Section II

Handling the Risks of Nuclear Waste. An Overview of Methods, Problems and Possibilities

3 Some Geological, Geodynamic and Geophysical Investigation Methods Used for the Siting of a Repository in Hard Rock

3.1 Introduction

The purpose of this chapter is to

- Provide an overview of important geoscientific investigation methods,
- Through a critical review, show whether one or more investigation stages or important information is missing in connection with site selection and whether additional investigation methods may have to be developed for the future siting work or in the forthcoming detailed characterisation phase for a repository for spent nuclear fuel.

This review will focus on geological, geodynamic and geophysical investigation methods that are considered to be of particular importance. A systematic review of evaluation methodology and modelling is not included. Furthermore, investigation methods based on Quaternary geology and pure chemical methods are not included in this review.

An important requirement in physical planning today and in the future is that the siting, engineering design and construction of facilities in rock, for example, for the disposal of spent nuclear fuel, should be performed in an environmentally sound and safe manner. This requires comprehensive and accurate information on the properties of the rock. Knowledge is also necessary to ensure an optimum design is achieved and that the construction work can be conducted in a manner that is technically and economically adequate, taking into account the environment and safety. The facility must be able to perform as intended throughout its envisaged “lifetime”, namely, for about 100,000 years in the case of a repository for spent nuclear fuel.

The primary task of the rock in a repository for radioactive waste is to ensure stable mechanical, hydraulic and chemical conditions that are favourable to the durability of the canister and clay barrier. Leaching of radionuclides from the spent fuel must be prevented and delayed as far as possible. The siting of a deep repository in suitable bedrock that fulfils these mechanical and chemical conditions is therefore crucial. To be able to evaluate mechanical stability, knowledge must be acquired of the bedrock and of its ancient and most recent geological history. In order to evaluate the chemical stability, knowledge of existing natural conditions and of the substances in the water that affect the stability of the buffer and canister. Furthermore, knowledge is required of the parameters that affect the evolution of water chemistry such as different groundwater types and their origin and of important reactive processes in the bedrock, in the soil cover and in the biosphere. Geological methodology, in the broad sense of the term, is therefore necessary in order to site the repository in a location that meets the safety objectives.

In the report *Site Investigations Investigation Methods and General Execution Programme TR-01-29* (SKB 2001), a system is described for collecting information on geoscientific conditions during different phases of the site selection work. An overview is provided with a flow chart for soils, rock type distribution, structure, hydrology and geochemistry.

The mapping approach involves direct ***geological investigation methods***, such as observations on exposed rock surfaces (outcrops), excavation and drilling. These methods are highly limited in the vertical direction and the investigation depth cannot be greater than the drilling depth. Problems also occur in the horizontal direction when observations from scattered drill holes must be linked. Fracture zones are often associated with movements in the Earth's crust. These displacement zones have been active during different geological periods. It can be assumed that individual zones are also active today. A characteristic of displacement zones is the patterns that can be seen in different types of data and which have arisen from intensive and lengthy shearing between large blocks of the Earth's crust.

The direct or indirect methods used to describe the geology are *static* in the sense that the conditions and properties of the bedrock are characterised in the present time. The dynamic aspect of geology, namely the evolution over time, requires ***geodynamic investigation methods*** in order to observe changes in specific natural reference structures or reference systems that are established for this purpose. This is of particular importance in the Swedish geological environment with very young sedimenttary deposits (soils) that were formed during and after the ice age, directly on top of very old crystalline bedrock types, which were formed over one billion years ago. The geological evolution, as can be seen in the soils, therefore encompasses a very short timescale (a few ten thousands of years at most) while the evolution that can be seen in the crystalline basement occurred an extremely long time ago. In spite of the fact that the soil stratification only contains traces from a short geological timeperiod, it is the only medium in which recent geological evolution can be observed. In order to predict the geological evolution during the lifetime of a planned nuclear waste repository, the geodynamic investigations must cover a timescale that is sufficiently long. For such studies, more tools exist today than were available when the question of nuclear waste disposal in bedrock was first discussed.

Due to erosion, fracture zones are mostly located in soil-covered depressions in the terrain and are therefore difficult to access for direct observations. Furthermore, the displacement indicators that can sometimes be observed are often very old and indicate the characteristics of the zone under completely different conditions than those that currently exist. Therefore, *indirect* mapping must also be made, based on the interpretation of aerial photography and ***geophysical investigation methods***. These methods are sensitive to contrasts in physical properties which characterise the transition from soil to rock or from one rock mass to another, such as in a fracture zone, but are also related to different water contents and water chemistry. Under favourable circumstances, the geophysical methods provide a systematic depth penetration, down to a depth of several kilometres, which is considerably deeper than can usually be achieved by direct observation. Furthermore, they reflect characteristics in conditions that are undisturbed by the investigation. The mapping methods for fracture zones also include the analysis of digital elevation data and aerial photographs. The bedrock in fracture zones is often disintegrated and can therefore be dispersed by weathering or be easily removed (for example by glacial erosion during an ice age) compared with unaffected bedrock. Depressions in the terrain and topographical escarpments can therefore represent the visible traces of fracture zones and they should be investigated by geophysical measurements in order to confirm whether the extent is significant.

However, there is a difference with respect to what geophysics and geology represent. Both approaches are applied to observe the same material in the same state and at the same time. However, each investigation method is limited to what can actually be measured although this is not necessarily what needs to be measured. Measurements require an analysis by which measurement values are transferred to models. These models are characterised by existing concepts, desired results and, above all, by assumptions concerning what it has *not* been possible to measure.

3.2 Geological Methods

3.2.1 Structural and Rock Mechanical Studies

The mechanical stability of the rock types is determined by the different mineral components and by the structural-geological history of the region. Rock structure can be plastic (for example, folding and foliation) or brittle (for example, joints, faults and crushed zones) (Berglund and Stigh 1998). These phenomena are usually included in the concept of *tectonics* and are a result of the geological evolution of the bedrock and of the original composition of the tectonically deformed material. Tectonic impact is therefore important from a repository perspective and the fracture pattern of the bedrock determines the ultimate design of the repository.

Studies of block tectonic patterns in areas of crystalline bedrock show that two types of bedrock blocks are common. *Shear lenses* are delimited by meandering shear zones and are formed in connection with horizontal block movements at depth. *Figures 3.1* and *3.2* show examples of lens-formed bedrock blocks. It is typical to find a meandering sequence of individual zones connected in a network with *shear lenses* located in between. The entire network can be hundreds of kilometres long and several tens of kilometres wide.

In addition more regular block patterns occur in the uppermost part of the Earth's crust due to the proximity to the free ground surface. *Plinths* are bordered by straight lineaments and are formed by fracturing and block movements in the uppermost part of the Earth's crust (an example of such blocks is shown in *Figure 3.4*). The analysis of seismic surface waves shows that the uppermost 1-2 kilometres of the crystalline crust has a lower seismic wave velocity, which can be explained by the occurrence of fractured and crushed zones (Åström & Lund, 1994). These patterns often overprint older deformations (for example, foliation and folding). The overprint can be discordant (cutting through older directions) although in large fracture zones, the new movements preferably follow the older zones of weakness in the crust. This causes a very complicated pattern which is also difficult to observe in the field since these parts of the zones that are most crushed have been eroded.

It is also difficult to drill through these zones and to obtain a sufficient number of drill cores to study the movement patterns in detail since crushed parts of the rock often result in drill core losses. The better-preserved parts of a shear zone are relatively older and the reference structures, which can be used to determine the movement are normally very old.

Figure 3.1. *Part of the topographic map of northeastern Uppland. The topographically visible Forsmark lens has been interpreted from elevation data. The lens is down-warped in the terrain and goes diagonally across the map view. The clear transverse step in the terrain in the middle of the lens is supposedly a fault with the southeastern block down-faulted. The Forsmark lens is 10 km long and 2 km wide (from Terrängkartan ©, Lantmäteriet Gävle 2004, permission M 2004/3790).*

The mapping of major fracture zones, shear zones and block shapes is conducted mainly by interpretation of digital elevation data and gravity, aeromagnetic, aero-VLF (Very Low Frequency) data. The mapping can be done, both on a regional scale and on a local scale. For small areas, the resolution is increased by the measurements in grids with a 10 or 20-metre point distance. In good conditions, such data allows the dip of the individual zones and the accumulated displacement with time and the direction of the block movements to be calculated.

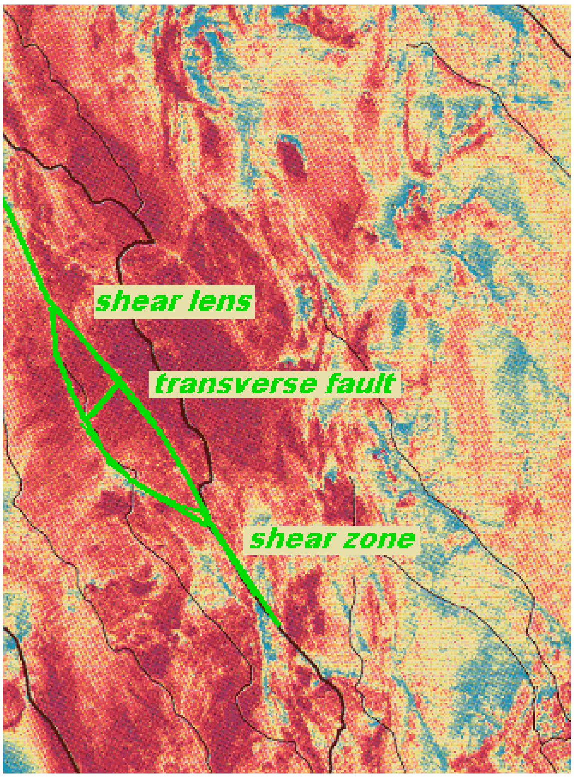

Figure 3.2. *Part of the aeromagnetic map of the inner part of Norrbotten. Shear zones are visible as low magnetic (light red) zones. The Murjek lens in the western part of the map area is interpreted from magnetic data. The transverse low magnetic zone in the middle of the lens is most likely a fault with the southeastern block thrusted over the northwestern block. The Murjek lens is 25 km long and 7 km wide (aeromagnetic measurement, data from Sveriges Geologiska Undersökning, (SGU), permission 30-915/2004).*

Facts

Fracture zone – region (1 m – 10 km) through the bedrock with a large frequency of fractures (0.1 mm – 0.1 m),
Crush zone – region with crushed rock caused by strong deformation,
Shear zone – region in bedrock (1 m – 20 km) with intense deformation caused by shearing,
Movement zone (fault zone) – region in the bedrock where rock blocks have been dislocated,
Shear lens rock block surrounded by shear zones,
Plinth – rock block surrounded by straight lineaments.

Facts

Movement zones in the bedrock described with respect to the *relative* displacement of blocks in the vertical and or in the horizontal direction:
Normal fault – one block is down-faulted,
Reverse fault – one block is pushed over the other,
Thrust – reverse fault at low angle with the horizontal plane,
Horizontal fault (strike-slip fault) – the opposite block has moved to the right (dextral) or to the left (sinistral).

Deformation zones around a rock body (a tectonic lens) can cause the tectonic lens to be less deformed than the surrounding bedrock and future deformation can be taken up in these zones. This requires a major difference in the deformability (competence) of the material in and around the lens. However, in crystalline bedrock, there is usually not a great difference in competence. With further deformation in the surrounding movement zones, the lens can become compressed or pulled apart with a risk of fragmentation. *Figures 3.1* and *3.2* show examples of lenses that have been intersected by fault zones. In certain cases, the lens can be favourable as a repository site if its vertical and horizontal range is adequate. However, rock stresses in such a tectonic lens can be high and this is a disadvantage from the siting perspective. *Rock stress measurements* are performed to investigate these stresses. Geodetic observation networks make it possible to determine the way in which the zones around the lens are active.

The orientation of stress in the bedrock in three dimensions (the *stress field*) can be calculated from registrations of major earthquakes and from measuring the shapes in deep boreholes. There is a considerable difference in depth between earthquakes (usually, deeper than 10 km) and drill hole data (usually less than 2 km deep). The results from such investigations have been compiled in a Neotectonic Map of Norway and Adjacent Areas (Dehls *et al.* 2000). The orientation of the stress field in Norway is different in coastal areas compared with inland areas. Blocks that have a similar stress field have a lateral length of about 250 km. This segmentation follows the coast and the large-scale morphology and the extension towards the northwest of major shear zones with a southeast-northwest orientation. In Sweden, a horizontal principal stress dominates in the southeast-northwest orientation although local deviations occur both horizontally and vertically (Amadei & Stephansson 1997). This orientation is suggested to be related to the pressure from the Mid-Atlantic spreading zone where the North American lithosphere plate separates from the Eurasian plate by about 2 cm per year. There is probably a similar segmentation in large present-day tectonic blocks in Sweden as in Norway. However, data from major earthquakes, which can be used to map this segmentation is lacking. Knowledge of the natural orientation of the stress field is decisive for forecasts of the tectonic evolution in an investigation area. In addition to the natural stress field, rock stresses will occur due to the repository itself and the increase in temperature around the repository that is generated by the radioactive waste. The long-term impact of the natural stress field on the site investigation areas should therefore be modelled together with the induced stress fields that arise as a result of the repository.

3.2.2 Drilling Methods, Borehole Measurements and Drillcore Analysis

The investigation of the properties of the bedrock is made by studying the available outcrops of the rock, with geophysical investigations and drilling. Normal hammer and core drilling is used for investigations in crystalline bedrock. Hammer drilling is carried out using bits with a diameter from 45 to 86 mm. The drilled rock is washed out of the borehole by air or water. The drilling process is logged and a diagram of the sinking of the drill hole is made. The diagram does not provide unambiguous information about the rock quality, especially when the rock is weathered or fragmented. Core drilling is made with rotation drilling and water flushing. The core drilling allows cylindrical drill cores to be recovered from the rock. Mineralogical and petrographical investigations are conducted at the site and on samples and microscope specimens (thin sections prepared by grinding and polishing) in the laboratory. More detailed studies of isotopes, physical properties and mineral composition are made on material from the drill cores. For projects in hard rock at depths greater than a couple of hundred meters, core drilling is the only drilling method used. Results from drill core mapping allow the fracture density of the bedrock to be estimated in several different ways.

The drill hole walls can be examined by TV camera. The camera can be lowered to great depth into water-filled holes. Several methods of measuring different conditions in boreholes are performed with borehole logging methods developed by various prospecting companies.

3.2.3 Rock Mechanics Testing and Rock Materials Testing

It is important to differentiate between rock types at a certain site or in a restricted area and the surrounding bedrock as a whole when the mechanical strength properties of the rock are analysed. The dimension of a rock sample or a small rock volume that is investigated has a considerable impact on the analysis results. The mechanical strength values for a large rock volume may be one-hundredth or less of the corresponding value of a small rock sample. When determining the mechanical strength values, existing stresses and moisture contents as well as the time-dependency that is so vital for deformation must be taken into account (Janelid 1965). Rock engineering leads to changes in stress, which can be of decisive importance for the stability in the short and long term. It is vital to know the state of stress of the undisturbed rock for planning activities. In order to achieve stability in the long term, the changes in stress and deformations that occur in connection with the construction of the repository must also be taken into account. If the factors that from a rock mechanical point of view affect planning are known, the influence of these factors must be determined, by measurements and modelling. The determination of the uni-axial compressive strength of the rock can be tested in the field on drill cores by uni-axial compression tests where the sample is pressed against a blade until a crack occurs (Andersson *et al.* 1984). Laboratories can also test the compressive strength under different load conditions (uni-axial, biaxial and tri-axial), different moisture contents and temperatures. The time factor is of importance since the deformation and creep properties of the rock are of decisive importance for the long-term stability. The stress state of the rock can be determined by analysis of major earthquakes, deformation measurements in large rock volumes or in drill holes. Measurement cells of different designs – from compliant deformation measuring cells (strain gauges) to stiff stress measuring – can be used for this purpose. Modelling can provide invaluable information for planning, for example, by optical stress investigations and load testing on scaled models under known or assumed conditions.

A field method for the determination of the stress state in the bedrock is *hydraulic fracturing* which is based on the measurement of the pressure that is required to create new or reactivate existing fractures. The orientation of the stress field is obtained from an investigation of the orientation of the fractures, which have been activated. In a recently conducted fracturing test in highly fractured crystalline bedrock at the Björkö island in Lake Mälaren, the horizontal stress field was less than previously found in crystalline bedrock areas in Sweden. The largest horizontal principal stress was in the northwestern-southeastern orientation, which is in agreement with other investigations (Ask 2003). Experiments show that the stress field has a local variation, which has also been noted in connection with the classification into tectonic blocks, based on earthquake analysis.

The bedrock characteristics are different in different directions. It is inhomogeneous with discontinuities. The rock varies from hard, massive rock types to rock types that have been weakened by different geological processes and by blasting. The characteristics of some rock material vary from being almost elastic to plastic. In view of this, statistic data must be obtained that is as representative as possible. The mathematical treatment of rock mechanical problems does not only include static stresses and related mechanical strength problems but also dynamic stresses which the rock takes up in connection with different types of deformations (Janelid 1965). If the mechanical strength and stability of the rock is initially inadequate, a certain improvement can be achieved by grouting, rock bolting and concrete injection.

3.2.4 Dating and Evolution Studies

With modern dating, important geological events can be dated. The crystallisation ages of magmatic rock types and the age of metamorphic events can be determined by measurement of isotopes from radioactive decay chains, such as uranium-lead. Zirconium is a suitable mineral for age determination, since it often has a core (which represents the crystallisation age) and an accretion zone around this core (which represents the metamorphic age).

By studying the formation sequence of fracture minerals, a relative age distribution can be obtained (*Figure 3.3*). It is also possible to determine the absolute age of certain fracture minerals with the help of radioactive isotopes and thereby increase the understanding of the tectonic evolution of the area. It is also possible to derive the pressure and temperature conditions under which the fracture minerals have crystallised.

The datable minerals are often considerably older than the intended repository lifetime. Processes that have occurred over the past 100,000 years do not leave any clear measurable traces in the materials that can be investigated. This condition underlines the importance of using geophysical and geodynamic observation networks in connection with site selection and of ensuring that more attention is paid to the youngest geological formations.

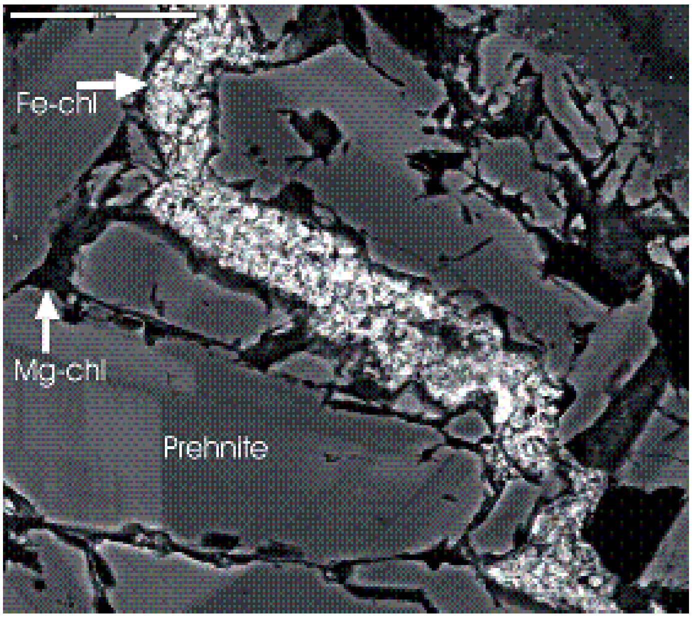

Figure 3.3. *Example of relative ages of healed fractures. (micro photograph). The thin section shows three different fracture minerals. The first generation (prehnite) is broken and the new fractures are filled with the mineral magnesium chlorite (Mg-chl). The youngest generation with iron chlorite (Fe-chl) as fracture fill mineral cuts the older patterns.*

3.3 Geodynamic Methods

Geodynamic processes are reflected in changes in the large-scale topography, the occurrence of land uplift, earthquakes and fault zones. The mapping of the range and intensity of geodynamic processes requires observations of deformations in reference structures or in grids over a longer timescale. However, currently, no measurement data are available which can determine the changes in large-scale topography. In order to achieve these, observations of the land surface and sea bottom changes over long timescales are required, probably for a several decades. Such observations are made in *geodetic* networks which are nationwide and where continuous measurements in comparison with satellites in the Global Positioning System (GPS) are applied. Information about land rise is also nowadays obtained with the same measurement system. Small-scale topography (for example elevated or depressed shear lenses) can indicate geodynamic processes. To study such changes, local geodetic GPS networks must be set up.

The brittle upper part of the Earth's crust is dissected by zones of movement. When adjacent bedrock blocks are displaced, this is referred to as a *fault* (see Berglund & Stigh 1998). Some of these zones were active just after the deglaciation. However, the movement must have displaced a datable geological structure, for example, an esker or a moraine ridge, a measurable distance in order to be able to observe the movement.

The major deformation zones are characterised by deviant topography, deviant land uplift, and the occurrence of earthquakes and systematic displacements of large blocks in the lithosphere. These different characteristics and how they can be studied are treated in greater detail in an ***appendix*** on ***geodynamic processes***.

3.3.1 Measurement of the Change in Gravity

Gravity can be measured with very great accuracy. By repeated measurements at the same sites, slow changes in gravity over time can be studied. The national land surveys in Norway, Sweden and Finland co-operate with measurements that follow the 63rd latitude (Ekman & Mäkinen 1996). The change in gravity is caused by the rising land surface and by the redistribution of masses that occurs at the same time in the lithosphere. This mass flow is controlled by the processes that operate. Measurements of both the land uplift and the change in gravity is an example of how it is possible to acquire better knowledge of geodynamic conditions with several independent methods.

3.3.2 Geodetic Networks

With the Global Positioning System (GPS), positions can accurately be determined in three dimensions. The system is therefore used in networks with GPS stations that register data continuously. If this is conducted over a long period, it is possible to determine how the site with the station has moved (relative to a reference point) due to various geodynamic processes. When the movements of the entire lithosphere plate are excluded from the data set, the differential movements caused by more local displacements can be studied. In Sweden, such a GPS network with 25 stations has been in operation since 1993. Measurement data are compiled at the Onsala Space Observatory. Co-operation is also in progress with nearby Norwegian and Finnish networks. The locations of the stations are determined taking into account land surveying applications. Certain areas may need to be supplemented (for example the Lake Vänern subsidence region and Kvarken) in order to make these data useful for geological applications.

GPS networks can also be designed for local surveys in order to monitor the movement of suspected fault zones or to study how individual tectonic blocks move in three dimensions in, for example, the Oskarshamn region (*Figure 3.4*). The measurement points must then be located on outcropping rock based on a tectonic analysis of the area. Furthermore, the measurement points must be designed so that the antennae can be placed on the point in a unique way (vertically and horizontally) and so that a large part of the horizon is visible. Values are registered for more than about 24 hours. Such local networks exist in Scania (about 50 km between the points), Norrbotten (over a number of shear zones with a distance of a few km between the points) and in the Stockholm area (with 3 points in a number of well-defined tectonic blocks). Personnel from the Royal Institute of Technology (KTH) measure them at intervals of a few years. The results that have been obtained so far are only preliminary. The large data sets mean that special computer codes must be used for the analysis and this is expensive. It is important to maintain the networks and to conduct measurements for as long a time period as possible in order to ensure that clear results are obtained. *Figure 3.5* shows the layout of a local GPS network. *Figure 3.6* shows the national GPS network, SWEPOS, and the change in the Scandinavian land surface.

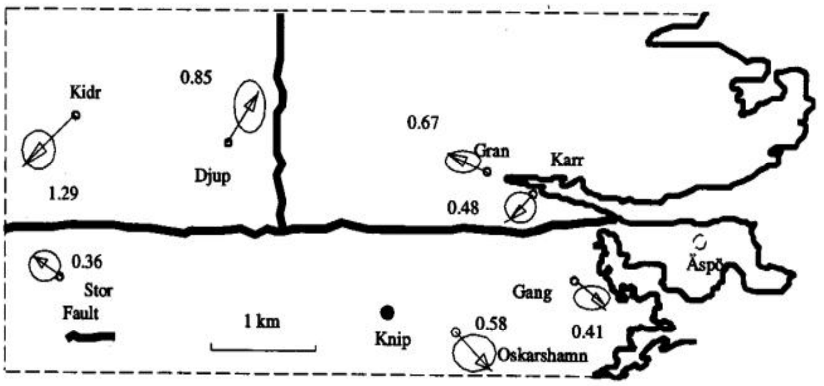

Figure 3.4. *The detailed GPS network in the Oskarshamn region for determination of block movements. The observation points have been given shortened place names. Fault zones are marked with a thick line. Arrows mark the displacement velocity in mm per year that have occurred during the observation period (from Sjöberg et al. 2002).*

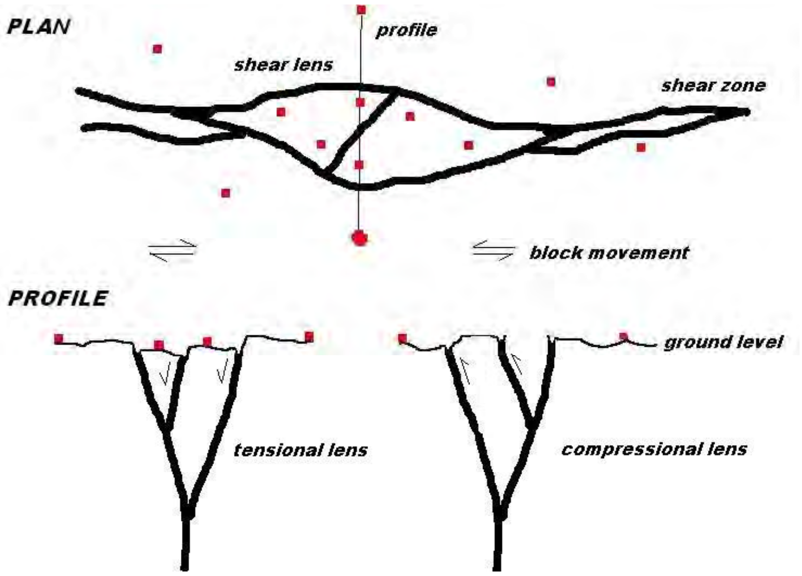

Figure 3.5. Block movements along a shear zone result in the formation of shear lenses. During compression (when the upper block is displaced towards the left), the free ground surface is elevated and during tension (when the upper block is displaced to the left), it is down-warped. The diagonal fracture is a normal fault in tension and a reverse fault in compression. The diagram also shows how a local GPS network can be designed across a shear zone and a shear lens. With three observation points (small squares) in every tectonic unit, the rotation and displacement in three dimensions can be calculated. With one observation point in every unit, only the horizontal movement relative to an external point can be determined.

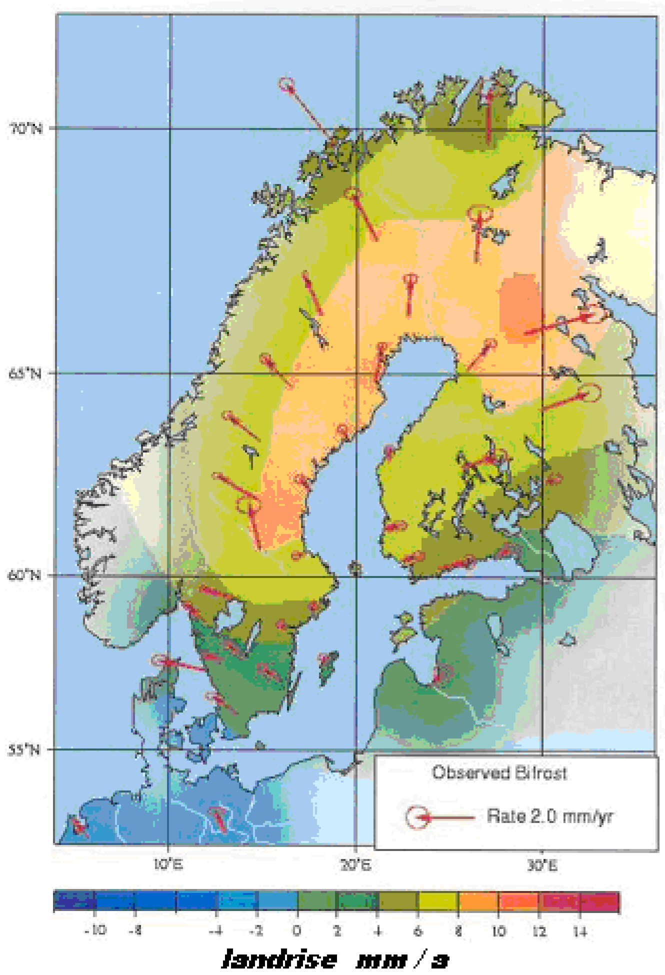

Figure 3.6. Change of the land surface derived from the GPS network SWEPOS in Sweden and Finland. The map shows the land rise with coloured contour intervals. Arrows indicate the lateral direction and the velocity of movement of the different GPS stations. (The ellipses mark the uncertainty range). With longer observation time a gradually more precise determination of the pattern of movement is obtained. (Scherneck et al. 2002).

3.3.3 Seismic Networks

Earthquakes occur when the Earth's crust is broken up by the sudden release of stress that has built up for a long period of time. Such stresses accumulate due to differential movement between crustal blocks along shear zones. The earthquakes are registered in national networks with seismograph stations.

In Sweden, a sparse network with seismograph stations has existed for a long time. These stations have mostly been used to register and analyse major earthquakes, which have occurred at remote sites. At the same time, the considerably smaller earthquakes in Scandinavia have also been registered. The long observation period means that there is an extensive catalogue of Swedish earthquakes to analyse. Furthermore, old observations exist which have been compiled from historical sources. This material shows that earthquakes in Sweden occur in two distinct areas: *Lake Vänern depression* and along *the Swedish coast of the Bothnian Sea and Bay* (especially around Luleå). During a few time-periods, seismograph networks have been established in small regions and a network in the coastal area of Norrland is currently being operated. From the registered data from these various sources, important information has been obtained concerning the position of the earthquakes in the crust and, in the case of large earthquakes, the orientation of the movement surface and stress field as well as the size and direction of the displacement. Seismic observation networks can, like GPS networks, be designed on a more local scale to monitor motions in the bedrock. A new seismic network has been established in 2000 focusing on the shore region of the Bothnian Sea and Bay, SNSN (2003), *Figure 3.7*. So far, over 1,000 earthquakes were registered in this network. A seismic observation network was previously located around the seismically active Lake Vänern depression. It is important to make the seismic observation network comprehensive and to ensure that the co-operation between adjacent countries is further developed. Together with

GPS networks, seismic networks are the only tools for monitoring the effects of the ongoing geodynamic processes.

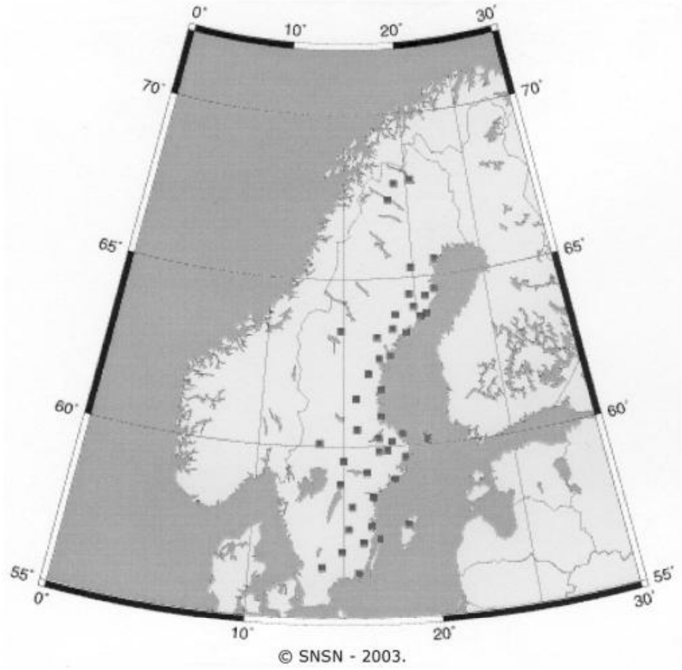

Figure 3.7. The Swedish National Seismic Network (SNSN). The squares indicate geophone stations. (SNSN, Uppsala University, Department of Earth Sciences).

3.4 Geophysical Methods

3.4.1 Problems and Objectives

Knowledge of deep conditions, without having to excavate or drill down to the area of interest, is necessary in order to solve a geological investigation problem. This can be achieved by the use of geophysical methods, which can indirectly provide such knowledge. The purpose of geophysical investigation methods is therefore to conduct systematic measurements of the conditions that cannot be directly observed, to present results in an informative manner and, guided by the measurement results, to construct models of the geological situation. Geophysical measurements are very accurate. However, the measurement results do not always have a unique geological cause, which leads to uncertainty in interpretation and modelling. For a calculated model to be able to reflect reality, several independent measurement data most often have to be combined in the same model. For this reason, several geophysical methods should be combined in an investigation to limit the interpretation alternatives. Furthermore, the physical characteristics of the geomaterials should be measured and used as boundary conditions for modelling. Boreholes and borehole investigations should be planned so that they can be used to calibrate and verify the models that have been created. In SKB (2001), the use of geophysical methods for site investigations has been indicated on the different flow diagrams. During the *analysis* stage, different structures are qualitatively and quantitatively determined. A quantitative determination should contain measures of a structure's horizontal and vertical extension.

