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Chemistry of deep groundwaters from granitic bedrock

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This report concerns a study which was conducted for SKBF/KBS. The conclusions and viewpoints presented in the report are those of the author(s) and do not necessarily coincide with those of the client.

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CHEMISTRY OF DEEP GROUNDWATERS
FROM GRANITIC BEDROCK

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SUMMARY

Water analysis data from Fjällveden, Gideå, Svartboberget and Kamlunge (7 different drilling holes, 26 sampling levels at vertical depths between 100 and 600 m) are discussed. Most of the waters are $\text{Na}^+ \text{-Ca}^{2+} \text{-HCO}_3^-$ -dominated with a total salt content of 200-300 mg/l and pH of 8-9. Intrusions of $\text{Na}^+ \text{-Cl}^-$ -dominated saline waters (up to 650 mg/l) are observed at great depth. The conditions are generally strongly reducing. The presence of clayish material (kaolinite, smectites) and zeolites in the fractures appears to have a large influence on the cation concentration ratios. The contents of organics, largely fulvic acids of intermediate molecular weight (<1000) is considerable (1-5 mg/l). Measurements on stable isotopes (^{18}O and deuterium) indicate a non-marine origin of the water and only minor exchanges with the surroundings. The presence of tritium is evidence of intrusions of young waters in some of the samples, probably due to the disturbances during drilling and sampling.

1. INTRODUCTION

A thorough knowledge of the general and site specific geological and hydrological conditions is required for the safety assessment of a potential repository for high-level radioactive waste in deep bedrock. The groundwater would constitute the mobile phase which acts as the transporting medium for any radionuclides that would eventually be released from the encapsulated waste. It is therefore essential that detailed information is available concerning groundwater chemistry and water-rock interactions in deep crystalline rocks.

Groundwater has been sampled and analyzed as a part of the geological and hydrological program within the Swedish nuclear waste project since 1977, and data are now available from ca 10 different sites (Finnsjön, Fjällveden, Forsmark, Gideå, Kamlunge, Kråkemåla, Sternö, Stripa, Studsvik, Svartboberget) (1-17). The present report summarizes some of the observations from Fjällveden, Gideå, Svartboberget and Kamlunge, essentially from the period 1982-83. Field data from these sites are previously reported in 12, 14, 16 and 17.

2. GEOLOGICAL SETTINGS

The test sites represent different geological environments within the Svecokarelian province of Sweden (Fig. 1).

The bedrock at Fjällveden is composed of a migmatized, veined gneiss of sedimentary origin. The composition is mostly granitic. The migmatization took place more than 1800 million years ago. Small bodies of amphibolite are also present. A foliated granite appears in the marginal parts of the test site. The youngest intrusion affecting the area is an east-west trending dolerite dyke (18).

The boreholes at Gideå were drilled in a migmatized veined gneiss of sedimentary origin mostly exhibiting a granodioritic composition. Thin dykes of a foliated granite is less frequent (19). The host rock has suffered Svecokarelian metamorphism more than 1800 million years ago. The area was intruded by vast amounts of post-Jotnian dolerite magma 1200 million years ago (20). This resulted in sheet-like dolerite bodies outcropping in the surroundings of the test site (21), but also in a dyke swarm of dolerites penetrating the bedrock of the area.

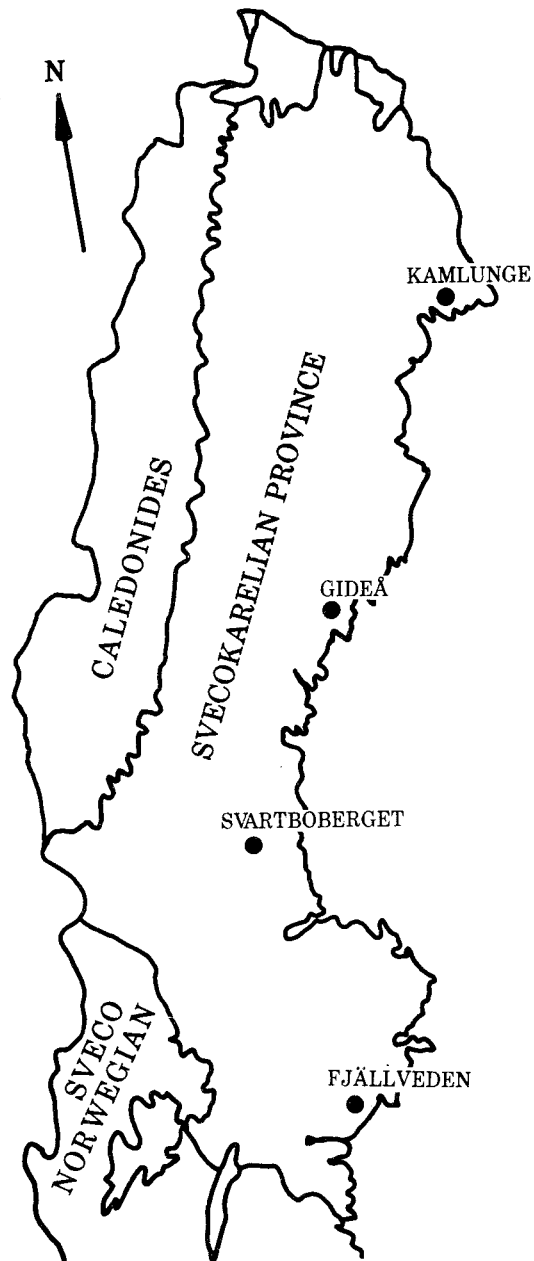


Figure 1 Map over the main lithological units and location of test areas.

At Svartboberget the bedrock is dominated by a migmatized paragneiss of the same age as that found at Fjällveden. The gneiss, which is granitic to granodioritic, is intruded by two small dolerite dykes within the test site. In the northern part of the area a 150 m thick sequence of a graphite bearing gneiss appears. Also small amounts of amphibolite as well as pegmatite can be seen (22).

At Kamlungekölen gneisses of sedimentary origin dominate. These are more than 1900 million years old and can be distinguished as two main types. One of them is a biotite rich variety; the other is a quartz dominated variety. In the southern part of the area a red, pegmatite rich granite appears which is younger than the paragneiss mentioned above. Small amounts of a granodioritic to dioritic rocktype appears as well as amphibolites (23).

The drilling holes and sampling depths at the four test sites are given in Table 1. Details concerning exact location, hydrological conditions etc. are given in 18, 19, 22 and 23.

3. CHEMICAL COMPOSITION

3.1. Sampling

The sampling procedure and equipment, Fig. 2, is described in detail elsewhere (12, 14-17, 24) and only summarized below.

The boreholes were evacuated with an air lift (using nitrogen) prior to the lowering of the equipment to the first sampling level. The selected fracture zone was sealed off by packers. Usually iodide (NaI, 0.01 mM) was added to the drilling water (water from percussion-drilled boreholes filtered through 5 μ m-filters) during the drilling phase which would allow estimations of the amounts of residual drilling water in the sampled groundwater. Generally long pumping times were used with periodic sampling (e.g. during a period of 2 weeks) at each sampling level. Thus, it was possible to follow changes in the composition that might be derived from the drilling water or intrusion of water from adjacent reservoirs. Typically the water volume pumped from each sampling level was 200-350 l/d, (except for Gi4:596 where only 1-2 l/d was obtained). The total amount of water sampled at each level would then be a few cubic meters (except at Gi4:596). The amount of drilling water that may have contaminated the groundwater

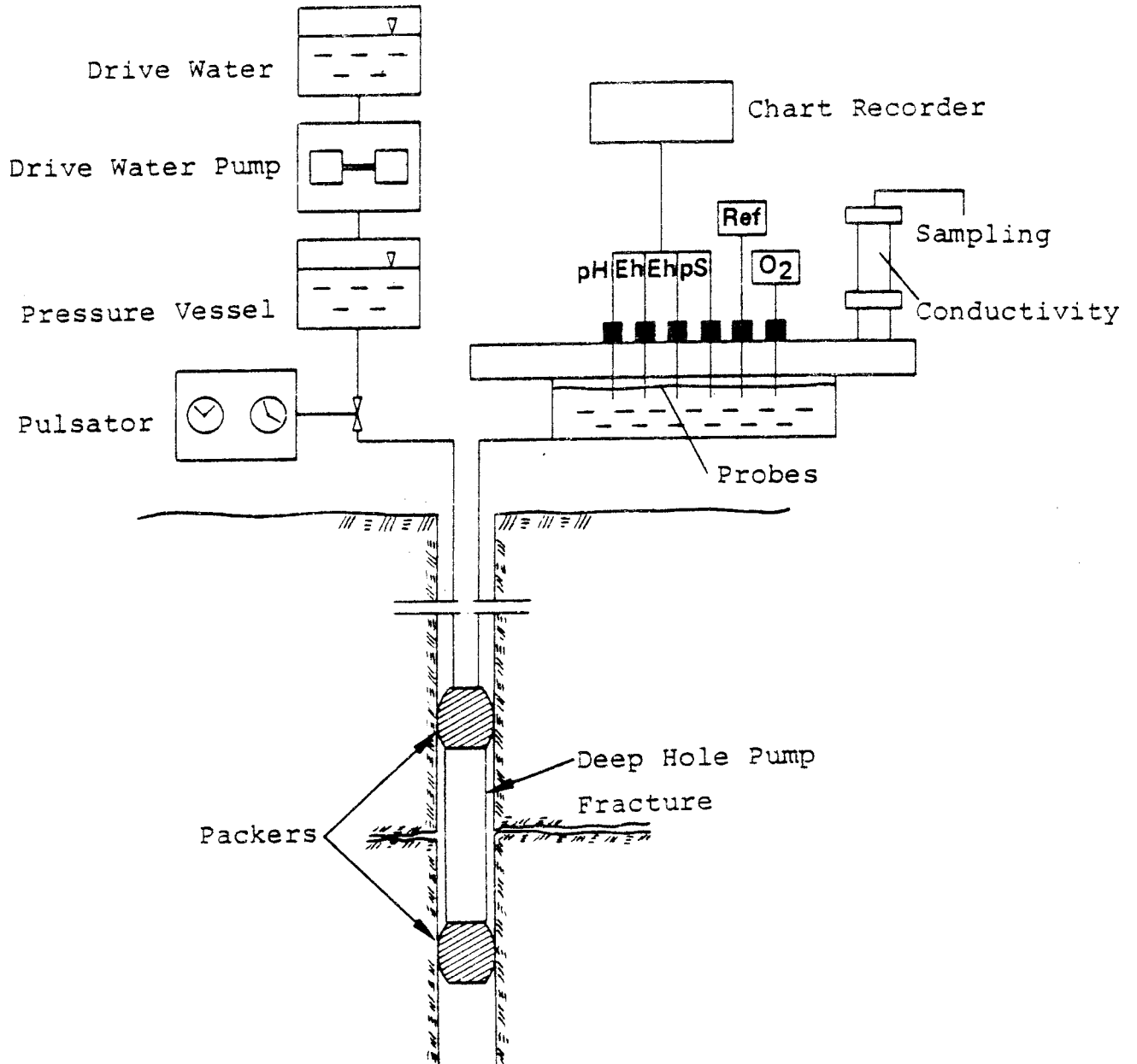


Figure 2 Schematic illustration of sampling equipment (from ref. 12, 14-17, 24).

samples were generally estimated to be a few percent, or below. However, unexpectedly high background concentrations of iodide prevented in some cases any firm estimates of the drilling water contamination.

Field measurements of temperature, pH, Eh (graphite and platinum electrodes) pS, O₂-concentration and conductivity were performed. The various probes used for these measurements were calibrated during an initial calibration phase.

Water was collected in glass bottles and plastic bottles for laboratory analysis of pH, Na, K, Mg, Ca, Fe(II), (preservation with HCl), Fe(tot), Mn, Al, Cu, Sr, lanthanides, HCO₃⁻, Cl⁻, SO₄²⁻, PO₄³⁻(tot), NO₂⁻, NO₃⁻, NH₄⁺, SiO₂(tot), S²⁻(tot), I⁻, TOC (total organic carbon) as well as of conductivity, turbidity and particulate matter (filtered water 0.45 µm millipore filter). (Swedish Geological AB, Uppsala; Hydroconsult, Stockholm; Swedish Institute for Water and Air Pollution Research, Stockholm; Swedish Geological Survey, Luleå).

Isotope analysis (²H, ³H, ¹⁴C, ¹⁸O) was carried out in some samples, as well as determinations of the natural concentrations of uranium, thorium, radium, radon and helium. (Laboratory for Isotope Geology, Stockholm; Institute for Radiohydrometry, Neuherberg; Mass Spectrometry Lab., Uppsala; Studsvik Energiteknik AB, Nyköping).

Any deviations from the general procedures outlined here, as well as discussions of methodology, accuracy, interlaboratory comparisons etc. are given in 12, 14-17 and 24. Only average values under stable conditions, with the uncertainties defined by the range of reported analysis data, are discussed in this report.

3.2. Total dissolved solids and inorganic constituents

The total amount of dissolved material in the various waters of Table 1 are given in Fig. 3. Most of the waters have total concentrations of dissolved components (TDS) between 200 and 300 mg/l (slightly over 100 mg/l at Kamlunge) fairly independently of the vertical depth. Exception are Fj2:506, Fj4:420 (initially), Gi4:385, and particularly Gi4:596, where waters with significantly higher TDS-levels are encountered (up to 640 mg/l, Gi4:596). These four waters are denoted "saline" in the subsequent discussion.

Table 1 Drilling holes and sampling depths at the test sites.

