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Ground water chemistry at depth in granites and gneisses

Gunnar Jacks

Kungl Tekniska Högskolan Stockholm april 1978

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GROUND WATER CHEMISTRY AT DEPTH IN GRANITES AND GENISSES

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GROUND WATER CHEMISTRY AT DEPTH IN GRANITES AND GNEISSES

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0. SUMMARY

Available data make it possible to asses the composition of the ground water at depth what concerns most of the components. The place considered for a storage is beneath a local water divide along the eastcoast of southern and central Sweden.

pH is fixed by the carbonate system and the ground waters can be expected to be saturated with respect to calcite. pH may vary from about 7,2 to about 8,5 with the most probable value around 8. The content of bicarbonate should be reversly proportional to the pH, and 3 meq./l or 180 mg/l can be expected as a mean value. Ca^{++} may vary from 10 to 100 mg/l with the most probable value at 40 mg/l. The content of Cl^- is difficult to asses as it is associated with the presence of relict sea water. Relict sea waters are linked to the postglacial clays and low sections of the terrain. Near local water divides the content of Cl^- is low approaching the atmospheric contribution or below 10 mg/l. At depth fluid inclusions in minerals may contribute some Cl^- when the residence time of the ground waters in the rock becomes drastically extended. The content of F^- is is restricted by the solubility of fluorspar and may be about 7 mg/l as a maximum, but normally 3,5 mg/l or lower. SO_4^{--} seems to be of the same order as the atmospheric contribution or about 15 mg/l. In the deep boreholes sampled so far the contents have been lower. This may be due to a lower atmospheric transport in the past or a microbial sulphate reduction. The latter is not unlikely. The deep ground waters ought to be examined for sulphate reducing bacteria. The partial pressure of oxygen in the ground water should be very low. An increasing iron-content towards depth is likely. Organic substance in the order of a few tenths of mg/l may be present. It is in the form of fulvic acids with a complex-binding capacity of 10-15 meq./g. Inorganic colloids as Fe- and Al-Si-precipitates are quantitatively of minor importance. Part of the iron may be attached to the organic substance.

The local heating of the water close to the storage may bring about precipitation of calcite while the subsequent cooling

when the water leaves the storage may result in precipitation of aluminiumsilicates. This may have a sealing effect on the rock.

1. INTRODUCTION

In connection with the final storage of nuclear fuel it is important to know the chemistry of ground water in order to be able to asses the risk of corrosion on containers and the possibilities of transport of radioactive components with the ground water.

The relatively extensive ground water chemical data collected by the Department of Land Improvement and Drainage, Royal Inst. of Technology, Stockholm, have been used for the purpose along with published analyses and other evidences.

Experiences from ground water chemistry at the depth of about 500 m are limited. They are essentially limited to drainage water in deep mines. It is however doubtful whether these conditions are representative. The mines have been drained during long periods of time and this has increased the turn over rate of the ground water. Such conditions will prevail only during the construction of the storage.

In the following most conclusions will be drawn from analyses from depths of 0-100 m. Some trends in these data can safely be extrapolated towards depth. In some connections however these waters are comparatively young which makes it hazardous to extrapolate. At the depth of 500 m the water may be 10-1000 times older than the water in the upper part of rock.

2. DISSOLVED SALTS IN GROUND WATER

The dissolved salts in the ground water from igneous rocks have essentially three origins. These are via the atmosphere, from weathering and relict sea water from the time when the land was submersed below the sea.

2.1 Atmospheric salts

The atmospheric salts come as wet and as dry fallout. The precipitation washes out the salt particles that are carried in over land from the sea. The dry fallout is essentially a catching effect by the vegetation affecting small salt particles (a few μ in size) The salts come essentially from the sea (mainly NaCl), but also to some extent from the continents ($Ca\text{-}Mg\text{-}HCO_3$). Some ions in the surface and ground water are almost entirely derived from atmospheric circulation. This is true for Cl^- and SO_4^{--}. SO_4^{--} comes from diffuse sources in the sea and on land. In our country we also recieve considerable amounts of antropogenic sulfur from the burning of fossil fuels.

2.2. Weathering

Rainwater has a very weak weathering action. Only when it has percolated through the soil it becomes aggresive. The soil atmosphere has a high content of carbon dioxide formed at the decomposition of organic matter. The carbon dioxide dissolves in the percolating water:

$$CO_2 + H_2O \rightleftharpoons H_2CO_3 \rightleftharpoons H^+ + HCO_3^-$$

The hydrogen ions make the water acidic with an initial pH of about 5. The water is however very rapidly neutralized when the hydrogen ions are used up in reactions with minerals in the soil. An example of such a reaction is the transformation of Na-feldspar (albite) into kaolinite:

$$NaAlSi_3O_8 + H^+ + 4{,}5\ H_2O \rightarrow Na^+ + 2\ H_4SiO_4 + 0{,}5\ Al_2Si_2O_5(OH)_4$$

Through weathering cations (Na^+, K^+, Ca^{++}, Mg^{++}) are released and the corresponding anion is according to the above reactions HCO_3^-. The weathering thus gives a bicarbonate water.

The weathering converts primary minerals to clay minerals. Which clay minerals are formed depends on the parent material and a number of environmental factors as the supply of carbon dioxide and water to mention a couple. After some time an equilibrium is established between the water and its mineral environment.

3. MINERAL ENVIRONMENT IN FRACTURES

As mentioned above clay minerals are formed through weathering. Clay minerals will thus cover the fracture surfaces in the rock. In the old precambrian basement clays are formed not only through weathering but also by hydrothermal solutions that has circulated through the rocks when they were deeper buried than now (Brusewitz et al 1974, Uusinoka 1975).

As is shown in the weathering reaction above where a feldspar is transformed to kaolinite, silica is dissolved along with the cations. Under extremely leaching conditions only aluminium is retained in the solid phase in the form of gibbsite. During less leaching conditions increasing amounts of silica is retained and finally also K, Na, Ca, and Mg which are built in into the clays. It is therefore possible to set up a sequence of clay minerals that characterizes different degrees of weathering intensity:

gibbsite - kaolinite - illite - montmorillonite

As the system water-minerals consists of several components, several phases can be stable simultaneously, that is to say one can find several of the above clay minerals at the same spot.

The clay minerals generally found in the Baltic shield are those to the right in the above sequence. This indicates that the water circulation was never very intense in the rocks. Brusewitz et al (1974) have thus found montmorillonitic minerals in tunnels in the Gothenburg region. Uusinoka (1975) gives similar statements. The drill-core analyses done by Swedish Geological Survey in connection with KBS selection of sites show illite, chlorite and smectite (common name for clays of montmorillonitic type) as common fracture fillings.