3.4.2 Processing and Presentation of Geophysical Data

During the last decade, considerable progress has been made in geophysics, primarily with respect to data processing and presentation. Measurement data from the different methods are treated with different forms of modelling techniques. Inverse modelling is often used, which means that the subsurface structure and characteristics are theoretically varied until they agree with the data obtained. A number of computer codes have been developed for these purposes. Ground penetration radar technology uses advanced signal processing. Even if data from several different methods are currently often combined, a method for co-processing is lacking. In certain cases, methods involving *neural networks* have been tested for these purposes.

Geographic Information Technology (GIT) has developed rapidly and is now a standard tool for the analysis of complex geoscientific data. Several Geographical Information Systems (GIS) are available on the market and they can now be run on PCs, as can 3D presentation programs such as CAD programs. The major breakthrough is due to the development of computer capacity, advanced visualisation technology and a multitude of tools for a combination of data, calculations and analyses as well as methods for decision support. Large data sets can be stored on CDs/DVDs. The increased 3D capacity means that data from, for example, borehole investigations, can be illustrated in three dimensions. It is of interest to present repeated or continuous geophysical measurements and take into account the time factor. Such 4D processing and presentation can be used in monitoring programs during the construction and operation stages to study the groundwater conditions and thermal conditions. It can also be used during the pre-investigation stage, for example, to study groundwater changes in heterogeneous environments during hydraulic testing or for the analysis of tracer experiments.

For model calculations, there is a need for knowledge of the physical properties, (i.e. *petrophysics*) of the geological materials (minerals, soils and rock types). The importance of knowledge of petrophysics is described by the following simplified formula for the relationship between measurement (anomaly A), cause (volume and orientation V, O), distance (d) as well as the contrast in the petrophysical property (K):

$$A = (1 / d)\ K\ f\ (V, O)$$

The measurement of A is determined by traditional geophysics and the description of volume and orientation by traditional geology. The relationship also includes a distance-dependent factor (1/d) which shows another typical condition in geophysics, namely the anomaly's or signal's decrease with increasing distance. The relationship shows that if the distance is great and the volume small, the anomaly will rapidly become so small that it can no longer be measured. The petrophysical contrast, K, is included in the relationship as a factor of great importance. The function of f of volume and orientation is not analytical and must therefore be approximated with mathematical methods.

Facts

The connection between geophysical method and characteristic property in crystalline rock:
Gravity – density – varies from 2.5 to 3.3 Mgm^{-3},
Magnetic field – magnetic susceptibility – varies from 10^{-6} to 10 SI,
Seismic – velocity of sound waves – varies from 5 to 8 kms^{-1},
Radar – dielectric property – varies from 1 to 80,
Gamma radiation – radioactive decay of uranium, potassium or thorium,
VES, VLF, slingram, MT, VLF-R – electric resistivity – varies from 10 to 10^5 Ωm,
IP (Induced Polarisation) – *chargeability* – varies from 0 to 20 %.

3.4.3 Measurements of the Physical Properties of Rock and Soil Materials

The physical properties of geological materials must be known in order to make it possible to interpret geophysical measurements. The measurements can be conducted in situ, directly on the soil or rock type, although in many cases, they are based on samples taken from the geological material. Such sampling must be based on statistical principles, which means that the number of samples is in relation to the variance of the property. It is not enough to have a single representative sample. The sampling of the bedrock is best done on rock cuts or drill cores to avoid the effects of weathering which affect the characteristics of the near surface.

The characteristics that are of interest to study relate to the geophysical method that will be used. Furthermore, the selection of a method is dependent on the geological question to be investigated. A short description of some important petrophysical parameters, the units in which they are expressed and how they can be determined, are presented below.

The *density* of geological materials is dependent on the mineral composition and porosity. The density of composite geological material is the sum of the densities of the components in proportion to their quantities. The density is determined by weighing and determining the volume of samples of the material or indirectly, by borehole logging. Knowledge of the density of the geological materials is required in order to calculate geological models based on gravity measurements. The unit for density is Mgm^{-3}.

The *magnetisation* of geological materials is the sum of *induced magnetisation* and the material's permanent magnetisation. It is also dependent on the occurrence of highly magnetic minerals. In Swedish crystalline bedrock, this mineral is usually *magnetite* and, in certain areas, *pyrrhotite*. The induced magnetisation is dependent on the *magnetic susceptibility* of the geomaterial and the *intensity* of the local geomagnetic field. The *magnetic susceptibility* can be measured directly on the geological materials in situ while the determination of permanent magnetisation requires sampling in the field and measurements in the laboratory. The calculation of geological models based on magnetic measurements requires knowledge of the combined magnetisation of the involved materials. *Magnetic susceptibility* is a dimensionless property and can be expressed as µSI.

The *electrical conductivity* (or the inverse – electrical resistivity) of geomaterials depends on the occurrence of electrically conductive minerals (graphite, magnetite and sulphides) and porosity (and the water that fills the pores). The conductivity can be measured in situ using electromagnetic or electrical methods, on drill cores or in boreholes with different types of logging. The interpretation of geophysical models based on electrical or electromagnetic measurements requires knowledge of the electrical conductivity of the materials. The resistivity is expressed in Ωm.

Geological materials can be electrically charged and this ability to be polarised is dependent on the occurrence of the electrically conductive minerals graphite, magnetite and sulphides. The *induced polarisation* can be measured in the field on drill cores or in boreholes with logging. It is an important method for determining the occurrence of electrically conductive minerals in the near field of the measurement and is therefore used in ore prospecting. The polarisation is dimensionless and is expressed as a percentage.

The capacitance per metre of different materials is called the *dielectric constant* (also called permittivity). It is important for the analysis of electromagnetic measurements in radiofrequency ranges and is to a large degree controlled by the water content of different geomaterials. The dielectric constant is often specified as *relative* to the conditions in a vacuum and is therefore dimensionless.

The velocity of propagation of *electromagnetic waves* varies with different geomaterials. Knowledge of this velocity is necessary in order to analyse radar measurements. Above all, it is dependent on the occurrence of water in the geomaterials.

The velocity of propagation of *sound waves* varies with different geomaterials. It can be measured directly in the field or in drill cores. There is a positive correlation between seismic wave velocity and density. The difference in wave velocity between crustal rock types and the upper mantle is so great that it is used as a criterion for the boundary between the crust and the mantle. Knowledge of seismic wave velocity is necessary in order to analyse seismic recordings. The unit used is ms^{-1}.

The *thermal properties* comprise heat production, thermal capacity and thermal conductivity. Crustal *heat production* is considerable due to the decay of naturally radioactive isotopes. This heat production varies from 2 to 20 μWm^{-3}.

Different geomaterials also have different *thermal conductivity*. This, and the crustal heat production and the heat flux from the mantle, determine how the temperature increases with depth in the upper crust. The heat flux from the mantle is about 60 mWm^{-2} and the *temperature gradient* in crystalline rock is 15-20 Kkm^{-1}. Areas with sedimentary rocks have a somewhat higher temperature gradient (the sediments function as insulation) and areas with bedrock rich in quartz have a somewhat lower temperature gradient due to differences in heat conductivity. Knowledge of the heat conductivity of geomaterials is important for forecasting the propagation of the temperature pulse that occurs during the storage of spent, but still radioactive, nuclear fuel.

The thermal capacity indicates how much thermal energy can be stored in a material in order to obtain a certain temperature increase. Water has a high thermal capacity, which is therefore much greater in water-saturated soils than in crystalline rock. This knowledge is important for the modelling of the thermal conditions surrounding the nuclear fuel and for the modelling of the temperature exchange with the biosphere. The thermal capacity is expressed as $WK^{-1}m^{-3}$.

The *gamma radiation* emitted from the ground depends on the content of radioactive minerals, with the components uranium, thorium and potassium. The content of radioactive minerals varies with the formation and age of the rock types. Measurements can be made from the air, on the ground, in boreholes or directly on samples. The radiation is often expressed as the calculated quantity (in ppm for uranium and thorium and as a percentage for potassium) of the different isotopes at the ground surface.

3.4.4 Strategies in Site Selection

Geophysical investigations for site selection start with an analysis of the site in question in relation to regional geological structures. At this stage, literature studies and map information that cover a large part of the country are required. The sites in relation to areas where geodynamic processes (see appendix on geodynamic processes) can be expected to affect the crust are an equally important and early part of site selection. In order to obtain knowledge of these conditions, geodetic and seismic observation networks are established in and around the area. Since it takes a long time to obtain data on changes, the networks should be set up at an early stage.

In the next step, the local conditions of the area are investigated, with the help of geological and geophysical mapping based on the databases (for example, airborne geophysical measurements), which already exist, and by supplementary investigations on the ground and from the air. At this stage, a large enough environment must be taken into account and the petrophysical properties of rock and soil material must be mapped. The extension of the investigation area must be at least 3 times as large as the extent of the area of interest in different directions. This means that an area that is about 10 times greater than a candidate site should be investigated with relevant measurements in order to understand the structural context of the area in relation to its environment.

Important structures are identified and followed up by more detailed ground geophysical measurements in a grid or in profiles. The methods for studying the bedrock are selected from among those that have suitable depth penetration and can cover the supposed investigation depth with a good margin. The methods to study the soil cover and the location of the upper surface of the bedrock beneath the soil cover are selected among those methods that have less depth resolution, see *Table 3.1*.

Based on these data, investigation drilling is ultimately required in order to do measurements and sampling in the boreholes. The depth of at least one borehole must extend into the saline groundwater region in order to enable the calibration of the electromagnetic methods used to map the transition to saline groundwater. When structures that are important for the stability of the area have been mapped, calculations are conducted of how displacement zones and the rock in between are affected by continued geodynamics and changes in rock stresses. Knowledge from geodetic and seismic observation networks and the existing rock stresses is necessary for these calculations.

The characterisation of soil types and the shape of the rock surface are important input parameters for the study of groundwater flows and groundwater recharge. Therefore, the methods that are applied for site selection cover a wide spectrum and it is an advantage if several methods are used in order to limit the possible interpretations. The selection of methods is also determined by the petrophysical properties that exist in the rock and soil material in an investigation area and by different types of natural or artificial constraints (for example, power lines).

3.4.5 Geophysical Measurement Systems

The various geophysical measurement methods can be classified in different ways. However, the measurement systems and the design of the measurements are similar within methods where measurements are taken from the *air*, directly on the *ground*, or underground in *boreholes*. For each measurement method, the measurement point distance is related to the size of the object. The measurement point distance should therefore be less than half of the size of the object. Corresponding data collection principles also apply to the selection of measurement data from a large database. Methods with a large-scale range are suitable both for general regional surveys and very detailed characterisations. Methods with limited depth penetration are suitable for investigating the soil cover and rock surface.

In order to establish the existence of a contrast, the object's surroundings must also be included in the measurement to an adequate extent. The surroundings included in a measurement should be as large as the specific area of interest. The measurement point distance and the area that is to be measured are directly related to the cost of the measurements.

Table 3.1 below provides an overview of geophysical methods and their applications, depth penetration and scale range. All of the methods are applicable in connection with site selection for nuclear waste disposal. The methods that are best combined partly depend on the geological conditions and, above all, on the petrophysical properties of the rock. Therefore, the starting point should always be the existing regional and local databases that occur in an investigation area in order to design methods and new investigations. If knowledge of the petrophysical properties is lacking, they should be measured at an early, initial stage of an investigation.

Table 3.1. Various geophysical methods and their fields of application.

METHOD (scale range in parenthesis)	**FIELDS OF APPLICATION**	**DEPTH PENETRATION**
Ground based geophysical measurements (1-100 km):		
Gravity (a)	Rock composition, large block movements	10 m – 10 km
Magnetic field (a)	Large fracture- and movement zones in magnetic rocks, block movements, bedrock mapping	10 m – 1 km
Electromagnetic methods (1 – 10 km):		
Slingram	Occurrence of conductive minerals	1 – 50 m
Radar (GPR)	Depth to bedrock and groundwater level	0.1 – 50 m
IP	Occurrence of conductive minerals	1 – 50 m
MT (a)	Vertical distribution of electric resistivity to large depth, level of salt groundwater	10 m – 10 km
VLF	Occurrence of fracture zones and their approximate dip	10 m – 1 km
VLF-R	Determination of soil and bedrock resistivity	10 m – 600 m
Electric methods (0.1 - 10 km):		
VES (a)	Determination of groundwater level, depth to bedrock, soil layering, level of salt groundwater	1 m – 1 km
Seismic methods (50 m – 1,000 km):		
Refraction (a)	Depth to bedrock and groundwater level, occurrence of steep fracture zones	1 m – 50 km
Reflection (a)	Depth to bedrock and groundwater level, layering in sediments, location of low angle fracture zones	0.1 m – 50 km

METHOD (scale range in parenthesis)	FIELDS OF APPLICATION	DEPTH PENETRATION
Airborne geophysical measurements (1 – 100 km):		
Magnetic field (a)	Orientation of large fracture zones in 3-dimensions, block movements, characterization of bedrock	10 m – 1 km
VLF (*)	Steeply inclined water containing fracture zones	10 – 100 m
Drill hole geophysical loggings (0.1 – 10 m):		
Water flow	Water flow in sections of the bedrock	
Electric resistivity	Porosity and occurrence of fractures	
IP	Occurrence of conductive minerals	
TV camera	Orientation of fractures in 3-dimensions	
Radar	Orientation of fractures in 3-dimensions	
Shape of drill hole	Orientation of the horizontal stress field	
Observation networks (1 – 2,000 km):		
Seismic	Location and orientation of displacements in the bedrock, orientation of the stress field	1 – 30 km
Geodetic (GPS)	Displacement and rotation of bedrock units in 3-dimensions in the uppermost crust, land rise	
Hydrological	Precipitation, run off, changes in groundwater level	

(a) Methods with great depth penetration, > 500 m.
(*) The airborne VLF method is direction selective depending on which transmitter that is used for the measurements.

3.4.6 Limitations Due to Terrain and Artificial Objects

All geophysical measurements are dependent on terrain variations. The more variable the terrain is, the greater the effects will be. This is taken into account when planning the measurements as well as in the analysis. With certain methods, the effect of the terrain can be reduced by applying corrections or by inclusion in the model. It is always suitable to study altitude data in parallel with the analysis of measurement data. This can be accomplished particularly efficiently by using digital data and Geographic Information Technology (GIT). Geophysical measurements can be performed on ice over water-covered areas. However, the increased distance to the soil cover or bedrock under the water reduces the signals to some extent. For certain measurements, constant altitude and the absence of topography over the measurement surface is an advantage. Geophysical measurements over water-covered areas make it possible to obtain more continuous information about rock structures. With methods based on electrical conductivity, water (especially seawater) and electrically conductive parts of the soil cover (such as clay) have a strong shielding effect.

Certain geophysical methods are sensitive to artificial (anthropogenic) objects. This particularly applies to electromagnetic measurements where secondary fields from power lines, telephone lines, large fences, pipes and telephone transmitters predominate the natural variations in the area close to such objects. In the case of large power lines, this can extend over several kilometres. Similarly, the environment around active telephone transmitters is severely disturbed. By using electromagnetic methods with controlled signal formation, these disturbances can be avoided in connection with measurements or they can be filtered out during data processing.

With magnetic measurements, it is large iron structures (such as power line poles and sheet metal roofs) instead that disturb the measurements in their proximity. Furthermore, direct current power lines result in magnetic fields that cause local disturbances. When conducting airborne measurements at low altitudes, large power lines and populated areas are avoided by flying at a higher altitude and the natural signals are thus weakened due to the increase in distance. Broad corridors can therefore occur around anthropogenic objects where the use of electromagnetic measurement methods is rendered difficult or impossible.

3.4.7 Airborne Geophysics

Airborne geophysical measurements are performed from satellites, aeroplanes or helicopters and, on the same measurement occasion, several different types of measurements can be performed simultaneously. The measurements rapidly cover large areas and, today, many countries have comprehensive airborne geophysical databases. Satellite-based measurements are internationally available and have global coverage. In Sweden the Geological Survey of Sweden is responsible for the design, processing and storage of airborne geophysical measurements. The compromise between cost and measurement point distance has so far favoured quite detailed measurements, which can be used for many issues. The measurements are also performed at a low altitude, about 50 metres, *i.e.* at a short distance to the geological structures in the bedrock. The measurement point distance is 20 metres along the flight lines but there are about 200 metres between the flight lines. For the analysis of measurement data, it is therefore important to know where the flight lines are situated, especially since modern interpolation techniques normally cannot reproduce the context of structures at a small angle to the flying direction. The measurement data are presented on maps in the scale 1:50,000, which follow the map sheet division of Sweden. Digital extracts of an optional geographical area can also be obtained from the database.

The measurements performed from airborne surveys in Sweden comprise *magnetic total intensity*, *gamma radiation* (represented as the equivalent content at the ground surface of the natural radioactive isotopes, potassium, uranium and thorium) and *electromagnetic secondary fields* from long-wave (VLF) transmitters. These measurements can be used for many purposes, including mapping of the extent of the rock types under the soil cover and under water, the mapping of large fracture zones in the bedrock, the investigation of radon risk (in soils, the bedrock and groundwater) as well as for prospecting for mineral deposits.

Radar measurements that are conducted from satellites and aircraft can be considered to be geophysical measurements. They are usually very detailed and are conducted in several frequency ranges. The direct geological use is for the mapping of fracture zones as well as the determination of soil water content. In areas with heavy vegetation and intensive forestry, the geological information is hidden by the traces left by the methods of land cultivation (like property boundaries and ploughing grooves) that are clearly visible in the radar measurements. The measurements are not dependent on cloud cover and the effects that occur from cloud and shadow on the ground in connection with aerial photography or satellite scanning in the visible wavelength spectrum can thereby be avoided.

Facts

Measurement direction, measurement spacing, measurement altitude
Airborne measurements – in east-west or north-south direction, 200 m distance between flight lines and about 20 m between measurements, elevation about 50 m above the terrain,
Ground based measurements – selected direction, distance between measurements and measured lines 5 – 20 m,
Regional gravity measurements – irregular net of measurements with 0.8 – 5 km spacing,
Profile measurements – oriented at right angle to the direction of the structure, measurement spacing 1 – 20 m.

3.4.8 Ground Geophysics

Purpose and Access to Data

Geophysical ground surveys have been carried out in connection with the prospecting of ore and industrial minerals and the mapping of gravel deposits as well as for specific investigations within the regular soil and rock type mapping at the Geological Survey of Sweden. From an early stage, ground geophysical surveys have been conducted for groundwater prospecting in Sweden and abroad. In recent decades, ground surveys have had an increased importance within different types of environmental studies.

Geophysical ground surveys, in connection with *underground construction*, such as for a repository for spent nuclear fuel, aim at developing a geological-tectonic model of the studied soil and rock volume. Furthermore, the aim is to increase knowledge of the composition and thickness of the soil cover, the physical properties, fracturing, water content and boundaries between different rock types. The ground surveys are non-destructive, but require considerable interpretation. In general, the resolution decreases with increased depth penetration. To study conditions at a depth of 500 metres, considerable changes in the physical properties (or large structures) are therefore required in order for them to be detected at the ground surface. Also near surface changes in the horizontal direction must be less than those in the vertical direction.

The measurements are performed on the ground, either in an irregular grid over large areas, in a systematic grid over a limited area or as profiles. The measurement point distance is determined by the purpose of the measurement and can vary between 1 metre (detailed profiles) and 5 kilometres (regional nationwide surveys). Measurement data are presented on maps or in profiles. For certain types of measurements, national databases exist that are managed by the Geological Survey of Sweden.

Regional, nationwide surveys currently exist for *gravity*, with point distances that vary between 0.8 and 5 kilometres. The measurements usually follow the road network. On large lakes and near-coastal sea areas, measurements have been conducted on ice.

Seismic surveys are sometimes conducted on a regional scale, especially in sediment-covered areas and for special projects. No complete overview exists of where such measurements have been conducted or of which company or institution that is holding the results.

Measurement Methods

The most important geophysical methods for investigations of structural conditions of the bedrock in the form of fractures and fracture zones are seismic, magnetic, electrical and electromagnetic measurements. Some of these measurement methods also provide information on rock types and rock type boundaries. All methods are sensitive to horizontal near surface variations while changes at depth are much more difficult to detect. It is also easier to detect steeply dipping structures and rock type boundaries than to map horizontal fracture zones and boundaries. In the case of refraction seismic and electrical methods, depth penetration also requires that instruments be arranged over long distances. *Figure 3.8* shows a series of measurements with different methods over a large shear zone in Norrbotten and how the results of the magnetic measurement can be used to determine the dip of the zone (*Figure 3.9*).

Seismic surveys are used to detect structures in the bedrock. The surface structures, for example the occurrence of fractured bedrock, especially steeply dipping fracture zones, can be interpreted from *refraction seismic surveys*, where the refracting part of the sound wave is followed by registering the time until it reaches the geophones that have been set up. The seismic signal velocity is considerably reduced in crushed rock. It is difficult to detect horizontal low velocity zones with refraction seismics. On the other hand, refraction seismics can be used to advantage to determine the depth to the bedrock under the soil cover. This has been carried out in connection with many large construction projects, for example, along the Bolmen tunnel where more than 200 km of refraction seismic profiles were evaluated (Stanfors 1987). With *reflection seismics* the part of the sound wave that is reflected at the interface to a material with a deviating sound velocity is measured. The method requires heavy equipment and more powerful computer processing. Therefore, it is more suitable for local surveys. A major advantage of reflection seismics is that the method is one of the few that can be used to identify rock structures with low angle to the horizontal at great depths, for example, horizontal fracture zones (Andersson 1993, Cosma *et al.* 1994). This is of decisive importance for the siting of a repository since it must be possible to take into account low angle structures when determining the position for the rock volume that can be taken into consideration. Seismic methods have also been used during the construction of underground facilities (Tunnel Seismic Predition, TSP) to predict the conditions and determine the need for reinforcement (Sattel *et al.* 1996).

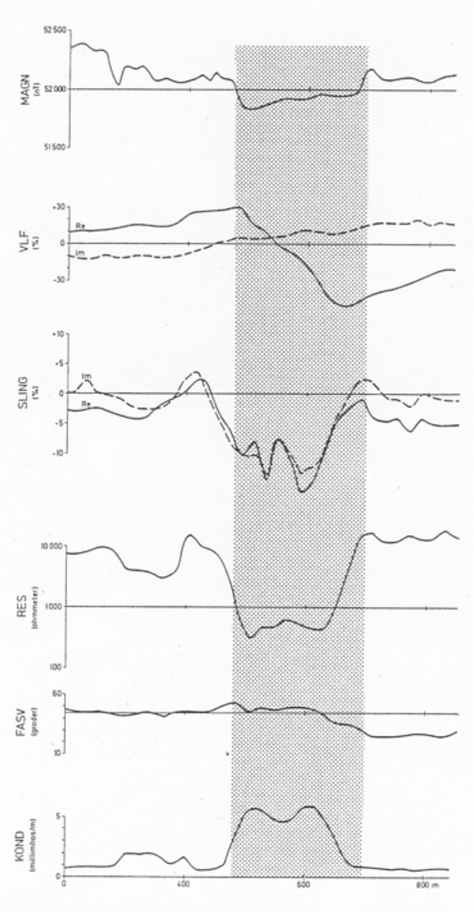

Figure 3.8. Example of the geophysical response from a large shear zone (marked with grey shading) in Norrbotten. The different methods show a clear response above the zone. From top: Magnetic (MAGN) low anomaly over the zone due to oxidation of magnetite, VLF horizontal component giving a typical anomaly over the zone, Slingram (SLING) gives a negative anomaly, VLF resistivity (RES) shows low resistivity, the phase angle (FASV) varies very little over the zone, and at the bottom a high conductivity (KOND) anomaly is seen. (from Henkel 1988).

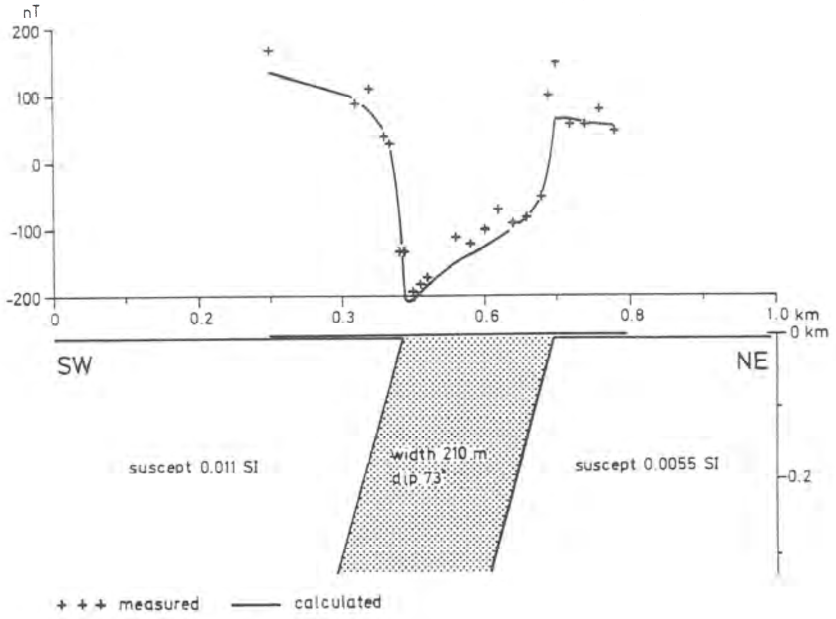

Figure 3.9. The magnetic anomaly seen on top in the previous figure has been used to determine the dip of the zone, which is 73 degrees towards the southwest (from Henkel 1988).

Ground Penetrating Radar (GPR) utilises similar reflection principles as reflection seismics but is based on the propagation of the electromagnetic waves through the ground. A reflection is obtained when the radar wave hits an object with deviant electrical properties. Today, pulse radar systems are often used, whereby electromagnetic pulses are directed by a transmitter antenna into the ground and a receiver antenna registers the reflected signals. The time delay for the reflected signals is measured in nanoseconds, 10^{-9}s. In recent decades, the method has become increasingly important for the mapping of superficial soil and rock layers and has been used for studies of soil layer conditions and geological evolution (Widén 2001, O'Neal & McGeary 2002, Helle 2004). GPR has also been used to study tectonic zones, both active (Rashed *et al.* 2003, Slater & Niemi 2003) and older neotectonic zones (Dehls *et al.* 2000, Tirén *et al.* 2001). Like with reflection seismics, the method can be used to identify low angle tectonic structures and is therefore of importance for studies of the near surface, generally more fractured rock, *Figures 3.10* and *3.11*. In soil-covered areas, the depth to the bedrock and flow-promoting structures in the soil-rock contact zone, that are important for groundwater recharge, can be mapped, *Figure 3.12*. GPR can also be used continuously during the construction phase, directly from the underground facility, to predict fractures, rock boundaries and other rock structures in order to establish reinforcement needs and to map the effectiveness of pre-grouting (Cardarelli *et al.* 2003).

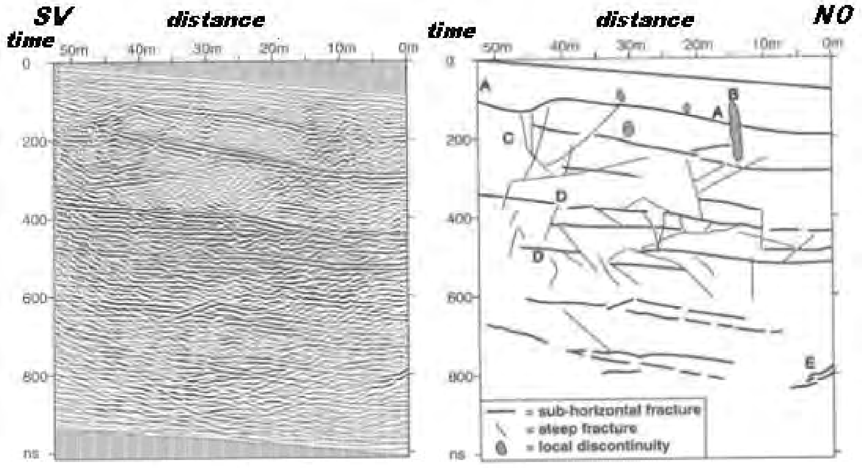

Figure 3.10. Example of an interpretation of fractures in the near surface bedrock using GPR measurements (Grasmück 1994). From the time scale, the depth to the reflecting structures can be calculated if the velocity of the signal in different geomaterials is known.

Figure 3.11. Example of a low angle fracture zone in the Forsmark area. Such fracture zones can be detected in near surface locations with ground penetrating radar and at larger depth with reflection seismic measurements (photograph by Kaj Ahlbom 2003).

If the crystalline bedrock is magnetic, magnetic measurements from the ground (or from an aeroplane) can be used to map large fracture zones. These zones are always low magnetic due to mineral alterations and can also be mapped beneath the soil cover and in water-covered areas. Through model calculations and with knowledge of the magnetisation of the surrounding bedrock, the dip of the zones can be established.

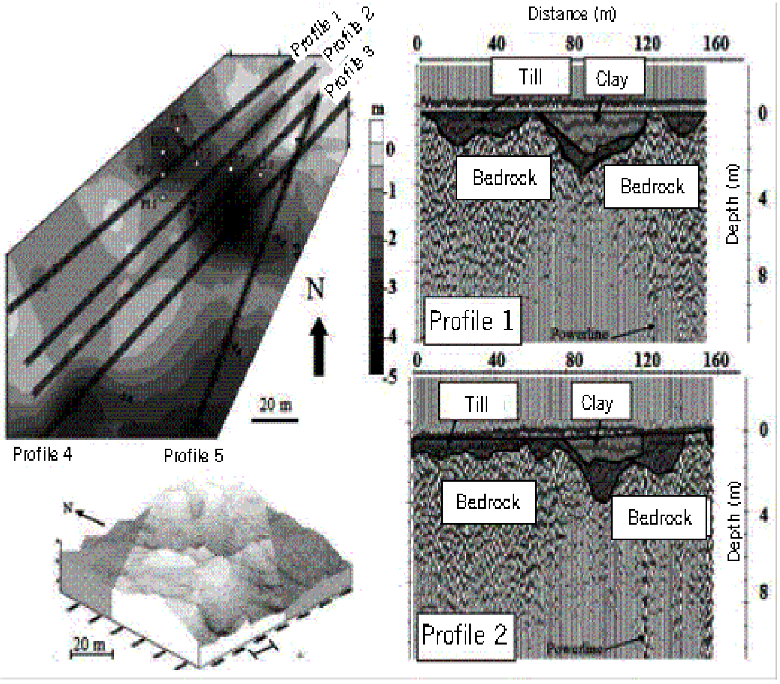

Figure 3.12. Example of interpretation of the depth to the bedrock based on GPR measurements in an area in southeastern Sweden (Olofsson et al. 2004).

Electrical measurements are based on electrical fields created in the subsurface between two current electrodes. The extent of the current field depends on the distance between the electrodes but is also affected by the conductivity of the soil cover. By measuring the voltage with potential electrodes, the *apparent resistivity* can be calculated. The resistivity depends on how the electrodes are arranged as well as on lateral and vertical changes in the electrical properties of the subsurface. Through *inverse modelling*, where the electrical properties of the subsurface and the thickness of the soil cover are varied until an agreement is reached with the measured values, an interpretation of the structure of the subsurface and its electrical properties is obtained. Most often, a multi-electrode system (Continuous Vertical Electrical Sounding, CVES) is used today with a large number of electrodes arranged along lines or in a network together with a computer that determines which electrodes are to be current and which are to be potential electrodes. Through inverse modelling, the resistivity distribution of the ground can then be two- or three-dimensionally calculated. Electrical measurements have become important for the mapping of soil and rock stratification and in determining groundwater surfaces. Other important applications are for environment-related investigations and for environmental control (Bernstone & Dahlin 1998, Aaltonen 2001), *Figure 3.13*. If fixed electrodes are set up in the ground, the method can be used for long-term monitoring, for example, around landfills where pollutants often have a high salinity (Aaltonen & Olofsson 2001) or for the monitoring of climate-related ground moisture conditions and groundwater levels.

Most of the multi-electrode systems occurring on the market only allow sensing to a depth of about one hundred metres. An interesting application of geoelectrical surveys is to map the occurrence of saline water at great depths with a several kilometre-long electrode separation. This is an excellent complement to deep drilling. However, in coastal areas, it may be difficult to avoid the short-circuiting effect of seawater on the measurements.

The *chargeability* of the ground can be measured by *induced polarisation* (IP). The method is based on a current field created over the ground that causes polarisation to occur in the subsurface. When the field is turned off, this polarisation continues for a certain time and can be measured. The method has a considerable potential for studies of polluted soils, like dispersed salt pollutants or oil spills (Dahlin & Leroux 2002, Sjögren 2004). Even without external current fields, a weak polarisation occurs due to the mineral content in the ground and the electrolyte properties of the ground fluid. The measurement of this natural *self potential* (*SP*) with sensitive non-polarising electrodes can, in the same way, be used in connection with ore prospecting and pollutant mapping. The method has also been used in connection with near-surface tracer experiments in rock (Nimmer & Osiensky 2002).