Area	Hole nr	Vertical depth, m
Fjällveden	Fj2	106, 293, 409, 506
	Fj4	131, 272, 349, 420
	Fj8	402, 562
Gideå	Gi2	157, 288, 353, 478, 528
	Gi4	91, 212, 385, 498, 596,
Svartboberget	Sv4	82, 324, 373, 551
Kamlunge	Km3	106, 376

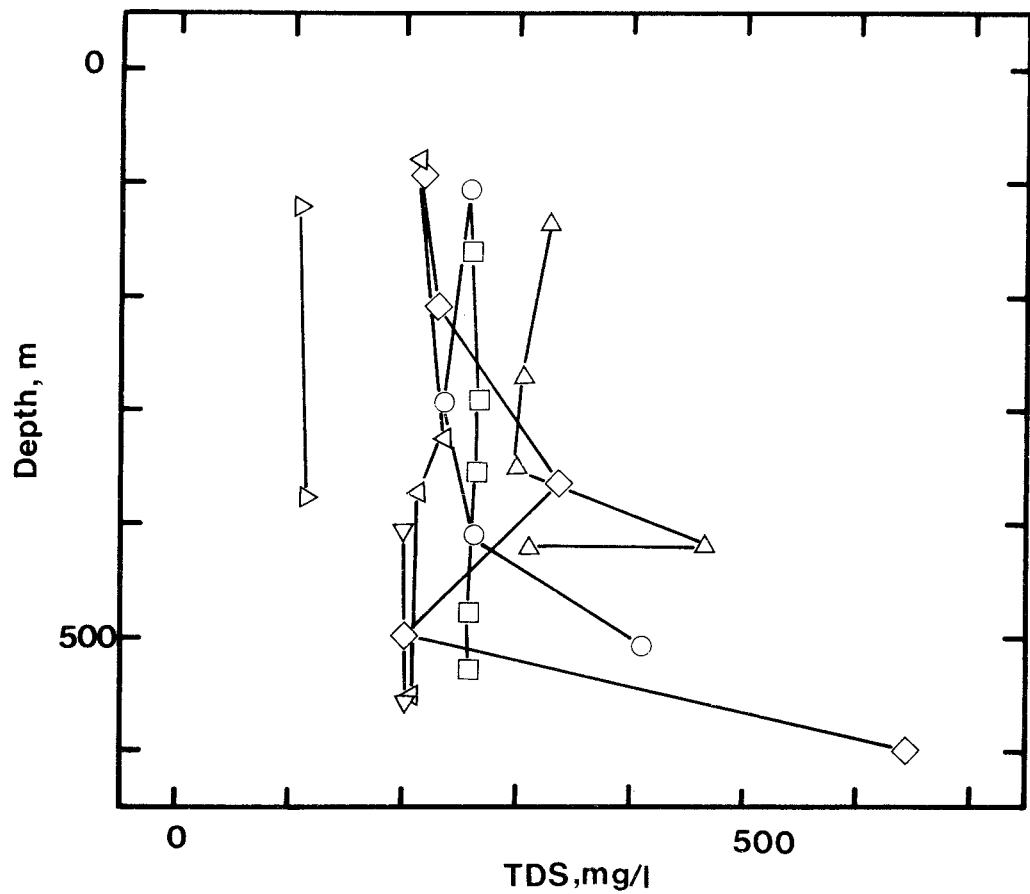


Figure 3 Total dissolved solid (TDS) in the sampled groundwaters
 ○ Fj2, △ Fj4, ▽ Fj8, □ Gi2, ◇ Gi4, ◁ Sv4, ▷ Km3.

The total concentrations of HCO_3^- , $\text{SiO}_2(\text{tot})$, SO_4^{2-} , F^- , HPO_4^{2-} , HS^- , Cl^- , TOC, Na, Ca, K, Mg, Fe(II), Mn and Al, as well as pH for the waters of Fig. 3 are given in Fig. 4-10, (average values with bars indicating the ranges of analysis data). The significant differences both between the various drilling holes within the same area as well as between sampling levels within the same hole (except possibly for Gi2) indicate that the exchange of waters even over fairly narrow horizontal and vertical distances generally is minor.

An effort to characterize and distinguish the various waters is given in Fig. 11. All the non-saline waters are bicarbonate dominated and with a high proportion of Ca^{2+} among the cations for the waters with the lowest total salt contents (100-200 mg/l; Km3, Fj8, Gi4). The waters with a higher proportion of Na^+ among the cations appear to constitute another water type, representing the non-saline waters with somewhat higher total salt content (250-330 mg/l; essentially Fj4, Gi2). These two water types are arbitrarily denoted GW1 and GW3 in Fig. 11. All other non-saline waters (GW2 in Fig. 11) are merely intermediates between GW1 and GW3, both in terms of the total salt concentration and usually in the average concentrations of individual components. Whether there exists a real difference between GW1 and GW3, justifying the division of the waters into two types (with one intermediate), e.g. representing different geologic environments or various ages, can not be established solely from the available chemical data. However, the ratios between cation activities corresponding to ion exchange equilibria with a solid phase indicate more uniform conditions (possibly higher degree of equilibrium - higher age) in the order $\text{GW3} > \text{GW2} > \text{GW1}$. (See 3.2.3.)

The saline waters (Fj2:506, Fj4:420, Gi4:385, Gi4:596), which are essentially $\text{Cl}^- - \text{SO}_4^{2-} - \text{Na}^+$ -dominated, constitutes a clearly distinguishable separate group, GW4, which appears to be even closer to an equilibrium involving all the cations $\text{Na}^+ - \text{K}^+ - \text{Mg}^{2+} - \text{Ca}^{2+}$ than GW3 (see 3.2.3.). The mere fact that water with high salinity is encountered very close to non-saline waters, and even at a level surrounded by non-saline waters (Gi4:385) indicate a very slow water exchange and possibly high age of the saline waters. The concentration relations of major components in the various water types indicated in Fig. 11 are further illustrated in Fig. 12.

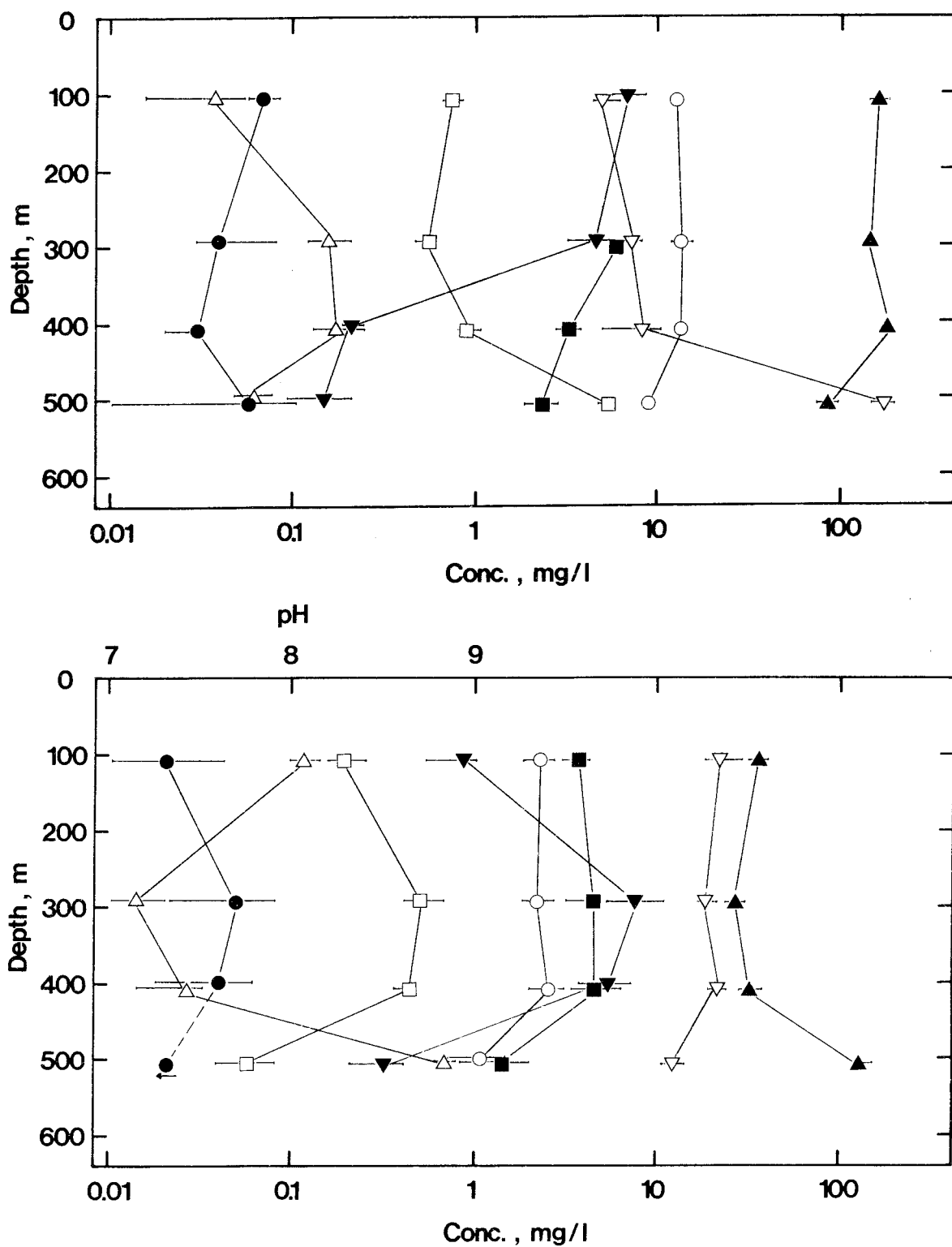


Figure 4 Dissolved constituents - Fj2
 a. ▲ HCO_3^- , ○ $\text{SiO}_2(\text{tot})$, ▼ SO_4^{2-} , □ F^- , △ HPO_4^{2-} , ● HS^- , ▽ Cl^- ,
 ■ TOC;
 b. △ pH, ▲ Na, ▽ Ca, ○ K, ■ Mg, ▼ Fe(II), □ Mn, ● Al.
 Values below the detection limits are indicated by ←.

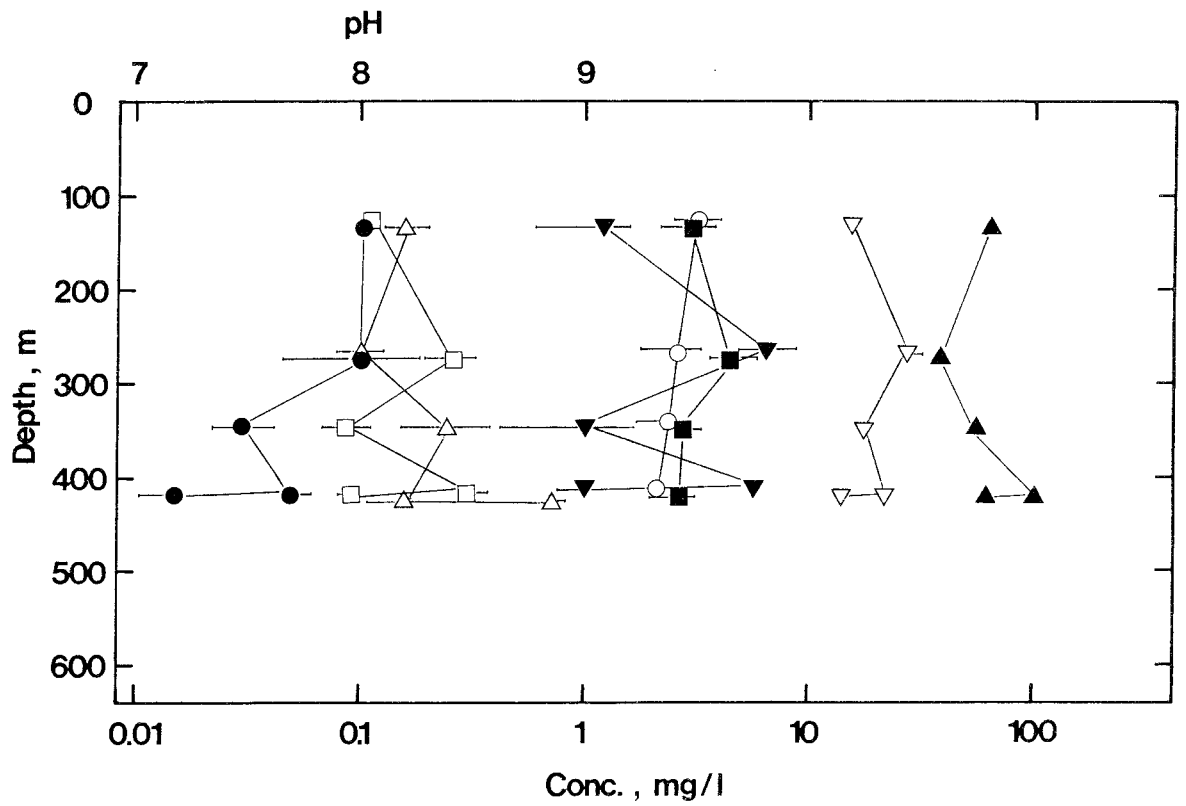
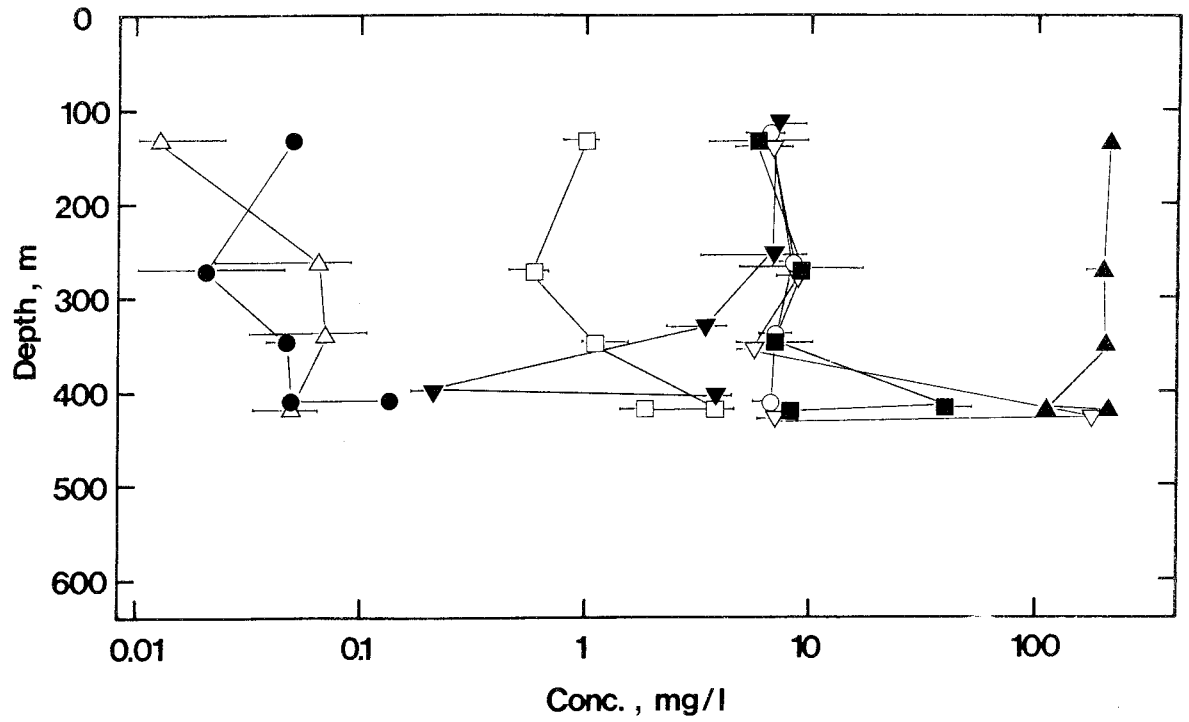


Figure 5 Dissolved constituents - Fj4
Symbols as in Fig. 4.