Among non-clay minerals calcite is very common (Brusewitz et al 1974, Uusinoka 1975). This calcite may be of hydrothermal origin, or it may be precipitated from ground waters.

A common mineral in fractures is fluorspar (CaF_2), especially in more acidic rocks. In rapakivi-granites fluorspar is ubiquitous.

Pyrite and a black pitchy matter of hydrocarbons is a not uncommon association seen in iron mines in middle Sweden (Geijer & Magnusson 1944).

Deep weathering zones have locally been found in the mining districts of middle Sweden where obviously a very intense water circulation has taken place (Magnusson 1973).

Primary minerals are also found in fracture fillings, but the water chemistry shows that they are not stable. Only in very tight rocks in newly drilled wells water has been found to be equilibrated with the primary minerals (Jacks 1973). Such waters show high pH, above 10.

4. GROUND WATER COMPOSITION

4.1. pH and the carbonate system

In most natural waters the pH is determined by the carbonate system, that is to say the distribution of the species carbonic acid, bicarbonate and carbonate. They are converted into each other by the uptake or release of a hydrogen ion and thus make up a very effective buffer system.

The carbon dioxide is as mentioned the most powerful agent for the weathering. An aggresive water reacts with its mineral environment until it has reached equilibrium with it. This reaction can be brought to an end while the water is still in the rootzone of the soil. This is called an open system, that is to say it is open in relation to refilling with carbon dioxide. The other possibility is that the water after passing the rootzone is still aggresive and that it continues to react with its surrounding as it moves downwards. This is called a closed system; it is closed for renewal of its carbon dioxide, as the refill is restricted to the rootzone of the soil. In nature these extremes are seldom found but rather intermediate conditions.

The two systems give what concerns pH and CO_2-HCO_3^--CO_3^{--}-distribution different results. In the case with an open system carbon dioxide is all the time refilled which results in a water with a pH just above 7 and dominated by the buffer CO_2-HCO_3^-. In the other case, the closed system, the pH will be higher and the carbonate system is displaced towards the HCO_3^{--}-CO_3^{--} side. On the way to an equilibrium the carbon dioxide is consumed which makes this displacement possible. pH may sometimes exceed 10.

Whether one gets an open or a closed system depends on a number of circumstances as amount of precipitation, permeability and the presence of weatherable minerals in the soil zone.

A number of borewells at Gnesta, Skokloster and within the Verka area (Nilsson 1973) were sampled at different depths. The sampling was guided by conductivity logging. Fig. 1 shows pH as a function of the sampling depth. Fig. 2 shows the negative logarithm for the partial pressure of carbon dioxide as a function of the depth.

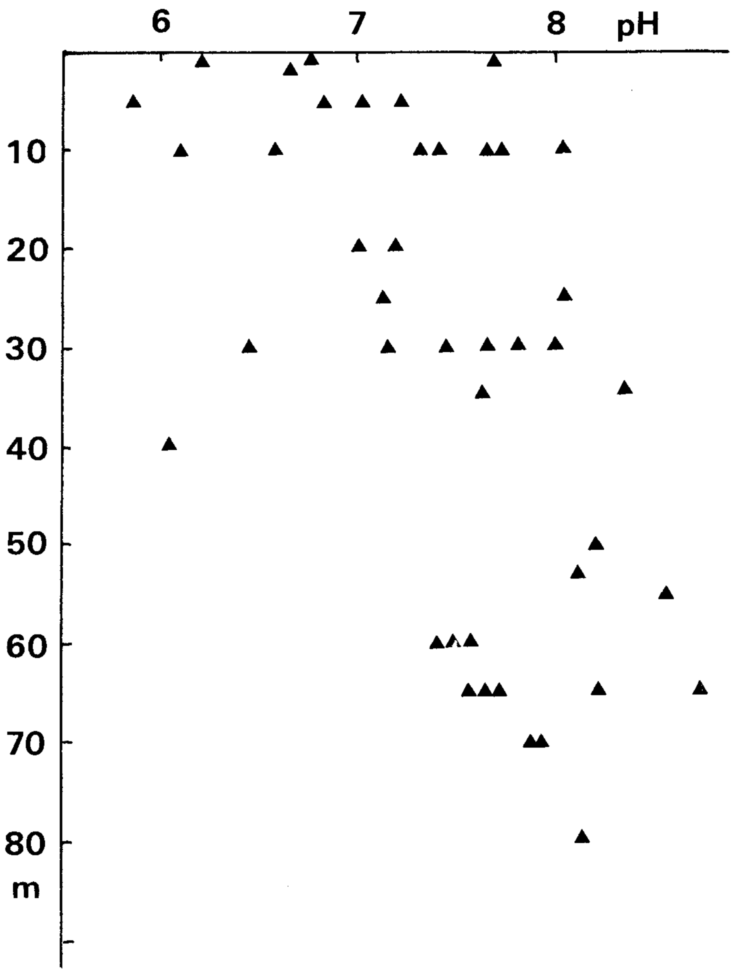

Fig. 1. pH as a function of sampling depth in wells at Gnesta, Skokloster and in the Verka Area.

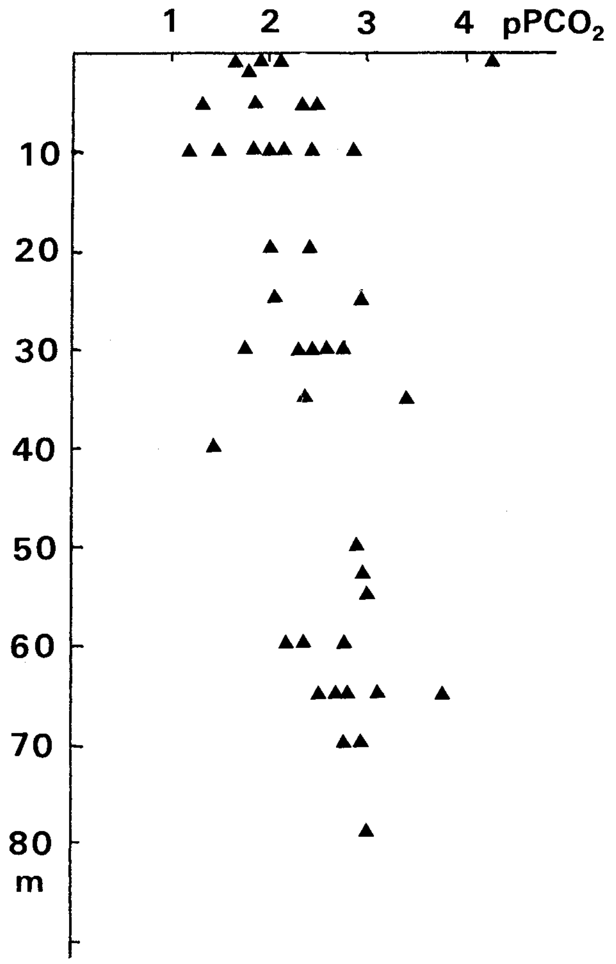

Fig. 2. Negative logarithm of partial pressure of carbon dioxide in ground water as a function of the sampling depth. Same wells as in fig. 1.