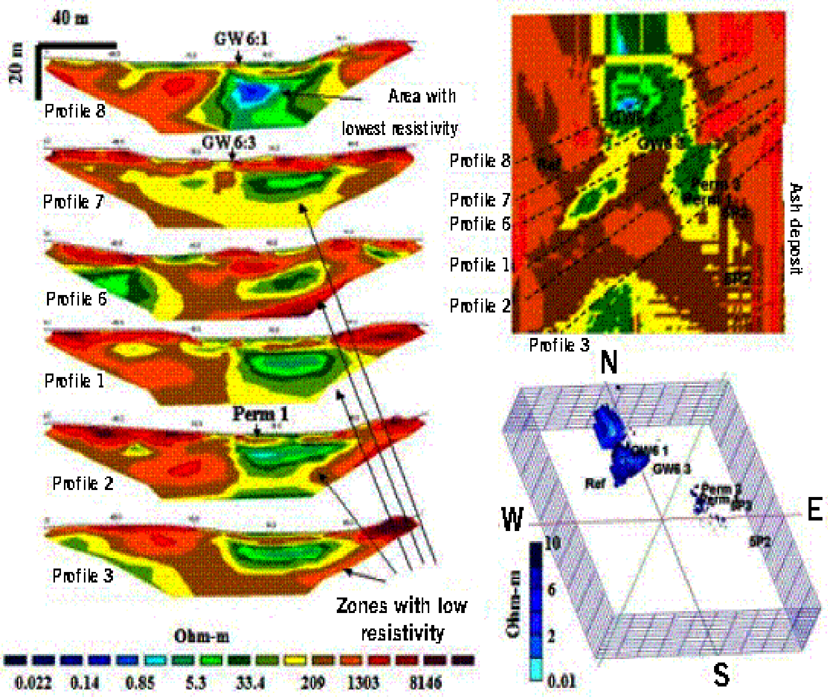

Figure 3.13. *Resistivity measurements for analysis of leacheates spreading from a waste deposit. The measurements are made with CVES technique and are modelled two and three-dimensionally, respectively. The results are presented as profiles (left), horizontal sections in the near surface bedrock about 12 m below the ground (upper right), and as three-dimensional low resistivity zones (lower right) (Olofsson et al. 2004).*

Electromagnetic induction means that a current field is created in electric conductors in the ground with the help of an external electromagnetic field. The secondary electromagnetic field, which occurs, can be measured at the ground surface. There are several frequency-controlled methods that either use natural electrical currents (MT measurements), in the frequency range from 10^{-4} to 1 kHz, or currents induced by radio signals (VLF measurements) in the frequency range from 15 to 25 kHz. Electromagnetic methods are also designed for fields created by a mobile transmitter (slingram) in the frequency range from 5 to 15 kHz. In general, the depth penetration is determined by the frequency of the electromagnetic field and the conductivity of the ground. There is always a high contrast in resistivity between unaffected bedrock and fracture zones in crystalline bedrock. VLF measurements are therefore an effective method of mapping fracture zones on land areas (but not over water-covered areas).

In recent years, electromagnetic measurement methods have been developed to describe the distribution of electrical resistivity down to a depth of several kilometres in the bedrock. A summary of VLF and MT methods is provided in Oskooi & Pederson (2004). With magnetotelluric measurements (MT), natural electrical currents occurring in the bedrock are used, *Figure 3.14*. Penetration depth is up to 10 kilometres and the measurement is conducted so that anisotropic conditions also can be investigated. The observation times are up to 12 hours. Interference from transmitters for mobile telecommunication and power lines can, in most cases, be filtered out. With measurements in a coarse network, three-dimensional electrically conductive structures can be identified. With this method, the depth at which the transition to saline groundwater occurs can be determined.

Electromagnetic measurement methods have been used for ore prospecting, investigations of water-bearing fracture zones in the rock and for studies of pollutant dispersion. The methods are based on complex theory. New instruments are developed for environmental applications, for example, EnviroMT (Bastani 2000).

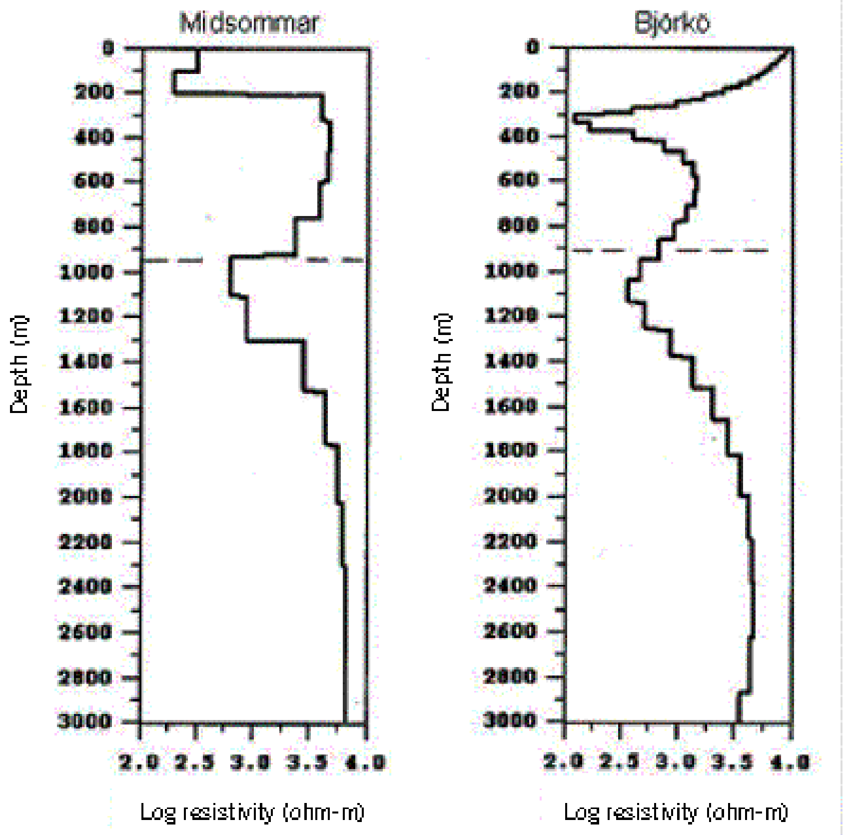

Figure 3.14. *Two models of the resistivity variation with depth based on MT measurements (from the islands Midsommar and Björkö in Lake Mälaren). (From Oskooi and Pedersen 2004).*

3.4.9 Borehole Geophysics

In most deep boreholes in Sweden, measurements have been conducted with different types of geophysical logging in order to determine the variation of the measured characteristic property with depth. Such measurements can be conducted at intervals that vary from 0.1 to tens of metres. Accurate methods have been developed to correlate the depth values obtained in connection with different measurements. However, in Sweden, there are very few boreholes with a depth exceeding 1 kilometre. Consequently, knowledge is lacking of where the transition to saline groundwater occurs in the bedrock and how deep the fracture zones extend into the upper part of the Earth's crust. Boreholes that are deeper than 2 kilometres in crystalline bedrock only exist in the central part of the Siljan structure in Sweden. In borehole measurements, the sensing distance in the horizontal direction is very small, from decimetres for certain methods and, under favourable conditions, up to tens of metres, in the case of borehole radar for example. The measurement of resistivity in boreholes only gives relative values and calibration is required to obtain values that are representative for rock types. This problem is treated in Löfgren & Neretnieks (2002).

The following types of borehole measurements are common and can be used in connection with site selection:

Methods that characterise rock types and rock type boundaries

Gravity measurements
Measurements of magnetisation
Induced polarisation (IP)
Gamma radiation (several different methods)

Methods that identify fracture zones and the occurrence of water

Measurement of electrical resistivity (several different methods)
Radar measurements
Water flow measurements
Measurement of borehole shape (calliper)
Temperature measurements
Video photography

Methods that identify stress and temperature conditions

Response to pressure changes
Measurement of borehole shape
Temperature measurements
Gamma radiation measurements (several different methods)

The methods are often used in combination and several sophisticated measurement probes have been developed for the electrical methods including a choice of different electrode configurations, *Figure 3.15*. Measurements involving radar, borehole shape and photography also provide information on the *orientation* of structures that have been detected. Water flow measurements are performed in limited sections, in response to pressure changes. The response in the bedrock displacement is measured and identified during the test period with the help of local seismograph networks. The connection between borehole data, which is very detailed (dm-scale) and continuous, with surface data that are dispersed (1 metre to 10 metre scale) and incomplete, is a difficult problem. The difficulty is related to how local phenomena can be distinguished from those that cover a large area. The distance dependence of the measurement method implies that the spatial resolution rapidly decreases with distance from the borehole and with depth in ground based surveys. The problem cannot be resolved with more frequent or more sensitive measurements – instead, more boreholes are needed – which however change the properties of the bedrock in an unfavourable manner.

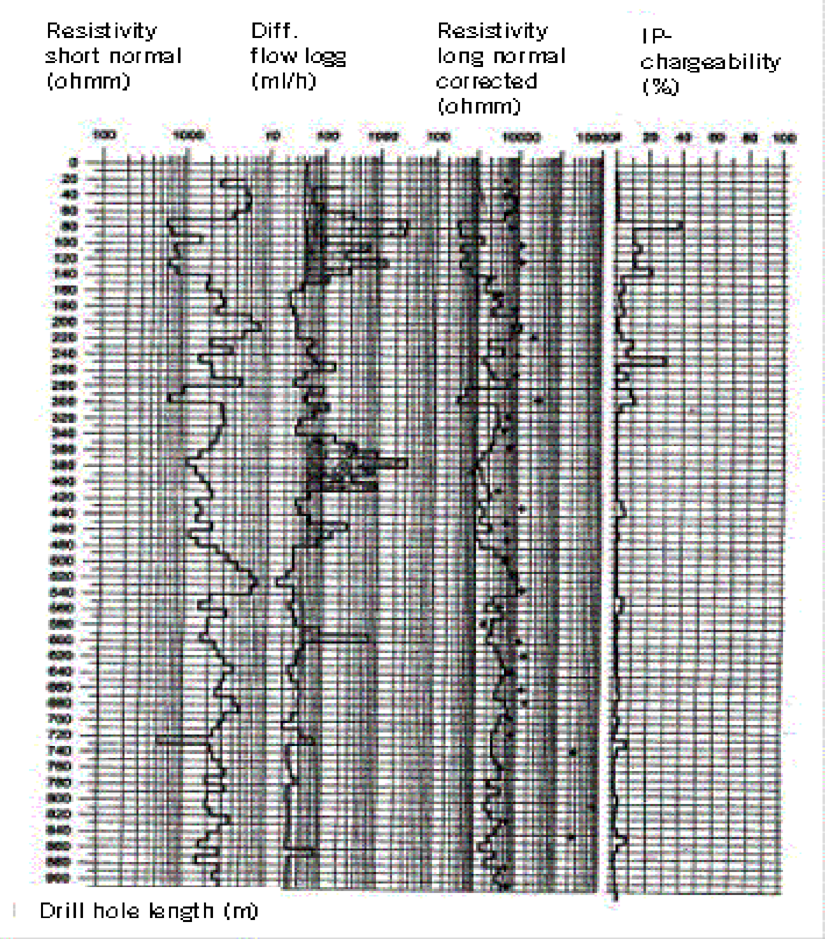

Figure 3.15. *Example of logging data from a 900 m deep drill hole (on Björkö in Lake Mälaren) with electric resistivity (two methods, short normal and long normal), water flow, and induced polarization (IP) generalised over 10 m intervals. (Sträng 2003). The variation in resistivity reflects the fracture frequency. Higher water flow indicates open fractures and increased IP effect indicates sections where electrically conductive minerals occur.*

3.4.10 Databases at SGU and the Swedish Maritime Administration

The Geological Survey of Sweden (SGU) holds a large number of databases with *digital* and *analogue* geoscientific information. They are available for research, prospecting, geotechnical and environmental investigations. A licence is required and fee must be paid to use the data. Digital data can be obtained for an optional geographical area and they are often directly suitable for analysis with Geographical Information Technology (GIT). Furthermore, geoscientific maps with different degree of detail occur all over the country.

Regional surveys are conducted systematically by SGU while detailed surveys are conducted by geological consulting companies, prospecting companies and geoscientific departments.

Queries about seismic measurements should be primarily directed to SGU. Several international projects, with the aim of determining the structure and thickness of the Earth's crust, have been based on seismic investigations covering many 100 kilometres.

In connection with prospecting for ore, large areas in Västerbotten and Norrbotten have been mapped in great detail with several types of ground geophysical measurements. These are documented at SGU.

The Swedish Maritime Administration handles bathymetric data, often with a high degree of detail, over coastal areas and inland lakes where shipping occurs. These bathymetric data can be used in the same way and in combination with altitude data to study the occurrence of fracture zones in water areas (or their extension from land to water areas). Such a combined study has, for example, been conducted for the Lake Vänern basin (Isaac 1992) and southern Björkfjärden (Chuang 2003).

3.5 Conclusions

Geological Methods

Swedish crystalline bedrock is a complex heterogeneous medium, formed by geological processes during more than 2,000 million years. Many of these processes are ancient but affect the stability and safety of a repository for radioactive waste. Other processes are active and gradually change the geological conditions. The geological situation in the shield with a very young soil cover over a considerably older crystalline bedrock means that the time-period during which ongoing geological changes can be studied is much less than the planned lifetime of the repository. Therefore, it is important to conduct systematic studies in the young geological deposits with respect to the effects from land uplift, earthquakes and fault movements. Many of these dynamic processes will probably continue for a foreseeable time in the future.

Geodynamic Methods

Much research has been conducted on the induced geodynamic changes that will occur locally through the construction of a repository for spent nuclear fuel, also coupled to the heat exchange that will occur between the repository and the environment. However, natural geodynamics has not attracted the same interest. Nevertheless, several indicators show that systematic movements between crustal blocks occur continuously. Such movements have been quantified in some cases with geological methods, with measurements in the Global Positioning System (GPS) and by analysing major earthquakes. The movements are located in limited areas or zones. These zones can be mapped geographically and in terms of depth using geological and geophysical methods. The movements along the zones are relatively small and, therefore, long observation periods are required in order to determine them with certainty.

The displacement zones function as part of a larger regional context and, at present, there is insufficient information on the way in which they function locally. Consequently, local systems must be built up and measured for a long time. The ongoing deformation is one of the key problems in making forecasts of the bedrock stability. Therefore, knowledge of the position and extent of the zones (horizontal and vertical), the velocity and direction of the motion, the function of the zones in time and their function in the regional and plate tectonic deformation is necessary. How plinths and shear lenses react to changes in the stress field should therefore be modelled. Such modelling can be made for observed structures in the investigation areas and their regional context.

The methods that must be further developed to provide such knowledge are both direct and indirect, for example measurement techniques with GPS and seismograph networks, age determinations of minerals and geological observations, in order to provide increased knowledge of the structure and performance of the lithosphere. The existing geodetic and seismic networks should be nationwide and co-operation over national boundaries should be developed to create databases that can be used for several geoscientific purposes. The GPS networks which were previously set up should also be measured in the future in order to obtain time series that are as long as possible. This also applies to measurements of the change in gravity.

Geophysical Methods

A large number of geophysical surveys have been conducted so far or are planned in connection with site investigations prior to the construction of a repository for spent nuclear fuel. The measurements have had different purposes and scales, from general airborne surveys to detailed characterisations in boreholes. In many cases, the aim has been to build up a geological/tectonic model over the area or to predict geological and tectonic changes during the construction phase, such as during the construction of the Äspö tunnel. A limited amount of research work has been conducted on the possibilities of transforming measurements to input variables for chemical dispersion models. Certain development efforts have been made, for example, resistivity measurements for determination of diffusion in massive rock (Löfgren & Neretnieks 2002).

Geophysical surveys are a very valuable tool since, in principle, they are the only methods that provide non-destructive measurements of the rock volume where the repository will be constructed. Therefore, it is of great importance that surface-based geophysical surveys should be conducted at an early stage. A combination of several methods with high data point density and determination of the physical properties of the geomaterials and control drilling is necessary in order to reduce uncertainty when interpreting the measurements.

A combination of magnetotelluric (MT) measurements, which have great depth penetration capabilities, and reflection seismic measurements are tools to determine the depth to saline groundwater and the occurrence of deep fracture zones. Such measurements must be made systematically and with sufficient coverage of the investigation area and its surroundings. Ground geophysical surveys are of particular importance, such as surface-covering measurements with ground penetrating radar in order to map the soil stratigraphy, the soil thickness, the contact zone between soil and rock and the fracture conditions of the near surface rock, as a basis for calculating groundwater recharge in the bedrock.

In the case of geological and geophysical investigations, the extent of the measurements must be large enough to include an adequate environment outside the actual area of interest is included. This also applies to water-covered areas. The extent should be about 3 times greater than the area of interest in all directions. The total investigation area should thereby be about

10 times greater than the area of interest. Reflection seismic and radar investigations should be conducted in a systematic manner in the entire investigation area in order to map the occurrence of low angle fracture zones since these can not normally be observed in outcrops or with other geophysical methods.

3.6 Appendix: Geodynamic Processes

The mapping of the extent and intensity of geodynamic processes requires observations of deformations in reference structures or in observation networks over a long time-period. The deformation of the crust over the past million years and the ongoing deformation of the crust are referred to as *neotectonics*. In Sweden, this particularly refers to the deformation that occurred after the last glaciation. The deformation that is currently in progress is almost unnoticeable over short time-scales (decades). However, it can accumulate to a considerable size during geological time periods (millions of years). Due to the constant movement of the global lithosphere plates, all parts of the crust are affected all over the Earth. Along the plate boundaries, the deformation is very great and causes severe earthquakes and volcano eruptions. Inside a lithosphere plate, the deformation is considerably less, hardly noticeable and does not cause catastrophes. The boundaries of our lithosphere plate (the Eurasian plate) are located in the middle of the North Atlantic (*Figure 3.16*), in the Arctic Ocean, along the Japanese island chain, Indonesia, the Himalayas, Anatolia, the Alps and the Atlas mountains. The Eurasian plate largely comprises continents and moves due to the growth of the Atlantic ocean crust by about 1 centimetre per year (i.e. the same order of magnitude horizontally as the land uplift). Several active deformation zones are located in this plate, for example, the graben system that stretches from the North Sea via the Rhine valley to the Rhone valley. Areas also exist in our vicinity that can be suspected, on good grounds, to be active deformation zones, such as the mountain belt, Lake Vänern and the Bothnian Sea and Bothnian Bay. The deformation zones are characterised by anomalous topography, anomalous *land uplift*, the occurrence of *earthquakes* and the systematic displacements of large crustal blocks. These characteristics, as well as how they can be studied, are treated briefly below.

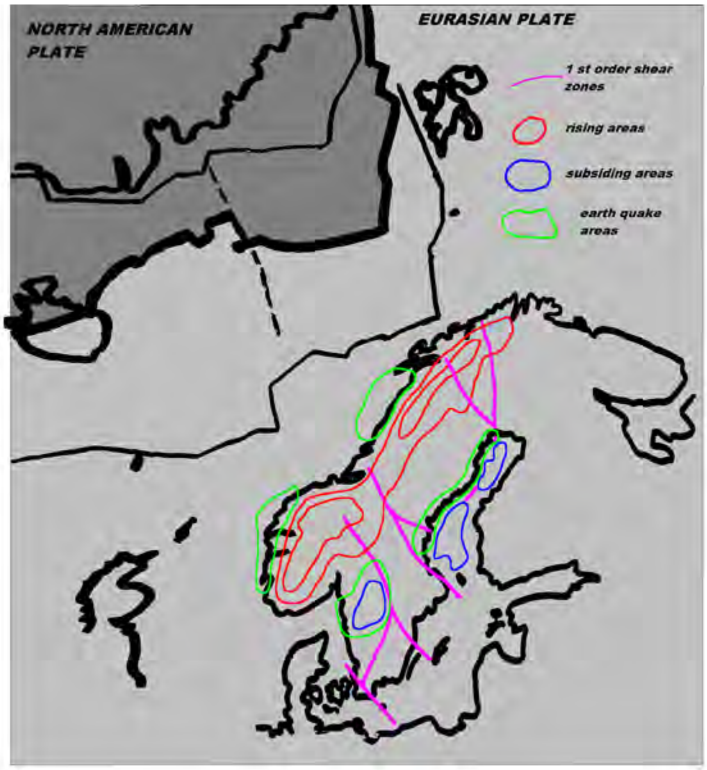

Figure 3.16. *The plate tectonic situation of Sweden. The Mid-Atlantic ridge is our nearest plate boundary between the North American (dark grey) and the Eurasian (light grey) lithosphere plate. The large-scale geomorphic regions are the rising area in the Scandian mountain belt (red lines) and the parallel, about 400 km to the east located, areas of down warping (blue lines) around Lake Vänern and the Bothnian Sea and Bay. Generalised areas with earthquakes are outlined green. The first order shear zones are marked with purple lines (Henkel & Roslund 1994).*

On a more detailed scale, neotectonics manifests itself by the occurrence of fault escarpments with varying height (from less than 1 metre to over 20 metres), landslides, slumping and liquefaction structures in soil layers, boulder fields and caves, and as displacements of glacially shaped outcrops. Some of these phenomena can also occur due to other geological processes and the connection between occurrence and cause requires extensive mapping over large areas. The terrain shapes that are associated with young fault zones were first discovered by studies of aerial photography in areas located over the highest shoreline and which, therefore, have not been exposed to seashore erosion. Landslides can be detected in the same way (and run the risk of erosion if they have been exposed to shore erosion). Therefore, these neotectonic indicators have so far mostly been found in northwestern Norrbotten and it is still unclear whether they indicate an anomalous neotectonic active area or whether they have also occurred in areas below the highest shoreline. Considerable new road construction work has created road cuts with sections that are suitable for detailed studies of disturbances in the soil stratification. These are common phenomena in mobile sedimentary environments and a determination of the boundaries of tectonically caused structures requires regional mapping and dating of the sediment stratification. Boulder fields and caves can be related to earthquakes but also occur through frost heaving and frost erosion. Minor displacements of glacially shaped rock outcrops across fractures are a clear indication of block movements that have occurred after the formation of the surface. An interruption in such surfaces, where one block is missing, occurs when the missing part is transported away by ice. In Mörner (2003), a thorough neotectonic interpretation of a large number of observations is made, which is connected to paleoseismic activity. Many of the observed phenomena are located in time to the deglaciation phase, which was a period of relatively major changes in the stress field.

Knowledge of the ongoing geodynamic processes is important for judging the long-term stability of a nuclear waste repository. Without actual measurements of geodynamic changes and knowledge of the underlying processes, predictions of future changes are based on assumptions.

3.6.1 Topography

Tectonic processes and erosion primarily cause the variation in the large-scale topography in Scandinavia. Elevated areas cannot last for long geological time-periods due to continuous erosion. In the same way, sinks cannot last for long periods of time due to continuous *sedimentation*. On the other hand, areas without significant topographical variation are relatively stable (for example, the Småland highlands and Finland). By studying elevation data (in the form of topographical maps or digital elevation data), older erosion surfaces can be reconstructed (Lidmar-Bergström 1988). However, only exceptionally can the age of these be determined. The large-scale topography in Scandinavia is young. Studies of sedimentation in the sea areas off the coast of Norway indicate increased sedimentation starting about 5 to 10 million years ago and distinct uplift areas have been identified in the mountain belt (Riis & Fjedskaar 1992). In Lake Vänern and the southern and northern parts of the Bothnian Sea and Bay, there is still no occurrence of thick young sediments despite ongoing mountain belt erosion and the fact that materials are being transported to these sinks by rivers. The cause of the young topography is not yet known. However, due to the large dimensions, it is likely to be related to plate tectonic processes. The natural evolution of the young oceanic lithosphere in the North Atlantic is gradually leading to the formation of a subduction zone at the edge of the continental lithosphere where the oceanic lithosphere is being submerged below the continental lithosphere. At a distance and in parallel with the subduction zone, a subsiding region would develop, as the lithosphere is dragged apart by an opposing current in the upper mantle. However, at present, no measurement data are available for determining changes in the large-scale topography.

In order to obtain such data, observations of the land surface and sea bottom changes are required over long periods of time, probably decades. Such observations are conducted in nationwide geodetic networks where continuous measurements are used against the satellites in the GPS system. Small-scale topography (such as elevated or depressed shear lenses) can also indicate geodynamic processes. *Figure 3.17* shows a cross-section of the Earth's crust from northwest Lofoten to central Finland (Henkel & Lund 2004). *Figure 3.18* shows a profile over a major shear zone in Värmland.

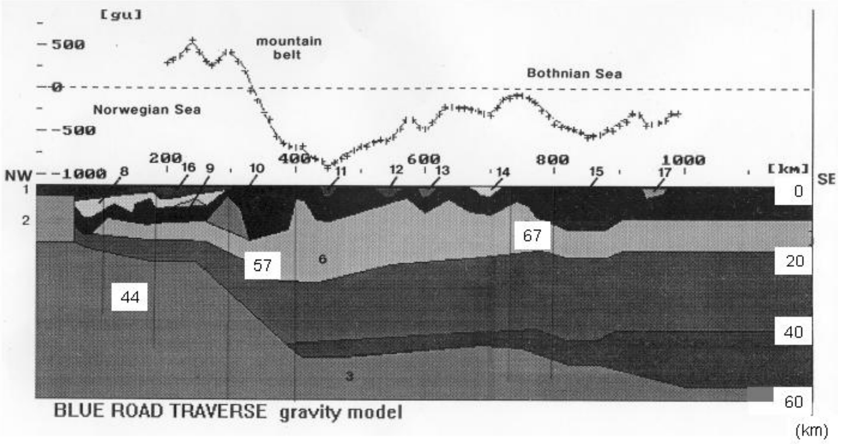

Figure 3.17. *Section through the Earth's crust from northwest of Lofoten to central Finland. The model is based on a combination of refraction seismic and gravity data. To the northwest, the change from the oceanic crust to the continental shelf is seen. The thickness of the crust is larger under the mountain belt and increases to its largest value in Finland. The number of earthquakes, which occur in three distinct zones (delineated by vertical lines), is presented with numbers in their approximate depth location. (The gravity anomaly is in gu (= 0.1 mgal). The small numbers mark areas of different density.*

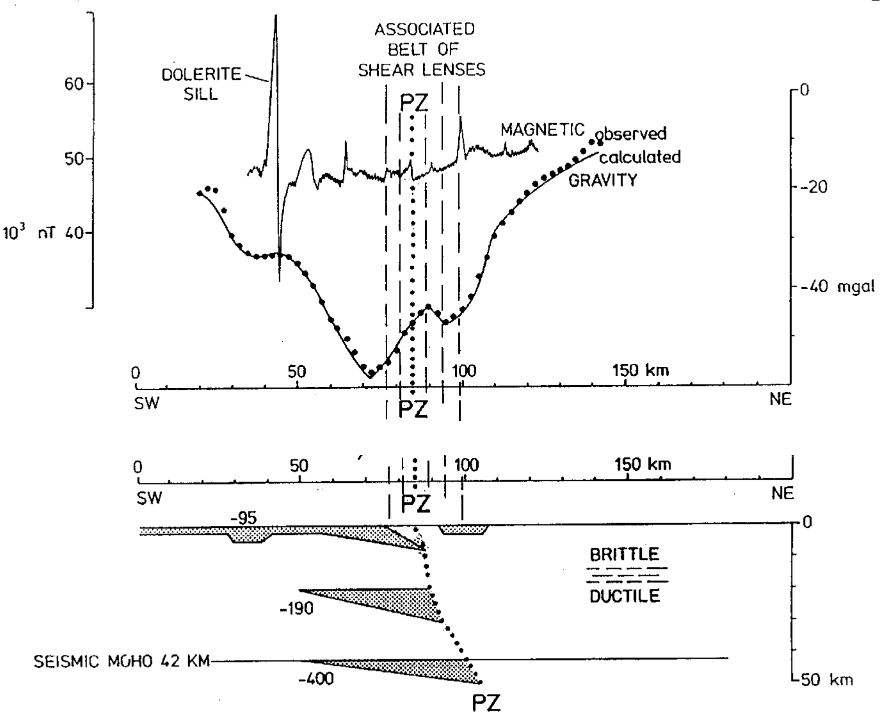

Figure 3.18. Profile across one of the first order shear zones in Värmland (marked as PZ), which cuts through the entire crust (here about 40 km thick) (Henkel 1992). In the upper part of the diagram magnetic measurements are shown which are the basis for calculations of the near surface dip of the zone. In the lower part of the diagram gravity measurements are shown which are the basis for calculations of the extent of the zone through the Earth's crust. It also shows down warping of different layers in the crust from west towards the zone, which dips steeply towards the east.

Facts

Plate tectonics – the deformation of the lithosphere due to heat convection from the Earth's mantle,
Lithosphere – the uppermost shell of the Earth, which is displaced as a unit in plate tectonic processes. Its thickness is about 250 km in central Scandinavia,
Mantle – the region between the lithosphere and the Earth's core, about 3,000 km thick,
Earths crust – the uppermost part of the lithosphere outside the mantle, the thickness in central Scandinavia is about 50 km,
The boundary between the crust and the mantle is the Moho (the Mohorovicic discontinuity).

3.6.2 Land Uplift

In Scandinavia, land uplift is well-known, has been measured for centuries and can be seen in the young geological deposits that are reshaped in shore zones and which have gradually ended up at increasingly higher levels above the present sea surface. Land uplift is currently the greatest, about 9 mm per year, in the vicinity of Umeå. It is zero close to the boundary of the crystalline shield against the surrounding sediment covered areas. In the area just south and east of the shield (i.e. in northern Germany, Denmark, the southernmost part of Scania, the Gulf of Riga and around Lake Ladoga) a slight land subsidence occurs. The cause of the land uplift is attributed to the deglaciation that occurred over 10,000 years ago. However, there are several indications that other forces are active. The extent of the land uplift is not compatible with the extent of the ice. Local deviations in the land uplift also exist (known as *differential* land rise) and there is a considerable difference in the land uplift gradient between the western and eastern part of the land uplift area. Areas with a significant deviation from the general land uplift (which can be connected to the deglaciation) show relative *rising areas* by more than 1.5 mm/year in the mountain belt and relative *subsiding areas* with corresponding deviations in the southern part of the Bothnian Bay and Sea (see *Figure 3.16*, areas within the red and blue lines respectively). The subsiding area extends further towards the northeast beyond the northern part of the Bay of Bothnia. It also includes northern Uppland (Fjeldskaar *et al.* 2000). Investigations of shoreline displacements in northeastern Uppland (Hedenström & Risberg 2003) show that the exponentially decreasing land uplift has turned into a linear trend about 5 500 years ago. This is a strong indication that other processes besides isostatic compensation after deglaciation are active.

The land uplift can be measured by recurring levelling of fixed points and such measurements provide the basis for knowledge of the present land uplift. However, since 1993, traditional levelling measurements have been replaced by data obtained from 25 permanent GPS reference stations placed all over Sweden, known as the SWEPOS network. After a long observation period, the relative movement of the observation points, horizontally and vertically, can be calculated from the measurement data.

3.6.3 Earthquakes

Earthquakes occur when the Earth's crust breaks apart due to the sudden release of stresses that have built up over a long period of time. Such stresses accumulate due to differential movements between crustal blocks along shear zones. In Scandinavia, only mild earthquakes occur and earthquakes with a magnitude of 5 or larger (on the Richter scale) are rare. The earthquakes occur in the brittle part of the crust at an average depth of about 18 kilometres along certain zones and in a few limited areas. The present-day seismic active areas in Sweden are especially the Lake Vänern depression and the Swedish coast along the Bothnian Sea and Bay (see map in *Figure 3.19*). The earthquakes are registered in a network of seismograph stations, which operate over a long period of time. The more dense the network, the better it is to locate and characterise also minor earthquakes. Registered data from average and large earthquakes can be evaluated with regard to the orientation of the stress field and the movement surface, its area and the displacement that has occurred. Such evaluations are conducted by the seismology division (Department of Earth Sciences) at the University of Uppsala, which also since the year of 2000 operates the new seismograph network, SNSN. So far, over 1,000 earthquakes have been registered in this network, *Figures 3.19* and *3.20*.

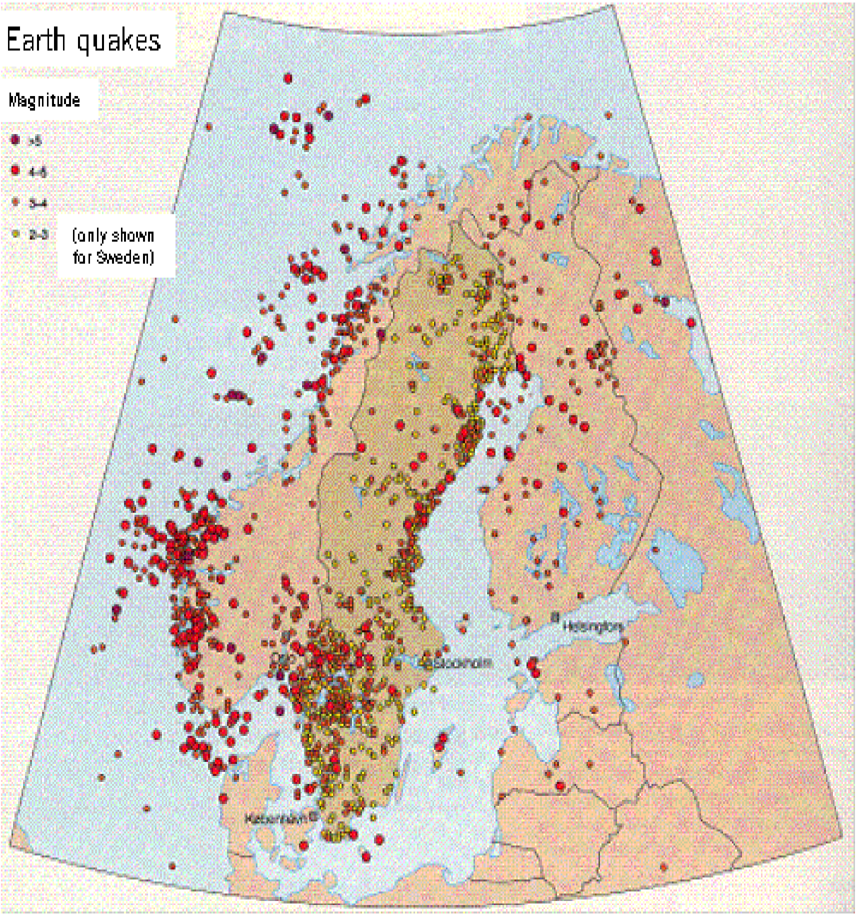

Figure 3.19. The occurrence of earthquakes in Scandinavia. The increased number of earthquakes in the four regions marked green in Fig. 3.16 is clearly visible (from Sveriges Nationalatlas ©, Lantmäteriet Gävle 2004, permission M 2004/3790).