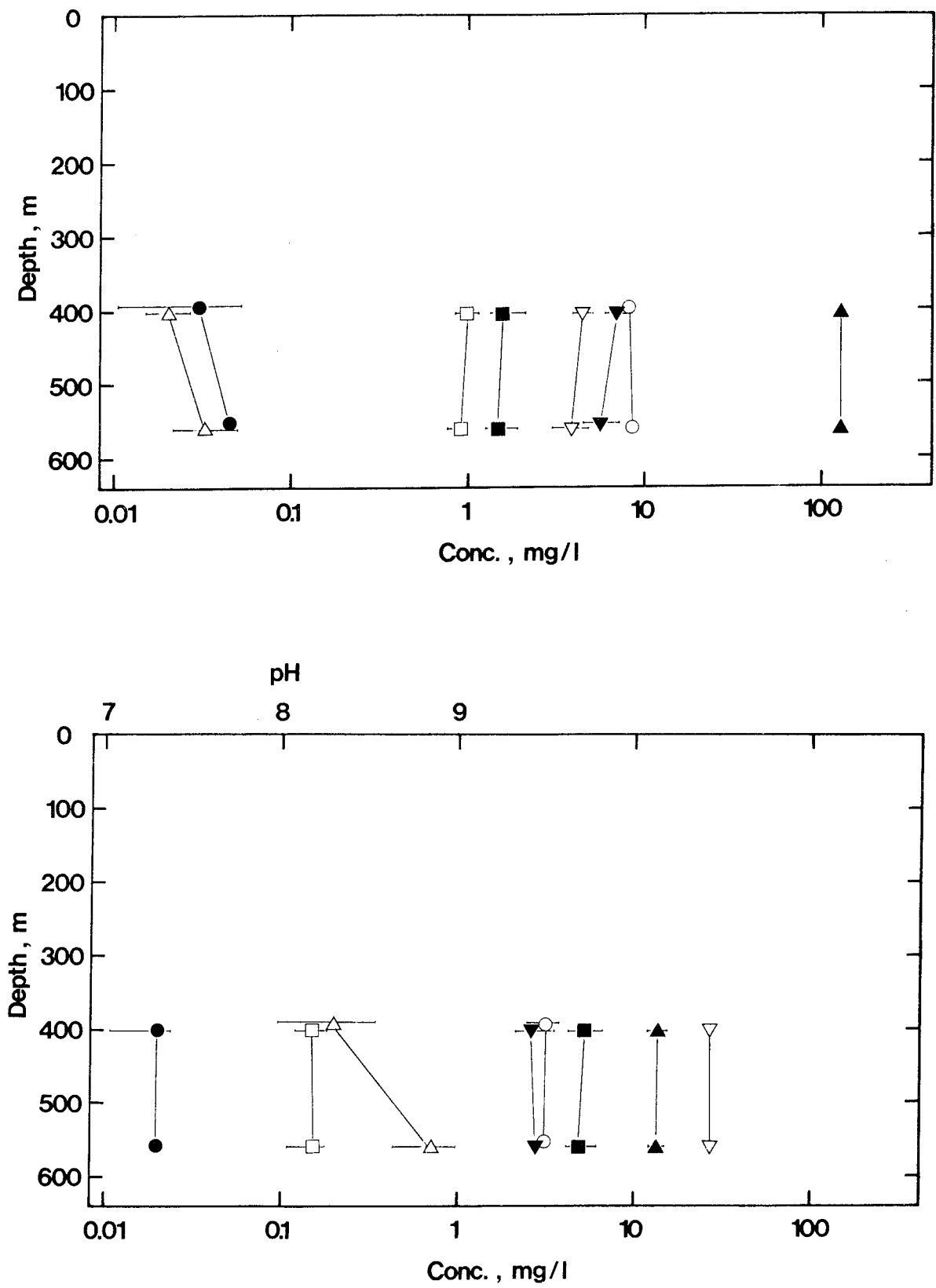


Figure 6 Dissolved constituents - Fj8
 Symbols as in Fig. 4.

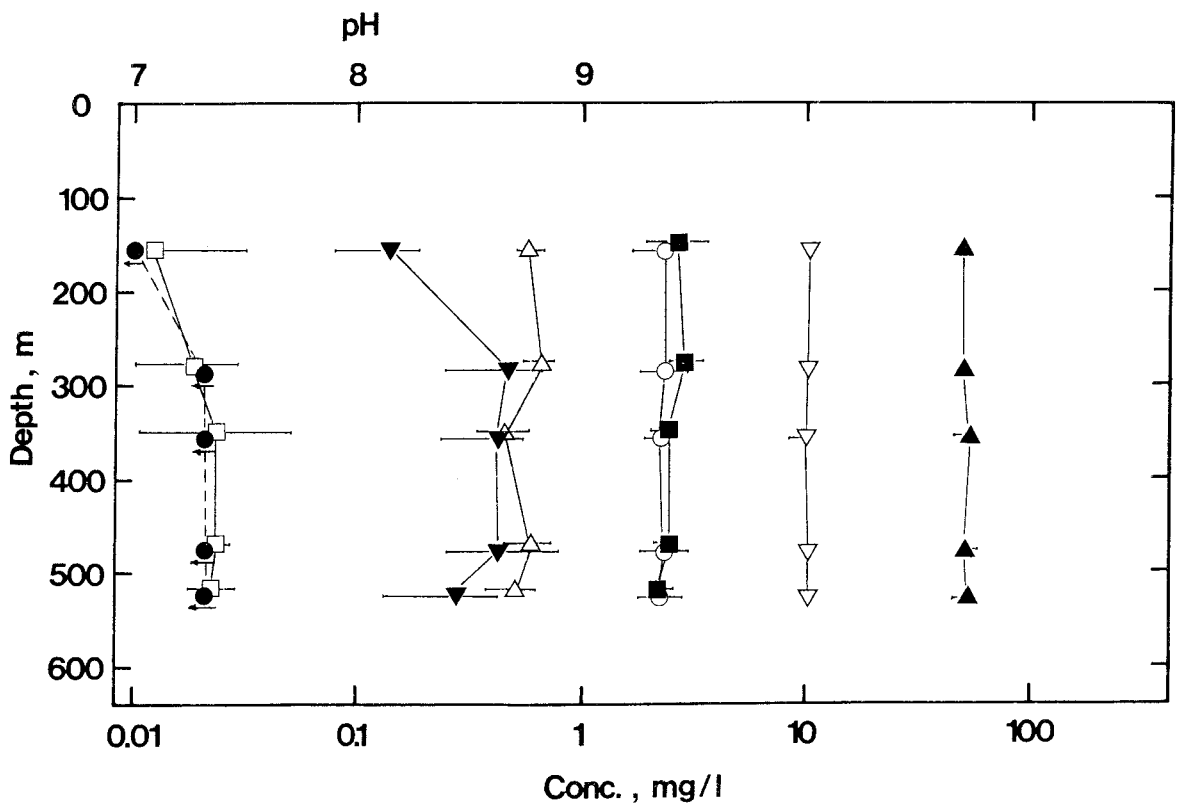
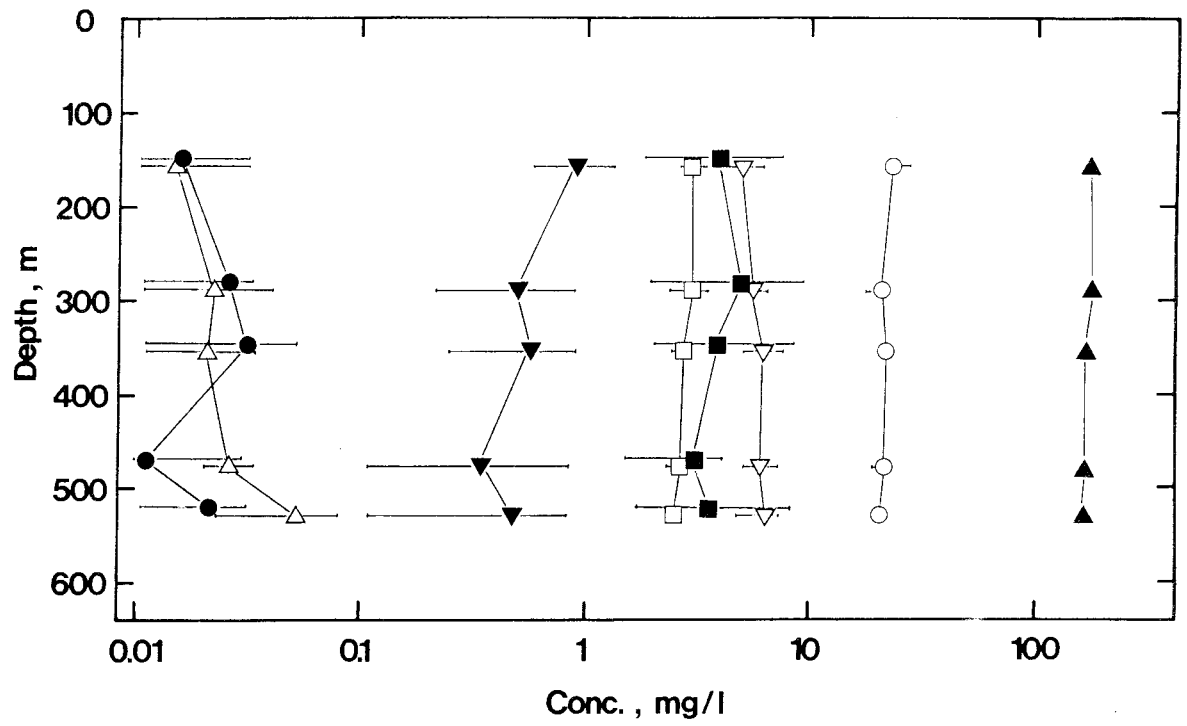


Figure 7 Dissolved constituents - Gi2
 Symbols as in Fig. 4.

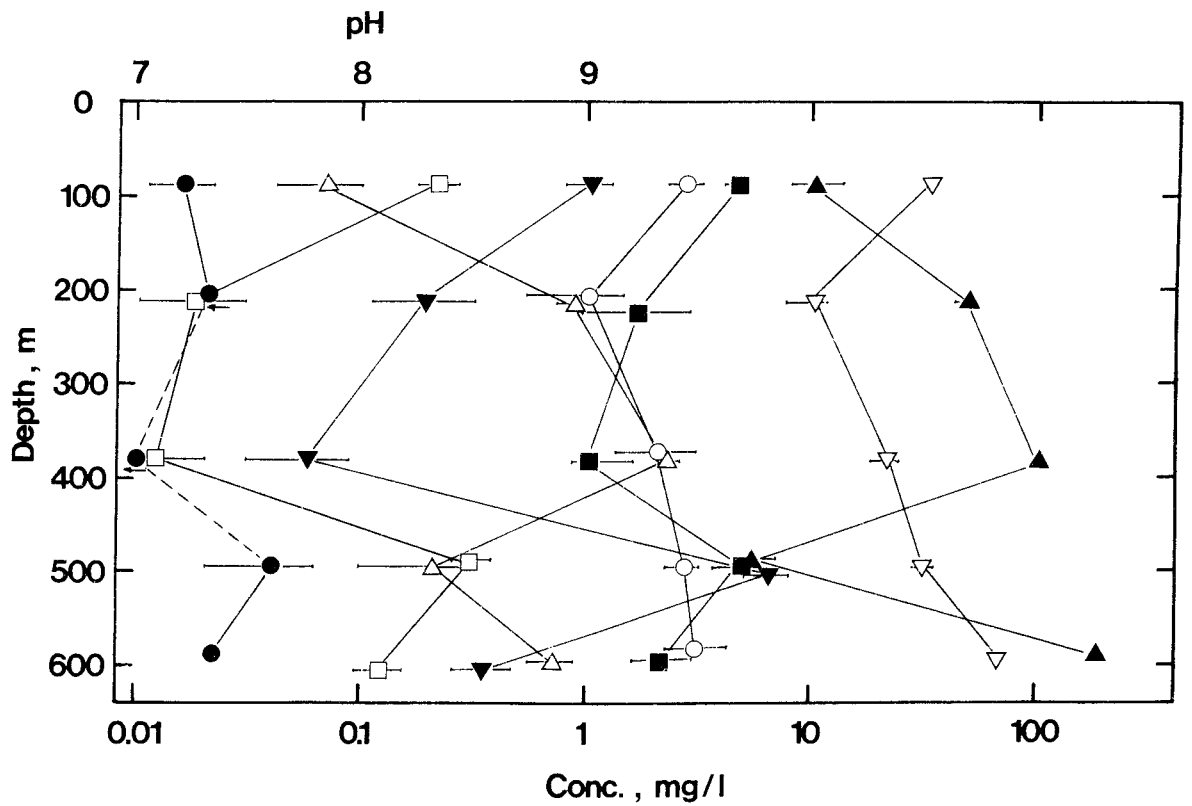
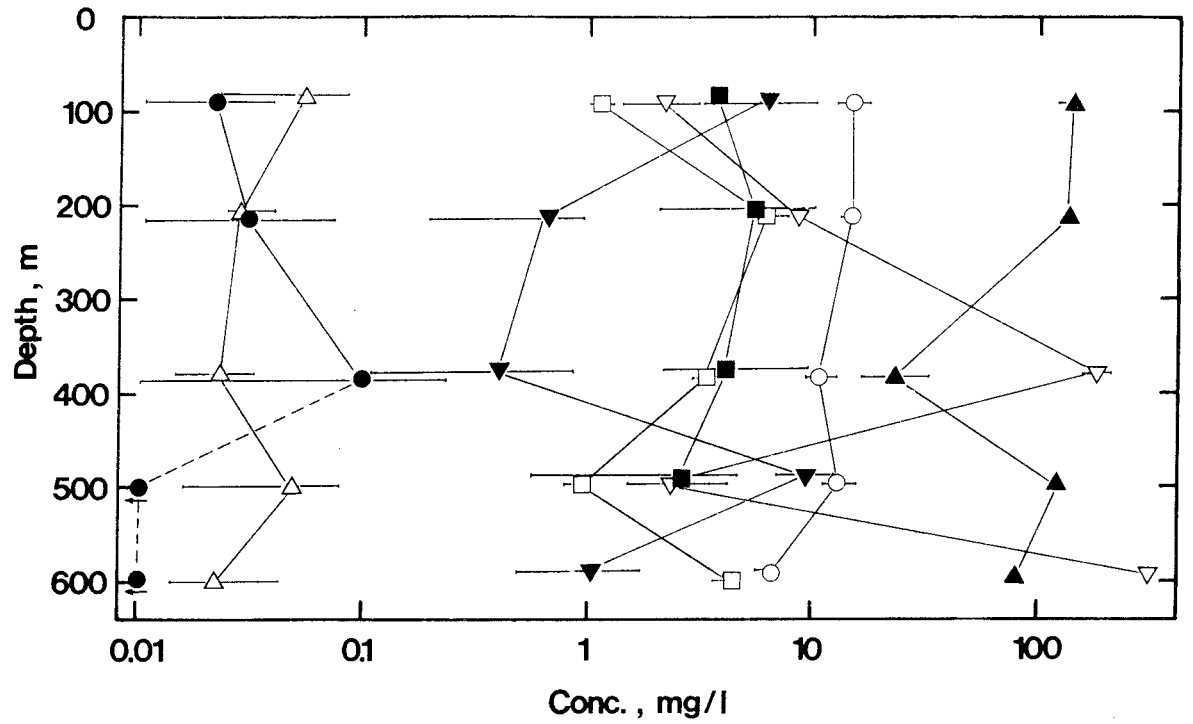


Figure 8 Dissolved constituents - C14
 Symbols as in Fig. 4.