These figures show a partially closed system. If the water-mineral equilibria are plotted as well it is found that the reactions ought to be brought to an end at about 50-70 m depth. The ultimate pH to be reached can be expected to be about 8,2. Out of totally 3 meq./l of alkalinity about 1,4 is produced in the soil zone while the rest is produced at depth. The sampled wells are situated in forest terrain at relatively high altitudes and they fairly well represent possible locations for nuclear waste storage beneath local water divides.

In the above wells the alkalinity was in between 1 and 5 meq./l, which means 60-300 mg/l HCO_3^-. The mean was 3 meq./l. The content of carbonate is of interest as uranium forms strong carbonate complexes. Further copper, a proposed material for containers, forms a complex with carbonate ($CuCO_3^0$), in an oxidizing environment.

In a more extensive collection of data it was found that the alkalinity varied within the same limits, 1-5 meq./l.

What concerns the relation of alkalinity to the above mentioned open and closed systems it should be true that an open system gives a higher alkalinity than a closed system. Thus one gets the combinations high pH-low alkalinity and neutral pH- high alkalinity.

Thus summarizing the above discussion:

Closed system	Open system
high pH	lower pH
low alkalinity	high alkalinity
high precipitation	low precipitation
permeable soil	tight soil
resistant minerals	easily weathered minerals
forest areas	agricultural land

Among minerals the presence of calcite is most important as calcite is the most easily weathered mineral in Swedish clays and tills.

Finally it should be mentioned that the partial pressure of carbon dioxide can be expected to be low at great depth or about 10^{-3} atm. The maximum partial pressure encountered is about $3 \cdot 10^{-2}$ atm. Considering a pH of 8 and 300 mg/l HCO_3^- this gives a partial pressure of carbon dioxide of $2{,}2 \cdot 10^{-3}$ atm or $pPCO_2 = 2{,}65$. This is equal to 5 mg/l CO_2.

4.2. Calcium and sodium

Ca^{++} and Na^+ are the two dominating cations in ground water. It is justified to discuss them in one connection as they partly have the same origins.

Ca^{++} as well as Na^{+} are added to the ground via the atmosphere. However out of the normal contents only a small fraction is atmospheric. For Sörmland-Uppland the contents in precipitation have been assesed at 0,9 mg/l Ca^{++} and 0,4 mg/l Na^{+}. With 50 % dry deposition and concentrating three times through evapotranspiration (Nilsson 1973) the atmospheric contributions should be 2 mg/l Na^{+} and 4 mg/l Ca^{++}. This should be the minimum amounts to be expected in ground water. However both Na and Ca are contained in relatively easily weathered minerals in most rocks, even granites and gneisses. The plagioclases are feldspars varying in composition between the two end members albite (Na-fp) and anorthite (Ca-fp). They are easily weathered (Grant 1963). The albite is associated with acid rocks while the anorthite is found in the basic ones. The sedimentary gneisses in Sörmland-Uppland usually contain oligoclases with Na and Ca in molar proportions of 75/25 (Stålhös 1969).

In acid granites Ca^{++} contents down to 10 mg/l have been found. Usually the contents are higher or about 40 mg/l. Na^{+} is present in similar amounts. This is true for Sörmland-Uppland, and it indicates that calcite in clays and tills give off considerable amounts of Ca^{++}. Especially in Uppland there is a remarkable content of calcite in the tills (Gillberg 1967).

Towards depth there is established a certain relation between the contents of Ca^{++} and Na^{+} . Probably this is due to an ion exchange on the clay minerals in the fractures in the rocks. Ion exchange of Na^{+} and Ca^{++} on montmorillonite can be written:

$$\text{Ca-mont} + Na^{+} \rightleftharpoons \text{Na-mont} + 0{,}5\ Ca^{++}$$

The law of mass action gives:

$$\frac{\left[Ca^{++}\right]^{0,5}}{\left[Na^{+}\right]} = K$$

If this expression is written in negative logarithms:

$$pNa^{+} + 0{,}5\ pCa^{++} = pK$$

According to Jacks (1973) this expression moves towards 0,5 at depth in borewells. This gives as a result that in relict seawater Na^+ is never found in amounts equivalent with Cl^- but is partly substituted by Ca^{++}.