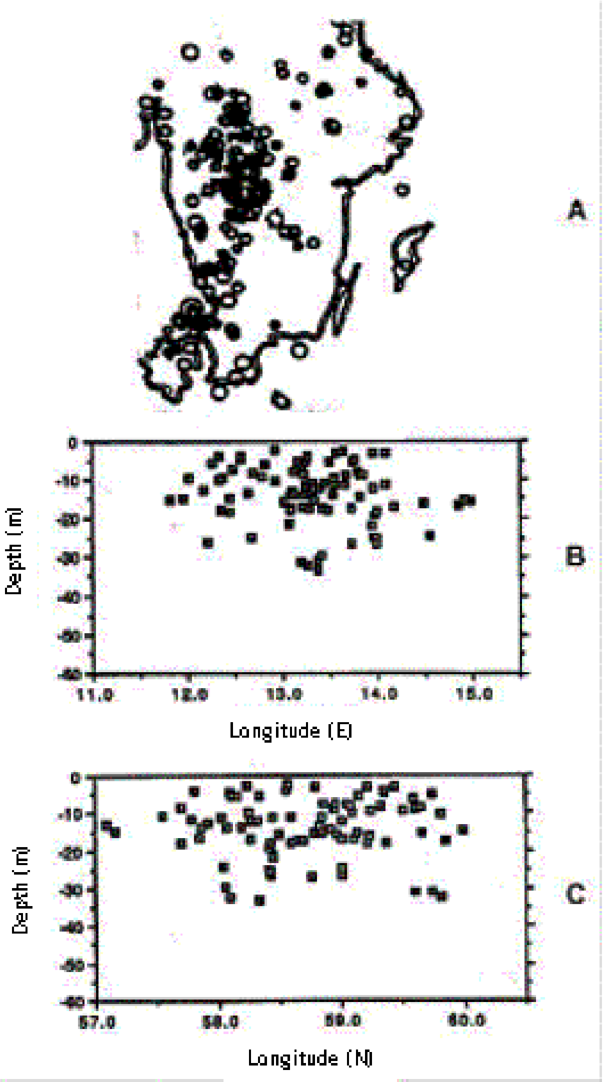

Figure 3.20. The distribution of earthquakes in the Vänern region shows an accumulation of earthquakes (map view A) and their distribution projected onto a west-east (section B) and a south-north section (profile C). (from Isaac 1992).

Traces of earthquakes can also be found in sediment stratification and in the bedrock. In the former case, the time of the earthquake can often be identified, while the age of the traces in the bedrock can seldom be determined. In the sediment stratification, the occurrence of *landslides* and *liquefaction* indicate earthquakes. In the bedrock, the occurrence of *friction melting* indicates the position and extent of fossil earthquakes. It has also been suggested that *bedrock caves* are caused by earthquakes.

3.6.4 Fault Movements

The brittle uppermost part of the Earth's crust is dissected by movement zones. When adjacent bedrock blocks are displaced, this is called a *fault* (see Berglund & Stigh 1998). Some of these zones have been active just after the deglaciation (*Figures 3.21* and *3.22*). However, the movement must displace a geological structure that can be dated, for example an esker or a moraine ridge, a measurable distance in order for the movement to be observable. If the movement has occurred in a completely homogeneous environment or is only very small, the movement cannot be determined. The geological environment must also be so stable that the displacements can be preserved. In recent years, methods have been developed that allow all systematic bedrock movements to be measured, for example, with repeated GPS measurements of fixed points positioned strategically with respect to the zones that are to be investigated (see *Figures 3.4* and *3.5*). The measurement series must be conducted over a period of at least 6 years in order to obtain interpretable results. Investigations so far conducted with GPS measurements show that lateral movements that are a few mm per year occur along the Tornquist zone in Scania (one of the first order shear zones – see *Figure 3.16*) (Pan *et al.* 2001). In the network in Norrbotten, where the observation time is only 5 years, it has not been possible to prove any movements with certainty

(Ågren 2001) and in that region, the measurements should be repeated several times. Information on deformations that have occurred very long ago has been compiled in Milnes (1998). However, their present function is still unclear. It is not well understood where, how and why present-day movements occur since this requires both detailed local investigations and a good knowledge of the movement pattern in the plate tectonic unit to which Sweden belongs. Furthermore, the problem would require a three-dimensional approach, which is difficult to achieve since the distribution of horizontal fractures are often unknown.

Figure 3.21. *Post-glacial faults in northern Scandinavia (red dashed lines), (from Lagerbäck (1988).*

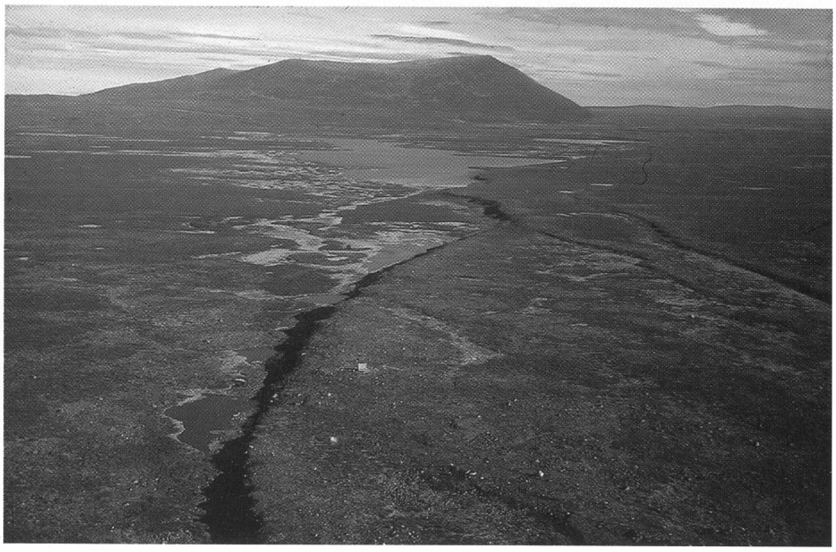

Figure 3.22. The Pärve fault – one of the large post-glacial fault zones in Scandinavia, view towards north (from Lindström et al. 2000), (photograph by J. Lundquist 1975).

When the position and movements of fracture zones have been established, questions arise concerning the future function of the zones. For example, which changes in the strength and orientation of the stress field can activate a certain fracture direction as well as how the stress field will change due to the plate tectonic evolution or due to future glaciations. In LaPointe et al. (2000), model calculations describe how earthquakes that occur in the vicinity of a repository affect the repository through the activation of existing fracture zones. With the same technology, it is possible to model the size of the change in the stress field that is needed to activate the fracture and shear zones mapped in the investigation area.

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4 Some Hydrogeological Methods for Determining Groundwater Recharge and Groundwater Flow

4.1 Introduction

The occurrence of groundwater, groundwater flow and the chemical composition of groundwater are of central importance for the siting and design of underground rock facilities, especially for the disposal of spent nuclear fuel. A high hydraulic conductivity makes rock engineering and disposal difficult. The groundwater can also attack barriers and waste containers, especially if the salinity is high, and can dissolve and transport hazardous components. The aim of this chapter is to present a few *hydrogeological investigation methods* used for determining groundwater recharge (quantity, spatial and temporal distribution) and its impact on groundwater chemistry as well as for studying groundwater flowpaths and their transport properties. Hydrological, hydraulical and chemical investigation methods and modelling methodology are not discussed here.

4.2 Hydrometeorological and Hydrological Data

Purpose

Hydrometeorological data are important in order to correctly calculate groundwater recharge in the ground and rock and to thereby provide general input data for flow potentials, for example, for numerical flow models. Hydrochemical data are also important for providing a basis for the calculation of chemical equilibrium reactions, mixing, groundwater age and flow patterns. In particular, isotope determination can be of importance for tracer studies, see Section 4.5. Hydrometeorological and hydrological data provide boundary conditions for modelling based on present-day conditions. For long-term, retrospective climate trends, geological and biological studies are required, for example dendrochronological studies (studies of tree rings), sediment studies with pollen and diatom analysis as well as studies of natural isotopes (for example, oxygen) in polar ice. These paleoclimatic studies can also provide valuable information on natural climate variations which provide input data for climate forecasts. Advanced computer models are currently used to calculate future climate conditions where anthropogenic effects (human impact) are of particularly great significance.

Data Access and Measurement Techniques

Hydrometeorological data series comprise temperature, precipitation, precipitation chemistry, relative moisture content, air pressure, wind direction, wind spred and global radiation. There are a large number of meteorological stations operating in Sweden from which data can be obtained. Most of these stations are run by the Swedish Meteorological and Hydrological Institute (SMHI). Furthermore, weather stations exist at airports, military air bases and other military facilities as well as along public roads for the purpose of road maintenance control. In addition, measurements (of precipitation, temperature and wind etc.) are conducted at nuclear facilities, for example, at Forsmark and Simpevarp, as well as at the Äspö Hard Rock Laboratory, southeastern Sweden. The frequency of measurement data collection and the parameters registered vary considerably. Major weather stations at SMHI are currently often completely automated. Information on suitable climate stations for regional and local investigations is provided by SMHI where data is currently stored digitally in several different large databases, *Svenskt klimatarkiv (KLAR)*, *Svenskt vattenarkiv (SVAR)* and for sea and oceanographic data, *Svenskt Havsarkiv (SHARK)*. Hydrometeorological data for Östhammar, Tierp and Oskarshamn have been compiled by SMHI on behalf of SKB (Larsson-McCann *et al.* 2002a, 2002b).

The *air temperature,* which is important for the calculation of evapotranspiration, is measured by mercury or resistance thermometers; the latter is preferable from the environmental standpoint. They are positioned protected from solar radiation, often about 1.5 metre above ground and at a distance from surrounding objects. The measurements are usually presented as weighted average values at different times of the day (Alexandersson 2002). In SMHI's assessment, error sources are small.

One of the most important and most common parameters which is necessary to provide input data for balance calculations and flow modelling is *precipitation*. The measurements are often conducted using a wind-protected precipitation gauge with a collection surface of 200 cm^2, placed 1.5 metres above the ground. In general, SMHI's stations perform measurements 1 to 2 times a day (at 07^{00} h and 19^{00} h, respectively). Precipitation collection is associated with significant uncertainty that is mainly due to turbulence around the gauge which causes the precipitation to miss the gauge. Significant measurement problems are associated with snowfall since the precipitation also has to melt. The losses vary with the wind strength and wind direction as well as with evaporation and adsorption on the walls of the vessel (Eriksson 1983). The measurements are often 10 to 25 % lower than the actual figures. Therefore, it is important for the precipitation values to be adjusted before use in, for example, water balance calculations. Statistical processing of long series of precipitation data is required to provide knowledge of the frequency of dry years and wet years. Since precipitation quantities can vary locally, it is important that several representative measurement stations are located in the areas under investigation.

Wind direction and velocity are measured and specified as a matter of principle at a height of 10 metres. Since the height and speed constantly vary, an average value is given as a rule, such as during 10 minutes. The wind is used to correct other data, including precipitation, although it also has considerable significance in other contexts, for example, for the calculation of airborne pollutants, including the spreading of salt to the environment around roads (Blomqvist 2001).

Air pressure is measured using different types of barometers and this is conducted at a large number of SMHI's weather stations as well as at airports and military air bases. The air pressure is important for the interpretation of surface and groundwater levels and other parameters and should therefore be taken into account in accurate calculations of small level changes. The combined effects of air pressure and wind can often have a considerable impact on surface water levels in lakes and seas. The effects can also be reproduced in the form of distinct level changes in groundwater levels where hydraulic connections exist. This should be taken into account in connection with groundwater level measurements in coastal areas.

Evapotranspiration is an important factor for the calculation of groundwater recharge. In practice, it is very difficult to measure, since it is dependent on many different factors, such as radiation balance, air temperature, air humidity, wind, type of ground, soil water and type of vegetation. *Potential evapotranspiration* can be calculated on the basis of climate parameters using Penman's formula or can be measured as evaporation from an open, water-filled vessel. However, since actual evapotranspiration is considerably less and is strongly affected by soil water and vegetation, it is difficult to measure. It should therefore be calculated, either as a loss item, if measurements of surface water runoff and precipitation are conducted, or be based on potential evapotranspiration and soil water content (Brandt *et al.* 1994).

Runoff is generally determined on the basis of data from the runoff stations operated by SMHI, among others. The mainland of Sweden is divided into 119 main catchment areas which, in turn, are divided into more than 13,000 sub-catchment areas (SMHI 2004). The water level in large lakes and runoff into many large watercourses are measured manually one or more times per day or, which is more common today, are continuously registered. On the other hand, information on runoff into minor watercourses is often lacking. Therefore, it is necessary to start measurements, as early as possible, of representative, small catchment areas in order to obtain a basis for local water-balance studies. The flow in watercourses can generally be calculated directly from the water level in a *measuring weir*, for example a V-shaped Thomson overflow.

Measurements of *precipitation* and *atmospheric chemistry* are conducted by SMHI and IVL Swedish Environmental Research Institute Ltd. Together with Statistics Sweden (SCB), they have formed a consortium for the collection of emission data and for the development of a national database. Deposition data are very important for groundwater chemistry modelling and can also be used to study infiltration and water transport to the groundwater (percolation). Previously, isotopes in the water were also determined in precipitation samples from several sites in the country. Since the isotope laboratory at the Department of Hydrology, Uppsala University, was closed down, such measurements are no longer conducted, which is a major disadvantage for studies of groundwater recharge (see Section 4.4).

4.3 Measurement of Surface and Groundwater Levels

Purpose

The measurement of surface and groundwater levels is an important part of hydrogeological investigations. The measurements can have many different purposes, including long-term measurements to determine long-term trends and seasonal patterns in the level fluctuations. They can also be conducted as general difference measurements to determine flow potentials and flow directions or as specific difference measurements to determine hydraulic relationships, for example, in connection with pump tests and other investigations that aim at determining the hydraulic properties of the ground. The measurements are often used within an environmental control programme, for example, in connection with underground construction in order to prevent environmental effects in the form of ground settlement and damage to constructions, groundwater supply and vegetation. The measurements are sometimes conducted for several purposes and this involves different requirements with respect to measurement frequency, accuracy and the length of the measurement series. Prior to the construction of a repository for spent nuclear fuel, level measurements that fill many simultaneous purposes are necessary. It is important that regular measurements in different types of geology and in different terrains are started as early as possible in the site investigation areas, in order to obtain long time series and to describe undisturbed conditions. Regular level measurements in observation tubes and boreholes are going on at Forsmark and in the region of Oskarshamn.

Data Access and Measurement Techniques

Long-term measurements of the groundwater level are regularly conducted within the Geological Survey of Sweden's (SGU) groundwater network, which comprises about twenty-five measurement areas spread over Sweden. In each measurement area, there are one or more specific measurement points that represent different aquifers, both tubes in soil as well as boreholes in rock. For groundwater monitoring, 82 areas with 120 stations also exist in the form of tubes, boreholes and springs for controlling groundwater quality. The size of the groundwater fluctuation and its temporal variation provide good information on the aquifers' properties, limitations, heterogeneity and hydraulic relationships. SGU's level measurements are generally conducted twice a month, and this is considered to be the minimum to obtain a clear reflection of the seasonal variations. High-resolution level measurements at som stations can be used to calculate the size of the groundwater recharge, see Section 4.4 (Johansson 1987, Healy & Cook 2002). Therefore, the evaluation requires a sufficiently high measurement frequency and, for aquifers with a small variation (for example, large aquifers or groundwater discharge areas), a high measurement accuracy is necessary.

In connection with major construction projects, separate control programmes are conducted for existing as well as newly installed measurement points. For example, major tunnel projects, such as the Bolmen tunnel which, during construction, involved measurements in more than 400 wells, tubes and boreholes as well as the Hallandsås tunnel, where the number of measurement points was close to 1,000 (Banverket 2000). However, for many of these measurement points, the measurements have only been conducted very occasionally or the measurement series are very short. Furthermore, in major cities, such as Stockholm and Gothenburg, specific monitoring programmes exist with a very large number of measurement points (almost 1,000 in Stockholm) which are, however, only measured a few times a year.

It has quite often been found that the *length of the measurement series* before the start of an underground construction project has been far too short for reliable assessments of groundwater impact to be made. The Swedish Environmental Protection Agency (1999) specifies an absolute minimum period of 6 months of measurement before construction start. Studies of *non-equivalent measurement series* (=unequal measurement frequencies) show that the measurement series should preferably be 15 to 18 months (Lundmark & Olofsson 2002) in order to determine minor deviations (<1 dm) from natural fluctuations. The length of the measurement series over several hydrological years and the use of equidistant measurements allow statistical *time-series analyses* to be used. If measurement points are used where groundwater abstraction occurs, for example, dug and drilled wells, the size of the groundwater abstraction must also be taken into account when evaluating the measurement series.

The measurements are either performed manually through sounding or continuously through, for example, pressure sensors and data logs. Continuous measurements are naturally preferable although they often result in large data sets. At present, there is a possibility of automating measurements from many points and of sending information via links to a data processing centre through which information can be obtained in real time from the measurement points. There is a considerable value in obtaining real-time information during the construction phase in order to allow for rapid measures and thereby prevent damage to buildings and vegetation. This approach has been successfully used in connection with underground construction in Norway in order to determine the need for leakage-mitigation measures in the underground facility (Randolph-Lund *et al.* 2003). It is also important that methods for analysis of groundwater level data are available in order to distinguish construction-related effects on the levels from natural variations. Systems for such statistical computer processing have been developed (such as *GCP – Groundwater Control Programme*) and have been used in connection with different underground projects, for example the Ormen tunnel in Stockholm (Cesano & Olofsson 1997), the Bolmen tunnel and the Hallandsås tunnel (Banverket 2000), see examples in *Figure 4.1*. Groundwater data are routinely collected in many construction projects, although this is often done without a structured analysis methodology. In this way, deviations have not been observed at all or have been detected at such a late stage that it has not been possible to prevent effects on the soil and vegetation.

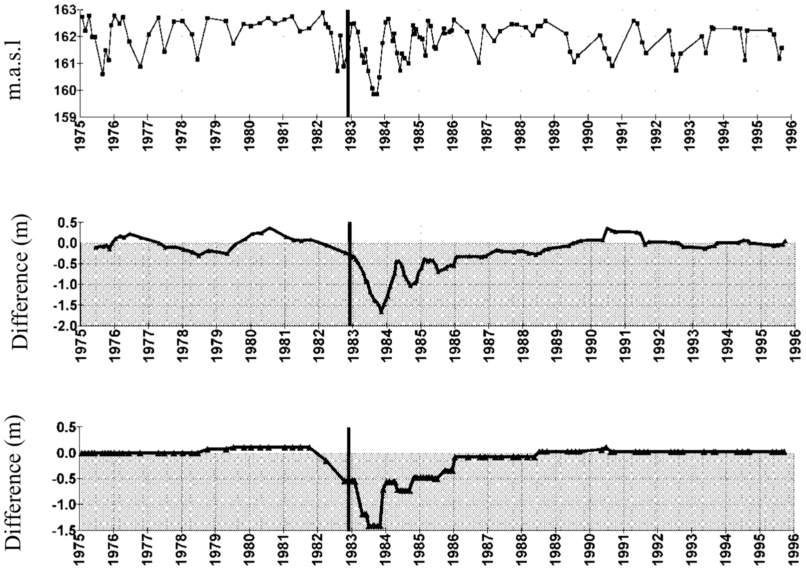


Figure 4.1. *Examples of how statistical calculations can be conducted on groundwater levels in order to obtain deviations from natural level variations in connection with underground construction. The uppermost diagram shows unprocessed groundwater level measurements. The middle curve shows deviations from natural level variations obtained using stepwise regression and the last curve shows deviations calculated using a modified double mass analysis. The vertical line shows the time when a tunnel was constructed close to the measurement point (taken from Olofsson in Knutsson & Morfeldt, 2002).*

The number of measurement points and their position are naturally very important for the usability of the groundwater data. Existing wells in rock and soil as well as springs are naturally used as measurement points which is particularly important in the initial stage as well as to investigate possible impacts during a later construction phase. The determination of the number of boreholes in rock that is necessary and the positioning of these boreholes in connection with construction projects, for example, during the construction of the Hallandsås tunnel and prior to the construction of the repository, is generally conducted on the basis of geological and geophysical investigations as well as the tectonic and geological models which are set up on the basis of these investigations. The borehole configuration is therefore highly dependent on the heterogeneity of the rock and the need to investigate specific geological rock structures. The number and positioning of soil tubes for investigation are similarly determined by the variation of the soil cover and topographical conditions. In order to capture the typical long-term level variations in an investigation area, soil tubes and boreholes must be located in different geological and topographical environments and the levels must be registered in recharge and discharge areas for different groundwater systems and at different depths (if the stratification involves several groundwater systems). Therefore, this is different from the siting of control and investigation holes that primarily aim at providing construction-related data or at investigating specific structures. In order to design a long-term control programme for groundwater levels, a good knowledge of the geology of the area is required. The more heterogeneous and geologically fragmented an area is, the more observation points are required for a good control of the level changes. It is very difficult to obtain an adequate control in crystalline bedrock, since two adjacent boreholes can demonstrate completely different or temporally displaced level variations. In construction projects, it is usual to underestimate the area of impact, especially along major conductive zones in the rock (Olofsson 1991, Banverket 2000). Existing measurement programmes can be made more efficient as measurement data are obtained, through different statistical methods, for example principal component analysis (PCA, Pearson 1901), from which the co-variation between different points can be determined. The methodology is independent of the spatial distribution of the points and only explores linear trends in the data set. For points with hydraulic connections, different variations of geostatistic methodology can be used, for example, kriging, in order to render the position and number of measurement points more efficient (Ackerberg 2002).

The measurement of surface water levels is important, as has been described above, for the determination of runoff in small watercourses and for the calculation of water balances and the interaction between the surface water and groundwater. In many cases, high-resolution registration is of considerable importance, with respect to time and level, and this can provide knowledge of the lake's or watercourse's hydraulic conditions. High-resolution pressure and temperature registration have been conducted in a few lakes in the Forsmark area and show that lakes in the same area can function very differently in hydrodynamic terms with respect to recharge and discharge conditions as well as with respect to hydraulic connections with the surrounding groundwater (Widén 2001).

4.4 Groundwater Recharge – Measurement Methods and Calculations

Background and Problems

Groundwater is the underground part of the water cycle and, thereby, the most difficult part to measure and investigate. Groundwater recharge is defined as the downward water flow that reaches the groundwater system in question. Knowledge of the quantity of the recharge and of its spatial and temporal distribution is of greatest importance, for example, in connection with siting, design and construction of underground facilities below the groundwater table as well as in connection with the siting and design of waste deposits and water supply wells. The impact of groundwater recharge on the groundwater chemistry (for example, through acid rain or pollutants) must also be taken into account, for example, by conducting a vulnerability analysis for existing or planned water supply wells.

Groundwater recharge can be *direct*, namely, the precipitation directly infiltrates through the ground to an open aquifer (aquifer=a permeable geological formation capable of yielding groundwater to wells and springs) or *indirect* through the inflow of water from surrounding elevated areas to a closed aquifer or through contact with other aquifers. Another indirect process is induced infiltration, namely leakage from adjacent lakes or watercourses to an open aquifer as well as infiltration in dry river beds which is common in arid climate areas after heavy rain. The infiltration conditions are very different in different rock and soil strata depending on their permeability and moisture content. The size of the infiltration naturally also depends on the weather conditions, primarily the nature, quantity and temporal distribution of the precipitation as well as the size of the evapotranspiration. In this way, the conditions for groundwater recharge are very different from year to year or from time-period to time-period depending on the changes in weather and climate, especially in an arid climate (Knutsson 1988). Therefore, it is very important to collect and process hydrometeorological and hydrological data (see above), also statistically, so that the frequency of “dry” years and “wet” years is determined as well as more long-term climate changes.

In connection with direct groundwater recharge, inflitration into the ground takes place within the elevated areas of the terrain, also known as *recharge areas*, from which the water flows all the way from small, superficial, local systems to large, deep regional systems (*Figure 4.2*). Through topographical variations as well as variations in the geology, such as the occurrence of horizontal or flat structures with considerable water permeability, such as sand and gravel layers in till, superficial, open fractures in the bedrock (such as in Forsmark, see *Figure 3.11* in

Chapter 3) or fracture zones at greater depths (such as at Finnsjön in Uppland, Sweden), the groundwater is then step by step linked to springs, wetlands, surface watercourses and lakes, also known as *discharge areas* at different levels in the landscape. In this way, only a small part of the water reaches deeper parts of the bedrock and flows to the regional systems. The small-scale topography which dominates both southeastern Sweden and northeastern Uppland is favourable for the occurrence of local flow systems and superficial groundwater recharge, although not for regional systems and groundwater recharge at deeper levels (Follin & Svensson 2003, SKB 2003). One difficult complication is if human intrusion should disturb the natural groundwater recharge, for example, by leading water away in connection with tunnel construction. This causes the groundwater levels to sink and the recharge area changes and this can lead to increased recharge, faster turnover and changes in the groundwater chemistry. In agricultural areas with irrigation, a small addition of (surplus) infiltrating water can be expected, as is the case in densely populated areas with leaking sewage and clean water pipes followed by subsequent changes in groundwater chemistry.

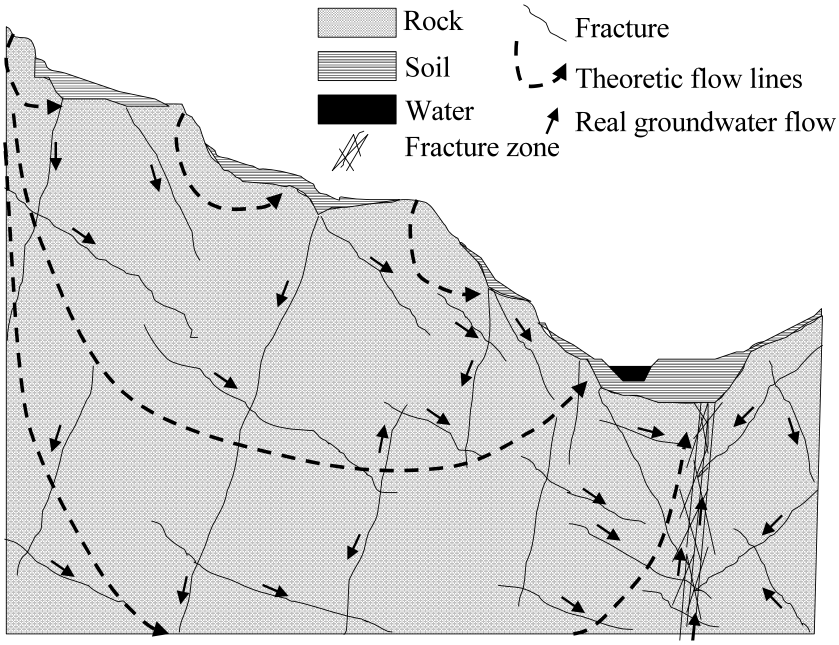


Figure 4.2. *Recharge and discharge areas as well as groundwater flow patterns in a valley with varying topography and thin soil cover on fractured, hard rock. The actual flow pattern deviates considerably from the theoretical depending on fractures and fracture zones (Olofsson et al. 2001).*

Important questions relating to groundwater recharge include the following:

- How and where does groundwater recharge occur? How is the chemistry affected?
- How great is the groundwater recharge in different aquifers and at different depths and what kind of hydraulic connection occurs with the surface water and between different aquifers?

- How does groundwater recharge occur in time with different weather conditions and climate changes?
- What kind of human intrusion can disturb groundwater recharge and change the chemistry?

Methods and Calculations

Overall assessments of groundwater recharge in a large area can be conducted in the form of a water-balance study based on precipitation and evapotranspiration data and taking into account geology, hydrology, topography and vegetation. The amount of groundwater recharge in a certain aquifer can be calculated using infiltration coefficients (the relationship between the quantity of infiltrated water and precipitation in a recharge area) for different rock types and soils if the geological and topographical conditions are very homogeneous and large-scale. However, this is seldom the case in Swedish terrain and consequently the method cannot be recommended.

Area and site-specific information on the size of groundwater recharge, its spatial distribution and temporal evolution requires detailed knowledge of geology, hydrogeology, land use and topography in the area as well as extensive measurements and calculations. Up-to-date information on precipitation and evapotranspiration is needed in the form of long series of or forecastes of climate data.

Different methods, based on different principles, exist for measuring and calculating groundwater recharge. Method selection should be conducted taking into account the purpose, time-scale, type of information desired (point or area data) as well as access to background information and resources. The use of the different methods must be based on good knowledge of the groundwater recharge processes and the existing geological and hydrogeological conditions. Therefore, it is important to initiate the study by setting up a *conceptual model* of the area of investigation. This entails a simplified, generalised description with a principle diagram (block diagram, cross section) of how the entire groundwater system functions as a whole *(Figure 4.3)*. The conceptual model is based on the water-balance calculations and on existing data on the geology, size and limits of the groundwater system as well as on where and how groundwater recharge occurs and flows and on whether human intrusion can be expected to disturb the natural processes. Based on the model and the criteria specified above, the most suitable investigation methods and computer models can be selected. However, remarkably few good examples exist of conceptual models that are openly described in the literature, especially for groundwater conditions in hard rock.

In principle, the methods can be classified according to where in the system the movement and quantity of the water is being studied:

- Recharge, for example, using tracers.
- Response within the system, for example using groundwater level analysis.
- Discharge, for example runoff measurements.

Preferably, several independent methods should be tested. The uncertainties in the calculations must be specified for each specific method.

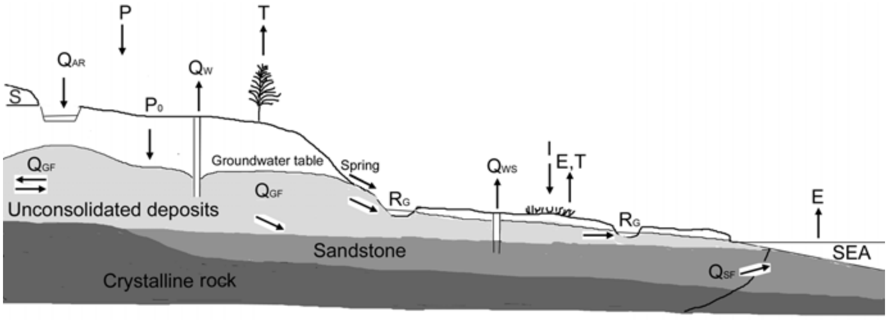


Figure 4.3. *Conceptual model for the groundwater conditions in the Nybro esker and its surroundings in a profile from the esker to the sea. E=Evaporation + T=Transpiration (350 mm/year), I=Irrigation, P=Precipitation (510 mm/year),* P_0*=Natural groundwater recharge (160 mm/year),* Q_{AR}*=Artificial recharge from basins (40 mm/year),* Q_{GF}*=Groundwater inflow and outflow,* Q_{SF}*=Outflow of groundwater from sandstone aquifer to the Baltic Sea.* Q_W*=Abstraction wells (60 mm/year),* Q_{WS}*=Abstraction of water from sandstone aquifer,* R_G*=Runoff in streams and drainage from groundwater, S=Sublimation and infiltration from snow. Note that the diagram is not to scale. The length of the profile is about 5 km and the maximum thickness is about 50 m (from Eliasson 2001).*

Recharge Methods

Groundwater recharge can be studied with the help of tracers and through modelling. Added tracers and natural tracers (see Section 4.5) have both been used to follow the path of the precipitation, such as snow melt water with a certain oxygen isotope composition through the unsaturated zone down to different depths below the groundwater surface for a couple or several years. Based on the average velocity of the water particles and the water content in an observed stretch, the size of the groundwater recharge is determined at 280 mm/year in sandy soil in the Uppsala district, south central Sweden (Saxena 1987). In Scania, south Sweden, and Denmark, tritium pulses have been followed for several years down through the thick soil cover to considerable depths in the bedrock. This has made it possible to determine parameters such as the time sequence for groundwater recharge to limestone at a depth of 150 metres on the Kristianstad plain to about 5 years (Engqvist 1991, see Section 4.5). Isotope data (deuterium, oxygen-18, carbon-14 and tritium) on the water from different depths at the Äspö Hard Rock Laboratory have provided important information on the origin of different types of groundwater, for example, that a low oxygen-18 content indicates that water from land ice melting has contributed to groundwater recharge deep in the bedrock (Laaksoharju 1999). These methods are of great interest for the development and evaluation of possible future scenarios for how a repository for spent nuclear fuel can be affected in connection with deglaciation after an expected, future ice age (see SKB 2003). It has also been possible to follow the changes in the original composition of the groundwater during the construction of the access tunnel to the Äspö Hard Rock Laboratory (see SKB 2003).