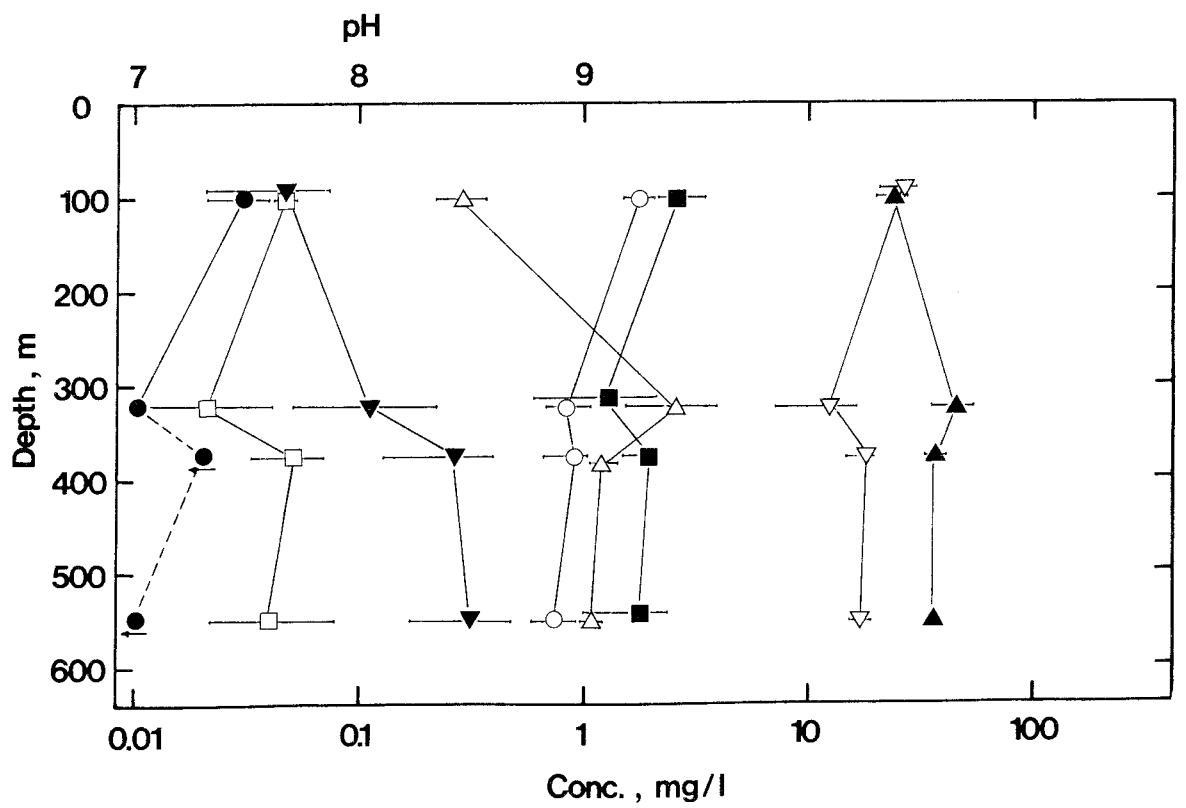
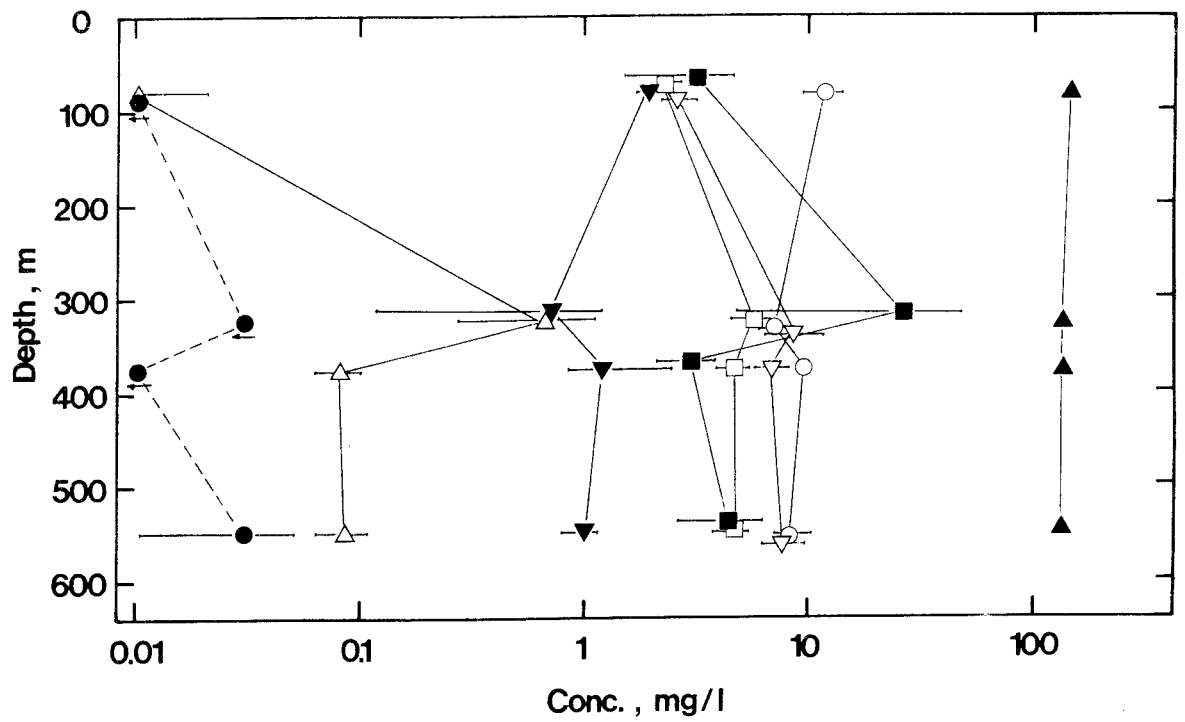


Figure 9 Dissolved constituents - Sv4
Symbols as in Fig. 4.

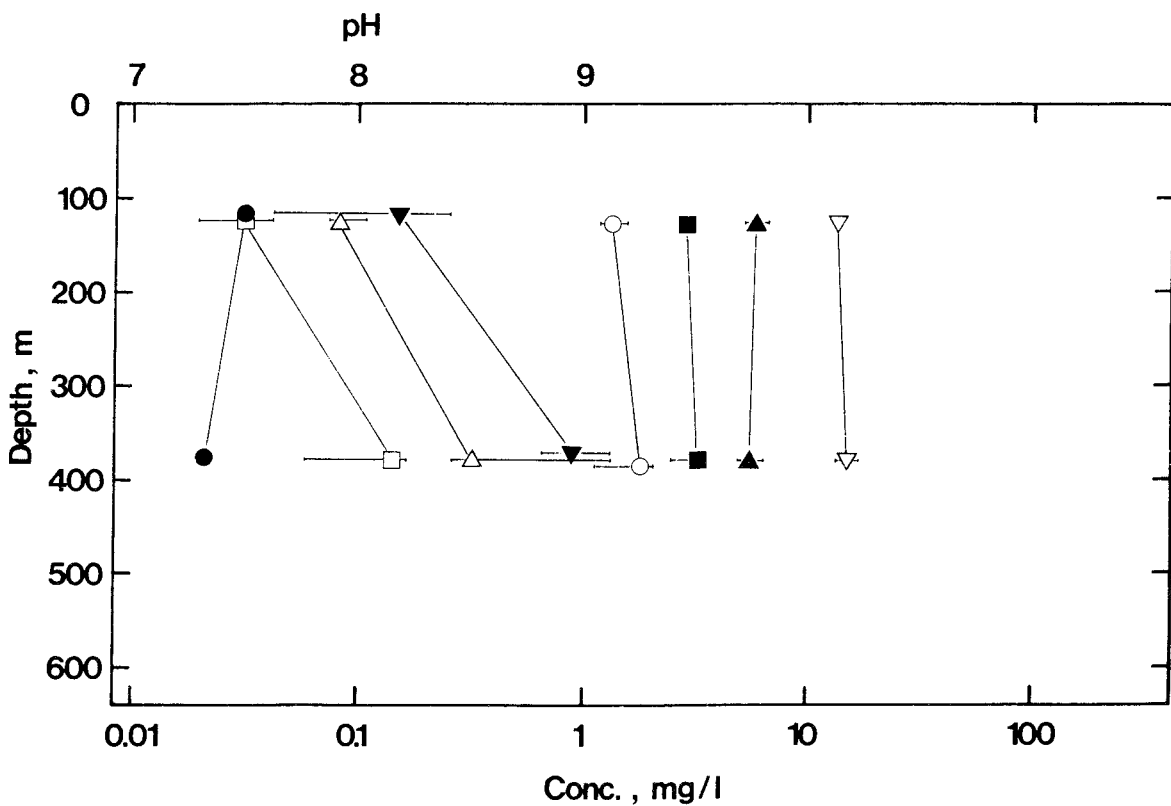
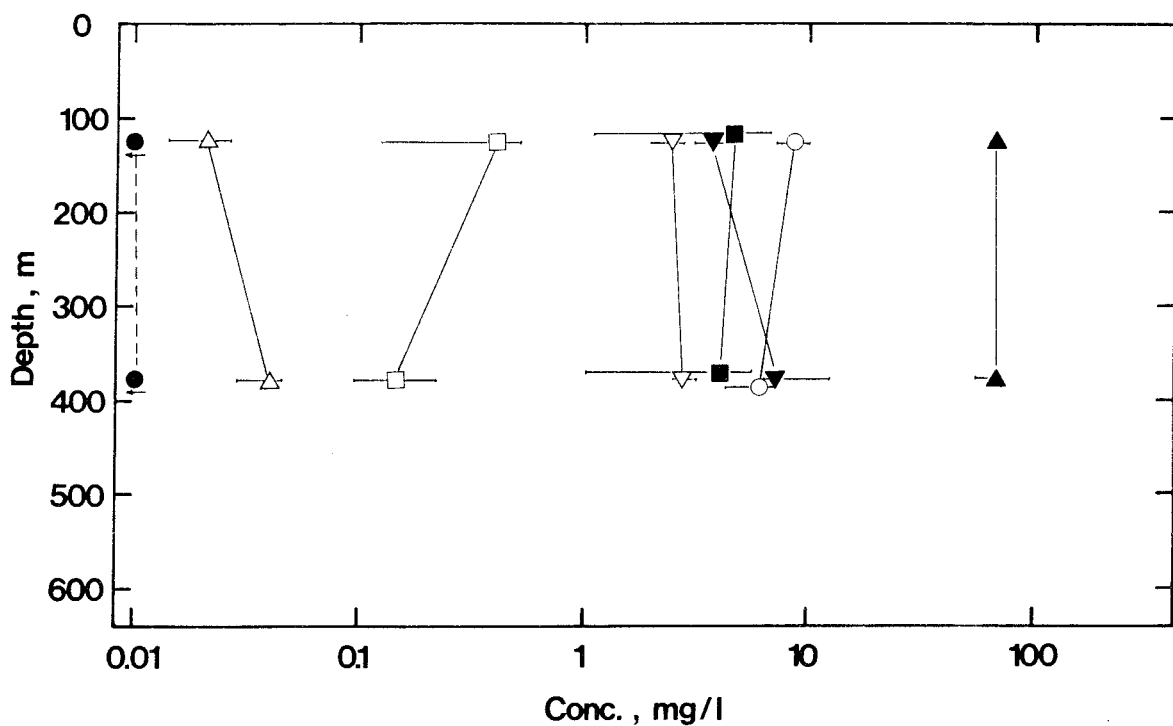


Figure 10 Dissolved constituents - Km3
 Symbols as in Fig. 4.

