently published) which indicates that past ice sheets have been largely cold-based. They only melted after considerable time had passed and a substantial thickness (> 2 km) had accumulated so that the pressure-melting point of ice (~ $-2^{o}C$ for this thickness) was reached. The assumption of rapid attainment of warm-based ice conditions and discontinuous or absent permafrost in SITE-94 effectively allows groundwater recharge to continue throughout most of the glaciation.

4. Hydrostatic heads have been assumed to be high in the SITE-94 study and related modelling due to the combined assumptions of the availability of basal melt-water and the load of ice. Recent literature suggests that heads are high in a melting glacier but they do not exceed the height of the ice surface (ie. they are not driven by distant upstream pressures). The more important parameter for determining if heads are causing groundwater recharge is hydraulic gradient, and this is only likely to be significant in areas where the ice sheet surface is steeply sloping (such as near the margin of an ice sheet).

5. The $O_2$ content of melt-water was estimated as 45 ppm by Guimera et al. (1998) based on a few results for polar ice. Recent studies have indicated that $O_2$ may be strongly depleted in dissolved gases in basal ice and, provided that contact with the atmosphere does not occur, much lower $O_2$ concentrations might be expected in melt-water recharging the bedrock.

6. The flow model and the central scenario used in SITE-94 and related models is believed to be disproportionately simplistic, overly conservative and unrealistic in several of its assumptions. The use of a high and constant permeability for the bedrock, the choice of an immensely long potential flow-path, the lack of structural boundaries on groundwater flow, and the use of an anisotropy that allows preferential horizontal instead of vertical flow, all combine to force the model to predict deeply circulating groundwater flow over a continental scale of distance under almost any climatic regime. Evidence from recent literature indicates that groundwater flow distances much greater than ~ 10 km are unlikely to occur in fractured crystalline rock in a low topographic environment (comparable to that at Äspö) because fracture zones tend to act as major conduits or flow boundaries. Any regional flow that does exist will not be significant over the time period considered for a nuclear fuel waste repository. In addition, the permeability of fractured rock is found to decrease in studies in Canada, Sweden and Finland to

less than $10^{-17}$ $m^2$ at repository depths (~500 m). The SITE-94 central scenario assumes permeabilities that are up to two orders of magnitude greater than this and that these persist, without reduction, to 10 km depth. The token attempt by Provost et al. (1998) to include these alternative scenarios are inadequate to address the importance of this issue.

Combining the SITE-94 flow model with the assumptions of thin or non-existent permafrost, a warm-based ice sheet and high hydraulic heads over much of the next 100,000 years inevitably predicts that oxygenated water will attain repository depths and thus jeopardise the stability of the spent fuel.

The investigations performed here present an opposing view to that of SITE-94. It is believed that a low permeability geosphere with discontinuous flow-paths is a more appropriate model and glacial cycles over the next 100,000 years will not significantly influence the safety of the repository because groundwater flow will probably only increase for a short duration during deglaciation, as the ice margin retreats over the repository site. In addition, the $O_2$ content of the recharging melt-water is expected to be lower than used in the transport model. Reduced minerals in the fracture flow-path will scavenge any $O_2$ that enters the system although the capacity for removal will likely be lower because the abundance of these minerals is believed to be less than previously estimated in SITE-94. Under this revised scenario, it is believed that $O_2$ will not penetrate to repository depths over the next 100,000 years thereby helping to ensure the geochemical stability of the repository. This work reaffirms the ability of the geosphere to act as a barrier to migration of nuclear waste.

# 7 Recommendations

Based on the above discussion and conclusions, the following recommendations are made:

1. The central scenario for climate change in the Äspö area should be redefined and the model rerun using more realistic (but still conservative) assumptions and make use of data describing the influence of permafrost, basal ice conditions, and hydraulic head gradients on groundwater flow. The flow model itself should use values of permeabilities and their depth distributions together with structural features and a regional flow system that better represent the conditions found at Äspö and in the surrounding area. Under these revised conditions, the flow model will probably show that melt-water penetration to repository depths will not occur over the next 100,000 years and that the geosphere is a significant asset in maintaining the stability of the spent fuel.

2. Additional efforts should be made to determine the stability of the geochemical system at repository depth using various geochemical and hydrogeochemical techniques. This evidence might include the presence or absence of isotopically depleted (ie. glacial-derived) groundwater; clay, calcite and iron oxyhydroxide fracture infillings that have stable isotopic compositions and radiometric ages indicating their conditions of formation and relative age. Such evidence would be much more credible than the results of a model with many questionable or overly conservative assumptions. To a large extent, much of this work is underway in the current EQUIP project. An important method that should be used is uranium-series disequilibrium analysis of clays and iron oxyhydroxides from all depth ranges at Äspö. This will indicate whether these minerals have formed more than one million years ago or whether they have been disturbed or even deposited in recent times.

3. The abundance and composition of reduced iron minerals in fractures should be determined on a more realistic and quantitative basis for the water-conducting pathways in the Äspö area. Previous estimates have been made using only three shallow boreholes. Boreholes from more representative depths (up to 1 km) should be used together with careful examination of the core to determine the distribution of $Fe^{II}$ minerals in open versus closed fractures. The mass transport model of Guimera et al. (1998) should be re-run using these more realistic estimates.

4. Examination of glacialogical and related literature should be continued to identify relevant analogues for the disposal repository scenario. Factors to be searched for include 1) the dissolved $O_2$ content of basal melt-waters, 2) the likelyhood of warm- versus cold-based ice during the various phases of a glaciation, and 3) the depth and influence of permafrost and its spatial continuity under an ice sheet, and the resulting impact on groundwater recharge and flow.

# 8 Acknowledgements

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