Experiments with added tracers result in pointwise measurements which can be difficult to transfer to larger areas or deeper levels. Furthermore, several of the mathematic models used are not suitable for determining groundwater recharge at depths since they have been developed for soil water studies. One complication in the use of recharge methods is if infiltration is affected by particularly permeable zones, for example, macropores in the soil cover or fracture zones in the rock. However, the results provide information on the quantity of water that is added to the surface groundwater system, namely the greatest possible groundwater recharge, which is of interest for further calculations using groundwater models (Olsson 2000). What are needed are methods to determine the groundwater flow from the soil cover to the bedrock on the basis of existing hydraulic heterogeneity, for example, for the determination of the flow from till to underlying fractured rock. This could be achieved using a combination of geophysical measurements for mapping conductive soil and rock structures as well as measurements of groundwater pressure levels and groundwater chemistry (including isotope analysis) in different geological environments.

Response Methods

The methods involve studies of how different parts of the groundwater system, for example groundwater levels, groundwater flow and groundwater chemistry, react to changes in the form of added water and chemical substances, in this case, through groundwater recharge or discharge/abstraction of water, which is compensated for by subsequent recharge. The latter continuous abstraction method has been tested at water supply wells with long-term abstraction, which often entails a change in natural groundwater recharge. The response methods provide information on the actual groundwater recharge to the system or to the level in question.

The analysis of groundwater level changes is the most immediately suitable method since groundwater levels are easy to measure and are often included in long measurement series, such as with respect to many municipal water supply wells, and in different control programmes and as a reference in similar groundwater environments in SGU’s groundwater network since the mid-sixties. A detailed description of the method with its different variations, for example, to calculate reservoir changes, is provided by Fealy & Cook (2002). The most common analysis involves transforming the groundwater level fluctuations in a number of representative observation tubes, wells or boreholes in an open groundwater reservoir to corresponding water quantities with the help of a value for the storage coefficient, also known as the specific yield of water. The storage coefficient is the quantity of water that is removed or added to the reservoir per unit area (for example, 1 m^2) in connection with the lowering or the raising of the groundwater level by one unit (for example 1 m). It is primarily determined by pump tests but can also be established on soil or rock samples in the laboratory. It should be known for different parts of the groundwater reservoir, which can be a demanding task, especially in fractured, hard rock. The groundwater level analysis method is otherwise best suited to groundwater levels that are fast-reacting and relatively deeply located, such as in the bedrock, where the level fluctuations are not affected by capillary transport and evapotranspiration (Johansson 1987) or ground frost. Important information which is also obtained through this method, includes knowledge of the temporal processes in groundwater recharge in relation to precipitation and climate changes and the effects of different activities that affect groundwater recharge.

The chloride balance or chloride concentration method is based on the relationship between wet and dry precipitation of chloride from the atmosphere and the chloride content in the groundwater. The chloride content usually increases with the infiltration of the water due to evapotranspiration but is then not changed in the groundwater zone. The method appears to be simple and inexpensive but has been found to contain significant uncertainties, particulary in the determination of dry precipitation and through the fact that the chloride content in the groundwater can be affected by both relict saltwater and pollutants. It is probably most suitable for rough estimates of groundwater recharge in large areas and over long periods of time. Gustafson (1988) has carried out such a calculation for crystalline bedrock in Sweden, divided into six regions with the support of existing data from SGU and SMHI. The results are interesting and show regional differences which appear to be reasonable, namely low values (24-28 mm/year) in eastern

Götaland and Svealand with a low net precipitation and high values in Scania (114 mm/year, compare with Hallandsås below) and in western Sweden (250 mm/year). The values correspond to groundwater recharge in relatively superficial parts of the bedrock since the calculation is based on data from local water supply wells which are usually 100 m deep, at most. The method has been much used in dry areas, where it is expected to be a useable supplement to other methods (Lloyd 1999).

The groundwater flow method is a more demanding method which applies both to input data on hydraulic parameters and boundary conditions and to calculations with analytical and numerical solutions, or nowadays, primarily with numerical modelling. An early use of a finite element model for two-dimensional flow was conducted in the sedimentary bedrock on Gotland in the Water Planning Official Report (Berggren *et al.* 1980). The groundwater recharge was calculated at between 10 mm/year in an area with low hydraulic conductivity and 80 mm/year in another area with higher hydraulic conductivity. The development of mathematical models has since then been extensive, including three-dimensional (3-D) flow, at the same time that increased computer capacity has speeded up calculations. In a 3-D model of northeastern Uppland, groundwater recharge at a depth of 500 m in crystalline rock is estimated at between 1.6 mm/year and 5.7 mm/year for different cases with a net precipitation of 250 mm/year (Holmén *et al.* 2003). In this case, it would be suitable to attempt to calibrate this modelling by measurements and calculations with other methods. Previously, groundwater recharge at Äspö has been estimated at 150 mm/year on the surface and 5 mm/year at great depth. In general, it is stated that a turnover of only 1-2 % of the surface groundwater recharge occurs in the deeper parts of the bedrock (SKB 2003). However, a disturbance in the form of construction work with subsequent groundwater lowering can essentially increase groundwater recharge. On the tunnel level in Hallandsåsen, the increase has been calculated at 25 % in connection with a maximum groundwater lowering of 100 metres (Anderberg 2000).

Discharge Methods

The methods are based on obtaining data on the quantity of water leaving the groundwater system either by direct measurements or by model calculations. The most simple method is *flow measurements from springs* on condition that the catchment area for the spring is well-defined and that no water passes by or below the spring. The method has been tested with great success in superficial systems, for example, springs in moraine areas (Johansson 1987) and is useable in sedimentary bedrock, above all in karst formations. On the other hand, it is difficult to apply to deep groundwater systems in fractured rock. Runoff measurements in surface water which drain a certain area and at the same time *isotope analysis* of oxygen-18 and deuterium in rainwater, groundwater and surface water from the same area have, however, been found to provide very valuable information, above all that the amount of groundwater in the runoff is much greater than previously assumed also at flood peaks in connection with snow melting or heavy rain (Rodhe 1987, *Figure 4.4*). However, the quantity of groundwater is probably dominated by superficial groundwater and it should be an important task to investigate, through additional isotope determination, whether the groundwater supplied from greater depths, for example from well-defined bedrock areas, can be separated.

Runoff models have been used to determine groundwater recharge in areas with consistent geology, such as the moraine areas and large glaciofluvial deposits in southeastern Sweden (Johansson 1987 and Eliasson 2001). In the first study, several different models and methods were compared. In both studies, different variations of the HBV model, developed by SMHI, which is based on easily available weather data, were used. The model gave reasonable results on the average, annual groundwater recharge for each area. In the second study, it was also possible to obtain a value for groundwater recharge in the sandstone aquifer situated below the glaciofluvial deposits (15 mm/year compared with 160 mm/year in the superficial layers, see *Figure 4.3*). Unfortunately, the study did not include the underlying hard rock, although the groundwater recharge can be estimated at only a few mm/year.

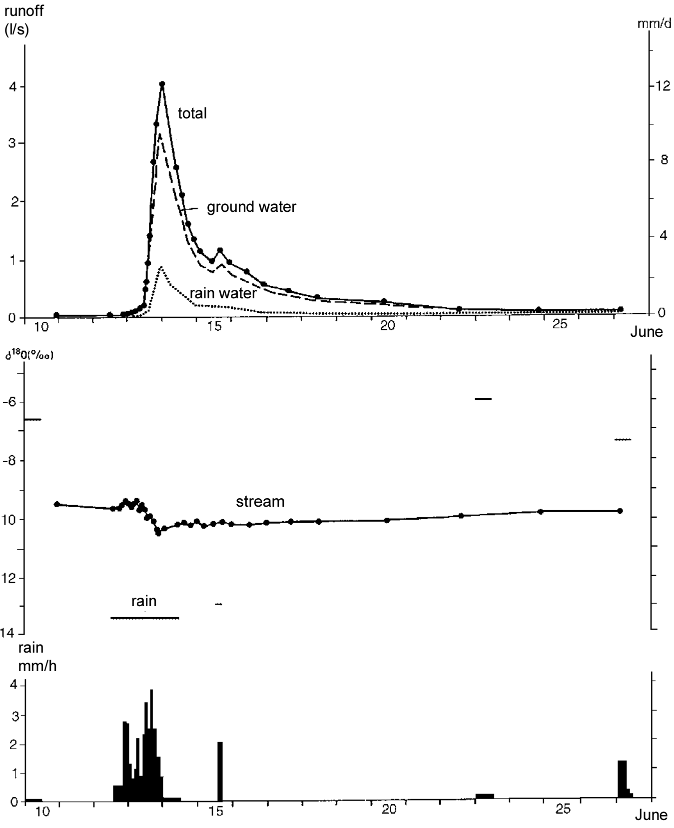


Figure 4.4. Diagram of runoff in a stream, the rain intensity and oxygen-18 content in rainwater at different precipitation times and in the stream water for the entire period. The low oxygen-18 content in the heavy rain does not have any particular impact on the oxygen-18 content in the stream water due to the fact that most of the runoff comprises "old" groundwater with a higher oxygen-18 content. It is forced out of the discharge areas near the stream of the infiltrating water upstream through the piston flow principle (based on Grip & Rodhe 1988 in Knutsson & Morfeldt 2002).

4.5 Tracer Methods and Isotope Techniques

Background and Problems

It is often of great interest to study groundwater flowpaths, flow velocities and transport pathways for pollutants or sorption of pollutants in the groundwater zone in connection with different types of groundwater investigations. The least controversial method to do so is to follow the path of the groundwater or pollutant through experiments and measurements of very small quantities of specific substances or isotopes that occur naturally or that are added to the soil and groundwater, namely, to conduct a tracer experiment. The results can then be used to determine hydraulic contexts and safety distances to sources of pollutants as well as to evaluate the results of modelling of for example, the transport and sorption of different radioactive substances. The usual problems that are studied are:

A. The groundwater

- flow between boreholes and wells, in boreholes in connection with packer tests and sampling or, for example, between sink holes and springs in a karst system
- flow direction and flow patterns in the bedrock fracture system or in soil layers
- flow rate between two points or, for example, within a protection area of a water supply well
- recharge, origin and age

B. The soil and ground

- dispersion properties, such as the dispersion plume of a pollutant
- hydraulic properties, primarily permeability/hydraulic conductivity
- sorption and ion-exchange properties, such as sorption of a pollutant

Requirements on Artificial Tracers

It is not difficult to find suitable tracers for investigations to determine a hydraulic connection between two points, since the behaviour of the tracer in the ground is not of decisive importance for the interpretation of results. The only important factor is to be able to detect the arrival of the tracer. In order to determine the flow direction of the groundwater, tracers, that to a certain extent are retained in the bedrock, can be used. The sorption of certain pollutants or ion-exchange in different rock or soil types, for example, fractured rock, can also be studied. This is the concept for a series of tracer experiments that are conducted in different parts of the world, such as at the Äspö Hard Rock Laboratory, prior to the disposal of spent nuclear waste.

For investigations to determine the actual flow rate of the groundwater and to determine hydraulic properties, there are major difficulties in finding suitable tracers. The ideal artificial tracer must fulfil certain requirements:

- It should follow the groundwater movement without being sorbed or delayed in the ground by ion-exchange.
- It may not react, for example, with microorganisms, or be affected by pH changes.
- It should be possible to detect the tracer in very low concentrations so that the physical and chemical conditions of the water are not changed.
- It may not be hazardous or damage plant or animal life, for example, in discharge areas and springs.
- It should be easy to acquire at a reasonable cost and should not entail high analysis and measurement costs.

The first two requirements mean that the difficulty of finding an ideal tracer is greatest in porous rock and soil types, where the contact surfaces between the particles in the rock or soil types and tracers are very large and, thereby, the sorption and ion-exchange processes are very active. The more fine-grained the rock or soil type, the greater is the effective contact surface for these processes. The mineral composition in the rock or soil types also play an important role for the scope of the processes as does the content of organic material as well as precipitation and weathering on particle or fracture surfaces. Quartz particles have the least impact, clay minerals and organic minerals have the greatest impact. This means that in the "cleanest" sandstones and sand deposits and in open fractures and channels in the bedrock, certain types of tracers are slightly or not at all affected, while the impact is great, for example in humus and clayey soil types, in clay-weathered zones and in fractures with precipitation in the bedrock (Knutsson 1971). The occurrence and role of the microorganism can have considerable importance for the decomposition of organic dyes, some of which are also sensitive to pH- and temperature changes as well as light.

The third requirement means that a tracer which must be added in large quantities to be detectable cannot be selected. Large quantities of sodium chloride have, for example, been added in karst areas, and a heavy saltwater stream has penetrated into deep cavities and thereby not participated in the natural flow process. In porous rock and soil layers, density stratification can occur. However, the problem has decreased as analysis techniques have evolved, which has also had a favourable impact on the fourth requirement which, during a period when radioactive tracers were preferable from the detection standpoint entails considerable limitations near to water supply wells. The fifth requirement can usually be fulfilled, even if the analysis costs for isotope determination are considerable. However, the dominant costs are often the experiment costs themselves since extensive drilling, measurements and sampling at the experiment site are required. Groundwater level measurements on a large number of points are therefore necessary both for the planning and performance of tracer experiments and for the interpretation of the results.

Different Types of Tracers

The following tracers have been used:

- Organic dyes with fluorescence, for example, Rhodamine, Sulforhodamine B, C, WT and Uranine.
- Salts, above all anions such as bromide, iodide, chloride and nitrate.
- Complex compounds such as fluorinated benzoates and stable metal complexes such as chromium-EDTA.
- Radioactive isotopes, primarily tritium and radioactive isotopes of anions and metal complexes (see above).
- Organisms, primarily bacteria, bacteriophagues and spores.
- Diverse chemical substances in the form of pollutants such as detergents, pesticides and chlororganic compounds (such as CFCs).

Organic dyes have been successfully used for a long time in karst areas and, recently, also in fractured crystalline bedrock, especially in connection with tracer experiments in the TRUE programme in the Äspö Hard Rock Laboratory (SKB 2001). In spite of the occurrence of both weathered feldspar on the fracture surfaces and mylonite, Uranine had the same transport rate as the anions, bromide and iodide as well as tritiated water, which indicates open fractures. However, the transport paths in this initial experiment were very moderate, about 5 m (SKB 2001). Therefore, it was not surprising that, in connection with the continued block-scale experiments on a 100-metre scale, which corresponds to the safety distance from a nuclear waste landfill to a major fracture zone, Uranine was significantly retarded in relation to bromide when in contact with different sorbing materials (Andersson *et al.* 2002). Similar results were obtained in connection with tracer experiments in a large fracture zone in hard rock in Germany in connection with a 295-metre flow path (Maloszewski *et al.* 1999). Normally since dye tracers undergo sorption and degradation in the soil cover, they cannot be recommended in such environments. Furthermore, they can probably not be recommended in porous sediment rock types. According to *Table 4.1*, it is on the whole difficult to identify a dye tracer which is not adsorbed, degraded or changed. It is remarkable that SKB has considered Uranine to be a tracer that is conservative (that cannot be affected), since it is pH and temperature-dependent and is easily adsorbed on humus and clay minerals.

Table 4.1. *Comparison between the properties of different fluorescing dyes (based on Tilly et al. 1999).*

	Detection limit, µg/1	*Tempe-rature dependent*	*pH-dependent pH 6–8*	*Photo-chemical degradation*	*Adsorption on humus*	*Adsorption on kaolinite*	*Cost*
BLUE							
Amino G Acid	0,51	little	little	moderate	big	relative little	high
Photine CU	0,36	little	yes	strong	very big	relative little	high
GREEN							
Uranine	0,29	moderate	yes	strong	very big	rather big	high
Lissamine FF	0,29	little	no	little	big	rather big	very high
Pyranine	0,087	little	yes	strong	big	relative little	high
ORANGE							
Rhodamine B	0,010	big	no	little	extremely big	extremely big	low
Rhodamine WT	0,013	big	no	little	very big	big	rather high
Sulpho Rhodamine	0,061	big	no	little	big	very big	rather high
BLUE-GREEN							
Na-Naphthionate			no	strong			

As far as *salts* are concerned, most cations can be excluded due to sorption and retardation by ion-exchange processes (although lithium has been used with a certain success). On the other hand, *anions* are only sorbed to a negligible extent or not at all, since the mineral particles are also negatively charged, as a rule. Bromide, iodide and chloride ions have been largely successfully used in a large number of experiments. Bromide and iodide have advantages since the natural concentrations are very low as a rule and, consequently, only very small quantities need to be added. However, they also have certain disadvantages, for example, the risk of sorption at low pH values, when the mineral particles are positively charged. Similarly, problems can occur in contact with iron precipitation in the B-horizon and below the groundwater table at low pH values (Tilly *et al.* 1999). Chloride appears to have given the consistently best results and is considered to be a conservative tracer which follows the water flow without being retarded. It has been used in a large number of experiments at municipal water supply plants in Sweden in order to determine residence times for water between infiltration basins and abstraction wells. In these contexts, it is significant transport distances (up to 2,000 metres) and long residence times (weeks to months), although on the other hand, the deposits are often very coarse-grained (Hansson 2000). However, sometimes chloride is less suitable, bearing in mind the fact that chloride can occur in varying amounts in certain geological environments and in places due to pollutants, for example from landfills and roads. Therefore, chloride is directly unsuitable for use at great depths in the bedrock where the chloride concentrations are often high. The use of chloride is also dubious in low-lying areas, below the highest seawater-line in Sweden with relict salt, which requires that large quantities of salt have to be added to obtain reliable results. However, the disadvantages are most often outweighed by the fact that chloride in the form of common sodium chloride is inexpensive and easy to handle and through the fact that detection in the field is simple. The conductivity is measured directly in boreholes, wells or springs or even from the ground surface using geoelectrical methods (see Section 3.4.8) as well as the fact that chloride analyses can be inexpensively conducted in laboratories.

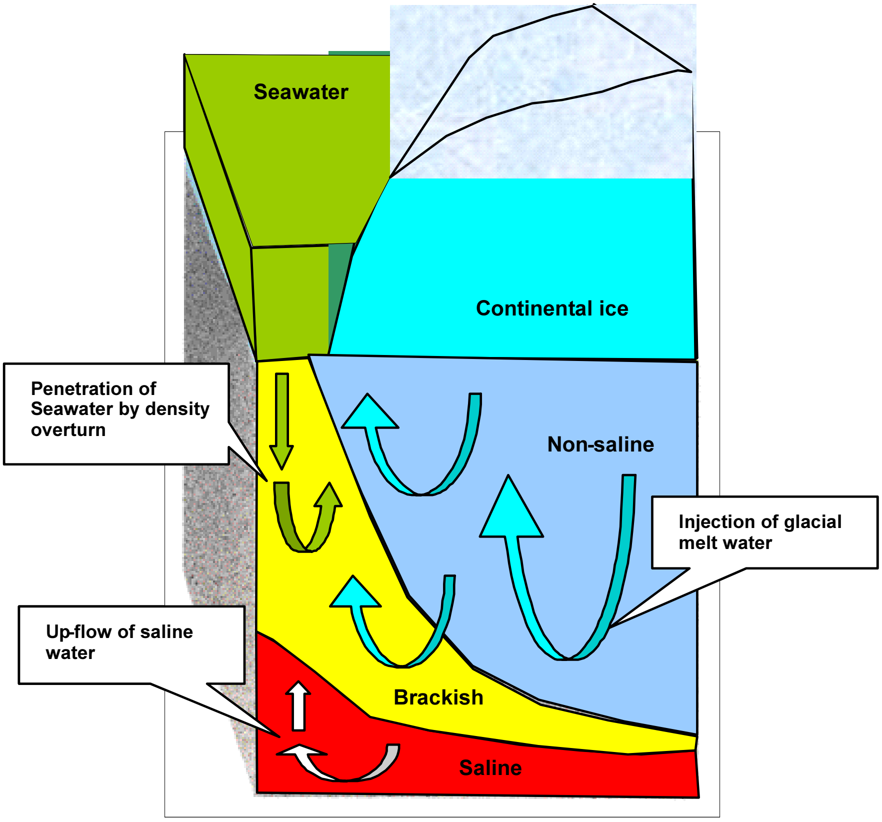
Stable metal complexes have also been found to be very useful as tracers. Suitable complexes exist among the metal chelates, of which the best known is ethylenediaminetetraacetic acid (EDTA). A chromium-EDTA complex has been tested in a very large number of laboratory experiments with different mineral mixtures, including different clay minerals, as well as several field experiments in rock and in the soil (Knutsson & Forsberg 1967). The chromium complex in dilution down to 0.0001 ppm is not sorbed or retarded in common minerals or in rock and soil types made of these minerals if complexation is complete. However, in contact with high concentrations of iron-bearing minerals and precipitation, for example, goethite and the B-horizon, certain iron and manganese-bearing silicates as well as clay minerals and clay mineral-rich rock and soil types, a certain retardation of the chromium complex occurs (Knutsson 1971). Other EDTA complexes and metal complexes have also been tested with favourable results (Knutsson 1970).

However, *tritium*, the radioactive hydrogen isotope, is the least controversial tracer since a small quantity of tritium is included in ordinary water (HTO) and tritium must be considered to follow the path of the water without sorption or retardation in most situations. Tritium has therefore been used extensively in a large number of experiments, especially in the Äspö Hard Rock Laboratory, as well as a reference tracer in connection with the testing of other tracers. However, in connection with such testing, with a 10 % addition of water-saturated bentonite (with montmorillonite as the main component) in quartz sand, tritium was found to be absorbed to these swelling clay minerals and tritium was retarded in relation to chromium-51-EDTA. Similar effects were not obtained in experiments with other clay minerals or other mineral mixtures. Furthermore, this was not the case in field experiments in different rock and soil types (Knutsson 1970).

By adding tritium, or previously, through the fluctuations in tritium content which occurred through hydrogen bomb experiments, it has been possible to follow a “pulse” of tritium from infiltration by water through the soil layers down to deep rock layers. In this way, the transport velocity of the water, or the time that it takes for the groundwater to reach a certain level in the bedrock, can be demonstrated. This has been studied in deep bedrock aquifers in Scania, south Sweden (Engqvist 1991) as well as in connection with nuclear waste disposal investigations. However, investigations conducted at the same time with other tracers have shown that a mixture of water of various origins can occur at great depths. A new modelling concept has therefore been developed within projects at SKB, the M3 model, through which it is possible to investigate the proportions in water of various origins (Laaksoharju 1999, *Figure 4.5)*.

Added tracers can primarily be used to determine the groundwater flow velocity between boreholes, in fracture zones or around water supply wells in order to determine the layout of the protection areas (particularly complicated in fractured bedrock) as well as to determine the residence time of the water in connection with artificial groundwater recharge as well as to map groundwater flowpaths from planned waste landfills. Investigations with added tracers are thus most suitable for small or medium scale experiments, where the residence times are moderate and the experiment times reasonable. On a regional scale, natural tracers should be used in the first instance or analyses of possible pollutants dispersed by man, such as freons and pesticides, should be tried, see below.

a)


Seawater
Continental ice
Penetration of Seawater by density overturn
Non-saline
Injection of glacial melt water
Up-flow of saline water
Brackish
Saline

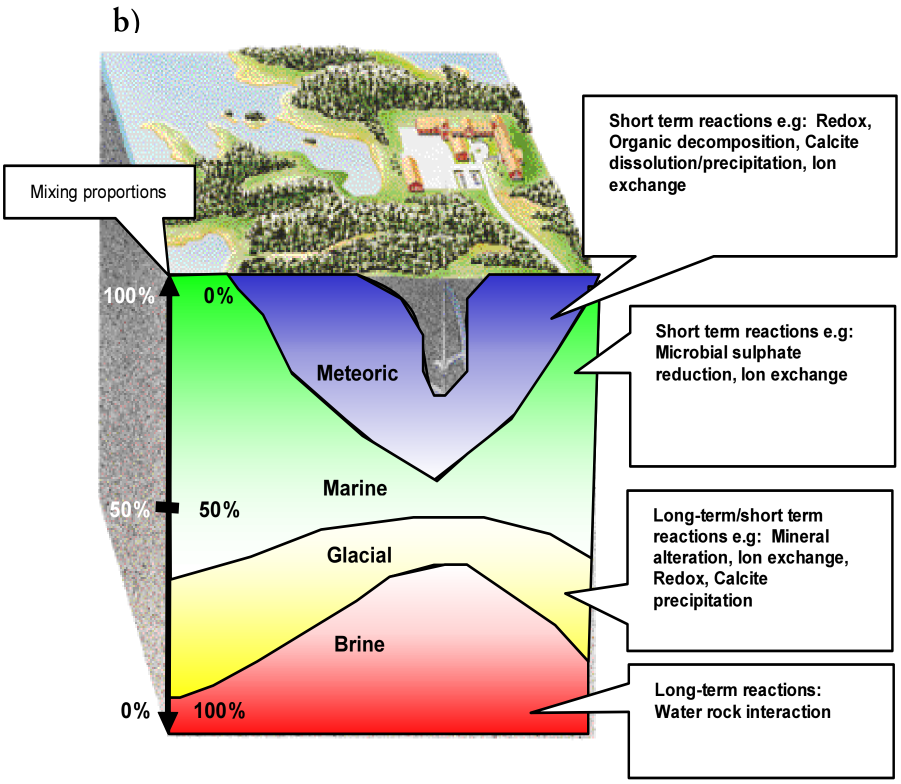


Figure 4.5. a) Conceptual model of different events in geological evolution since the ice age, which have affected groundwater chemistry at Äspö. ***b)*** *The calculation of mixing portions between different types of water at Äspö as well as dominant mass balance reactions with the help of the M3 model (from Laaksoharju, 1999).*

A special application of added tracers is to use the conservative tracer elements such as bromide, iodide and HTO together with a number of sorbing elements, common cations, such as sodium and calcium, and radioactive cations such as cesium and strontium. In this way, flow conditions and hydraulic parameters can be reliably determined through measurements of the conservative tracers and sorption and delay (through ion-exchange and diffusion) of different cations can be studied under controlled forms. The methodology has been tested in a large number of experiments conducted in a number of SKB projects, previously in Stripa and Finnsjön and, in recent years, at the Äspö Hard Rock Laboratory, where several tens of experiments have been conducted with highly interesting results (SKB 2001, *Figure 4.6*).

A new type of tracer which enables groundwater dating and groundwater recharge determination to be conducted is to measure the content of chemical products which started to be manufactured in recent years and which are used in liquid form, such as for agricultural purposes (pesticides) or released as gases in the atmosphere (freons, namely chlorofluorocarbons /CFCs/). The assumption is that they are not degraded or that the decomposition products can be measured. Freons appear to be the most useful. Freon manufacturing started in the 1940's and, since then, they have accumulated in the atmosphere. They are water soluble, are added through precipitation and act as tracers. The determination of the Freon content in groundwater at varying depths can therefore show with great accuracy when the water in question came into contact with the atmosphere. The method was developed in the USA in the 1970's and has been used in Germany and Denmark and other countries as well as on the Kristianstad plain and in southern Scania (Barmen 2001).

A group of researchers in Uppsala has started to use the method to determine the age of the groundwater in fractured rock (Bockgård 2000). The concentrations of CFC-12 and tritium at different depths in three boreholes at Finnsjön show an increasing age with depth and a mixture of water of different ages (Bockgård *et al.* 2004). As the use of freons ceases, the method will become less useful. Pesticides have been found in deep aquifers in Denmark as well as in some wells drilled in the rock in Sweden. The difficulty often lies in determining when the pesticides were brought to the surface.

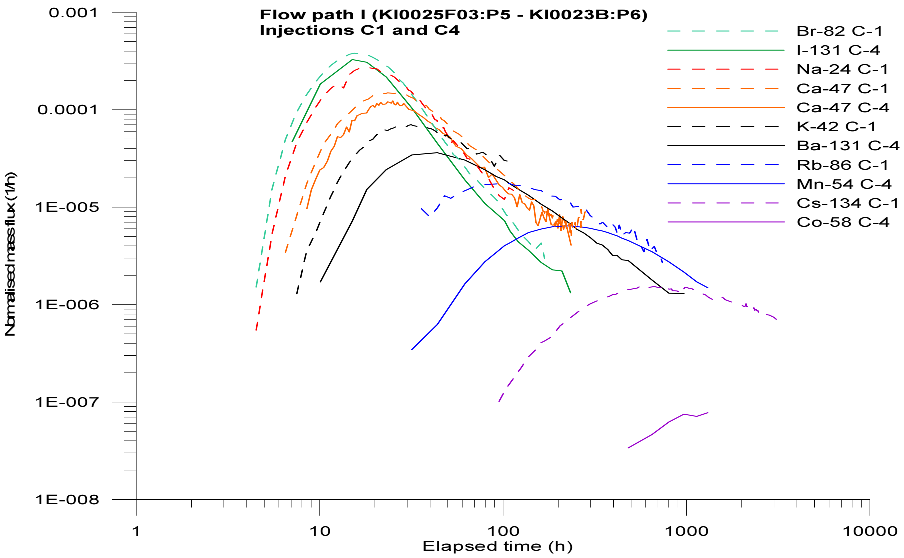


Figure 4.6. *Normalised breakthrough curves for all tracers in the C1 and C3 tests in the TRUE block-scale experiments at Äspö. Note the difference in recovery and transport time between the conservative tracers, Br-82 (bromide) and I-131 (iodide) and the sorbing tracers Cs-134 (cesium) and Co-58 (cobalt), which are retained and retarded to a large extent (from Andersson et al. 2002).*

Natural Tracers

The following natural tracers can be used:

- Radioactive isotopes, above all tritium, carbon-14 and chloride-36.
- Stable isotopes, above all deuterium, oxygen-18/oxygen-16 as well as sulphur-34/sulphur-32.
- Noble gases such as argon, helium and radon (primarily radon-222).

Tritium and *carbon-14,* which are produced in the atmosphere through cosmic radiation, are the most important radioactive isotopes for determining the age of the groundwater. This can vary from a few weeks to many thousands of years and is of interest to know in several practical contexts, for example, in order to determine whether groundwater at great depth is fossil-based and without turnover occurring in connection with present-day conditions which can be favourable for the disposal of hazardous pollutants but unfavourable if groundwater abstraction for water supply is planned. Before the first hydrogen bomb explosion, it was possible to determine the actual age of the groundwater to a certain level, although certain complications arose due to the mixture conditions between different types of groundwater. At that time, precipitation had a certain concentration of tritium (4 to 20 tritium units /TU/ depending on the season) and no additional tritium was supplied during infiltration. The knowledge that tritium has a half-life of 12.3 years meant that the age of the groundwater could be calculated fairly accurately. After the hydrogen bomb tests, the tritium concentrations in the precipitation increased very rapidly with the highest values at about 10,000 TU for 1963 and 1964 in certain locations in Europe. This was like an enormous tracer experiment in the whole of the northern hemisphere. Since then, the concentrations have successively decreased so that they are now at natural levels apart from in some fairly old groundwater with residues of bomb tritium and where local sources of pollutants occur (IAEA 2000). In spite of pollutants in the groundwater system, one way of determining the actual age is to determine the relationship between the concentrations of tritium and its daughter, helium-3. Helium starts to accumulate in the groundwater zone when tritium-bearing groundwater reaches the zone. The method is expensive and so far little used (Bockgård 2000).

Tritium determination can also be used to determine whether the groundwater at different depths is of the same origin and whether the groundwater at a certain depth is fed by groundwater from another area or is connected to the surface water. This has been successfully utilised for practical purposes, in a large Swedish mine in order to trace the origin of large flows of mine water, partly in connection with tunnel engineering in the Gothenburg area to determine whether the water in lakes could be connected to the groundwater in bedrock where tunnels would be built (Knutsson & Morfeldt 2002).

The determination of the age of very old water can be conducted with the help of carbon-14 which has a half-life of 5,730 years or chlorine-36 with a half-life of about 300,000 years. The determination of carbon-14 in groundwater carbon dioxide has been used since the 1950's in many parts of the world, and very high ages have been measured in groundwater in deep aquifers, for example, in Florida and in Nubian sandstone beneath the Sahara desert as well as at great depths in Swedish crystalline bedrock within the SKB projects and at great depths in the Kristianstad plain where mineral water is abstracted which, according to the carbon-14 determination, was formed during the bronze age. However, the use of carbon-14 is problematic and complex due to the fact that the carbon dioxide content of groundwater can have different origins: from the atmosphere, from fossil organic material as well as from carbonate minerals. Major progress in resolving this problem has been made in a SKB project through the development of a method of measuring the carbon-14 concentration in groundwater in the very small occurrences of humus in groundwater at great depths by enrichment in ion-exchange columns (Petersson & Allard 1991). The deep groundwater ages calculated by previously used methods in Swedish crystalline bedrock were found to be too high.