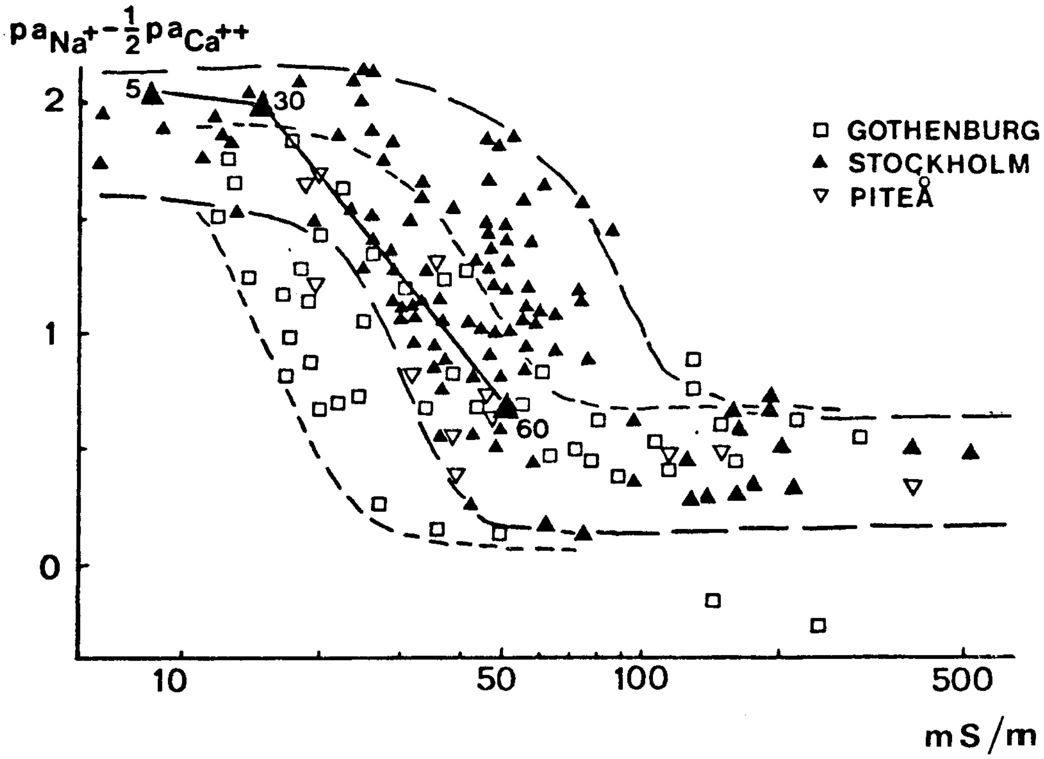

Fig. 3. Ion exchange on clays illustrated by the expression $pa_{Na^+} - 0{,}5pa_{Ca^{++}}$ plotted against electric conductivity. Interconnected triangles with figures 5-30-60 means water sampled in one well at 5, 30 and 60 m depths.

In addition to the equilibration with respect to montmorillonite as evidenced by the water chemistry, the water also becomes saturated with respect to calcite (fig. 4.). The diagram is valid for Sörmland-Uppland. The saturation may be reached at a slower rate for other areas (Wenner et al 1974). Ground water below the depth of 100 m may be regarded as being saturated with respect to calcite however. This is evident from the water chemistry as well as from studies of fracture fillings (Brusewitz et al 1974, Olkiewicz et al 1978).

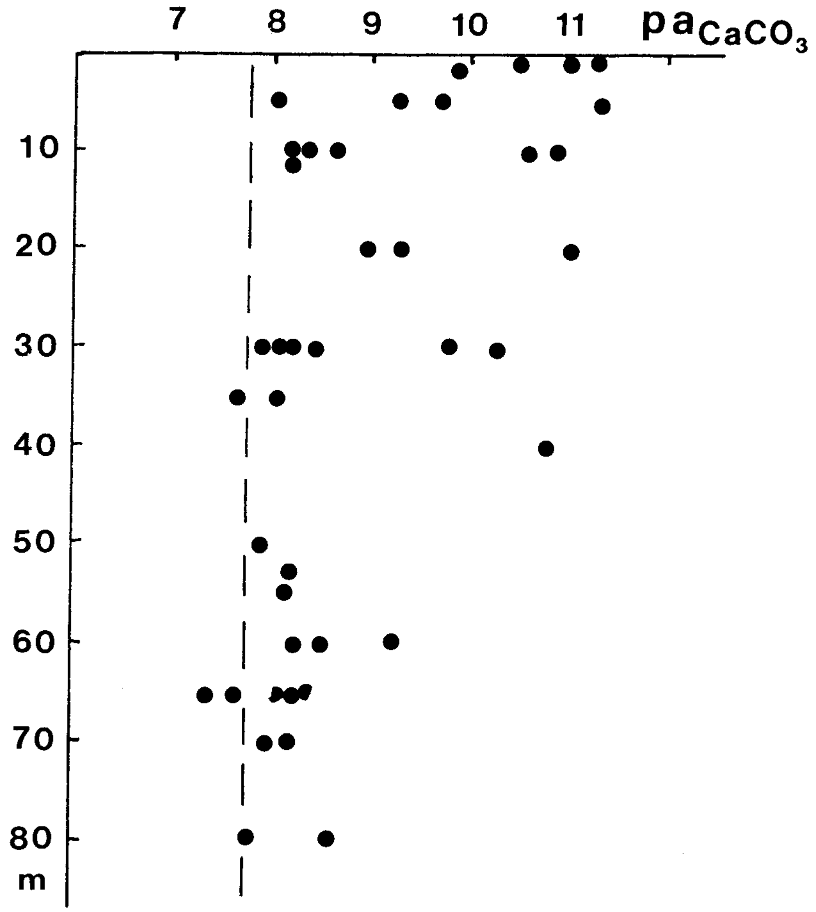

Fig. 4. Activity product for $CaCO_3$ in ground waters plotted against depth of sampling. Broken line indicates saturation with respect to calcite.

Considering a number of probable combinations of pH and alkalinity it is possible to calculate the Ca^{++} content of the ground water at saturation with respect to calcite. Activity factors are set to 0,9 and 0,7 for HCO_3^- and Ca^{++} respectively. This is valid for an ionic strength of about 0,01 or an electric conductivity of about 70 mS/m. In the table below three combinations are treated.

pH	HCO_3^- mg/l	Ca^{++} mg/l	$pPCO_2$	CO_2 mg/l
7,5	300	86	2,14	15
8,0	180	46	2,87	2,7
9,0	60	14	4,34	0,1

The second alternative ought to be the most probable, but also the other ones may be found depending on conditions of infiltration as is discussed above in chapter 4.1.

4.3. Potassium

The atmospheric contribution of potassium to the ground water is about 1,2 mg/l. The content actually found is about 3 mg/l and very seldom above 5 mg/l. The small increment due to weathering is explained by the resistence of the potassium silicates like K-feldspar and K-mica. Besides that K^+ is fixed in weathering products e.g. illite.

4.4. Magnesium

The atmospheric contribution of magnesium is about 0,75 mg/l. In ground water 5-15 mg/l are usually found. Mg^{++} is derived from ferromagnesian minerals like pyroxenes, hornblende and biotite. They are fairly easily weathered. Mg is however retained in secondary minerals as chlorite and montmorillonite. Possibly the presence of chlorite along with kaolinite may act as a check on Mg^{++} content in ground waters (Jacks 1973). It is usually found that Mg^{++} on a molar basis is about 1/3 of the Ca^{++} content. This is persistent towards depth and may be a manifestation of an ion exchange equilibrium on some of the solids formed.

4.5. Chloride

Chlorine occurs in small amounts in granites and gneisses, about 200 ppm are found according to Wedepohl (1970). In biotite chloride may be found in the form of small fluid inclusions. The amount is in the order of 2000 ppm (Wedepohl 1970). Normally this content is of little significance as has been found in chemical balances for areas studied during the IHD work (Eriksson & Holtan 1974). In Stripa however there is observed an increase of Cl^- in ground water with depth (P. Fritz, personal communication, Olkiewicz et al 1978). Those ground waters have been subject to a very deep and slow circulation and it is possible that part of their chloride content comes from a slow diffusion from the mentioned fluid inclusions.