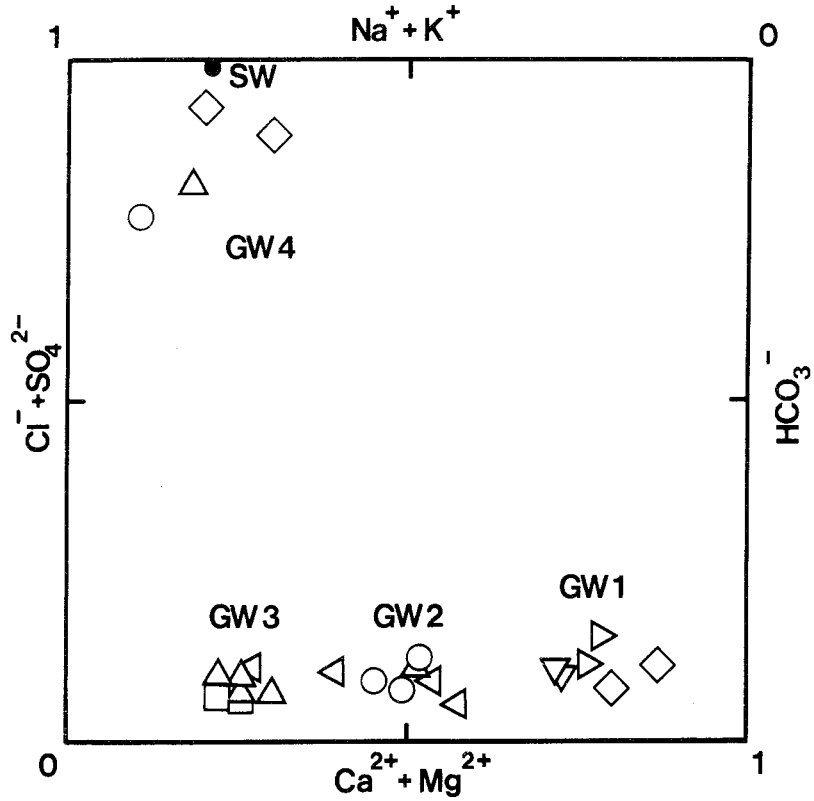


Figure 11 Modified Piper plot of major groundwater components (moleqv-% of anions vs moleqv-% of cations)
 Symbols as in Fig. 3.

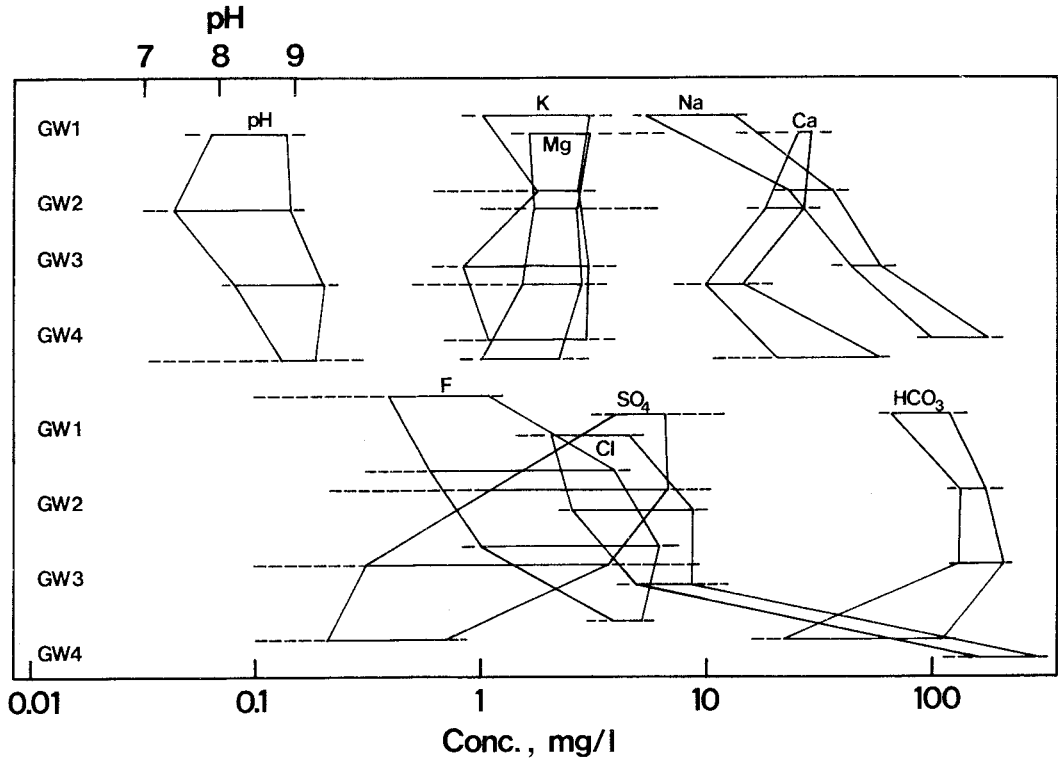


Figure 12 Dissolved major constituents (concentration ranges) in GW1, GW2, GW3 and GW4 (as defined in Fig. 11).

3.2.1. Carbonate and pH

The $\text{CO}_2/\text{H}_2\text{CO}_3$ -system is strongly affecting and buffering pH to a high level. This is primarily due to protolysis in the H_2CO_3 -system and according to

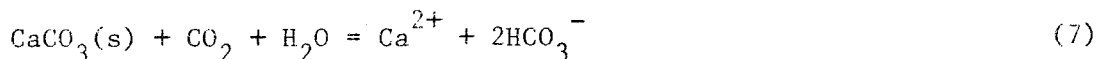


(cf. discussion in ref. 9).

The presence of CO_2 -charged waters, e.g. in open systems, will lead to an increase in both pH and alkalinity, according to



or



In closed systems a calcite saturation would be expected, according to (5).

The observed pH - HCO_3^- - Ca^{2+} -relations are in fair agreement with what can be expected, considering the reactions (1)-(7) for GW1 and to some extent GW2 but not always for GW3, (cf. Fig. 12). Both GW1 and often GW2 are generally slightly undersaturated with respect to $\text{CaCO}_3(\text{s})$ -precipitation, while some waters of GW2 and essentially all of GW3 and GW4 are saturated or supersaturated, Fig. 13 and 14.

For most of the saturated and supersaturated waters, particularly among GW4, both Ca^{2+} and pH are higher than for the other waters, while HCO_3^- is low. A continuous addition of Ca^{2+} from some other source than the calcite dissolution (saline waters or weathering processes and ion exchange, see below) would give such an effect. Thus, an increasing portion of hydroxid is contributing to the alkalinity. This is evident in Fig. 15, where the waters of GW4 are markedly different from those of GW1 - GW3, for which an

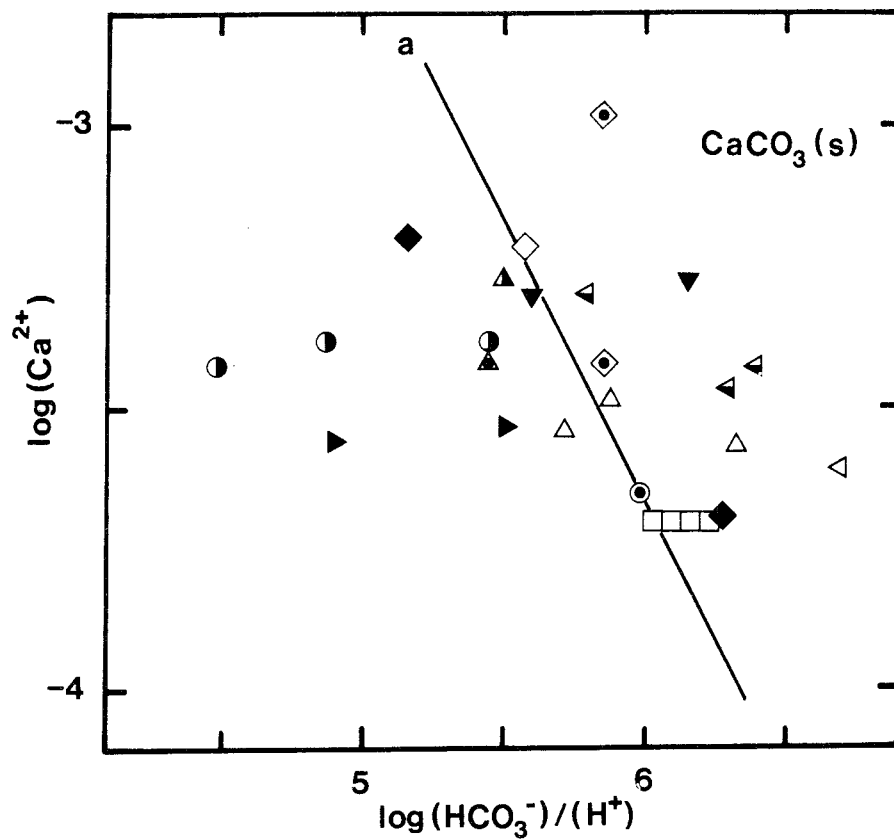


Figure 13 $\log(\text{Ca}^{2+})$ vs $\log(\text{HCO}_3^-)/(\text{H}^+)$
 a. $\log(\text{Ca}^{2+}) = -\log(\text{HCO}_3^-) - \text{pH} + \log K_s$; saturation line at 10°C ,
 $\log K_s = -8.15$.

Parantheses denote activities.

Symbols:

	GW1	GW2	GW3	GW4
Fj2		●		⊙
Fj4		▲	△	▴
Fj8	▼			
Gi2			□	
Gi4	◆		◇	
Sv4		◀	◁	◈
Km3	▶			

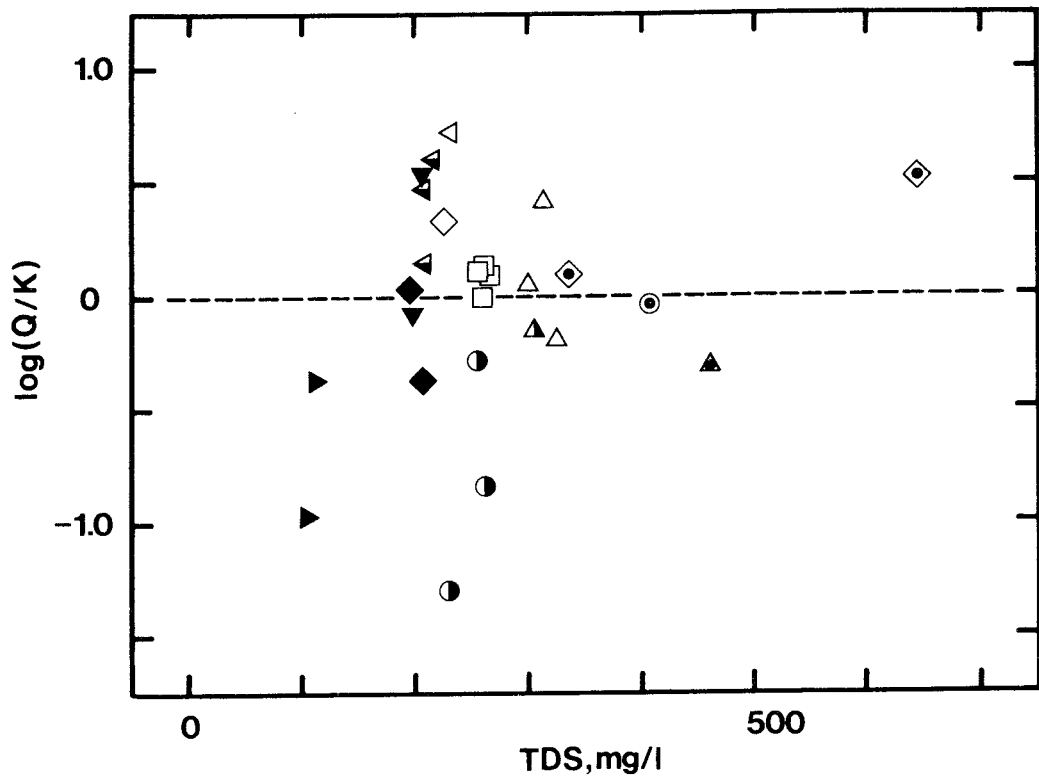


Figure 14 Calcite saturation
 $Q=(Ca^{2+})(CO_3^{2-})$; $K=K_s$
Symbols as in Fig. 13.

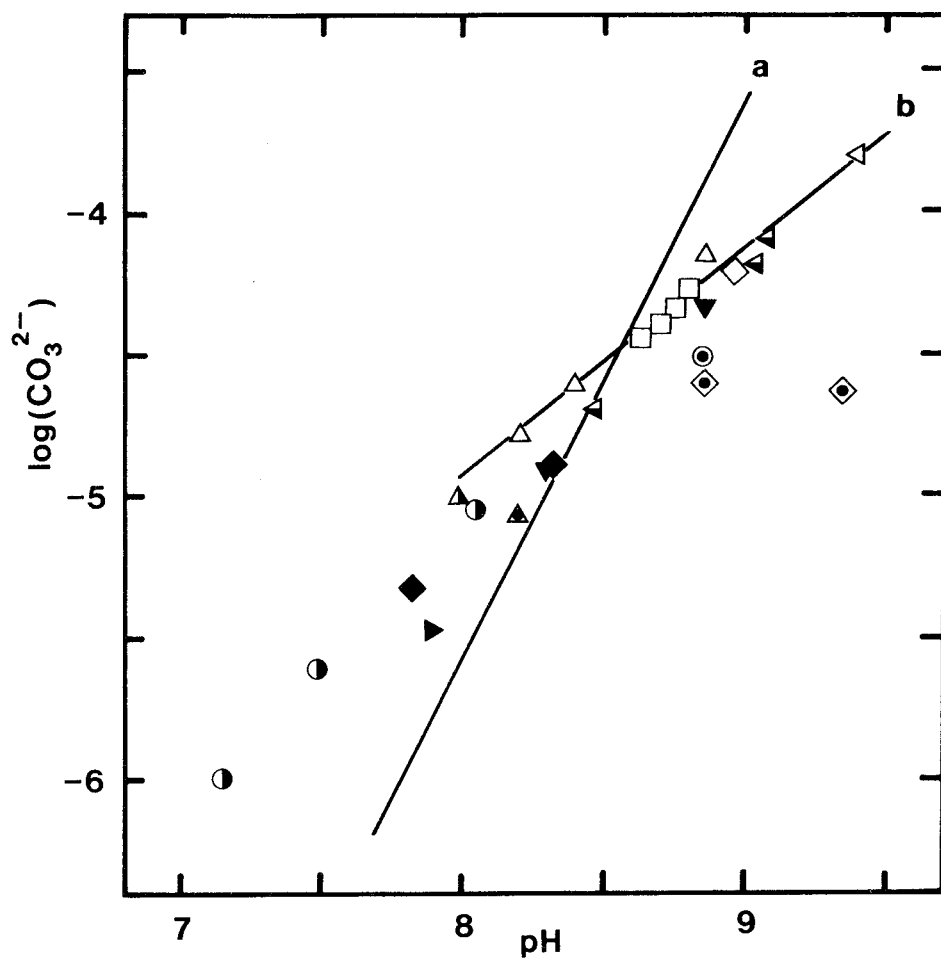


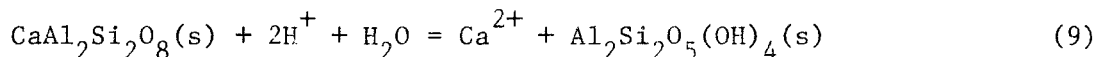
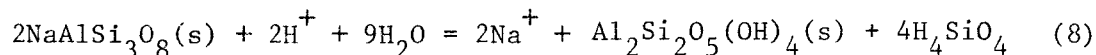
Figure 15 $\log(\text{CO}_3^{2-})$ vs pH

a. $\log(\text{CO}_3^{2-}) = 2\text{pH} - 18.1 + \log P(\text{CO}_2)$; saturation with air,
 $P(\text{CO}_2) = 10^{-3.5}$

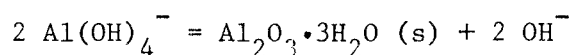
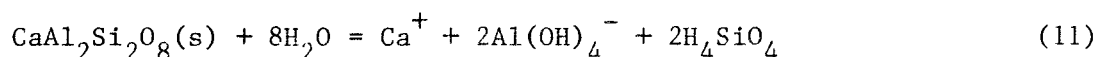
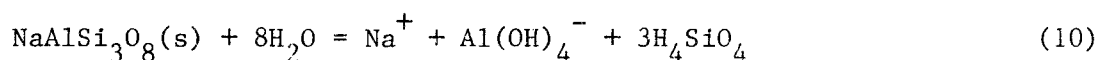
b. $\log(\text{CO}_3^{2-}) = 0.79\text{pH} - 11.25$; empirical line limiting the maximum
carbonate activities at $\text{pH} > 8$

Symbols as in Fig. 13.