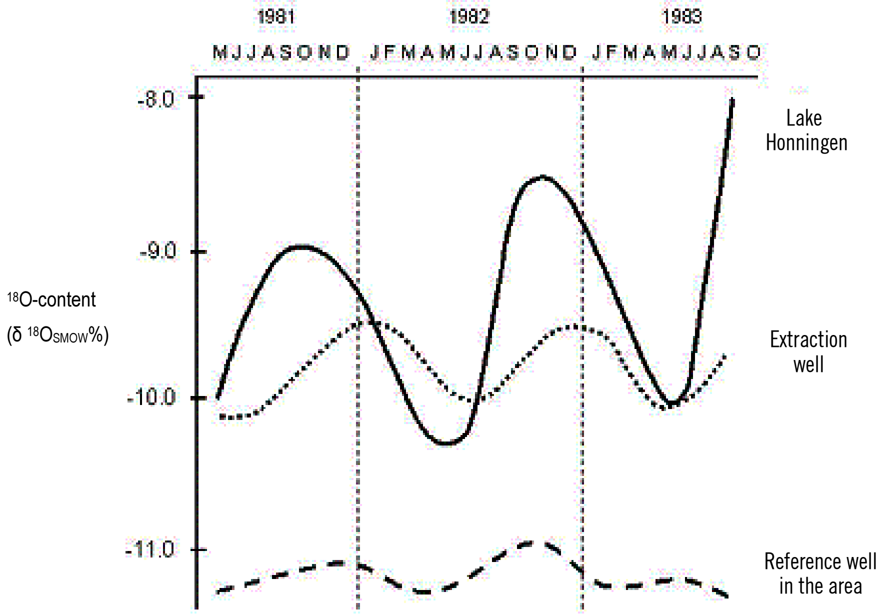
Deuterium (D) and oxygen-18, which in very low concentrations are included in the water molecule, are of great interest in order to determine the residence time and origin of the water. The possibility of using these stable isotopes arises from the fact that an isotope fractionation (see Chapter 5) occurs in water through the fact that during each evaporation process, the enrichment of the lighter oxygen-16 isotope occurs in relation to the remaining liquid phase. The vapour that forms over the sea therefore has a lower oxygen-18 content and deuterium content than the seawater. The fractionation process is affected to a high degree by the temperature conditions prevailing during evaporation and condensation which leads to seasonal variations in temperate climates and with increasing altitudes over the sea. Norwegian investigations have found very small variations on the coast but major seasonal variations in upland areas in the interior of the country. It has been possible to determine residence times for water infiltrating into different groundwater systems (Haldorsen 1994) as well as the amount of induced surface water in connection with the pumping of groundwater in crystalline bedrock (*Figure 4.7*). In Greece, the geographical origin of groundwater recharge for different springs and boreholes in a rock area is identified through differences in the oxygen-18 concentration due to the effect of altitude (Leontiades and Nikolau 1999). The method has also been used to obtain data for the previously mentioned modelling conducted at Äspö (*Figure 4.5*) as well as in connection with experiments with added tracers at Äspö.

Helium has started to be used to study diffusion in the bedrock matrix (Andersson *et al.* 2002).

Radon has been successfully tested to study the exchange between surface water and groundwater.

4.6 Conclusions and Recommendations

Hydrometeorological and hydrological data series are necessary for the calculation of the water balance and groundwater recharge in an area where underground facilities are to be sited in the rock. Statistical processing of long data series is necessary in order to obtain the frequency of, for example, “dry years”, which is the design basis for the removal of groundwater, bearing in mind the environmental consequences to fauna and flora as well as for the local water supply.



SMOW=Standard Mean Ocean Water

Figure 4.7. Variations in the concentration of oxygen-18 in a well in hard rock and in lake water in Rakkestad, Norway, which shows induced infiltration in rock with a residence time of 2.5 months and 77 % mixing of lake water (based on Hansson 2000).

The local variations in, for example, precipitation and temperature can, however, be considerably dependent on, for example, altitude effects and location, in relation to the coast. Therefore, it is necessary to supplement the national measurement stations by local and regional measurement stations for hydrometeorology and hydrology as well as extensive networks of measurement stations for groundwater level/groundwater pressure and groundwater chemistry in different hydrogeological environments and at different depths. The data are successively processed statistically and correlated with the data series from the national measurement stations.

Conceptual models of the groundwater conditions on a regional and local scale must be set up and reported openly for each investigation area to provide a basis for method selection and for computer models. There are several investigation methods that can be used to determine groundwater recharge and considerable knowledge has been obtained in the past few decades, although unfortunately, not much has been obtained regarding the size of groundwater recharge at large depths in hard rock. Knowledge must therefore be improved by testing of several independent methods on the same area, for example, response methods in combination with natural isotopes and freons as well as computer models. By using different methods, results can be checked and compared and different types of information can be obtained on groundwater recharge in time and space. This has been found in an analysis of ten different methods which were tested at Yucca Mountain in the USA (Flint *et al.* 2002). It is also of great importance to develop methods for the measurement of the groundwater flow from the soil layer to the bedrock, which can, for example, be achieved by combinations of geophysics, the measurement of groundwater pressure and groundwater chemistry including isotopes. In this, and in most other contexts, it is a disadvantage that the determination of natural isotopes in the water and certain other isotopes is no longer conducted in Sweden. The use of natural isotopes has decreased in Sweden unlike, for example, Norway, with its “domestic” laboratory.

Large-scale tracer experiments (safety distance to regional fracture zone) with several different conservative tracers (not dyes) are necessary in order to characterise the groundwater flow, not only in fracture zones but also in the entire bedrock.

Experiments to determine the sorption and retardation of different radioactive substances should be conducted in parallel as should diffusion experiments.

Calculations using different computer models must naturally continue, in order to predict relevant groundwater recharge and groundwater chemistry conditions in the site investigations and to explore different future scenarios, such as different climate situations (greenhouse effect, glaciation) in these contexts during the repository construction phase and during long-term disposal.

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5 Analysis and Fractionation of Isotopes

5.1 Introduction

The ability to measure extremely low concentrations of ions and other dissolved substances in the groundwater around a deep repository for used nuclear fuel is an essential requirement for future safety analysis. This concerns not only the measurement of total concentrations, but also the speciation and transport through the natural and technical barriers that will surround the copper capsules deposited in accordance with the KBS-3 concept. Of particular concern in these respects are the radionuclides, which are isotopes of the elements that decay, emitting electromagnetic or particulate radiation, and thus may be hazardous to the environment. Although many such isotopes occur naturally, it is quite conceivable that they could be released from nuclear waste deposits should the protective barriers fail to function as planned.

Since most radionuclides are more or less soluble in water, transport pathways will be dependent on the presence and the characteristics of the water in the repository environment.

Freely flowing water should, of course, be avoided, as this would facilitate rapid transport of dissolved species, as well as colloids. Colloids may consist of precipitated radionuclides or particles from the bentonite barrier together with adsorbed species.

However, transport of ions and neutral species will even occur in stagnant water, as a result of, e.g., diffusion. One of the driving forces for the latter process is known as chemical potential, which strives to eliminate differences in concentration. Consequently, ions or other mobile species flow from regions of high concentration to regions of low concentration. Capsule breach, followed by dissolution of exposed radionuclide compounds, presents such a scenario where diffusion would become operative.

There is also an additional array of chemical processes that affect transport, e.g., precipitation, dissolution, complex formation, oxidation/reduction and adsorption on surfaces in the local environment. All of these processes can interact with transport, the dominating mechanism depending on the chemical characteristics of the species, as well as water parameters such as pH, ionic strength, redox conditions and the presence of other dissolved substances or bacteria. Investigations concerning the analysis and transport of radionuclides therefore constitute prioritised areas of research for the Swedish Nuclear Fuel and Waste Management Company (SKB).

For these purposes, a variety of analytical techniques have been applied, ranging from simple measurements of electrical conductivity to diverse chromatographic methods. Atomic absorption spectrometry, as well as the more modern and more advanced technique of inductively coupled plasma mass spectrometry (ICP-MS), has also found application. The latter technique offers the distinct advantage of furnishing isotope-specific information. This enables its use in, e.g., determining the age and origin of groundwater, or tracing the sources of possible heavy metal and radioactive substance contamination.

Such measurements often assume that isotope ratios are constant, which has proven to be a rule with many exceptions. In fact, it has been observed that many, if not all, of the aforementioned processes lead to changes in the original isotopic composition, an effect termed fractionation. This is an important reason for KASAM to give an account of the current state of knowledge about isotope analyses, and to give examples of processes leading to fractionation.

This chapter will begin with a general introduction to the elements and their isotopes, as well as a description of certain characteristics of the latter.

5.2 The elements, isotopes and mass numbers

Atoms of any given element are characterised by a specific number of protons in their nuclei, defining the atomic number, normally denoted by the letter Z. For example, pure copper consists entirely of atoms of atomic number 29. Each element has been assigned a name and a defined place in the periodic table; all atoms of the element have essentially identical properties.

Even though all atoms of a given element have a defined number of protons, the number of neutrons can vary, within certain limits. This leads to the occurrence of isotopes. One example is element 1, hydrogen, which besides containing a single proton in its nucleus, may also possess 0, 1 or 2 neutrons, i.e., hydrogen has three isotopes with atomic masses of 1, 2 or 3, denoted by the symbols H, D (or ^{2}H) and T (^{3}H), respectively.

Several important properties of the isotopes are, on closer inspection, dependent on mass number, which is the sum of the numbers of protons and neutrons, denoted A. The mass number gives an approximate value of the atomic mass of an isotope, whereas the atomic weight of an element is determined by the composition of the isotopic mixture in question.

To specify a certain isotope of an element, the quantities A and Z, in addition to the chemical symbol, are written in a defined fashion. As examples, the following notations

$$^{63}_{29}\mathrm{Cu},\ ^{235}_{92}\mathrm{U},\ ^{1}_{1}\mathrm{H}$$

correspond to three specific isotopes of the elements copper, uranium and hydrogen, namely copper-63, uranium-235 and hydrogen-1, or simply hydrogen.

5.2.1 What is fractionation?

Two different isotopes of the same element therefore have distinct mass numbers but essentially identical chemical properties since these are determined by the number of electrons. Certain chemical effects can, however, arise because of differentces in mass number. Different isotopes may exhibit slightly different equilibrium constants for a given chemical reaction, which can result in so-called fractionation. The extent of this effect can be expressed in terms of the fractionation factor, α, also known as the separation factor or enrichment factor. α is defined by the quotient between isotope ratios describing the compositions of two different chemical compounds or phases, i.e.,

$$\alpha = \frac{\left({}^{h}N/{}^{l}N\right)_1}{\left({}^{h}N/{}^{l}N\right)_2}$$

where ${}^{h}N$ and ${}^{l}N$ are the abundances of the light and heavy isotopes, respectively, present in the two forms denoted by the subscripts 1 and 2.

To obtain more convenient numbers (with fewer decimal places) the following relationship is often used. The fractionations given in the rest of this chapter have been calculated using

$$\delta^{h,l}N = \left[\frac{\left({}^{h}N/{}^{l}N\right)_{\text{sample}}}{\left({}^{h}N/{}^{l}N\right)_{\text{standard}}} - 1\right] \cdot 1000‰$$

yielding the relative change in an isotope ratio between two forms, in this case a sample and a reference, expressed in per mil units (‰). An example of an equilibrium reaction that results in fractionation is the precipitation of calcium carbonate ($CaCO_3$) from an aqueous solution. ^{18}O is enriched, relative to the most common isotope, ^{16}O, in the precipitate by 25‰ (2.5% or $\alpha = 1.025$). The magnitude of the fractionation factor is temperature dependent, permitting measurement of oxygen isotope ratios in $CaCO_3$ to be used to determine the temperature of the water at the time of precipitation. This is the principle for the oxygen isotope geothermometer.

Photosynthesis is an example of a process where the lighter isotope, in this case ^{12}C, is enriched relative to the heavier ^{13}C. Cellulose and lignin in wood have in this way been enriched in ^{12}C by somewhere in the region of 2.5% (or $\delta^{13,12}C = -25‰$). This is actually an example where kinetic effects lead to fractionation, since the lighter carbon-12 isotope moves more rapidly through the processes of cellulose or lignin formation, thereby becoming enriched in the final products of the reactions.

Physical processes such as evaporation, condensation and diffusion can also result in pronounced fractionation. In this way the lighter oxygen isotope, ^{16}O, becomes enriched in water vapour from the oceans. At the same time, since the heavier ^{17}O and ^{18}O containing water molecules are enriched by condensation, atmospheric water vapour becomes even more depleted in these heavier isotopes. Through evaporation and condensation processes at the equator and poles, water deposited in Polar Regions is enriched in ^{16}O by up to about 5%.

The fissionable uranium isotope, ^{235}U, can be separated and enriched relative to the more abundant, non-fissile isotope ^{238}U, by virtue of minor differences in transport rates arising when gaseous $^{235}UF_6$ and $^{238}UF_6$ diffuse through porous barriers (see section 5.5).

5.2.2 Radioactive isotopes

Only a small fraction of all known isotopes are stable, whereas the vast majority changes spontaneously by radioactive decay. A radionuclide decays ultimately to one or more stable isotopes with the release of energy. This may be exemplified by the radionuclide tritium (^{3}H or T, as mentioned previously), which always converts to helium-3, ^{3}He, by release of a β-particle, which is nothing more than an energy-rich electron.

Under normal conditions, each type of radioactive isotope decays at a well-defined and characteristic rate. This means that, in the absence of any new source of formation, it is only a matter of time until all radionuclides disappear. However, certain isotopes decay so slowly that they still persist on Earth some 4500 million years after their formation. Examples of long-lived radioactive isotopes are potassium-40, ^{40}K, rubidium-87, ^{87}Rb, neodymium-144, ^{144}Nd, thorium-232, ^{232}Th, uranium-235, ^{235}U, and uranium-238, ^{238}U.

It might therefore seem surprising that short-lived isotopes, such as radon-222, ^{222}Rn, and carbon-14, ^{14}C, are so common on Earth. The reason is that the amounts of these isotopes are continuously renewed by special nuclear reactions, ^{222}Rn by radioactive decay of uranium and ^{14}C by cosmic radiation. Nuclear weapons testing and nuclear power plants also give rise to a multitude of radioactive isotopes.

5.2.3 The isotopic composition of the elements

Since the end of the 1930's, geochemists, astrophysicists and nuclear physicists have combined talents in an attempt to explain the observed isotopic composition of different elements. Hydrogen and helium, the two lightest elements are now assumed to have been formed in the "Big Bang". The relatively rare isotopes with mass numbers 6-11 (lithium, beryllium and boron) have partly originated from the influence of cosmic radiation. The heavier elements are believed to derive from nuclear reactions occurring in stars, resulting in the isotopic composition of the elements known today. Consequently, practically all iron on

Earth and in meteorites has been shown to contain about 5.85% ^{54}Fe, 91.75% ^{56}Fe, 2.12% ^{57}Fe and 0.28% ^{58}Fe.

The fact that the isotopic composition of different elements is relatively constant has enabled the tabulation of average atomic weights. Atomic weights are of the utmost importance in all chemical calculations.

5.2.4 The properties of isotopes

Generally speaking, all differences between the properties of different isotopes of the same elements can be related to two factors – differences in mass or differences in nuclear structure. The first is usually called the isotope effect, whereas the second has various names depending on the nature of its effect.

Helium consists of two stable isotopes, ^{3}He and ^{4}He, both of which exist as gaseous atoms under normal conditions. At given temperature and pressure, ^{4}He will have 33% greater mass than the same volume of ^{3}He, thus conferring a greater density. If the hydrogen isotopes (H) are completely replaced by deuterium in water, the result is so called heavy water, with a density some 10% greater than that of normal water.

A further difference in properties, which also depends on isotopic mass, concerns the mobility of atoms. Gaseous ^{3}He atoms move with an average velocity some 15% greater than ^{4}He at the same temperature. Additional properties that depend on average velocities, and hence mass, include thermal conductivity and gaseous viscosity.

As mentioned above, certain properties are dependent on nuclear structure. Radioactivity is one of these, and is the result of an interaction between the forces acting on protons, neutrons and electrons. For example, ^{6}He is radioactive whereas ^{4}He is stable.

Nuclear spin is another property of the isotopes that depends on the number and structural arrangement of neutrons and protons in the atomic nucleus. This means that atomic nuclei behave like minute magnets, which can, e.g., interact with electromagnetic radiation. This property is exploited in nuclear magnetic resonance (NMR) spectroscopy, which is employed in research, medicine and a range of technical applications.

The distribution of neutrons and protons in the nucleus also affects the surrounding electrons. The presence of an extra neutron in a certain isotope changes the distribution of protons and thus the shape of the atomic nucleus, in turn affecting the energies of electromagnetic radiation that can be absorbed or emitted by the electrons.

5.2.5 Fissionable isotopes

None of the elements with atomic numbers greater than 83 (bismuth), i.e., $Z>83$, possess stable isotopes, and therefore are subject to radioactive decay. Those elements primarily of interest for application in the nuclear power industry are the actinides with $Z\geq 90$, since some of their isotopes are fissile.

Among the actinides are the only known fissionable isotopes with their enormous potential for energy production, but also with attendant, long-term risks for the environment. Uranium, with atomic number 92, has a pair of fissile isotopes, ^{233}U and ^{235}U. Plutonium, $Z=94$, also has two such isotopes of considerable importance, namely ^{239}Pu and ^{241}Pu. These are formed as unwanted bi-products in nuclear reactors from ^{238}U, itself non-fissile, via the capture of neutrons released during ^{235}U fission. ^{235}U is present at a relative abundance of only about 0.7 % in naturally occurring uranium ores, and must be enriched to concentrations in the vicinity of 2.8 % before a nuclear reaction can be initiated (Spiro & Stigliana, 2003a).

Heavier actinides, $Z>94$, are mostly of scientific interest, although they have found some, albeit limited, application in cancer therapy. ^{232}Th has great potential economic value since it can be converted to ^{233}U, which is in turn fissionable.

Even though relatively few isotopes are actually fissile, considerably more are instable and radioactive. As a matter of fact, all elements have at least one radioactive isotope. As mentioned previously, the lightest of all elements, hydrogen, has three isotopes, of which the heaviest, tritium, is radioactive. More than 1,000 radioactive isotopes are currently known, about 50 being found naturally and the rest artificially synthesised. In excess of 500 radionuclides are produced in nuclear reactors.

Radioactive isotopes are utilised in a range of applications in medicine and technology: radioactive tracers for imaging and functional diagnostics, e.g. technetium (^{99}Tc) phosphate complexes for skeletal scintillography; further ^{99}Tc labelled substances for the diagnosis of heart, kidney, lung, etc. conditions; and ^{18}F labelled glucose for tumour diagnosis.

Radioactive formulations are also used for localised radiation therapy, e.g., ^{125}I and ^{192}Ir for tumour treatment. ^{60}Co is employed as a radiation source for cancer therapy and ^{131}I to locate brain tumours, whereas ^{14}C is utilised for studies of diabetes, gout, anaemia, etc.

^{241}Am has found application in fire alarms, ^{3}H in luminous evacuation signs, and both ^{210}Po and ^{238}Pu in batteries for the space industry.

5.3 Analytical methods and their limitations

5.3.1 Mass spectrometry

Mass spectrometry (MS) is the technique that has provided most of the experimental evidence on which our understanding of the nature and, indeed, the very existence of isotopes is based.

J. J. Thomson is credited with constructing the earliest form of instrument designed to separate atoms on the basis of their mass-to-charge ratios. Experiments with this instrument led to the discovery of the first two isotopes of any element, ^{20}Ne and ^{22}Ne of the noble gas neon in 1913 (Rouessac & Rouessac, 2000b). This work followed Thomson's receipt of the 1906 Nobel Prize in physics, for studies demonstrating the particulate properties of the electron.

A colleague of Thomson, F. W. Aston continued this pioneering work and discovered over 200 of the naturally occurring isotopes, including a third minor isotope of neon, ^{21}Ne. For his outstanding achievements, Aston was awarded the Nobel Prize for chemistry in 1922. Platzner (1997; Chapter 1) gives a brief historical account of the early development of MS in a recent book, which is also a good source of literature on the subject of isotope measurement in general.

Over the century since its conception, a plethora of instruments for MS has been designed (Platzner, 1997), certainly too numerous to describe in detail here. Therefore, we will confine ourselves at this point to a discussion of the magnetic sector type of instrument, as used in the original device constructed by Thomson, and still in use today. The basic premise in MS is that the motion of charged particles in a vacuum can be manipulated by application of magnetic or electric fields. As illustrated in *Figure 5.1*, ions having different mass-to-charge ratios will subscribe circular trajectories of differing radii in a magnetic field. Another important feature is that ions of different mass-to-charge ratios will be brought to focus along a plane. By positioning an array of detectors along this focal plane, it is therefore possible to simultaneously monitor a suite of ions and thus measure isotope ratios with very high levels of precision.

Once ions have been generated in a suitable source, vide infra, they are sampled by applying a large potential gradient, of opposite sign to the ionic charge, between the source and the mass spectrometer. In this way, the ions are accelerated to high velocities, preventing them from simply diffusing to surfaces inside the instrument where they would be neutralised.

A prerequisite for the technique is that the ions survive transport from the source of their production to the detector. Operating the mass spectrometer under vacuum conditions facilitates their survival. As the pressure is lowered, collisions between particles become less and less frequent. Collisions are undesirable for two reasons, the first being that the ion may lose its charge by abstracting an electron from its collision partner. Uncharged particles cannot be detected and so collisions resulting in charge transfer will result in a reduction in the number of ions surviving transport to the detector. The second effect of collisions is to scatter ions, diverting them from the trajectory that leads to the detector, again resulting in losses before detection. (This latter effect can be visualised as a billiard ball grazing another ball on its way to the pocket. The collision will obviously cause the moving ball to change direction and the shot will be missed.) Scattered ions tend to collide with parts of the instrument, such as the magnet poles, where they are neutralised.

Most of the residual pressure in a mass spectrometer results from leakage of atmospheric gases into the instrument. These gases, mainly oxygen (O_2), nitrogen (N_2) and argon (Ar), are very light, i.e., have relatively low masses, and therefore tend to scatter lighter ions to a greater degree than heavier ions. (A stationary billiard ball will scatter a moving ping-pong ball much more effectively than a moving bowling ball.) Consequently, heavier ions are more likely to survive transport through a mass spectrometer than lighter ions, which means that the detection efficiency increases with mass. It should be noted that there are also other effects that tend to accentuate this problem, which is termed instrumental mass discrimination or mass bias. The result of this effect is that, when an isotope ratio is measured experimentally, it will not correspond to the true value actually present in the studied material. Experimental MS data must therefore always be corrected for instrumental mass discrimination. Correction is based on experimental measurement of the mass discrimination using a sample of known isotopic composition. However, herein lies the dilemma, since isotopic compositions are determined using MS! The key to overcoming this problem is to use synthetically prepared mixtures of pure isotopes.

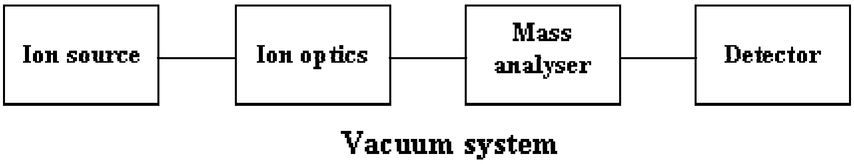
Although more efficient means for the isolation of pure isotopes are now available, MS itself can be used for this purpose. Historically, large sized mass spectrometers, known as calutrons, were developed for the electromagnetic separation of ^{235}U, the uranium isotope employed in the manufacture of the first atomic bomb in the Manhattan project (Rouessac & Rouessac, 2000b). Assuming that sufficiently pure samples of two or more isotopes are available, mixtures of known composition can be prepared. Comparison of measured (R_{meas}) with known or true (R_{true}) isotope ratios can then be used to calibrate the mass spectrometer, i.e., determine the instrumental mass discrimination correction factor (K):

$$R_{true} = R_{meas} \times K \; ; \; K = R_{true}/R_{meas}$$

It should be mentioned that K is dependent on mass, and must therefore be determined for each element, and perhaps even for each pair of isotopes under study (Woodhead, 2002). The functional form of the correction factor has also been the object of considerable investigation (Russell et al., 1978).

Since a complete mathematical treatment of mass discrimination awaits a more thorough understanding of the underlying physical phenomena, K remains a purely empirical correction factor, despite widespread reference to various “laws” in the literature. For this reason, there is a growing need to ensure that isotope ratios measured and corrected in one laboratory, can be reproduced elsewhere. This necessitates the availability of reference materials that can be used as international standards for the calibration of mass spectrometric measurements. Reference materials with certified isotopic compositions may be obtained from such authorities as the Institute for Reference Materials and Measurements, Geel, Belgium, and the National Institute of Standards and Technology, Gaithersburg, USA.

(a)



(b)

$$m/z = \frac{B^2 \cdot r^2 \cdot q_e}{2 \cdot V}$$

m = mass of isotope (kg)
z = ionic charge (dimensionless)
B = magnetic field strength (T)
r = magnetic sector radius of curvature (m)
q_e = electronic charge (1.60×10^{-19} C)
V = accelerating potential (V)

(c)

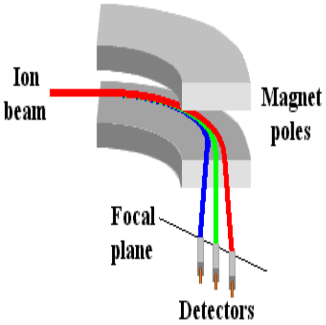


Figure 5.1. *Schematic diagram showing: (a) The lay-out of a mass spectrometer; (b) The governing equation showing how mass separation is achieved. Ions generated in the ion source have characteristic mass to charge ratios (m/z). The ions are accelerated by a potential applied across the ion optics, and injected into the mass analyser. Since the magnetic sector has a fixed radius of curvature (r), ions of different m/z are brought to focus by varying either the accelerating potential (V) or the magnetic field strength (B). Increasing B or decreasing V will bring heavier ions to focus at a given point on the focal plane shown in (c); (c) A detail of the magnetic sector mass analyser, showing how ions of differing m/z are focused at different positions. The left (blue) beam represents the path of the lightest ions; the right (red) beam is that of the heaviest ions. Moveable detectors are positioned at intervals along the focal plane to permit simultaneous collection of several ion beams and determine isotope ratios.*

5.3.2 Infrared spectroscopy

Although MS is undoubtedly the first choice of technique to use for the measurement of isotopic abundances, it is not the only one. The major advantage of MS in its various forms is that it is applicable to essentially all elements and compounds. On the other hand, as noted in the preceding paragraphs, mass discrimination can be a source of considerable error if not adequately corrected for. Thus there is an incentive to be able to measure isotope ratios by other techniques, as this provides an independent means of checking the data. One such independent technique is infrared (IR) spectroscopy.

As far as the measurement of isotopes is concerned, IR spectroscopy is still very much in its infancy. Unlike MS, which can measure isotopic compositions for samples in almost any form, IR spectroscopy is, at least at present, only applicable to compounds that can be introduced to the instrument as gases. Consequently, isotope ratio measurements by IR spectroscopy have found greatest application in the study of atmospheric gases, carbon dioxide (CO_2) being a popular choice (Becker et al., 1992; Esler et al., 2000). The greatest advantage of IR spectroscopy over MS, other than the lack of mass discrimination problems, is that the instrumentation is less expensive, simpler and portable. Therefore, atmospheric monitoring probably comprises the most important area of potential application.

Esler et al. (2000) have remarked that IR spectroscopy is capable of resolving the symmetry isotopomers of gases such as ozone (O_3) and nitrous oxide (N_2O) – a gas implicated in the environmental issues of global warming and the destruction of the stratospheric ozone layer (Spiro & Stigliani, 2003b). [Isotopomers are chemical compounds in which one or more of the constituent atoms may exist as a mixture of isotopes. Since there are two stable hydrogen isotopes, ^{1}H and ^{2}H, the simple molecular species hydrogen (H_2) exists in three isotopomeric forms ($^{1}H–^{1}H$, $^{1}H–^{2}H$ and $^{2}H–^{2}H$) representing all the possible combinations of two hydrogen atoms.] In the case of N_2O, two such isotopomers, $^{14}N^{15}N^{16}O$ and $^{15}N^{14}N^{16}O$, have identical molecular weights, since they contain equivalent isotopes, and thus cannot be distinguished by MS (Yung & Miller, 1997).

Considering how the atoms are connected provides the key to the resolution of this and other pairs of equal mass isotopomers by IR spectroscopy. Although mass discrimination is not of direct concern, IR spectroscopy still requires calibration in order to be able to convert instrumental signals to concentrations of isotopomers. For this reason, gas standards with known composition must be available for calibration purposes. Such standards are often analysed by MS (Esler et al., 2000). Fortunately, synthetic standards can also be prepared using isotopically enriched starting materials and purified products, thus relaxing the reliance on complementary measurements by MS.

It may come as some surprise to note that most car owners will unwittingly come into contact with IR spectroscopy, sooner or later (Rouessac & Rouessac, 2000a). Such instruments are namely used routinely to measure exhaust gas emissions of atmospheric pollutants such as carbon monoxide, unburnt fuel in the form of hydrocarbons, etc.

5.4 Applications of isotope ratio measurements

Isotope ratio measurements have numerous areas of application in a variety of scientific disciplines (Platzner, 1997). Two particularly relevant fields of study concern the dating of groundwater and the isotopic analysis of the actinides, especially uranium (U) and plutonium (Pu), as exemplified in the following paragraphs.

5.4.1 Dating of groundwater

Mass spectrometry has an important role to play in the selection of sites for nuclear waste storage.

One important criterion for the selection is that, in the event of leakage from the deposited capsules, the released radioactive material will be isolated by geological barriers and hindered from entering the biosphere (KASAM, 2001a). In effect, contaminated depository water should not be able to flow unrestricted into neighbouring water bodies. Injecting stable isotopes or long-lived, radioactive isotopic tracers and monitoring their progress through geological barriers and appearance in recipient water bodies can be used to investigate water flow patterns in boreholes. Changes in isotopic composition can then be employed to infer rates of transport through the bedrock. Such experiments have recently been initiated at the Äspö laboratory.

A more traditional and non-invasive means to assess the efficiency of the bedrock as a barrier to radioactive waste dispersion is provided by dating techniques. These are performed by measuring specific isotopes in water samples collected from boreholes at prospective repository sites. Certain radioactive isotopes are naturally produced by the interaction of cosmic rays with atoms present in the Earth's atmosphere. Such so-called cosmogenic radionuclides are distributed in the atmosphere, gradually being removed by rain and show, thus entering water bodies at the Earth's surface. One chlorine radionuclide, ^{36}Cl, shows particular promise for groundwater dating (Faure, 1986). This isotope has a half-life of 3.08×10^5 years, and the chemical properties of the chloride ion largely ensure that deposited ^{36}Cl will remain dissolved. Therefore, losses of ^{36}Cl are only by radioactive decay (and not by precipitation reactions as may affect other cosmogenic radionuclides), the time-scale of which allows water that may be millions of years old to be dated.

Measurements of ^{36}Cl in modern Antarctic ice samples have shown that the concentration is about 2.5 × 10^6 atoms kg^{-1} ice. If a volume of water, or a block of ice, is isolated from further input of recently formed ^{36}Cl, the concentration will decay exponentially, as shown in *Figure 5.2*. From the known initial concentration present in water, and measurement of the current level in a sample collected from, e.g., a borehole, the age of the water can be calculated. For example, if the measured ^{36}Cl concentration is 1.25 × 10^6 atoms kg^{-1} (1.25 mega atoms per kg = 1.25 Matoms kg^{-1}), then one half-life has expired in the sample, i.e., the water is about 0.3 million years (0.3 Myear) old. If water sampled at a site has been isolated from the in-flow of younger, fresher water for extensive periods of time, then it is likely that the geological formations will also be able to limit the out-flow of any accidentally released radioactive material in the event of any of the capsules being breached.

Further examples of the use of radioactive isotopes in connection with the dating of groundwater and groundwater flows are given in Chapter 4.

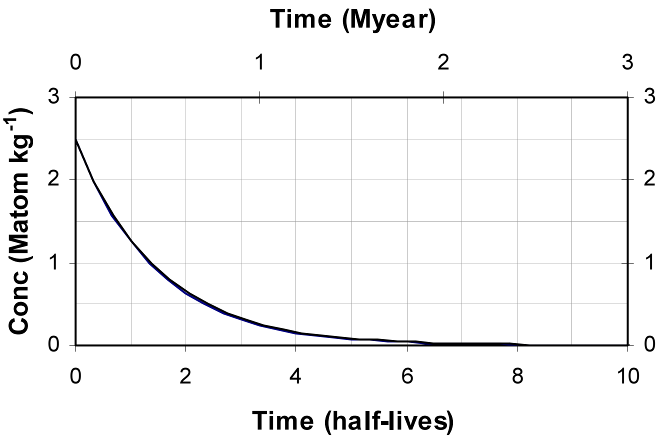


Figure 5.2. *Change in the concentration of the cosmogenic radionuclide ^{36}Cl in groundwater as a function of time. It is assumed that the initial concentration is 2.5 × 10^6 atoms kg^{-1}, as has been measured in recent samples of Antarctic ice (Faure, 1986). After 3.08 × 10^5 years (one half-life), the initial concentration will have halved. Measurement of the ^{36}Cl concentration in groundwater by mass spectrometry therefore allows the age of the sample to be determined.*

5.4.2 Tracing radioactive sources

The uranium found in all naturally occurring minerals has an essentially invariant isotopic composition, consisting of 0.005% ^{234}U, 0.720% ^{235}U and 99.275% ^{238}U (Richter et al., 1999; Desideri et al., 2002; Cobb et al., 2003).