The lowermost Cl^- content to be expected is the Cl^- brought in via the atmosphere as wet and dry fallout. For an area covering Sörmland-Uppland the precipitation has been observed to contain about 0,6 mg/l Cl^-. To this should be added the dry fallout, possibly about 50 % of the wet fallout. The resulting Cl^- deposition is then concentrated by evapotranspiration. According to Nilsson (1973) the runoff is about 200 mm out of 600 mm precipitation. This gives a factor of concentration of 3 and a minimum content of 3 mg/l should be expected in the ground water. The actually found contents are in many cases only slightly higher.

Considerably higher contents of Cl^- are found when relict sea water from the Litorina sea is left over in the rock (Lahermo 1971, Jacks 1973, Wenner et al 1974). The question how common such relict waters are is difficult as there are many factors that influence their washing out. Among such factors permeability and hydraulic gradients may be mentioned. Several investigators (e.g. Wenner et al 1974, Agerstrand personal communication) forward the relation between fine sediments (postglacial clays) and salty waters in the fractures of the rock. This seems logical. Probably there was no extensive penetration into the fractured rock as long as the sea covered the land. The free water surface gave no gradients for flow. The differences in density were probably not effective in replacing the sweet water with salty. However in the Litorina sea clays were deposited with salty pore waters. When the uplift of the land moved these clays above the sea level gradients were formed and the salty pore water began to move, sometimes finding its way into the fractured rock. This washing out is still active and the position of the salt water front depends as mentioned on gradients and permeabilities. When sediments were thin and placed at some altitude the wash out may be completed. Deep down in the rock however there may still be salt water under elution. This makes it delicate to asses how common such relict waters are. A few criteria can however be listed. The probability of striking relict salt water increases with the following factors:

- presence of postglacial clays
- flat topography
- decreasing altitude
- decreasing permeability

If one wants to avoid relict waters one should thus avoid areas with thick postglacial clays. As the storages are intended to be placed under water divides there should be good possibilities of avoiding salt water. Problems may arouse during the time the storage is constructed and when there is in principle a reversed flow pattern around the site.

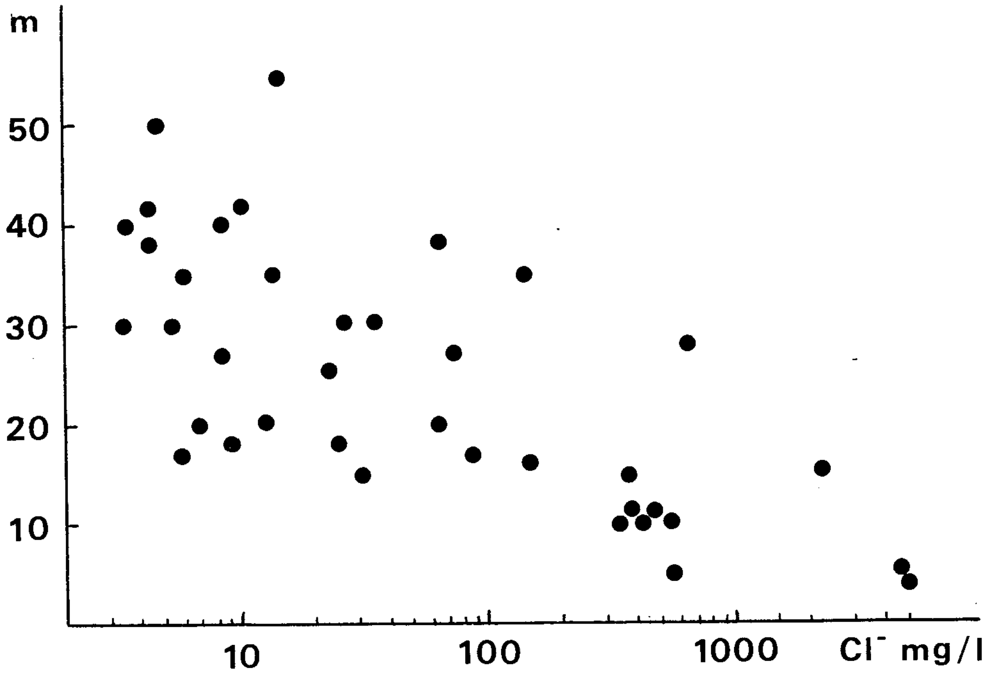

Fig. 5. Cl^- in wells in relation to the altitude (ground level).

Fig. 5. shows the content of Cl^- as a function of the altitude (ground surface) of a number of wells. The most salty waters are found close to the present level of the Baltic. The highest levels of Cl^- are about 5000 mg/l. Svenonius in Sandegren et al (1922) reports 5115 mg/l from the Rörberg iron mine in northern Uppland. 5000 mg/l is also found on Värmdö (Jacks 1973). From Finland is reported 6300 and 4900 mg/l by Lahermo (personal communication). A number of wells with about 2000 mg/l are listed by Engqvist (1969) and Lahermo (1970).

Fig. 6. shows two bardiagrams illustrating the content of Cl^- in borewells situated in agricultural land and forest respectively. The median content in wells in forests is only about half of that in agricultural land. This is due to the presence of clays in the latter and a lower topographic setting.