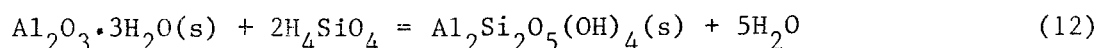
almost linear relationship between $p(\text{CO}_3^{2-})$ and pH is observed. Contributions from feldspar hydrolysis could involve the degradation to e.g. kaolinite, as for the plagioclases:



Another set of pH-affecting reactions would be the formation of gibbsite through



and the gibbsite - kaolinite transformation



The reactions (8)-(12) could serve as examples of pH-increasing processes. In the presence of CO_2 the released OH^- would lead to an increased HCO_3^- level instead:



Other similar reactions involving the hydrolysis of Na- and Ca-feldspars as well as K-feldspars and leading to the formation of micas or other clays than kaolinite, e.g. montmorillonite, are feasible.

For Ca^{2+} and to some extent Mg^{2+} the formation of sparingly soluble carbonates (calcite, dolomite, magnesite) would represent a sink for the total dissolved carbonate.

It is evident from the examples given on feldspar decomposition that changes of distribution and concentrations of cations e.g. due to mixing of non-saline waters with highly saline waters, would have a large effect both on weathering processes as well as on the final groundwater composition

(including pH) at equilibrium with the solid phases. Sources of saline waters are briefly discussed in 3.3.

3.2.2. Silica and silicates

The total SiO_2 -concentration is between $10^{-3.4}$ and $10^{-4.1}$ M for all the groundwaters which is above the saturation level for quartz (SiO_2) but below the level for amorphous silica (H_4SiO_4). Protolysis according to



would be negligible unless pH is close to or above pK, (around 9.9).

The major source of aqueous silica is the aluminosilicates rather than quartz. The systems $\text{M} - \text{SiO}_2 - \text{Al}_2\text{O}_3 - \text{H}_2\text{O}$, where $\text{M} = \text{Na}, \text{K}, \text{Mg}$ and Ca , are illustrated in Fig. 16, according to ref. 26. It appears, that kaolinite, smectite, and possibly leonhardite (laumontite) could be thermodynamically stable phases in equilibrium with the sampled groundwaters. However, predictions of predominating secondary minerals can not be made solely from this type of diagrams (see 3.3.3).

3.2.3. Major cations (Na^+ , K^+ , Mg^{2+} , Ca^{2+})

Most weathering processes would lead to the release of cations and many of the intermediate or end products in weathering sequences would have a much higher ion exchange capacity than the starting material. An increased pH would also lead to enhanced cation exchange capacity (CEC). Thus, unaltered feldspars usually exhibit a CEC below 10 meq/kg at pH 8, while kaolinite could have a CEC up to 20-30 and montmorillonite up to 700-800 meq/kg (25). The selectivity of cation exchange depends on the charge properties of the solid matrix and the solution layer on the solid. For clay minerals the exchangeability is usually in the order



i.e. decreasing with decreasing size. For zeolites the order is usually the opposite.

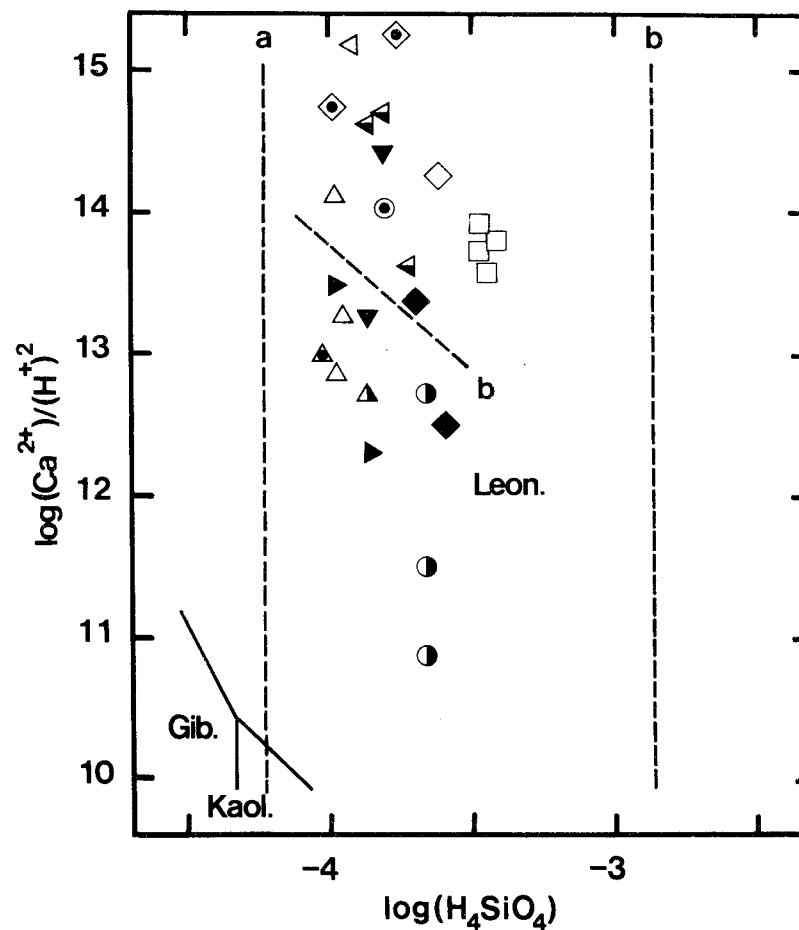
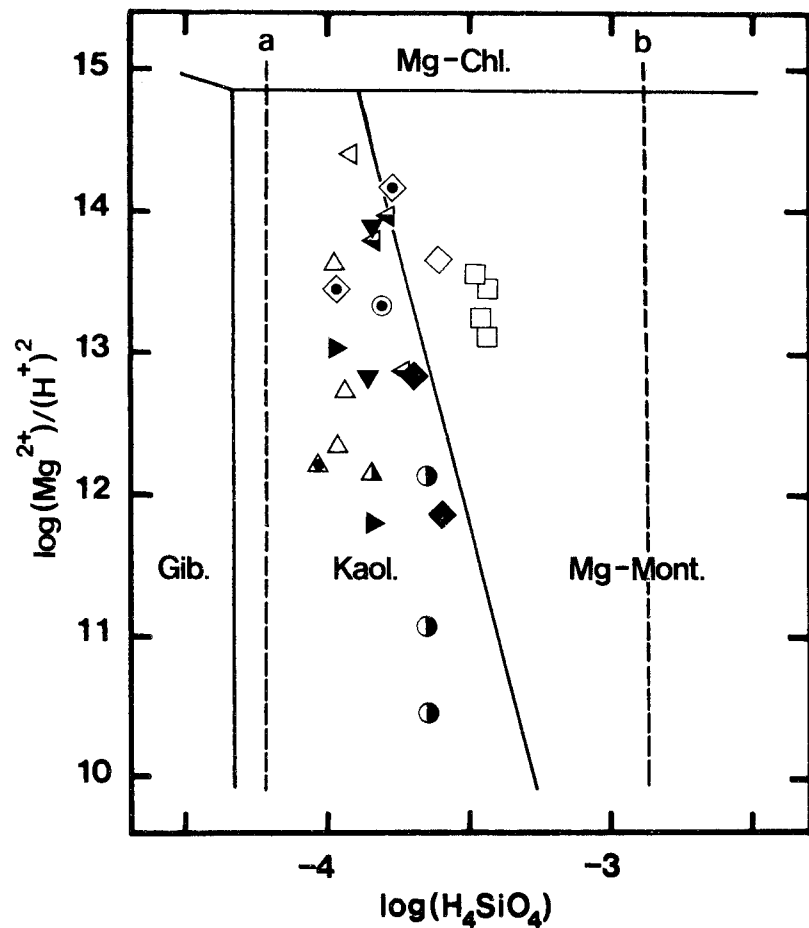


Figure 16 Stability diagrams for Na-K-Mg-Ca-silicate phases (10°C).
 Alb.=albite, Chl.=chlorite, Gib.=gibbsite, Kaol.=kaolinite,
 Leon.=leonhardite (laumontite), Micr.=microcline,
 Mont.=montmorillonite, Musc.=muscovite, Pyroph.=pyrophyllite

a. Saturation line, quartz
 b. Saturation line, amorphous silica
 c. Aprox. saturation line, calcite
 Symbols as in Fig. 13.

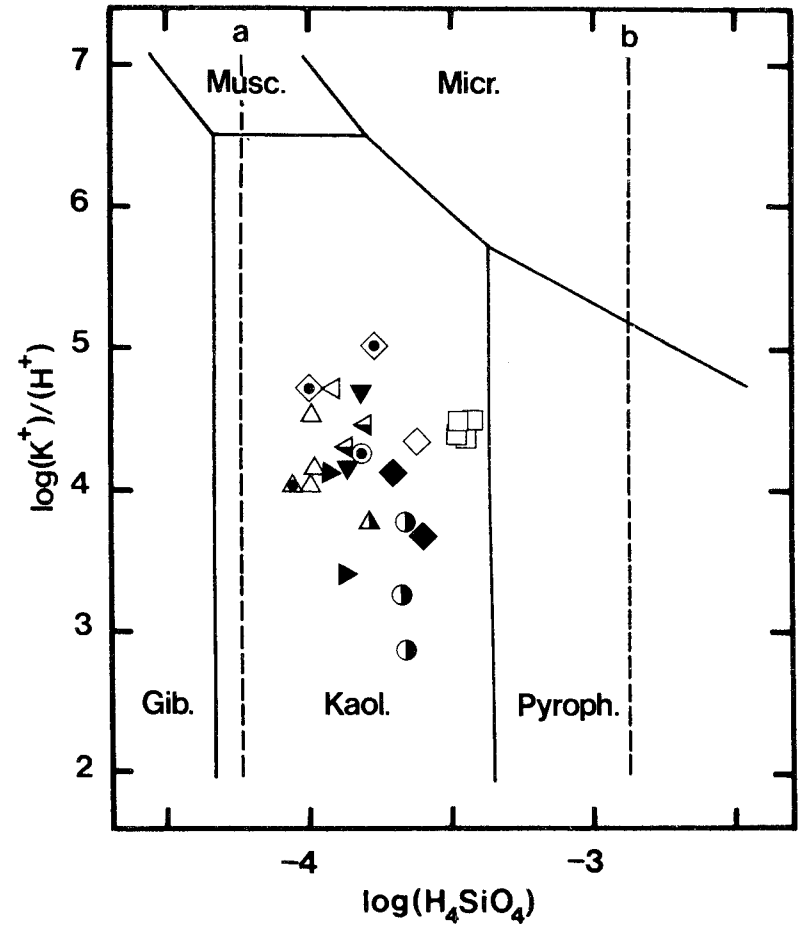
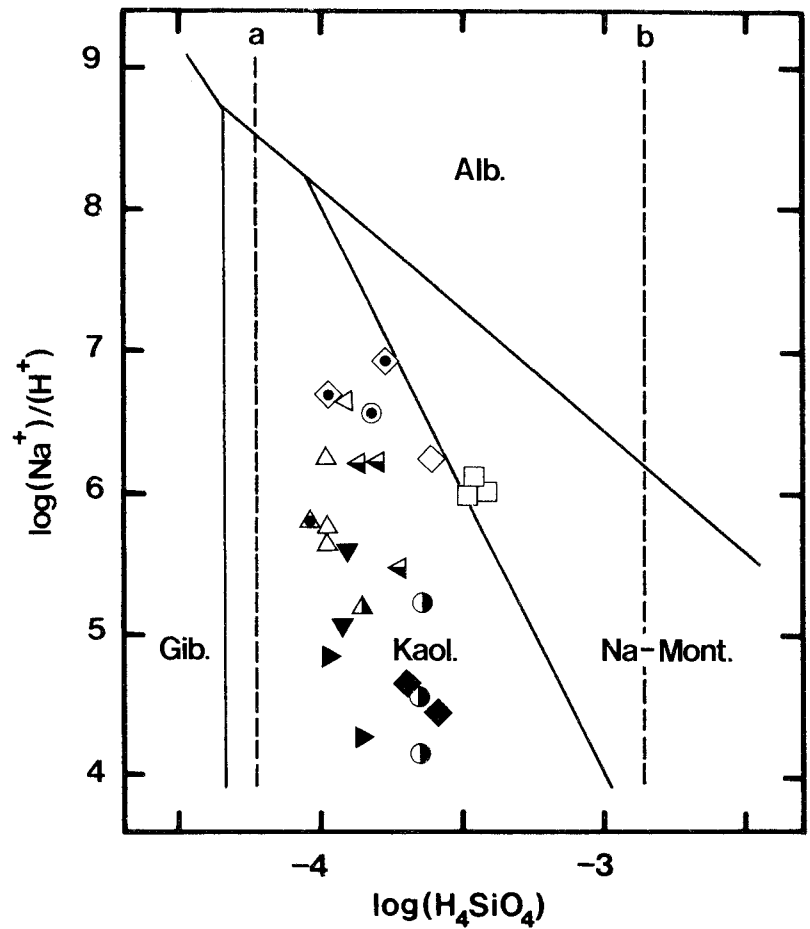


Figure 16 (continued)

It is likely, that cation exchange processes on water-exposed fracture minerals, which could be high-capacity weathering products, rather than the slow diffusion controlled weathering processes would govern the distribution of cations in the groundwater. Thermodynamic analyses of the type discussed in 3.2.2. will of course tell whether a certain solid phase would be thermodynamically stable or not in a certain environment, but could not serve as the basis for calculations of the groundwater composition with respect to the exchangeable cations and pH.