The resultant ratio, $^{235}U/^{238}U \approx 7.25 \times 10^{-3}$, is therefore that expected in, e.g., biological tissues and body fluids collected from flora and fauna exposed only to natural, environmental sources of uranium. However, the isotopic composition is radically altered by industrial processes geared to the production of enriched uranium suitable for nuclear-fuel or -weapons manufacture. Both applications require enrichment in the abundance of ^{235}U, although to rather different extents. For use as fuel in light-water nuclear reactors (the kind used in Sweden), the atomic abundance of ^{235}U must be at least 2.8%, whereas enrichment to some 93% is necessary to produce weapons-grade uranium (Spiro & Stigliani, 2003a). A by-product of the enrichment process is the infamous depleted uranium (DU).

In the wake of the Gulf War and the more recent Balkan conflict, concerns grew that exposure to DU-containing debris from spent armour-piercing ammunition could constitute a health hazard (Sandström, 2002). Some soil samples collected from Kosovo in the aftermath of the conflict revealed altered uranium isotope ratios, providing evidence for the contamination of the region by DU originating from munitions (Boulyga et al., 2001). Urine samples collected from inhabitants of a suspected DU contaminated urban area, on the other hand, exhibited uranium isotope ratios consistent with natural sources (Tresl et al., 2004), as illustrated in *Figure 5.3(a)*.

In 2001 a study was conducted on participants of the Swedish peace keeping force. Before departure for Kosovo, and again after six months of service, urine samples were acquired and analysed by MS. It was observed that the concentrations of uranium in urine were, on average, 10 times lower at the later date, as evident in *Figure 5.3(b)* (Sandström, 2002). Thus the application of MS contributed to dispelling fears concerning the exposure of Swedish servicemen to DU in Kosovo.

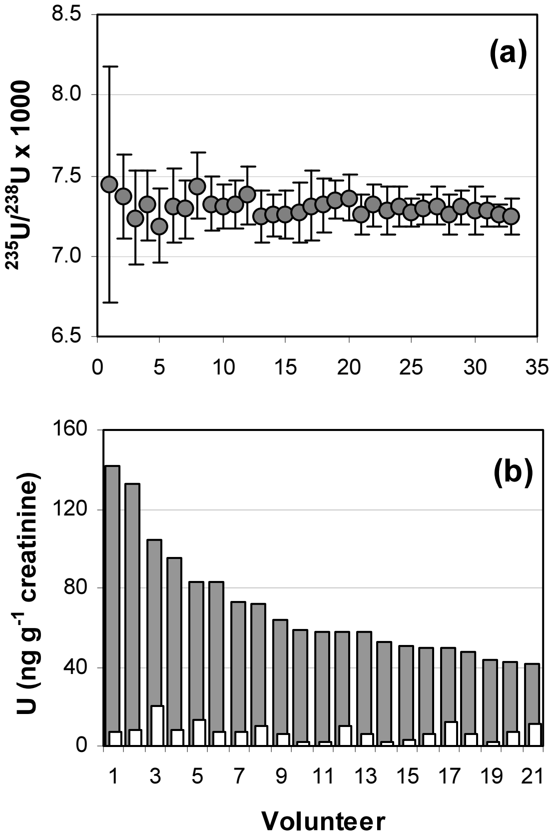


Figure 5.3. (a) *Uranium isotope ratios measured in the urine of local inhabitants of an urban area suspected of being contaminated with depleted uranium. Uncertainty bars are drawn at a 95% confidence level; data have been gleaned from Tresl et al. (2004). (b) Uranium concentrations present in the urine of KFOR personnel before leaving Sweden (shaded bars) and after six months service in Kosovo (open bars). The data have been extracted from Sandström (2002).*

It should be mentioned that spent fuel from nuclear reactors still contains ^{235}U. By reprocessing, this ^{235}U can be re-enriched up to about 4% and used to fuel a reactor (Desideri et al., 2002). However, during the chemical reprocessing of spent fuel, both the enriched uranium and the DU by-product are contaminated with artificial isotopes including ^{236}U, ^{239}Pu and ^{240}Pu.

Mass spectrometric analyses have provided irrefutable proof that the armour-piercing ammunition employed in the Balkans contained at least some DU derived from re-processed nuclear fuel, and thus spread artificial radioactive isotopes in the environment. On the other hand, the amounts of radionuclides actually released are considered to be insignificant in comparison to other sources, such as fallout from Chernobyl, and consequently, their toxicological effects are deemed negligible (Desideri et al., 2002).

Mass spectrometry also has an important roll to play in differentiating the sources of nuclear contamination in the environment. During the latter half of the 20th century, nuclear weapons detonations in the stratosphere, conducted by the United States and the former Soviet Union, spread radioactive material across the face of the Earth, deposition reaching a maximum in the period 1963-1964.

This resulted in global fallout inventories in the range 50-100 Bq m^{-2}, expressed as the sum of the most prevalent isotopes, ^{239}Pu and ^{240}Pu, and denoted $^{239+240}$Pu. This may be augmented by local or regional sources, such as leakages or authorised discharges from nuclear power plants and fuel reprocessing facilities, as well as reactor or satellite accidents (Warneke et al., 2002). Although a local or regional source might conceivably be detectable on the basis of an elevated $^{239+240}$Pu inventory, more definitive evidence can be obtained from isotope ratios, each source having a characteristic isotopic composition (Kelley et al., 1999; Warneke et al., 2002; Ketterer et al., 2004).

Note that the use of $^{239+240}$Pu results from the common application of a-spectrometry to measure plutonium inventories in the environment. For most instruments, the energies of the α-

particles emitted by decay of ^{239}Pu and ^{240}Pu are too similar to be resolved, and so, in effect, the sum of the activities of both isotopes is measured (Mitchell et al., 1997).

Data for the plutonium isotopes are collected in Table 5.1, where it can be seen the ^{240}Pu/^{239}Pu ratio differs greatly between sources, thus providing an excellent means of revealing the origins of environmental contamination. This possibility is exemplified by the data shown in *Figure 5.4* (adapted from Ketterer et al., 2004), where soil sampled from southern Poland is seen to be contaminated with plutonium from northern hemisphere fallout. Samples collected in north-eastern Poland, on the other hand, have also been subjected to pollution from the Chernobyl accident. The plutonium present in the latter samples therefore represents a mixture of material derived from these two different sources.

It is important to realise that isotope ratios will be altered over the projected time scale for nuclear waste storage in the deep repository, because of differences in the half-lives of the radionuclides, such as ^{239}Pu and ^{240}Pu, as evident from *Table 5.1(a)*. Nevertheless, differences between the isotopic signatures of the sources will be preserved, and thus isotope ratio measurements will continue to provide a means of identifying the origins of radionuclides in the environment, far into the future. However, in about 65,000 years time, i.e., after 10 half-lives of ^{240}Pu, the concentration of ^{240}Pu will be only 0.1% of the current level, and therefore extremely difficult to detect, at least with today's technology.

Table 5.1. (a) *Half-lives (Kelley et al., 1999; Ketterer et al., 2002) and abundance ranges of Pu isotopes in weapons-grade plutonium (Mitchell et al., 1997). (b) Atomic abundance ratios of the two most common Pu isotopes originating from various sources.*

(a) Isotope	***Half-life (year)***	***Abundance (atom %)***
^{238}Pu	87.74	<0.005 – 0.04
^{239}Pu	24 119 ± 27	93.3 – 97.0
^{240}Pu	6 564 ± 11	2.9 – 6.0
^{241}Pu	14.33 ± 0.02	0.12 – 0.58
^{242}Pu	376 000	–

(b) Source of Pu	***^{240}Pu/^{239}Pu***	***Reference***
Global fallout (northern hemisphere)	0.166 – 0.194	Kelley et al., 1999
Weapon production	0.01 – 0.07	Warneke et al., 2002
DU from nuclear fuel reprocessing	0.12	Desideri et al., 2002
Chernobyl accident	0.37 – 0.41	Muramatsu et al., 2000, Boulyga & Becker, 2002

5.5 Processes leading to isotopic fractionation

In the so called LTDE (long term diffusion experiment) project, SKB is currently assessing the extent of diffusion of radioactive species through bedrock. In the parallel LOT (long term test of buffer material) project, SKB is also studying the diffusion of radionuclides through the bentonite buffer material.

These projects, to study the transport of radionuclides through the bedrock at Äspö and bentonite, raise two potentially complicating factors for the interpretation of the results. As the groundwater flow rate through the bedrock at any finally selected site must be essentially zero, to prevent the spreading of any accidental radioactive waste leakage, transport of material will be driven by diffusion.

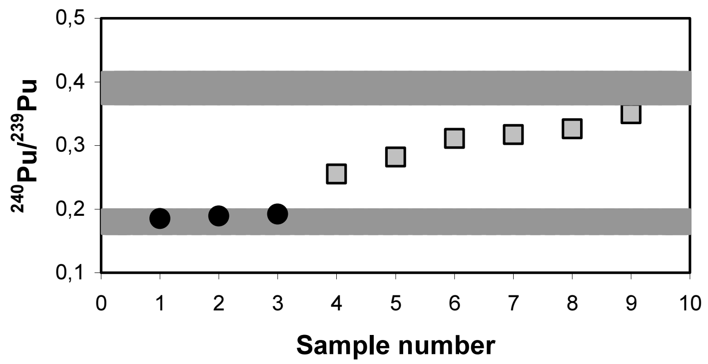


Figure 5.4. Plutonium isotope ratios in soil samples collected in Poland, illustrating the ability to discriminate between sources. The lower shaded region covers the range of $^{240}Pu/^{239}Pu$ typically found in northern hemisphere fallout. The Pu present in samples 1-3 collected in southern Poland (filled circles) clearly originates from this source. The upper shaded region encompasses the composition interval of Chernobyl-derived Pu. Samples 4-9, acquired in north-eastern Poland, exhibit isotope ratios that are characteristic of two-component mixing between northern hemisphere fallout and Chernobyl Pu. Data adapted from Ketterer et al. (2004).

The first question that follows is whether diffusion transports different isotopes of the same element in groundwater at the same rate or not. The second concerns the extent to which chemical reactions, such as precipitation and complex formation, might induce fractionation of isotopes in the deep repository environment.

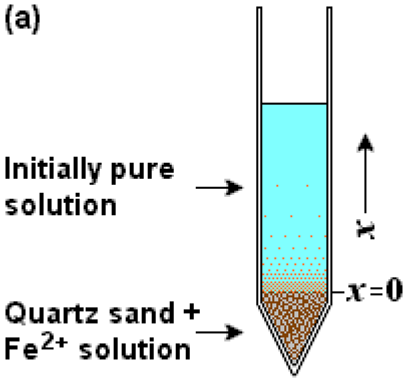
It is well known that, in the gas phase, lighter atoms and molecules diffuse faster, this principle being exploited for the enrichment of fissionable ^{235}U, as mentioned above. This process requires that uranium be converted into the volatile compound,

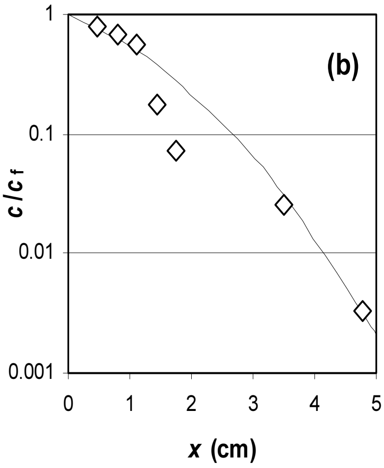
UF_6. On passing through each of a series of porous diffusion barriers, the gas becomes enriched in the lighter isotopomer, $^{235}UF_6$, by a factor equal to the square root of the $^{238}UF_6/^{235}UF_6$ mass ratio. As each fluorine atom has a mass of 19, the factor is equal to $\sqrt{[(238 + 6 \times 19)/(235 + 6 \times 19)]} \approx 1.004$, so literally hundreds of diffusion barriers are required to enrich natural uranium, containing 0.720% ^{235}U, to fuel-grade uranium, with at least 2.8% ^{235}U (Spiro & Stigliani, 2003a).

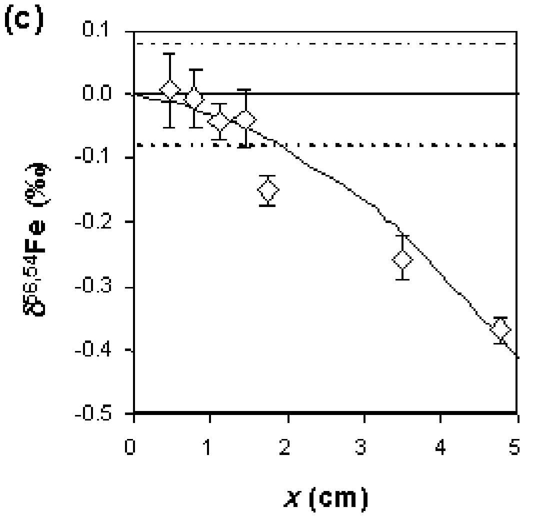
Although isotopic separation by gas phase diffusion is a well-understood process, the question of whether similar effects might be observed in solution has only recently been addressed (Rodushkin et al., 2004).

As shown schematically in *Figure 5.5(a)*, when a solution containing dissolved iron (Fe^{2+} ions) is brought into contact with another, iron-free solution, Fe^{2+} will diffuse into the pure medium. Gradually, Fe^{2+} migrates deeper into the initially pure solution, causing a concentration gradient to develop, as illustrated in *Figure 5.5(b)*. The concentration of iron in the solution drops by factors of 10 and 100 at distances (x) of about 2.7 cm and 4.2 cm, respectively, from the boundary between the two solutions, located at $x = 0$. These observations are in perfect agreement with the behaviour expected according to theory (Noggle, 1996).

In *Figure 5.5(c)*, the effect of diffusion on the isotopic composition of the iron sampled at various distances from the initial boundary is depicted. After 72 h and about 5 cm from the initial boundary, a $\delta^{56,54}$Fe-value of –0.4‰ (–0.04%) is observed, i.e., the solution has become enriched in the lighter isotope, consistent with ^{54}Fe-species diffusing more rapidly than the corresponding ^{56}Fe-containing ones. *Figure 5.5(d)* demonstrates that consistent behaviour is also obtained for a third iron isotope, ^{57}Fe.


(a)
Initially pure solution
Quartz sand + Fe2+ solution
x
x=0


(b)
1
0.1
0.01
0.001
c/c f
0
1
2
3
4
5
x (cm)


(c)
0.1
0.0
-0.1
-0.2
-0.3
-0.4
-0.5
$\delta^{56,54}$Fe (‰)
0
1
2
3
4
5
x (cm)

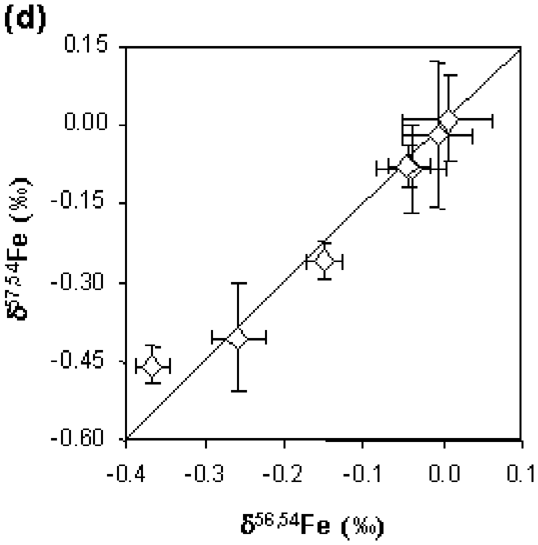

(d)
0.15
0.00
-0.15
-0.30
-0.45
-0.60
$\delta^{57,54}$Fe (‰)
-0.4
-0.3
-0.2
-0.1
0.0
0.1
$\delta^{56,54}$Fe (‰)

Figure 5.5. *Effect of diffusion in solution on the concentration and isotopic composition of iron. (a) Experimental set-up with 1 ml iron solution covered by quartz sand to provide a mechanical barrier. After careful addition of 9 ml of pure, iron-free solution, the set-up was allowed to stand undisturbed for 72 h. Then the solution was removed, from the top, in portions of 0.5 to 1.0 ml. (b) With increasing distance (x) from the source of dissolved iron, the concentration drops in accordance with a pure diffusion model, illustrated by the solid line. The data are given as measured concentrations (c) divided by the final concentration (c_f) that would be reached once all iron has been uniformly distributed throughout the solution volume. (c) Changes in the isotopic composition of the iron measured at various distances from the source. The solid horizontal line shows the initial isotopic composition, the parallel dotted lines representing the range of uncertainty in the ^{56,54}Fe ratio. The data points (diamonds) with uncertainty bars trace the change in isotopic composition with distance. After about 1.5 cm, the isotopic composition has changed significantly from that of the iron source. The solid curve is a mathematical model of the effect of diffusion on the isotopic composition. (d) Three-isotope plot showing that the change in isotopic composition is dependent on the masses of the diffusing isotopes. As the mass differences between the isotope pairs ^{57}Fe – ^{54}Fe and ^{56}Fe – ^{54}Fe are 3 and 2 atomic mass units, respectively, the data points should plot on a line with a slope of about 3/2. Clearly, all but one of the data points fit the theoretical slope (solid line).*

The latter is known as a three-isotope plot (Zhu et al., 2001), and provides a useful check on the consistency of experimental data. Considering the isotope pairs ^{57}Fe – ^{54}Fe and ^{56}Fe – ^{54}Fe, the mass differences are 3 and 2 atomic mass units, respectively. Any mass-dependent process causing fractionation between ^{56}Fe – ^{54}Fe (*Figure 5.5(c)*) would be expected to induce a proportionately greater effect on the ^{57}Fe – ^{54}Fe isotope pair, by virtue of the greater mass difference. In the simplest terms, we expect that the ratio of δ-values, $(\delta^{57,54}\mathrm{Fe})/(\delta^{56,54}\mathrm{Fe})$, should be roughly equal to the ratio of the mass differences, 3/2, as verified by the results shown in *Figure 5.5(d)*. Analogous results have also been obtained for zinc isotopes in the same experimental set-up.

Clearly then, diffusion will cause fractionation of dissolved species in any environment, representing a potential source of error in the interpretation of isotopic measurements. On the other hand, the magnitude of the observed effect is very small as shown by *Figure 5.5(c)*, and thus should not have any significant effect on the safety analyses performed on the deep repository for nuclear waste. Pescatore (2002) has called for more careful consideration of the potential isotope fractionation effects that may occur in the setting of radioactive waste disposal. To this end, a new model accounting for the effects of chemical potential gradients as well as Brownian motion has been proposed (Pescatore, 2002).

It should be noted that based on the theory of Brownian motion, there is a simple relationship between the diffusion coefficient ($D/\mathrm{cm^2 s^{-1}}$) and the distance (x/cm) that an isotope (or any other diffusing species) will travel in a given time (t/s), as expressed by the Einstein-Smoluchowski equation (Atkins, 1990): $D = x^2/(2\ t)$. If we assume a diffusion coefficient of 1.0×10^{-5} $\mathrm{cm^2s^{-1}}$, which corresponds to a somewhat lighter and faster moving species than iron ($D \approx 0.6\times10^{-5}$ $\mathrm{cm^2s^{-1}}$), the Einstein-Smoluchowski equation tells us that travelling average distances of 1, 10 and 1,000 m by diffusion alone would take 16, 1600 and 16 million years, respectively. Diffusion in solution is evidently an extremely slow process, which is why you must stir your coffee or tea after adding a lump of sugar!

Regarding the second concern alluded to above, there is a growing body of evidence that fractionation is a rather commonplace accompaniment to chemical reactions in a diverse range of settings. Therefore, the discussion will be confined to some preliminary experimental results obtained following the addition of individual, transition metal solutions to fresh, powdered samples of bentonite clay (Forsling et al., 2004).

After standing overnight, samples of solution remaining above the swollen bentonite were taken for concentration measurement and isotopic analysis. The results are summarised in *Table 5.2*, and indicate that bentonite efficiently removes most of the dissolved metal ions from solution. This is one of the attractive features of bentonite as a technical barrier in the KBS-3 concept for nuclear waste storage (KASAM, 1998).

One interesting observation made during these experiments was that, during water (and metal ion) uptake, the degree of bentonite swelling was noticeably diminished by the presence of copper ions (Cu^{2+}) in solution. A similar effect has previously been noted for calcium ions (Ca^{2+}), i.e., the degree of swelling for sodium-bentonite is significantly greater than that of calcium-bentonite (Abdullah et al., 1999; Hoeks et al., 1987). During the repository lifetime, sodium-bentonite (the preferred barrier material in the KBS-3 concept) would be converted to the calcium form by ion exchange processes occurring following the influx of Ca^{2+}-rich groundwater (KASAM, 2001b).

Calcium-bentonite cannot adsorb as much water as the sodium form, and will therefore contract, possibly creating fissures in the material. This could potentially impair the efficiency of bentonite as a barrier to the dispersion of radioactive waste in the event of capsule damage. Corrosion of the copper cladding, and subsequent uptake of Cu^{2+} by the clay, might similarly compromise the integrity of the bentonite layer.

The % sorption results in *Table 5.2* show that the efficiency of copper uptake is lowered at the higher initial concentration, suggesting that the bentonite buffers capacity to take up Cu^{2+} has been exceeded. This indicates that Cu^{2+} is very strongly bound to the surfaces of negatively charged bentonite particles, in turn causing inter-particle attraction and aggregation of the clay, effectively limiting the space available for water molecules to be accommodated (KASAM, 2001b). This interpretation explains the observation that dissolved copper reduces the degree of bentonite swelling, and is supported by the isotopic data presented in *Table 5.2*, as will become apparent below.

Table 5.2. *Uptake of cadmium, copper and zinc from solution during the swelling of initially dry, powdered bentonite. The δ-values are expressed as average values per atomic mass unit to facilitate comparison between isotopic pairs for different elements. The isotope ratios measured were* $^{111}Cd/^{110}Cd$, $^{112}Cd/^{110}Cd$, $^{113}Cd/^{110}Cd$ *and* $^{114}Cd/^{110}Cd$ *for cadmium*' $^{65}Cu/^{63}Cu$ *for copper and* $^{66}Zn/^{64}Zn$, $^{67}Zn/^{64}Zn$ *and* $^{68}Zn/^{64}Zn$ *for zinc.*

	Concentration (mg l^{-1})			
Element	***Initial***	***Final***	***Sorption (%)***	***δ (‰ per amu)***
Cadmium	500	18.9	96.2	+0.02 ± 0.04
Cadmium	50	2.0	96.1	+0.13 ± 0.04
Copper	500	55.1	89.0	–0.06 ± 0.10
Copper	50	0.2	99.6	–1.64 ± 0.09
Zinc	50	0.35	99.3	+0.52 ± 0.14

First though, we must consider the processes occurring during hydration of bentonite clay. During water uptake, water molecules and dissolved metal ions interact with surfaces and diffuse into pores in the clay mineral particles. If diffusion to particle surfaces and within pores were the only processes taking place, then we would expect that the fraction of ions remaining in solution would be over-represented by heavier isotopes. As shown in *Figure 5.5(c)*, diffusion favours the lighter isotopes, which would thus be able to migrate into pores more readily than their heavier counterparts, leaving the latter behind. This is what appears to happen in the cases of cadmium and zinc (*Table 5.2*). At both initial cadmium concentrations, the fractions sorbed are statistically identical. While this demonstrates that the capacity of bentonite for cadmium uptake has not been exceeded, it also indicates that the Cd^{2+} resides within water channels inside, and between particles, and is not strongly bound to surfaces like copper ions.

Other than transport phenomena, chemical reactions may occur during the hydration process. The products of reactions are the most stable compounds that can be formed from the given starting materials. Heavier isotopes form more stable bonds than lighter isotopes (Fujii et al., 2002; Schauble et al., 2001; Weston, 1999), thus chemical reactions may be an important source of fractionation. As copper is suspected of being strongly bound to bentonite particles, the reaction should lead to preferential removal of the heavier isotope (^{65}Cu) from solution. In other words, the $\delta^{65,63}Cu$-value measured for the copper remaining in solution should be negative, as indeed is observed (*Table 5.2*). Isotopic measurements can therefore be used to shed light on processes occurring in the deep repository.

5.6 Conclusions

There are a variety of processes with the potential to disturb the natural isotopic compositions of different elements. Such changes, which can be documented by measuring isotope ratios, are termed fractionation effects.

Many of the processes inducing isotopic fractionation have been known for decades, such as those observed for oxygen and carbon in natural cycles or caused by radioactive decay. The fractionation occurring during natural cycling of the elements is, to a large extent, a result of differences in diffusion rates between isotopes in the gas phase.

This chapter demonstrates that there are additional chemical and physical processes that may cause fractionation, e.g., in aqueous solutions, and thereby affect the transport of radionuclides through natural and technical barriers in the deep repository. These processes include diffusion in solution, which favours transport of the lighter isotopes of a given element, whereas many chemical reactions, such as precipitation, complex formation and possibly adsorption as well, lead to enrichment of the heavier isotopes.

Measurements of the isotopic composition of a specific element have traditionally been used to trace pollution sources, but the fact that many chemical reactions along transport pathways may alter isotope ratios, suggests that such methods may provide ambiguous results.

As illustrated by our own experiments, changes in isotopic compositions resulting from various chemical and physical processes can be exploited in different ways. Clearly, careful measurements of isotope ratios can provide important information on the underlying mechanisms for transport of various elements in the deep repository.

This is a field that SKB should investigate further in the future.

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6 Copper Canisters – Fabrication, Sealing, Durability

6.1 Introduction

Based on available information on canister fabrication methods, strategies and industrial experience, this chapter provides an account of the fabrication processes, methods and techniques which are generally known in the industry and which are reliable with a broad base of experience. Processes, methods and techniques that are new or customised for this application will also be examined. A comparison between this fabrication project and other similar industrial manufacturing projects is conducted, particularly with respect to design, welding and residual stresses. Furthermore, requirements on methods for Non-Destructive Testing (NDT) are compared with other industrial applications. Metallurgical characteristics that arise as a result of varied manufacturing techniques are evaluated and the impact of these characteristics on the long-term properties of the repository is examined.

The sealing of the repository is examined by evaluating its long-term properties and by posing the question of whether there are any canister fabrication processes that could degrade mechanical properties or cause long-term corrosion. Methods, procedures and models that are usually applied in the industry to evaluate long-term corrosion or creep are discussed in a long-term perspective. Furthermore, the extent to which different models can predict phenomena such as corrosion and creep over very long timescales is treated.

According to the KBS-3 method, the canister, which comprises different parts (*Figure* 6.1), is an important barrier inside the repository, since the canister prevents groundwater from coming into contact with the radioactive spent nuclear fuel. The outside of the canister is a copper shell which covers a nodular cast iron insert – combining external resistance to corrosion with internal load bearing capacity. Both of these components are therefore important for isolating the spent nuclear fuel from the groundwater over very long timescales (>100,000 years), which is much longer than the lifetime of any other industrially fabricated product. To prevent radioactive substances from leaking out of the canister, the fabrication methods must allow a defect-free canister to be manufactured where the materials properties of the copper and cast iron are guaranteed and optimised against all relevant damage mechanisms, such as forms of corrosion, creep, rupture etc.

Figure 6.1. *Dimensions and weights for canisters with wall thicknesses of 50 mm and 30 mm (RD&D Programme 2001).*The

fuel from both boiling water reactors (BWRs) and pressurised water reactors (PWRs) will be deposited in the repository. The total quantity of spent nuclear fuel to be deposited depends on the total number of reactor operating years. For example, in a scenario with 40 years of reactor operation, the quantity of BWR fuel from Swedish reactors is estimated at about 7,000 tonnes and the quantity of PWR fuel at about 2,300 tonnes. In addition to this, 23 tonnes of MOX fuel and 20 tonnes of fuel from the Ågesta reactor will be deposited. The hypothetical repository is designed for 8,000 tonnes of BWR fuel, which corresponds to 4,000 canisters in Sweden. The canisters weigh about 25 tonnes each when filled with four PWR or twelve BWR fuel elements. This means that one canister can hold about two tonnes of spent fuel. (RD&D Programme 2001)

In Finland, three different canister models are needed, one each for BWR fuel, VVER 440 fuel and OL3 fuel. After 50 to 60 years of reactor operation, the quantity of spent BWR and VVER 440 fuel is estimated at 6,000 tonnes, which corresponds to 3,000 canisters in Finland. The current estimate of spent OL3 fuel is about 2,000 tonnes, which corresponds to 1,000 canisters. (TKS-2003)

The copper shell (50 mm wall thickness) protects the spent nuclear fuel against corrosion. From the standpoint of fabrication, a thinner wall thickness (30 mm) would be better, but the wall thickness must be acceptable from the corrosion standpoint, especially from the perspective of local corrosion. A 30 mm wall thickness facilitates Non-Destructive Testing (NDT). With thinner material, finer grain sizes are also achieved and the canister microstructure is easier to control.

It is expected that the canister will be fabricated either from drawn seamless tubes or by welding together two tube halves of rolled plate. A bottom will be welded to the copper tube either through electron beam welding or Friction Stir Welding (FSW).

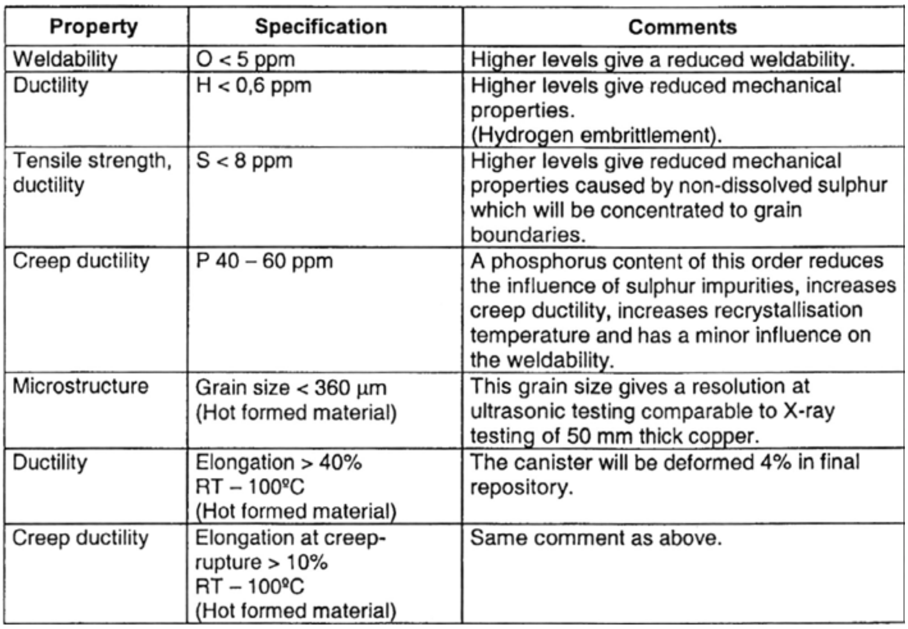
Figure 6.2. Example of fabricated canister components and objects (RD&D Programme 2001).

After the fuel has been placed in the canister, the insert is sealed with a lid that is screwed. The lid is then welded onto the copper shell and leaktightness testing is then conducted using NDT methods. The quantity of copper needed for the finished canisters is about 40,000 – 60,000 tonnes in Sweden and about 25,000-30,000 tonnes in Finland.

The copper material must first and foremost meet the requirements in accordance with the ASM UNS C101000 (Cu-OFE) or EN133/63:1994 Cu-OF1 standards. Other requirements besides these are: O<5 ppm, P 40-60 ppm (in the future, possibly 30-70 ppm), H<0.6 ppm, S<8 ppm and the grain size <360 μm in all conditions after fabrication. See *Table 6.1* for the quality requirements for copper (Andersson, 2002).

The fuel channels in the insert are fabricated in the form of an array (cassette) of square tubes. The walls and bottom of the insert are then manufactured by casting nodular cast iron around the channel array. The cast iron must comply with the requirements of the EN-GJS-400-15U standard. The insert is the heaviest component of the canister.

Table 6.1. Requirements on materials composition and comments on different properties of the copper canister material (Andersson, 2002; Appendix 2, Technical Specification no. KTS001).



Property	Specification	Comments
Weldability	O < 5 ppm	Higher levels give a reduced weldability.
Ductility	H < 0,6 ppm	Higher levels give reduced mechanical properties. (Hydrogen embrittlement).
Tensile strength, ductility	S < 8 ppm	Higher levels give reduced mechanical properties caused by non-dissolved sulphur which will be concentrated to grain boundaries.
Creep ductility	P 40 – 60 ppm	A phosphorus content of this order reduces the influence of sulphur impurities, increases creep ductility, increases recrystallisation temperature and has a minor influence on the weldability.
Microstructure	Grain size < 360 µm (Hot formed material)	This grain size gives a resolution at ultrasonic testing comparable to X-ray testing of 50 mm thick copper.
Ductility	Elongation > 40% RT – 100ºC (Hot formed material)	The canister will be deformed 4% in final repository.
Creep ductility	Elongation at creep-rupture > 10% RT – 100ºC (Hot formed material)	Same comment as above.

Three methods of NDT are used for the canister. X-ray radiography allows pore defects to be detected while ultrasound inspection allows defects that do not occupy volume to be detected, such as incomplete penetration, and eddy-current testing allows near-surface defects to be detected. Since the weld surface must withstand corrosion, it is important that the finished canister should not have any surface defects. Acceptance criteria will be established in the future for all of the parts of the canister, including the welds. The other destructive test methods make it possible to determine whether the test criteria are fulfilled by the NDT methods.