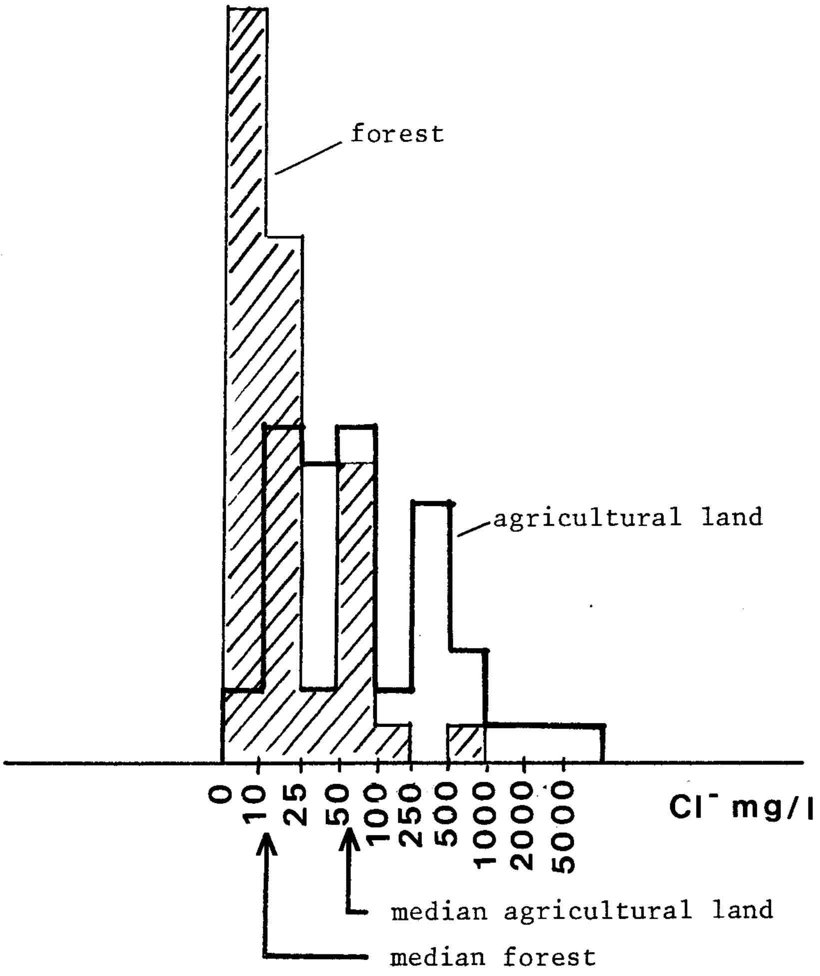

Fig. 6. Contents of Cl^- in wells situated in agricultural land and in forests (hatched).

In Finland a brine has been discovered during drilling in Outokumpu coppermines (Maijala 1969). The content of Cl^- was 16 g/l and the water was accompanied by methane and other gases.

4.6. Sulphate and sulphide

SO_4^{--} like Cl^- is a constituent that circulates via the atmosphere. In representative areas investigated during the IHD it has been found that the addition of SO_4^{--} from pools in the ground is small if none (Eriksson & Holtan 1974). For SO_4^{--} as well as for Cl^- it should be true that the lowermost content should be equal to the deposition. For Sörmland-Uppland the content of SO_4^{--} in precipitation during the period 1955-66 has been measured at 3,2 mg/l. With 50 % addition from dry fallout and a concentration three times through evapotranspiration the final content in ground water should be 14,4 mg/l. Fig. 7 illustrates contents of SO_4^{--} in ground water in the mentioned area .

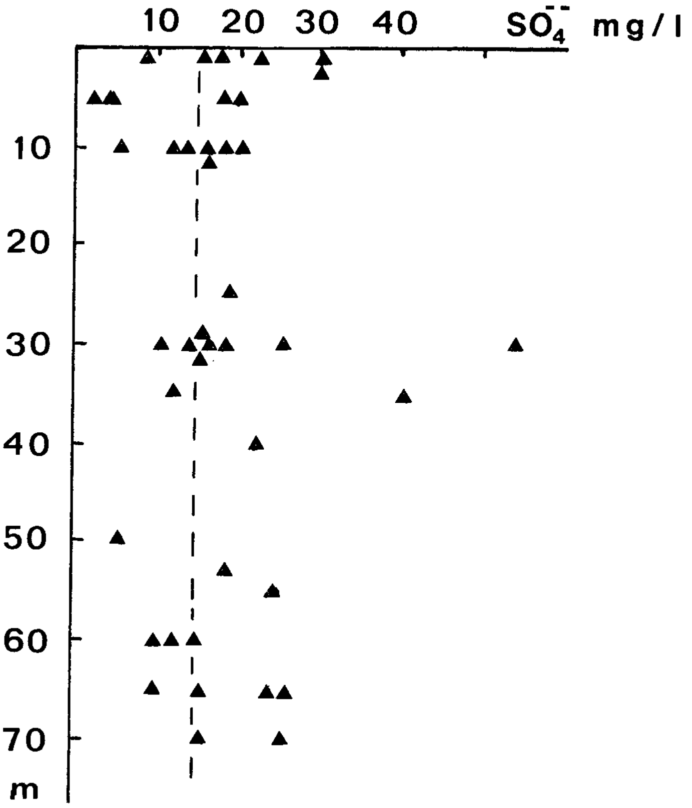

Fig. 7. SO_4^{--} in ground water as a function of sampling depth. Hatched line = atmospheric contribution.

Contents lower than the calculated atmospheric contribution are not uncommon. This may be due to several phenomena. Transport of sulphur via the atmosphere was lower a few decades ago. Sulphur is also a biogenic element which may be accumulated in the organic matter. Sulphate reduction is still another possibility. The smell of H_2S may occasionally be observed from borewells in precambrian rocks. Rennerfelt (1977) has detected sulphide in deep boreholes. Whether this sulphate reduction has taken place during the infiltration in the soil zone or it has occured in the fractured rock is as yet not known. Both possibilities must be considered. Organic substance as a substrate for microbiological growth is present in the form of humic substances in deep ground waters. When drifting at depth in mines in Bergslagen organic precipitates are observed on the walls (Wesslén personal communication). The humic substances are however not easily biodegradeable. By cultivation on a substrate containing sulphate and lactate it is possible to confirm the presence of sulphate reducing bacteria (Bastin 1926).

Higher contents of SO_4^{--} are related to relict sea waters and the same discussion as concerns Cl^- can be applied here. The maximum SO_4^{--} content observed is 800 mg/l. However this is an extreme. Usually the deep ground waters are depleted in SO_4^{--} if compared with the present day composition of the Baltic sea. This is most probably due to sulphate reduction either at the sea floor or at depth.

4.7. Fluoride

Fluoride in ground water is derived from several minerals. The most easily soluble fluorine in granites and gneisses is in the form of fluorspar (CaF_2). Considerable amounts of fluorine are present in micas and pyroxenes in -OH positions (Koritnig 1950). The fluorapatite is the most insoluble among common fluorine minerals.

In natural waters the solubility of fluorspar is usually limiting the fluoride content (Handa 1975, Jacks 1973):

$$CaF_2 \rightleftharpoons Ca^{++} + 2\ F^-$$

In consequence of this reaction soft waters should be those having highest content of fluoride. This is also found. Such waters are found in acid granites where the plagioclase is a pure albite. Rapakivigranites are usually acid and in addition to that they usually carry fluorspar as an accesory mineral.

Fig. 8. shows F^- as a function of Ca^{++} in a number of ground waters from southern central Sweden. The plot scatters just under line depicting the solubility of CaF_2.