The contributions for exchange reactions in the groundwaters are illustrated in Fig. 17, illustrating the relations between the major cations and pH corresponding to



where -S indicates the existence of the cation in a solid cation exchanging matrix.

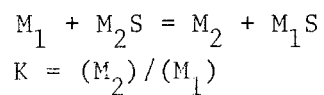
It appears, that the activity ratios, corresponding to the equilibrium constants for reactions level-out when going from the non-saline water GW1 → (GW2) → GW3 to the saline GW4. This constant level could possibly be representative of a condition close to equilibrium with some cation exchanging solid phase. The general approach to what appears to be a constant ratio when going from GW1 to GW4 is also some indication of an increasing age (and increasing degree of equilibrium) in that same order.

A summary of all the apparent activity ratios corresponding to ion exchange reactions between cations are given in Table 2. A similar value ($\log K = 0.5$) has previously been observed for the Na/Ca-exchange in deep groundwaters (3).

3.2.4. Sulfate, fluoride

The sulfate provided by meteoric water would be around 5-15 mg/l (3). The measured concentrations are generally in the lower range of this interval,

Table 2 Ion activity ratios corresponding to the reaction.



M_1	M_2	log K
K^+	$0.5 Ca^{2+}$	2.7
K^+	$0.5 Mg^{2+}$	2.2
Na^+	$0.5 Ca^{2+}$	0.6
Na^+	$0.5 Mg^{2+}$	0.1
Ca^{2+}	Mg^{2+}	1.0
K^+	Na^+	2.1

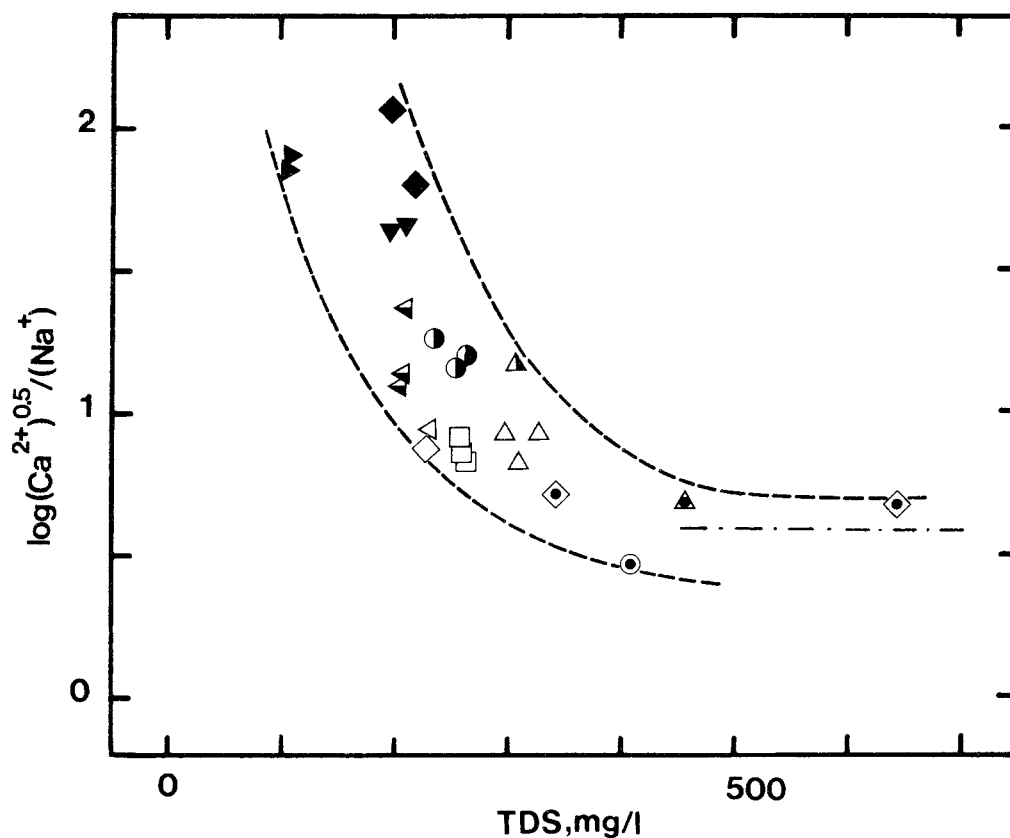


Figure 17 Cation ratios corresponding to ion exchange reactions
 Symbols as in Fig. 13.

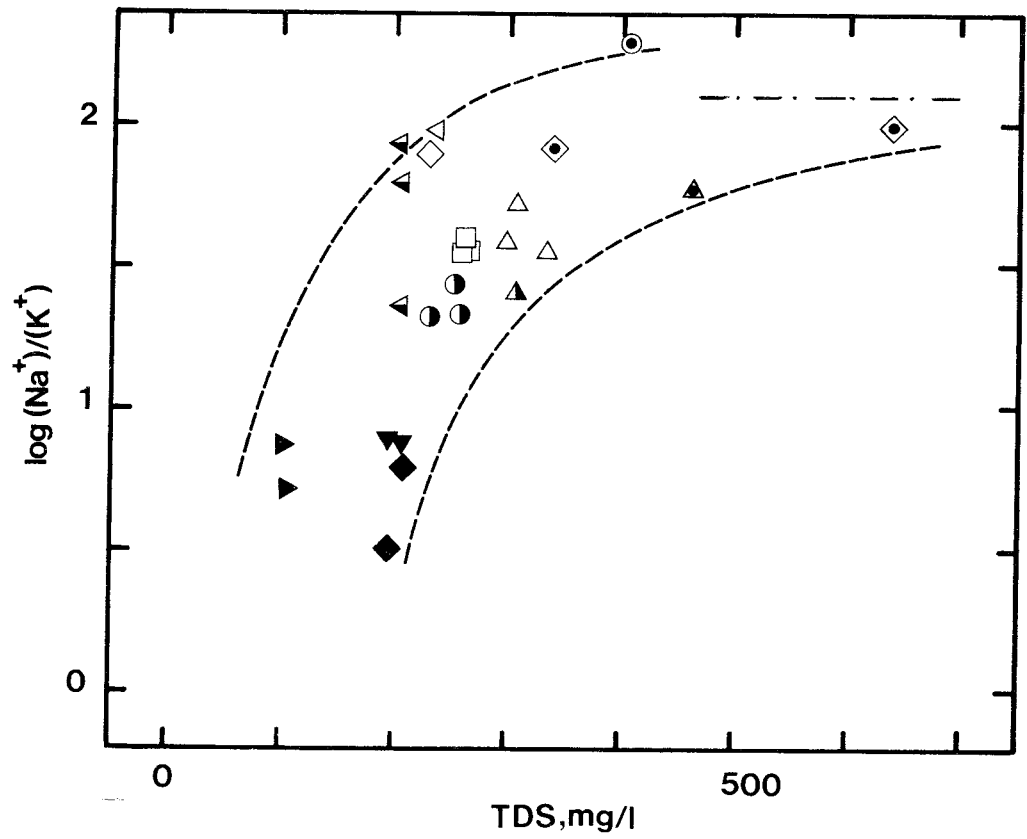
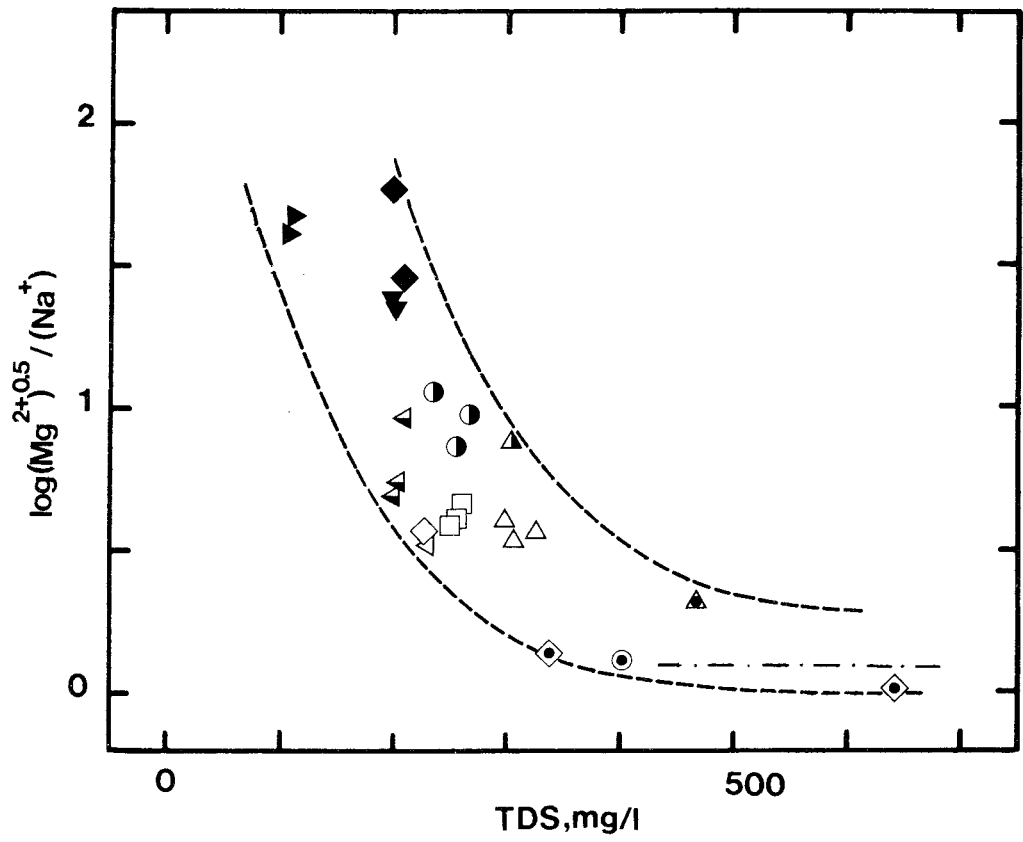


Figure 17 (continued)

and not seldom significantly lower, particularly in GW4, (see Fig. 12). Evidently, the high chloride concentrations in GW4 are not combined with high sulfate concentrations. Since all the waters are very far from saturation with respect to $\text{CaSO}_4(\text{s})$, Fig. 18, the reduction of the sulfate concentration must have other causes.

Possibly this is an indication of highly reducing conditions, leading to a reduction of the sulfate to sulfide, which would partly precipitate as pyrite.

Most of the waters are slightly undersaturated with respect to $\text{CaF}_2(\text{s})$, Fig. 19, although the saline waters in GW4 as well as GW2- and GW3-waters from Gi4 and Sv4 are fairly close to saturation.

3.2.5. Chloride, saline waters

Chloride concentrations in the groundwater would hardly be above 5 mg/l, if meteoritic water is the sole chloride source. Most of the waters in GW1 and GW2 are in fact close to this level although GW3 exhibits somewhat higher concentrations (see Fig. 12). However, in GW4 the chloride concentrations are up to two orders of magnitude higher than in e.g. GW1. The chloride is closely associated with sodium, as shown in Fig. 20.

The origin of the salinity in GW4 is unclear. Possible sources of sodium chloride could be

- o present seawater or Baltic Sea water
- o relict seawater or Baltic Sea water
- o residual sodium chloride in e.g. post-glacial clay formations (pore waters)
- o fluid inclusions or residual hydrothermal fluids
- o brine solutions associated with salt formations.

Much of the information on deep saline groundwaters comes from the thorough analyses of the Stripa waters, e.g. in 5 and 11. Some representative element ratios in saline waters are given in Table 3. The origination from relict Baltic Sea-water, possibly with minor contributions from residual hydrothermal fluids is probable. In any of these cases the residence time of the water would be 3-4000 years or more.

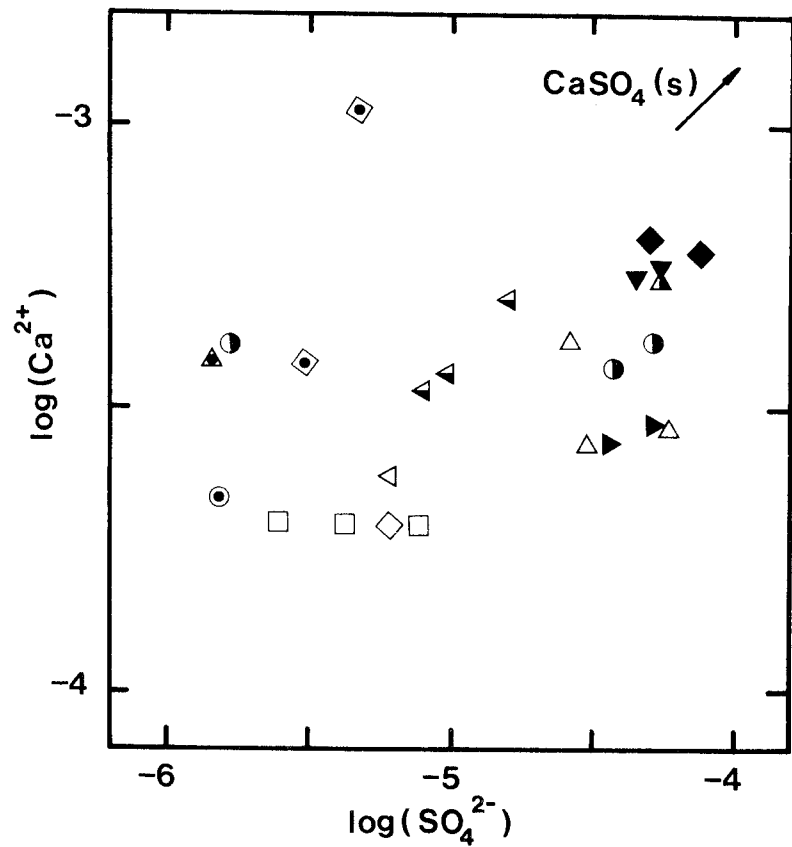


Figure 18 $\log(\text{Ca}^{2+})$ vs $\log(\text{SO}_4^{2-})$
 Symbols as in Fig. 13.