6.2 Fabrication

6.2.1 Copper Shell

Forging

The forging of copper tubes is a possible alternative to canister manufacturing. However, this process has not been completely investigated and developed. A large number of lids and copper bottoms have been manufactured through hot forging of continuous cast parent material. Homogeneous and defect-free material has been obtained for the finished components. The structure of the material is more coarse-grained than the material in extruded or pierce and draw processed copper tubes but meets the grain size requirement of the Swedish Nuclear Fuel and Waste Management Co's (SKB) specification (360 μm), see *Table 6.1*. Further development work is needed to optimise the forging process with respect to materials microstructure and usage.

Roll Forming

By roll forming 12 full-scale copper tubes have been longitudinally welded with electron beam welding for SKB. The weld technique has been improved and the weld quality has increased with time. Consequently, the method can probably be further developed, especially for thinner (30 mm) wall thicknesses, as a suitable alternative for tube manufacturing.

Extrusion and Pierce and Draw Processing

Extrusion and pierce and draw processing are two different methods for the manufacturing of drawn or seamless tubes. Both methods are currently suitable for canister fabrication. At least 14 seamless tubes have been manufactured, 11 by extrusion and three by pierce and draw processing, for SKB. 5 tubes have been fabricated for Posiva in Finland.

Hot Isostatic Pressing (HIP)

The HIP process is currently only a theoretical possibility since the largest HIP furnaces are only 3 m high. The HIP process can provide a small and even grain size compared with the other manufacturing methods, although the properties can only be moderately improved due to the absorption of oxygen during the process.

6.2.2 Cast Iron Insert

Many attempts (over 20 in Sweden and two in Finland) have been made to cast full-scale inserts. Since the mechanical properties of the cast iron are highly dependent on the dimensions of the body of casting, the materials testing must be conducted on the finished inserts. So far, these studies have shown a wide scatter in tensile testing results. This has been caused by both casting defects and microstructural inhomogeneity. The probability of obtaining a defect of a critical size increases with the size of the component and under the assumption that the greatest defect determines the canister's load-bearing capacity, the maximum permitted load – the size effect – in large components decreases. The casting process and the specification of the cast iron, EN-GJS-400-15U (EN 1563), (see also Andersson, 2001), must be optimised due to the above investigations of inserts. For the cast iron insert, reliable materials data are required as input data in connection with final mechanical strength calculations.

The canister with its cast iron insert must withstand considerable stresses under the hydrostatic pressure that can occur in a deep repository, for example, the groundwater pressure at a depth of more than 500 m in the rock, the pressure from the swollen bentonite buffer surrounding the canister and from a 3 km thick ice sheet (glaciation). Altogether, there might be a maximum pressure of about 45 MPa (450 bar) on the canister. In a recently conducted test, the model canister – with a cast iron insert with defects – managed a hydrostatic pressure that was three times as high, 130 MPa. Investigations of deformations and possible canister cracking are underway. On the basis of these studies, it will be possible to state, with a high degree of certainty, whether the canister will meet the requirements with an adequate margin (Nilsson and Burström, 2004).

The cast iron insert must be manufactured with high tolerance requirements as must the copper tube for the canister. Copper tubes manufactured by roll forming and longitudinal welding require more material for the final machining than the other manufacturing methods.

6.2.3 Lid Welding

Electron Beam Welding

Electron beam welding is a fusion welding method which in a vacuum (or under low pressure) with a strong electron beam melts the material through local heating. The method has several advantages: thick objects can be welded without consumables and weld parameters are programmable and reproducible. The weld has the same composition as the parent metal, but the oxygen concentration in the copper, in particular, has a negative impact on the weldability and the oxygen concentration must therefore be controlled. Even with a high-energy method, such as electron beam welding, the welding of copper is difficult to conduct due to the high thermal conductivity of the material and the low viscosity of the melt. Therefore, the electron beam welding method needs to be further developed with respect to equipment and welding parameters in order to achieve a stable process with high reliability (Claesson & Ronnetag, 2003). In particular, the seal weld must meet the requirements on long-term properties and durability.

Friction Stir Welding (FSW)

The principle for Friction Stir Welding is relatively simple. A rotating tool is pressed into the joint between the parts that are to be welded. The copper around the tool is heated up by the friction to over 800°C and becomes soft. The tool is then moved in the direction of the joint and the two metal parts are joined together. The fundamental difference, compared with electron beam welding is that the material does not melt during welding. A new large welding machine for FSW has been taken into operation at SKB's Canister Laboratory in Oskarshamn in 2003. The design is such that the welding head rotates during the process around the stationary canister. When the FSW technique is developed, both the lid and the base are welded to the tube and the copper tube can also be manufactured of two halves of roll-formed plate, especially if 30 mm thick copper is to be used. The development of the tool, together with the optimisation of the design is particularly necessary in order for the technique to function reliably. When full-scale welding is conducted on a 50 mm thick copper canister (3.3 m long weld) with a welding speed of about 100 mm/min, the welding time is up to an hour for the entire canister circumference. The welding temperature can be up to 950°C and the welding forces are high (Andersson *et al.*, 1999; 2000; Cederquist, 2003).

Narrow Gap (NG) Welding

Narrow Gap (NG) TIG welding is currently used to a large extent for the manufacturing of nuclear power components of steel. For several different reasons, for example, the fact that the thermal diffusivity of the copper is 10 to 100 times higher compared with steel and nickel-based alloys, this technique cannot be applied to thick-walled copper products since the heat transfer must be very high and the welding speed becomes slow (Pohja et.al., 2003).

6.2.4 Residual Stresses

After all manufacturing stages, with forming, machining and welding, residual stresses will occur in the material. These residual stresses must be measured and modelled. The residual stresses have a major impact on creep and stress corrosion. The highest permitted value for the residual stresses, which should be below half of the yield point, must be determined. Furthermore, the need for different techniques to reduce the residual stresses, for example, through stress relief annealing or mechanical surface treatment methods, must be evaluated.

6.2.5 Non-Destructive Testing (NDT)

The copper canister has defects after manufacturing, but only a few (0.1 %) of the canisters are allowed to have greater defects than those allowed by the acceptance criteria for the NDT (RD&D Programme 2001). The acceptance criteria have not yet been specified. One assumption is that these unacceptable defects can cause water leaks in the canister in 100,000 years' time. Bowyer (2000) has compiled an overview of all possible materials or manufacturing related defects and residual stresses that can occur in copper canisters and in cast iron inserts. Above all, defects in the canister lid weld are important. From the standpoint of corrosion, it is important to minimise the occurrence of fabrication-related defects. Therefore, it is essential that the size and form of various initial defects can be measured as accurately as possible. The requirements on the maximum grain size are important to facilitate ultrasonic testing. The acceptance criteria for initial defects must be based on the best available NDT methods. The sensitivity of the NDT methods must be verified with the help of metallography and microscopic investigations of defects and POD (probability of detection) diagrams for defects of different sizes, forms and positions must be generated. Further qualification of the NDT methods that will be used in the final process during the manufacturing and sealing of canisters must be conducted.

6.2.6 Encapsulation Plant

Trial manufacturing of thirteen full-scale canisters with cast iron inserts has so far been completed (Andersson, 2002). Five of these have already been used in different research projects.

The layout of the canister manufacturing plant was planned in Sweden (Andersson, 2001). The plant is expected to produce more than 200 canisters per year and contains equipment for machining canister shells and lids, welding of copper bottoms, machining of cast iron inserts, quality control and the final assembly of canisters. Finally, the finished canisters are delivered to the encapsulation plant. The handling of the canisters during manufacturing, transport and emplacement in the repository is critical for the subsequent corrosion behaviour of the canisters. Manufacturing methods, equipment and organisation still have to be established in order for canister manufacturing to be conducted as required to achieve a high level of productivity and quality in manufacturing. Further investigations concerning choice of method are needed with respect to welding processes and copper cylinder machining, in particular.

6.3 Durability

6.3.1 Corrosion Properties

The copper canisters will be affected by both general and different types of local corrosion in the complex chemical, microbial and mechanical environment of the repository, which varies in time and space. The probability of corrosion penetration in the canister should be very low in a 100,000-year perspective. During the first hundred or two hundred years, the copper shell will be deformed under compression. During the same time, oxidising corrosion conditions will occur in the repository. The risk of stress corrosion during this stage must be thoroughly evaluated. The threshold for the initiation and crack propagation of stress corrosion in copper must be measured in the repository environment under different modes of loading. There is a considerably better understanding of other corrosion mechanisms, both with respect to general and local corrosion (pitting and crevice corrosion), due to laboratory investigations and experience from marine and archeological copper discoveries. Considerable progress has also been made with respect to the modelling of these corrosion forms. However, a fundamental problem is the fact that the corrosion rates are based on short-term experiments. Therefore, it is uncertain whether these results are relevant to very long timescales. All known corrosion mechanisms with respect to copper have been summarised in a state-of-the-art report (King *et al.*, 2002). The corrosion properties of weld metals, where the microstructures vary and are quite different compared with the base metal, have been investigated to a limited extent.

When the copper shell has been penetrated due to some corrosion and rupture mechanism, the water will penetrate into the damaged canister and into the gap between the copper shell and the cast iron insert. The copper and cast iron are in contact with each other and galvanic corrosion occurs in the cast iron which causes hydrogen gas to form and leads to increased pressure inside the canister. Under anaerobic conditions, the rate of cast iron corrosion is still very low, less than 1 μm/year. The galvanic contact with copper in oxygen-free water will only cause a marginal increase in the corrosion rate. Verified experiments should still be conducted to show that galvanic corrosion is not probable in a repository environment for the canister configuration in question. After some time, water will come into contact with the spent nuclear fuel and tube material of zirconium and the actual fuel material will also be attacked by corrosion. At this stage, a number of corrosion mechanisms are active and modelling must be based on many different assumptions (e.g. Shoesmith, 2000). Due to the complexity and the possible interaction between different mechanisms, in order to better model how corrosion damage evolves in the damaged canister, empirical studies must also be conducted under realistic conditions in the future.

6.3.2 Creep Properties

After manufacturing, there is a gap of about two mm between the copper shell and the cast iron insert (depending on tolerances). This means that the copper must be capable of deforming about 4-5 % in the repository. Slow deformation, in the temperature range of 75 to 90ºC, which occurs in the repository under residual stresses together with the hydrostatic pressure and the pressure caused by the swelling of the bentonite buffer, generates creep in the copper shell. The copper which is used must have a creep ductility (maximum strain before rupture) of at least 10 % even after long timescales, both in the base metal and the weld metal. The importance of the phosphorus alloying (50 ppm) in the base metal for the creep-rupture strength and creep ductility of pure copper must be explained mechanistically. Information is also needed about the mechanisms for the long-term extrapolation of available data. It is important to explain the creep properties of weld metals, both for the electron beam and FSW welding, which have very different creep properties compared with the parent metal due to their highly variable grain size, which is of importance to creep. When the creep data for all of the canister materials is available, it is possible to carry out a Finite Element Modelling of the deformation of the entire canister.

6.4 Summary

The canister design has already been specified with high precision and the design principles can be considered to be good. However, flexibility must be maintained when ultimately selecting the manufacturing methods, such as lid welding. Full-scale manufacturing is probably easier with extrusion and draw and pierce processing compared with roll forming and longitudinal welding, which cause greater residual stresses in the canister. When selecting the manufacturing method, economic factors should not only be taken into account but also, for example, the long-term properties of the canister. The above-mentioned methods are known in the steel industry on a large scale, but have not been previously used for copper products. As a result, a very small number of companies are expected to be adept at these techniques on a large scale for copper.

The insert, which is of nodular cast iron, has not yet shown such acceptable mechanical properties and, therefore, the casting process must be analysed and better controlled or some other type of cast iron must be used. Casting defects must be more thoroughly analysed. Casting simulation can be a support when planning improvements in casting processes and for the design of different forms of casting. Also, a more accurate specification of the casting process (downhill or uphill) and requirements for the insert are necessary.

Welding methods, electron beam welding and FSW are potentially acceptable for high quality welding of canisters. Both methods, especially FSW, should be further developed. Electron beam welding is known to be suitable for steel products on a large scale and FSW has been previously applied to thick aluminium structures. FSW is a completely new technique which has never before been used for welding 50 mm thick copper. These methods should be further studied since both methods may be needed, especially for repair welding. Under all circumstances, a very deep understanding of the mechanisms that cause weld defects must be developed and the planning of repair welding must be started at an early stage. For this, different NDT methods are required in order to detect defects and to verify the quality of the canisters. It is also very important that no macro-defects, which can rapidly penetrate the canister, should occur during manufacturing and remain undetected. It is necessary to thoroughly follow the development of new NDT methods and to determine their limitations for different defects (Stepinski *et al.* 2004).

More research work, focusing on the long-term properties of the copper canisters with different manufacturing methods and conditions, is required to better predict future scenarios. More corrosion research is necessary, especially focusing on stress corrosion and microbial corrosion of the copper canister; at an initial stage, under laboratory conditions but also over a longer timescale and, if possible, also *in situ* in the actual repository.

To guarantee reliability throughout the canister manufacturing process and the final disposal period, acceptance criteria for all of the components of the canister, including welds, must be developed. These criteria should take into account material properties and defects, both surface defects and defects inside the material, in the copper shell and in the cast iron insert. Altogether, consequence analyses must be performed in order to predict possible processes when the canister does not meet the requirements that have been established. It is also important that the acceptance criteria can be verified by NDT methods and that a quality system for canister fabrication will be formulated.

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7 An Attempt at a Comparable Classification of Radioactive Waste and Hazardous Chemical Waste

7.1 Introduction

The quantification and classification of risks are currently being conducted in different ways for different activities. This means that the risks to which human beings are exposed are difficult to compare and the overall situation is difficult to evaluate. From the standpoint of society, it is desirable to obtain as many comparable assessments as possible. This chapter attempts to compare the risks of radioactive waste with the risks of hazardous chemical waste.

Waste is material that is considered to have too low a value to justify further use and must therefore be disposed of in some way. Waste therefore imposes an economic burden on the producer and society. However, it cannot be excluded that the waste could have a value in some other application than that where it was produced or in a future activity. Waste, which contains toxic substances, either in the form of radionuclides (radioactive substances) or toxic chemicals, is created by many human activities. Waste can arise as a result of the increased availability and concentration of natural substances and materials (mining activities, petroleum extraction, uranium extraction), industrial activities (melting facilities, metal industry, chemical industry, pharmaceutical industry), agriculture (weed killers, plant protection agents) or in households (batteries, fire detectors, electronics, medicines).

Many of the most important radioactive substances, from the standpoint of waste, are heavy metals and have chemical properties that are similar to those of non-radioactive (stable) heavy metals. They are non-volatile and less soluble in water than several other pollutants. It is also important to remember that radioactive substances with long physical half-lives can be more chemically toxic than radiotoxic. This applies, for example, to natural uranium in human beings and, probably also to radioactive iodine, ^{129}I, in soil.

Unlike organic pollutants – and like metals – radionuclides cannot be destroyed or degraded. Therefore, waste management is based on methods such as separation, concentration, volume reduction, fixation and isolation.

Radionuclides have the advantage over stable heavy metals that the amount of a radioactive substance is reduced through radioactive decay, even if the process occurs slowly in some cases.

Over the years, radioactive and chemical waste have come to be viewed differently. Classification systems and regulations are currently strongly associated with the source of the waste.

The difference in views has meant that the public often judges radioactive waste as hazardous while planners often have the view that safety margins are large with respect to radioactive substances. Similar risk-based classification systems and regulations for radioactive substances and chemically toxic substances would simplify environmental impact statements. For environmental and economic reasons, society would gain from a harmonised view based on evaluations that are made on principles that are as similar as possible. Such an approach would also broaden the perspective in the debate on a repository for spent nuclear fuel and would bring to the fore the need for a corresponding repository for heavy metals, such as mercury (SOU, 2001).

7.2 Proposal for a Comparable Classification of Radioactive and Chemical Waste

The National Council on Radiation Protection (NCRP, 2002) in the USA has recently proposed a system for classifying all types of waste containing either radioactive or chemically toxic substances. This chapter describes the proposal and discusses it in the light of the current system, which is different for radioactive substances and toxic chemicals. The proposed system is interesting in principle, since it provides the necessary basis for making the above-described similar evaluations of different types of waste.

In Europe, limited activities are conducted in the area within the OECD/NEA and within the European Union's Fifth Framework Programme (www.riskgov.org).

The system proposed by NCRP:

- can be used for each type of waste that contains radionuclides, hazardous chemicals or a mixture of these,
- has a classification that is based on a determination of the health hazards to the population resulting from the waste,
- has an exemption class for waste that involves such a low risk that it can be handled as non-hazardous waste.

The system comprises 3 waste classes:

High-level hazardous waste
Low-level hazardous waste
Non-hazardous waste

The system is based on the following principles:

- A linear dose-effect relationship without a threshold for cancer-inducing substances and with a threshold for non-cancer-inducing substances.
- The term "dose" should be given a uniform meaning. "Dose" currently means different things for radioactive substances

and for hazardous chemicals. In the case of radiation and radioactive substances, the "dose" is the absorbed dose (often average absorbed dose to the entire specified organ or tissue) or effective dose. The biological effect can be assessed from such dose data. The "dose" in the case of toxic chemicals (as for pharmaceuticals) is the amount of substance taken in.

- A uniform risk concept. The risk figures that have so far been used for ionising radiation and radioactive substances refers to the number of cancer fatalities while, for chemically carcinogenic substances, the number of cancer cases (incidence) is usually cited.
- The way of estimating health hazards is also different in the case of radionuclides and hazardous chemicals through the varying degree of caution that is incorporated into the postulated probabilities for undesirable health effects per dose unit and through taking into account different numbers of risk organs in the body.
- A number of exposure scenarios are assumed when estimating the risk that human beings are exposed to (exposure situations that can be used for each waste repository).

7.3 Designations for Risks to Individuals

For ionising radiation, the following risk assessments are often made

- *Unacceptable risk,* which must be reduced, regardless of cost or other conditions. This assessment is made if the radiation caused the extra risk of dying of cancer during the rest of the individual's life to be greater than an "acceptable" level (often a value in the range of 0.1-0.001 or greater or, in other words, between 10 % and 0.1 % or greater; where in the range it is, often depends on the exposure situation).

- *Acceptable risk* (risks under unacceptable levels as well as ALARA – "as low as reasonably achievable"). Risks just below unacceptable levels are considered to be just about acceptable and should be considerably reduced by applying the ALARA principle.
- *Negligible risk* is considered to be so low that further effort to reduce the risk (in accordance with ALARA) is not justified. This risk usually comprises an extra fatal cancer risk of 0.0001-0.000001 (0.01-0.0001 %) or less for the rest of the individual's life.

What we currently call "acceptable" risks or doses in the case of toxic chemicals corresponds to what is called "negligible" risk in the case of radionuclides, while "acceptable" risks or doses in the case of radionuclides may be far above negligible levels, providing that they are ALARA. In the case of hazardous chemicals, "unacceptable" is essentially the same as "non-hazardous". In the case of radionuclides, "unacceptable" refers to doses and risks that are far above negligible levels that cannot be tolerated under normal conditions.

Table 7.1. *Differences in the interpretation of "acceptable" and "unacceptable" risk (dose) for radionuclides and hazardous chemicals*

Risk description	*Interpretation for radionuclides*	*Interpretation for hazardous chemicals*
”Unacceptable”	Intolerable risk. This risk must be reduced, regardless of cost.	The risk exceeds negligible levels. Risk reduction must be considered but is only required to an appropriate extent.
”Acceptable”	Risk below intolerable levels and ALARA.	The risk is negligible. Further risk reduction is not necessary.

Risk, in the case of radioactive substances, is considered to be unacceptable even if it is below unacceptable levels, but not ALARA. This difference in interpretation between toxic chemicals and radioactive substances in risk assessment causes considerable difficulties for decision-makers and the general public as well as for those who have to provide information to these groups.

7.4 Proposed Risk Index for Waste Classification (NCRP)

"Risk" generally refers to the probability of damage occurring combined with the degree of severity of the damage (for example, death, reduced life expectancy, reduced kidney, liver or thyroid function).

For waste, the NCRP normally states the risk as the probability that something will happen to an individual or as the frequency of events in a population group. The following are required in order to calculate the risk

1. The probability of events resulting in a radioactive release
2. The probability that the individual or population group will be exposed to the release
3. The probability that exposure will result in damage

The risk from a repository/landfill can be expressed as a dimensionless risk index (RI). The risk index for the *i*th hazardous substance (R_i) is defined as the risk that arises from the disposal of the substance in question relative to a determined allowable risk for an assumed waste system.

$$\mathrm{RI}_i = \mathrm{F}_i \, \frac{\text{(risk due to disposal)}}{\text{(allowable risk)}}$$

F_i is a modifying factor for substance i and may depend on the design of the waste facility, the packaging of the waste, the uncertainty in the risk assessment etc.

For each substance where the risk can be assumed to be proportional to the dose (substance quantity/activity quantity) without a threshold, RI_i can be written as follows:

$$RI_i = F_i \frac{\text{(dose due to disposal)}}{\text{(allowable dose)}}$$

The difference in the meaning of "dose" for radionuclides and chemicals is uninteresting as long as the same meaning is applied to a given substance in the numerator and the denominator.

If we assume that the risk from an individual source is additive, the following is required

$$\sum_i RI_i < 1$$

namely, that the sum of all contributions may not exceed the permissible dose (or risk).

Adding the risks for non-carcinogenic substances requires caution, bearing in mind the fact that the dose-effect relationship does not have to be linear and that interacting (multiplicative) factors between different chemical substances cannot be excluded.

The advantage of the proposed risk index is that all toxic substances are treated in a similar manner.

7.5 A Risk-based Waste Classification System

The NCRP proposes that all types of waste should be classified into three classes.

I Waste Excluded from the Regulations

For non-carcinogenic toxic chemicals, the NCRP recommends that a negligible dose should be defined as a small fraction (for example, 10 %) of a certain threshold value for deterministic (predictable) effects in human beings. For radionuclides, it is recommended that an annual effective dose of 0.01 mSv should be considered to be a negligible individual dose. This dose corresponds to an estimated lifetime risk of cancer mortality of about $4*10^{-5}$ (0.004 %) for an assumed exposure time of 70 years (5 % per Sv). This dose is also the dose that the IAEA uses to define an exemption class for radioactive waste.

What is meant by negligible risks or doses for radionuclides and chemical carcinogens can also be discussed in relation to risks from natural background radiation (1 mSv/year) that cannot be avoided. Since the lifetime risk from exposure to natural background radiation and the natural occurrence of chemical cancer-inducing substances is about 1 %, a negligible risk could be determined as a part of this average background risk (for example 1 % of 1 %). In the long-term, such a “negligible risk” would be less than the variation in background risk, which arises as a result of differences in living habits.

II Low Risk Waste

Low risk waste can be deposited in a special landfill for hazardous waste. It should be possible to derive the limit for concentrations of hazardous substances by establishing that the risk or dose for an unintentional intrusion should not exceed acceptable (just tolerable) levels.

Acceptable (barely Tolerable) Risks or Doses

For non-carcinogenic toxic chemicals, an acceptable dose should be established at the threshold for the deterministic effect on humans or just below the threshold (for example, by a factor of 2 or 3) if an additional safety margin is desired.

For radionuclides, the limit for the annual effective dose to individuals among the general public is 1 mSv, which corresponds to an estimated risk of dying of cancer during the remaining lifetime of about 4×10^{-3} or 0.4 % (for an assumed exposure time of 70 years). This can be compared to the risk of dying of cancer from causes other than radiation which is just over 20 % or a little more than one out of every five people. Acceptable risk or doses can, as above, also be related to the unavoidable risk from the natural background.

III High-risk Waste

This waste cannot be deposited in landfills but must be deposited beneath the surface of the earth. Geological repositories have so far been the solution for high-level radioactive waste. This type of disposal is also now recommended for mercury (SOU, 2001).

7.6 Risk Estimates and Risk Comparisons

It is possible to, at least theoretically, estimate the risk from low radiation doses (ICRP, 1991). It is of course necessary to discuss this risk in the light of the risk that we accept, without further thought, in our daily lives, for example, risk from natural background radiation. On the other hand, the existence of other risks does not entitle exposure to additional radiation. However, the risk that we have previously accepted provides a framework for gaining a perspective on the risk of an additional exposure to radiation from radioactive waste.

Another way of gaining a perspective on the radiation doses to which we are exposed is to compare the hazards of radionuclides with the hazards of chemicals. There is a basic difference between radioactive substances (ionising radiation) and chemicals with respect to dose-response calculations. The dose-response calculations for radiation can be based on an estimated absorbed dose to organs and tissue in the body. Furthermore, the relationship between the dose and the response which was obtained from studies of groups of individuals exposed to radiation is applied to all radionuclides and most exposure situations. Thus, separate studies do not have to be conducted for each individual radionuclide as is required for each individual chemical. In the case of chemicals, the situation becomes more complex because there are about 30,000 substances, of which perhaps 20-25 % may cause cancer, damage to the embryo/ foetus and genetic effects (Bengtsson, 2002). For toxic chemicals, no units have yet been defined that correspond to the absorbed dose or equivalent dose, even if much work is currently being done to develop a “dose measure” (Törnqvist and Ehrenberg, 2001). The dose response relationship for specific toxic chemicals must therefore be based on studies of the specific substance.

Predictable (Deterministic) Tissue Effects

A basic principle in protection work is to prevent predictable (deterministic) tissue effects (for radiation: skin damage, cataracts; for chemicals: kidney and liver damage, neurological effects etc.) occur both as a result of radioactive substances and chemicals. For chemicals and radiation, the dose-effect curve is expected to have a threshold for deterministic effects. For each substance, the assumed threshold value is based on data for the most sensitive organ or tissue. However, there are important differences between radioactive substances and toxic chemicals in the way these threshold values are calculated and then applied for radiation protection work.

In radiation protection, dose limits for predictable effects (skin damage, cataracts) are only based on data from human beings and are normally established at a factor of 10 below the assumed threshold values. This safety factor is intended to ensure that deterministic effects are excluded for practically all individuals, including those who could be unusually sensitive to radiation. With respect to toxic chemicals, an even more conservative approach (which probably overestimates rather than underestimates the risk) is used. This is partly due to the fact that the toxicity of the substances has only been studied in animal experiments. Limits for acceptable doses are often defined by "reference doses", which are usually derived from the lower value of the uncertainty range for the assumed threshold values in the way that they are represented by the NOAELS (No Observed Adverse Effect Level) or lower confidence limits, for the benchmark dose (the dose where 10 % of the tested animals show an effect) by adding a great number of safety and uncertainty factors, most often of a minimum of a factor of 100, in order to obtain a reference dose. These safety factors can be as high as 5000 for some substances.

The reference doses for toxic chemicals therefore probably most often give a significantly greater safety margin than the dose limits for radiation-induced predictable effects.

Random (Stochastic) Effects

The basic principle for protection against both radioactive substances and toxic chemicals is that the probability for random effects, primarily cancer, should be limited to an acceptable level for the individual and society, seen in the light of the advantages that the activity generates or has generated. For each substance that causes random effects, a linear dose-response relationship without a threshold is generally postulated for health effects. This approach is well established in radiation protection and is gaining increasing acceptance with respect to estimating the risk of cancer from carcinogenic chemicals in the environment (Bengtsson, 1998; Granath and Ehrenberg, 1997; Duggan and Lambert, 1998; Granath et al., 1998; Törnquist and Ehrenberg, 2001).

The specified probability values for radioactive substances and chemicals that result in stochastic effects differ in two important respects. Firstly, the dose-response relationship for radiation and associated probability coefficients is based on the best possible estimates. On the other hand, as far as chemicals causing stochastic effects are concerned, the corresponding data are often intended to provide an upper boundary (the upper boundary in the uncertainty interval). In animal data, such a value can be 5 to 100 times greater than the best estimate. Secondly, the primary measure of random effects of exposure to radiation and radioactive substances has been the number of fatalities for the rest of life. On the other hand, in the case of chemicals, the measure has been the incidence, namely the proportion of those becoming ill or injured in a population as a result of exposure to a carcinogenic substance, which is explained by the fact that, in the latter case, the estimate is based on animal testing.

There is another difference between radiation and chemicals. Radiation is a more general carcinogen which can result in cancer in many more organs and tissues than chemicals. In radiation protection, the effective dose measure takes this into consideration. In the case of most of the toxic chemicals, only a single risk organ or tissue is taken into consideration and the rest of the body is ignored. The development of biokinetic models for toxic chemicals provides a corresponding possibility. However, such models have not yet been prepared to any great extent.

7.7 Calculation of Risk Figures

Different measures of cancer risk are established for radionuclides/radiation and chemicals. In order to classify radioactive waste, the risk figure of 0.05 per Sv (5 % per Sv) can be used, which is the figure that is normally used for the radiation protection of the public (ICRP, 1991). This figure has been derived from the best adaptation to epidemiological data for high doses, above all from Hiroshima and Nagasaki, and has then been further adapted for low radiation doses and dose rates (factor: 0.5). For chemical carcinogenic substances, the risk figures come from the upper value of the uncertainty interval for observed effects at high doses (mainly from animal studies). In several studies, the adjustment at the upper boundary of the uncertainty interval is 10 times higher than that obtained with the best possible adjustment.

For risk estimates, the risk figure mentioned above is applied to the effective dose. The risk figures for chemical carcinogenic substances are based on the observed effects on an individual organ or on a special tissue (often in animals). Attention is seldom paid to the possibility of effects on several organs. Risk figures for low doses of carcinogenic chemicals are more conservative (it is more probable that the risk is overestimated) than the risk figures for radioactive substances.

For radionuclides, the dose constraint at an effective dose of 1 mSv limits the deterministic effects. In the case of toxic non-carcinogenic chemicals the threshold for deterministic effects on humans is estimated from benchmark doses which, to an increasing extent, are used to obtain values for permissible doses of non-carcinogens. A benchmark dose is, as has already been mentioned, a dose that belongs to a specified effect level in a population that is studied (for example, a 10 % increase in the effect). The lower boundary of the uncertainty interval for the benchmark dose (which belongs to the 10 % increase) is then used as a basis for obtaining the permissible dose. In order to obtain a dose that will provide a safe protection for all human beings, it is recommended that a dose that is 10 times lower than the lower confidence level should be used for the "benchmark" dose which is obtained in connection with a well-conducted study of human beings and 100 times lower than the lower confidence level for a benchmark dose that is obtained in connection with a well-conducted study on animals.

In order for the proposed standardised classification system to be usable, risk figures for a very large number of chemicals must be obtained. The limited availability of such risk figures currently limits the applicability of the proposed system.

7.8 Examples of Comparative Limits for Radiation, Asbestos and Nickel

In this section, the limits that have been obtained for exposure to radiation, asbestos fibres and nickel compounds are compared (Schneider et al., 2000). Both epidemiological studies and animal experiments have clearly shown that asbestos fibres and certain nickel compounds can cause cancer in the same way as radiation. Epidemiological studies have shown that there is a relationship between relatively high exposure levels and an extra incidence of cancer. There is also cause for a linear, no-threshold relationship to be assumed between exposure and risk. The comparison between risk and permissible levels is based on the rules applied in France (which, at least with respect to radiation, are the same as in Sweden) and are illustrated in *Table 7.2*.

Table 7.2. Comparisons between risk estimates and permissible exposure levels for ionising radiation, asbestos and nickel. In this comparison, an exposure time of 40 years has been used.

	Extra risk of mortality during the expected remaining lifetime	*Permissible exposure level*	*Risk in connection with permissible exposure level for 40 years*
Ionising radiation, occupationally exposed persons	4 % per Sv	100 mSv/5 years	3 %
Asbestos, occupationally exposed persons	0.04 % per fibre/ml years	0.1 fibre/ml	0.16 %
Ionising radiation, public	5 % per Sv	1 mSv/year	0.2 %
Nickel, public	14 % 10^{-6} per ng/m^3 year	Tens of ng/m^3	0.01 %

The dose limits for ionising radiation, asbestos and nickel are based on extrapolations from known dose-risk relations at high exposure levels. In order to arrive at permissible exposure levels, a comparison with the risk of mortality in other occupations considered to be safe has been conducted for occupationally exposed persons. For the public, comparisons with natural background radiation have been conducted.

7.9 Consequences of the Proposed Classification System

The proposed classification system is applicable to every type of waste containing radioactive substances, toxic chemicals or a mixture of these two types of waste. The system is based on an assessment of the health risks to the public as a result of waste disposal. The system contains an exemption class, which contains waste which entails a very low risk and which can be handled as non-hazardous material. The other two classes are low-risk waste (can be stored in near surface facilities) and high-risk waste (requires an underground repository in bedrock).

In future work, the limits between the different waste classes can be quantified in terms of limits for concentrations of different substances so that the exemption group will contain such as low concentrations that the substances do not entail more than a negligible risk for a hypothetical "intruder" into a waste repository. The low-risk facility may not cause more than an acceptable risk for an intruder. Waste with a higher content than can be dealt with the two types of near surface facilities mentioned are classified as high-risk waste and need to be deposited in the bedrock.

The fact that considerable quantities of waste that contain small amounts of radionuclides or toxic chemicals can be excluded from the regulations simplifies handling and makes it cheaper. The high-risk waste will mostly comprise high-level radioactive waste, transuranic waste and long-lived radioactive waste with a lower activity. Chemical waste containing high concentrations of heavy metals (lead, cadmium, mercury) belongs to the same group. The current classification system for chemical waste does not contain such a class. It is assumed that the proposed system will be advantageous compared to the current system. It is simple and easy to understand. The clear connection between the classification and the requirements of protecting the public health will hopefully increase public confidence in waste management and disposal. The system has obvious advantages when it comes to handling mixed waste. The system provides the necessary conditions for a more just assessment of the hazard of different types of waste than the current system.

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