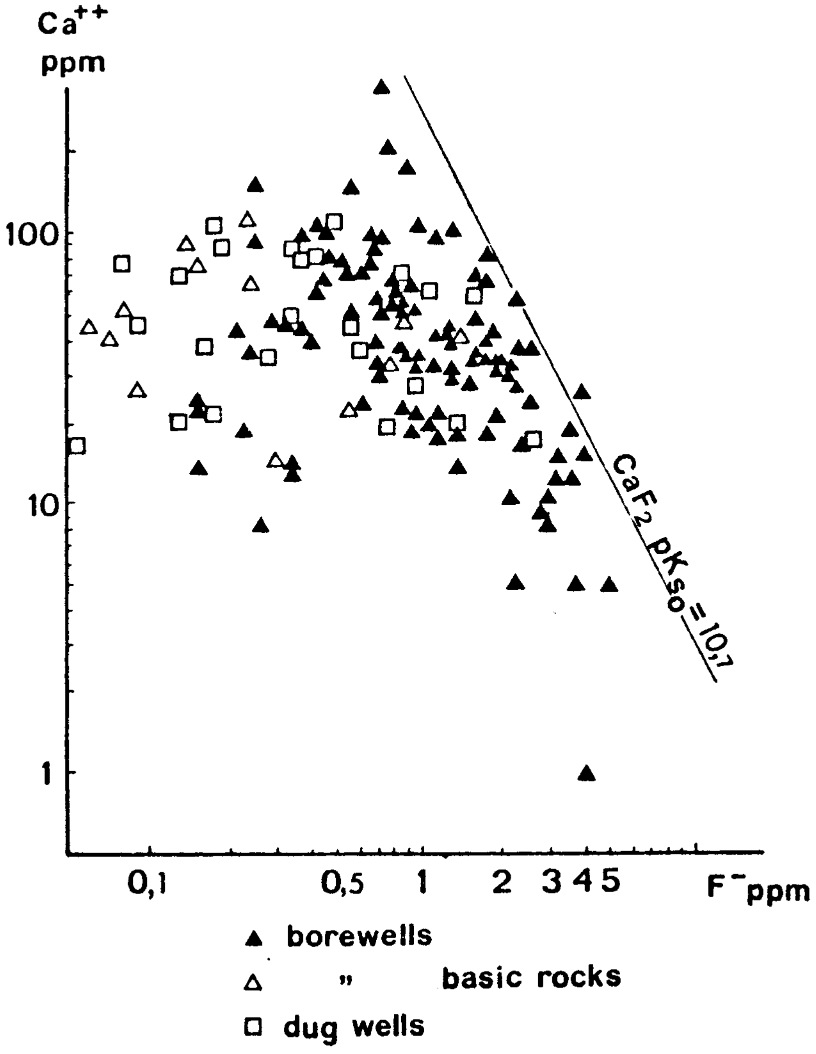

Fig. 8. Relation between Ca^{++} and F^- in ground waters.

The maximum contents found are about 5-7 mg/l F^-. Usually the ground waters are slightly undersaturated with respect to CaF_2. The ground water from the deep borehole in Kråkemåla (Gidlund 1978) in connection with site selection by KBS is however slightly oversaturated. The rapakivigranite there contains fluorspar as a fracture mineral. The saturation with respect to fluorite is rapid, while the saturation with respect to calcite is achieved slowly afterwards by dissolution of calcium-silicates. This gives a secondary oversaturation with respect to fluorspar.

Assuming that calcium is present only in the form of fluorspar it is possible to calculate the contents of Ca^{++} and F^- at saturation with respect to fluorspar. If the electric conductivity is 70 mS/m and ionic strength 0,01, activity factors for Ca^{++} and F^- are 0,7 and 0,9 respectively, their solubilities becomes 7 mg/l,equal for both ions. At the normal content of 40 mg/l Ca^{++} and the same ionic strength the solubility of F^- becomes 3,5 mg/l.

4.8. Nitrate and ammonium

The content of NO_3^- is of interest as it may act as an electron acceptor in microbial oxidations.

The content of NO_3^- varies within wide limits as a result of human influence in some way or other. Within the Verka area in Uppland 24 wells show a mean value of 1,3 mg/l NO_3^--N or 5,8 mg/l NO_3^-. The highest value was 40 mg/l as NO_3^-. These wells are situated in an area dominated by agricultural land. For wells in forest areas (Sörmland-Uppland) a mean value of 0,3 mg/l of total-N is found, with less than half being NO_3^--N. The mean content of NO_3^- was 0,6 mg/l. Ground waters infiltrated in forest areas can be expected to be low in NO_3^-. This is also found by Fleetwood (1969) from collection of seepage water in a tunnel north of Stockholm.

The distribution between NH_4^+-N and NO_3^--N does not change towards depth in the studied wells indicating that no biochemical conversions concerning nitrogen take place in the fractured rock.

In this connection it may be mentioned that ground waters in the Alnarp valley (Scania) subject to sulphate reduction did not show any signs of nitrate reduction. There was no correlation between SO_4^{--} and NO_3^- (Jacks 1977). Thermodynamically NO_3^- should be more easily reduced than SO_4^{--}.

4.9. Phosphate

Phosphate is of importance as it is a micronutrient for bacteria. Phosphate is found in amounts of around 0,01 mg/l. Hydroxyapatite is a common mineral in most igneous rocks and should furnish a control on the phosphate content (Stumm & Morgan 1970). The low phosphate contents however does not restrict the limited bacterial activity that may occur at depth in the ground water. The restraint should rather be the presence of suitable organic matter.

4.10. Silica

The solubility of quartz is about 6 mg/l as SiO_2 and that of amorphous silica just above 100 mg/l. In ground waters in granitic rocks the content of SiO_2 is usually in between the two figures or 10-60 mg/l, with 10-25 mg/l being the most frequent interval. It is likely that the assembly of secondary clay minerals act as a control on silica content rather than quartz or amorphous silica (Stumm & Morgan 1970). However the content of dissolved silica is lower than would be expected from e.g. the formation of kaolinite from feldspars. This may be due to the formation of an amorphous precipitate of aluminiumsilicate early in the weathering process (Pačes 1973).

Towards depth only slight changes can be expected as the mineral-water equilibria are established usually within the upper 100 m of the crust.

4.11. Oxygen and redox-conditions

Thermodynamic considerations make it likely that the ground water at depth has very low partial pressure of oxygen (Eriksson & Khunakasem 1970, Brotzen 1978 in print). From practical points of view it is free of oxygen. The considerations are based on the equilibration between the water and common minerals like pyrite and magnetite. The low oxygen contents to be expected cannot be measured. Rennerfelt (1977) has with a refined technique measured the oxygen content in the deep boreholes made by KBS and he has found contents below or close to the detection limit of 0,01 mg/l O_2.

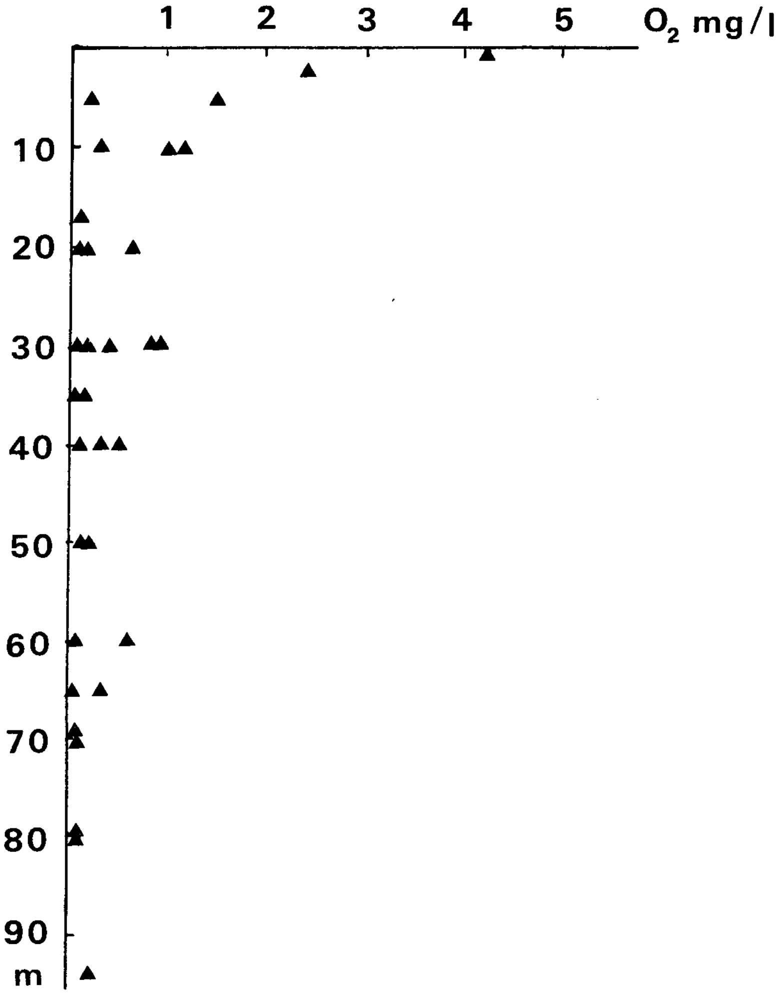

Fig. 9. Content of O_2 as a function of sampling depth for wells at Gnesta and Skokloster.

According to fig. 9 the oxygen content in wells at Gnesta and Skokloster shows a decrease towards depth. Conventional Winkler sampling was used. Contents of 0,2 mg/l O_2 and below may represent oxygen free water. Measured redox potentials also show a decrease towards depth from roughly 0,3 V to 0,1 V at 70-80 m depth. Grenthe (1978) has measured redoxpotentials carefully in deep ground waters (400 m) and found about -0,17 V with an Au-electrode. Contents of Fe^{++} increase towards depth in the above wells, indicating more reducing conditions.

The mere presence of Fe^{++} indicates that PO_2 is very low (Eriksson & Khunakasem 1970, Hem 1977). Iron given in analyses may however be complexed by organic substances (see 4.12.).

That the O_2 content of the ground water is slowly depleted towards depth is made plausible by several authors who states that borewells have higher contents of iron than wells in the quaternary (Arrhenius 1954, Hedin 1967).

4.12. Organic matter and colloids

Ground water in hard rock has been found to contain organic substance in the order of a few tenths of mg/l (Rennerfelt 1977). Uusinoka (1975) has three analyses from the depth of about 45 m showing 15 mg org. C/l, corresponding to about 30 mg/l of organic substance. The organic substance is of importance because it can support microbial activity and it can through its complexing character contribute to the transport of radionucleides by the ground water. The organic substance present is in the form of fulvic acids. Fulvic acids consists of benzene-rings with attached active groups, essentially being carboxyls. These fulvic acids have through gelfiltration been found to have certain spectra of molecular weights from about 700 to 50 000 (Hall & Lee 1974). The most important complexing groups in fulvic avids are carboxyls and the capacity is in the order of 10-15 meq./g of fulvic acid (Gamble & Schnitzer 1973).

Whether the organic substance can support microbial growth is not clear. Sulphate reducing bacteria can exist at very low concentrations of dissolved organic matter in the form of fatty acids (Kutznetsov et al 1963). According to calculations done on the conditions found in the Alnarp ground water body the content of organic matter suitable for Desulfovibrio need not be higher than 10^{-20} M (Jacks 1977).

Colloids are essentially of two kinds, Fe-hydroxides and Al-Si-colloids. Differences in content of total iron and Fe^{++} may indicate the presence of a few mg/l of ironhydroxides. The Fe not being detected as divalent may however also be complexed to the organic substance. The latter alternative is actually the more probable one. The amount of Al-Si-colloids may be estimated from the differences in dissolved Al usually found when filtering with successively finer filters (Hem personal communication 1977). Discrepancies what concerns dissolved Al are in the order of a few tenths of ug/l as a maximum. This may indicate the presence of a few tenths of ug/l of Al-Si-colloids in the ground water.

The presence of organic matter and colloids, being of importance as carriers of radionucleides, may be summerized as follows:

substance	magnitude
fulvic acids	tenths of mg/l
Fe-hydroxides	$<$ mg/l
Al-Si-colloids	µg/l

Recently the organic matter in ground water from a 500 m deep borehole at Finnsjön in northern Uppland has been examined at the Department of Chemistry, Agricultural University of Sweden in Uppsala (Andersson 1978). The investigation was carried out by means of filtration on CPG-170 (Controlled Glass Pore, Electro-Nucleonics Inc., Fairfield, N.J., USA). The filtration showed two sizes of macromolecules, one being fulvic acid the other possibly being some iron precipitates. The content of organic carbon was 15 mg/l corresponding to about 35 mg/l of fulvic acid or 0,5 meq./l.

5. EFFECTS OF TEMPERATURE AND PRESSURE

The temperature has a certain influence on the water-rock equilibria that are established. The increased pressure at the actual depths does not appreciably change the equilibria.

The ground water temperature is in middle Sweden 6-7 °C close to the ground. The geothermal gradient is about 15 °C/km which is observed in boreholes and mines (Nämnden för Energiproduktionsforskning 1977). At 500 m depth the temperature will be about 15 °C.

The solubilities of most silicates increase with the temperature while the carbonates show a decreased solubility (Helgeson 1969, Bernhardsson 1977). This may bring about a slow deposition of calcite from the ground water and a similar slow dissolution of silicates during the movement downwards.

The above mentioned effects are accentuated close to the storage due to the local heating caused by the decay of the nuclear waste. On approaching the storage the water is heated and calcite is precipitated while silicates may dissolve. On leaving the storage the water is cooled and the reverse process is occuring. Aluminiumsilicates in crystalline or amorphous form may precipitate. Possibly the precipitates will tend to cement the fractures and cause decreased permeability.

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