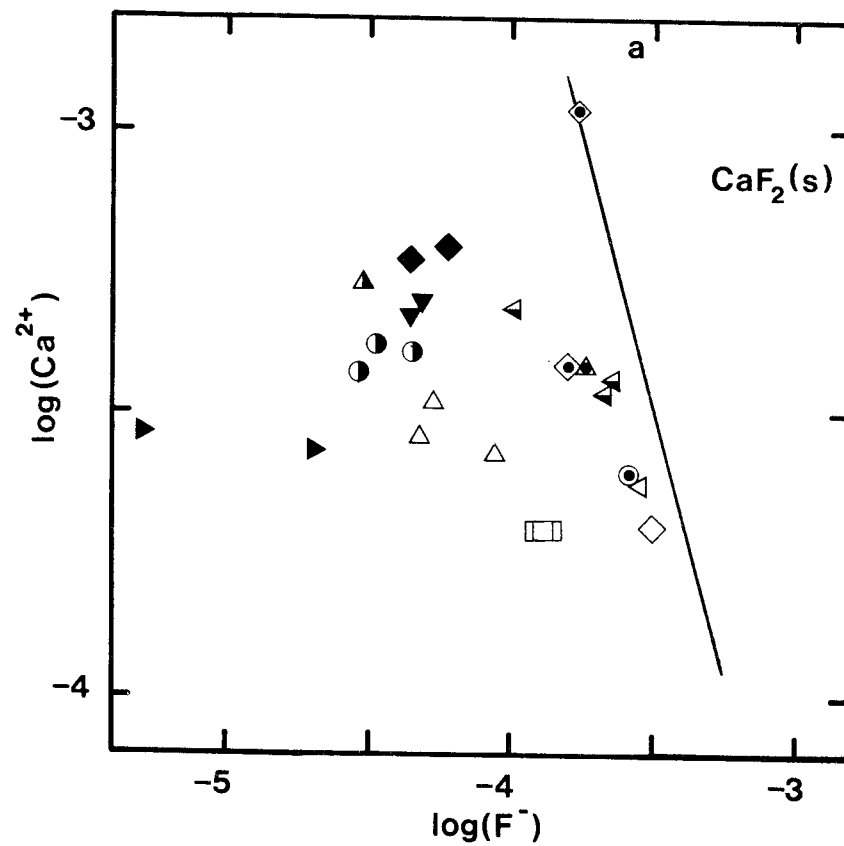


Figure 19 $\log(\text{Ca}^{2+})$ vs $\log(\text{F}^-)$
 a. $\log(\text{Ca}^{2+}) = -2\log(\text{F}^-) + \log K_s$; saturation line at 10^0C ;
 $\log K_s = -10.54$
 Symbols as in Fig. 13.

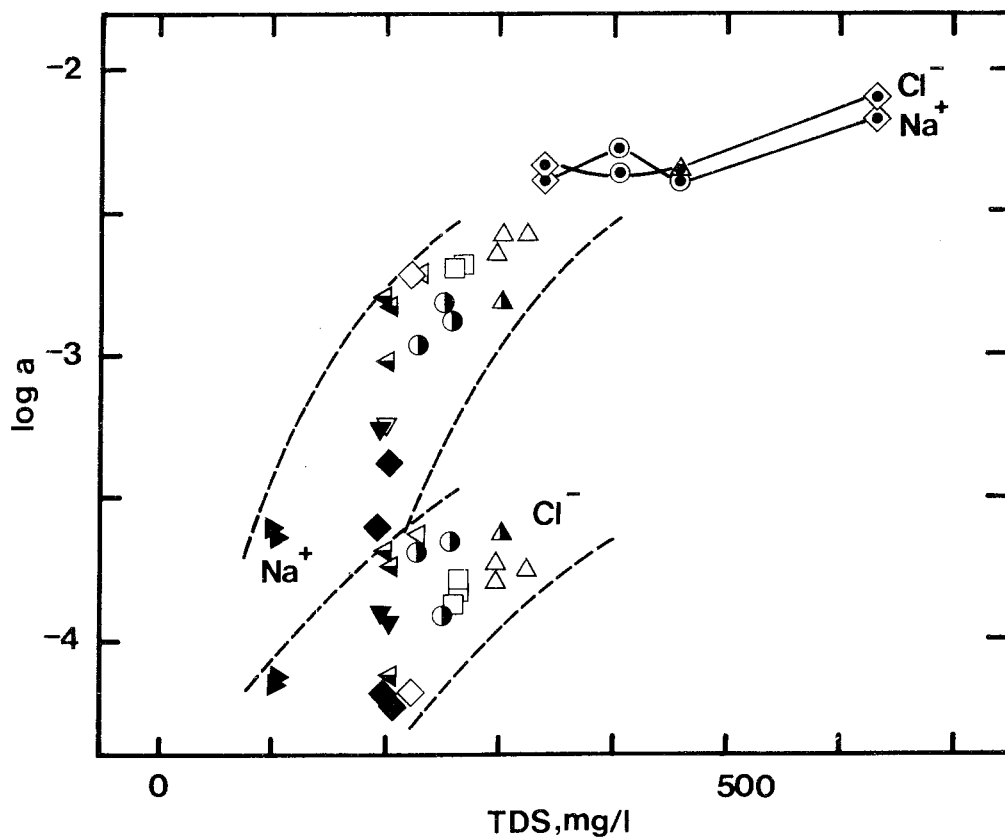


Figure 20 $\log(\text{Na}^+)$ and $\log(\text{Cl}^-)$ vs TDS
 Symbols as in Fig. 13.

Table 3 Element ratio characteristics (by weight) in saline water (GW4)

Sample	Na/Cl	K/Cl	Mg/Cl	Ca/Cl	Ca/Na	Ca/Mg
Fj2:506	0.76	0.0066	0.0084	0.071	0.095	8.6
Fj4:420	0.59	0.017	0.013	0.12	0.21	9.1
Gi4:385	0.58	0.012	0.0058	0.12	0.21	21
Gi4:596	0.56	0.0095	0.0063	0.21	0.37	33
Seawater	0.55	0.021	0.067	0.021	0.038	0.32
Baltic Sea						
Relict seaw.	0.4-0.6	0.009-0.01	0.008-0.02	0.2	0.3-0.5	20-24
Relict	0.3-0.9	0.003-0.07	0.05-0.08	0.04-0.5	0.3-0.6	1-57
Baltic Sea						

The occurrence and origin of saline groundwaters in Sweden are further discussed in Appendix 1.

3.2.6. Other inorganic constituents

The redox potential sensitive systems (Mn, Fe(II), HS^-) as well as the measured redox potentials are thoroughly discussed in ref. 15 and briefly summarized in 4.

The total phosphate concentrations are with few exceptions below 0.1 mg/l.

The total nitrate-nitrite is usually also below 0.1 mg/l and the ammonium slightly above 0.1 mg/l. The uncertainty in these data (see discussions in 12, 14, 16 and 17) are, however large, and reliable conclusions, e.g. on the correlation with sulfate or total organic carbon can hardly be made. These systems are not further discussed in this report.

The measured aluminum concentrations are generally low; only occasionally above 0.5 mg/l and frequently below the detection limit 0.01-0.02 mg/l. The uncertainties in these determinations as well as the high detection limit largely prevent any final conclusions from the measured concentrations. However, the amount of aluminum in particulate matter is frequently much higher than the soluble fraction (see 3.5.).

3.3. Organic constituents

The organic contents in the deep groundwaters are surprisingly high, usually in the range 2-4 mg/l, but occasionally much higher. Fractionation analysis of the organic materials has been performed, and the possible composition of the various fractions is discussed in detail elsewhere (10,27) and summarized in Table 4.

Most of the identified material appears to be fulvic acids of intermediate molecular weight (700-1000) or higher, and only occasionally high-molecular humic acids (e.g. in Km3:376).

The complexing properties of these organic acids are not known. However, a capacity of 2-3 meq/g would be possible for the high-molecular weight fractions (28), which in fact would potentially constitute a total complex-

Table 4 Fractionation analysis of organic material (27).

Sample	HA ^a	FA ^b					TOC mg/l.	
		MW fractions					c	d
		>1000	1000-700	700-300	c 300	c		
Fj2:409	0	28	63	8	1	4.3	3.3	
Fj2:506	0	18	63	15	4	2.3	2.3	
Fj4:349	0	48	47	5	0	4.5	6.7	
Fj4:420	0	53	37	9	1	6.8	7.9	
Fj8:402	0	12	67	18	3	1.1	1.6	
Fj8:562	0	23	46	17	14	1.6	15	
Gi2:478	0	43	47	10	0	1.8	3.0	
Gi2:528	0	55	34	6	5	1.9	3.5	
Gi4:385	0	21	68	11	0	1.7	3.9	
Gi4:498	0	29	40	19	12	1.2	2.6	
Sv4:551	0	41	40	9	10	2.1	4.4	
Km3:376	31	69	0	0	0	0.4	4.0	

^a HA = humic acid

^b FA = fulvic acid; includes all pH 1 - soluble fractions

^c Ref 27, corresponds to all fractions in the table

^d average values, see Fig. 4-10; from ref. 12, 14, 16, and 17.

ing capacity comparable to that of carbonate in many of the deep groundwaters. Thus, the influence on trace metal chemistry by the presence of natural organic material in deep groundwaters would not be negligible for e.g. lanthanides and actinides, as well as for many hydrolyzable transition metals.

3.4. Particulate matter

Particulate matter was removed from groundwater by filtration through a 0.45 μm membrane filter during the field sampling. Most of the waters appeared to have a particulate content of 0.1-0.2 mg/l, corresponding to about 50-500 particles/ml (2-80 μm). Higher values (0.3-0.4 mg/l; up to 5000 particles/ml) were occasionally observed, as well as a few waters with very high particulate content (11 mg/l, 38000 particles/ml in G14:512) (see ref. 12, 14, 16 and 17, as well as Appendix 2). It is likely that the higher values merely represent crushed material that has been generated and released during the drilling operation, as indicated from the size-distribution measurements, which frequently showed a maximum in the frequency of various particle sizes around 10 μm or above. The lower values might be more representative of the natural conditions. Iron and aluminum constitute minor fractions of the particulates, while possibly the bulk of the material could be silica (not analyzed).

4. REDOX CONDITIONS

Minerals containing ferrous iron (Fe(II)) present in granitic rock will interact with the groundwater to give it a reducing character. Laboratory experiments have e.g. shown that the reducing capacity of the rock is fast for the reduction of dissolved oxygen (29). The subsequent discussion summarizes the observations on the redox conditions in the sampled waters. A more detailed analysis is given elsewhere (15).

4.1. Redox couples and equilibria in solution

The groundwaters contain a number of redox sensitive elements and compounds. The most important of these are oxygen, iron, manganese, uranium as well as sulfur and nitrogen containing species. From all the various redox couples present in the waters it is possible to calculate a redox potential (E_h) according to the Nernstian equation:

$$E_h = E^{\circ} + \frac{RT}{nF} \ln \frac{a_{\text{ox}}}{a_{\text{red}}} \quad (20)$$

where E° is characteristic of the redox couple and a_{ox} and a_{red} are the activities of the oxidized and the reduced form, respectively, of the element.

Calculations of Eh made on this basis would give as many different results as the number of redox couples used for the calculation. This simply indicates that no true redox equilibrium exists in the groundwater. The reason for this might be a relatively late mixing of the water or a continuous mixing of different types of waters, but would also reflect the large uncertainties in the analytical data (e.g. for the concentration of Fe(III) in solution). Also the exact chemical composition has to be established for the members of the redox couple.

The various redox couples may also be far from equilibrium (slow kinetics of the reactions). Therefore only those reactions having reasonably fast kinetics should be used when measured E_h -value is interpreted in terms of equilibria between various species, or when Eh-values are calculated from analytical data.

4.1.1. Dissolved oxygen

Oxygen is a strong oxidant in the E_h -regions of natural groundwaters. The presence of measurable amounts of oxygen in deep groundwater is an indication of a non-realistic composition of the water. Thus, the oxygen present in the field monitoring equipment has most likely leaked into the sampling system, especially during the calibration procedure. Even when the amount of dissolved oxygen is below the detection limit the amount can be high enough to influence the E_h -measurements (cf. 4.2.).

The oxidation of Fe(II) by oxygen is a rather slow process. Still simultaneous presence of Fe(II) and traces of oxygen is not likely in groundwaters with long residence times; instead it would be an indication of a recent mixing of a deep groundwater with oxygenated water, (e.g. the drilling water).

4.1.2. The Fe(II)-Fe(III) couple

The Fe(II)-Fe(III)-ratio is thought to be the prime redox determining reaction in most natural groundwaters. This system could also be used for the calculation of theoretical E_h -values of the waters (see 4.3.).

The solubility of Fe(III) is so low under the conditions of the investigated waters, that usually no dissolved Fe(III) can be quantitatively determined in the sampled waters.

All the sampled groundwaters contained Fe(II) which is a direct indicator of the reducing ability of the waters.

4.1.3. Uranium and manganese

The uranium and manganese contents in the water can be used as indirect E_h -sensors. The manganese is insoluble in the oxidation state IV, whereas it is soluble in the II-state. Measurable amounts of Mn(II) indicate an E_h of less than about 0.34 V.

Uranium in the VI-state is soluble as carbonate complexes, whereas uranium in the IV-state has a very low solubility (30). The very low uranium concentrations obtained in the sampled waters indicate that uranium is not oxidized to the hexavalent state. Consequently the E_h is less than about -(0.15-0.2) V under the conditions of the sampled waters.

4.1.4. Sulfide and others

The presence of sulfide in the water is a good indicator of a reducing environment. The sulfide is easily oxidized to sulfate by dissolved oxygen, whereas the opposite reaction only takes place in the presence of bacterial activities. Hence no calculation of E_h should be made on the SO_4^{2-}/HS^- couple.

Also the NO_2^-/NO_3^- -system as well as certain dissolved organics would affect the redox conditions of the water.

4.2. Measured and calculated redox potentials

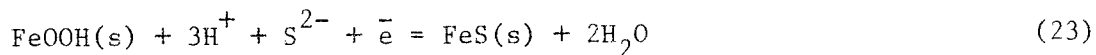
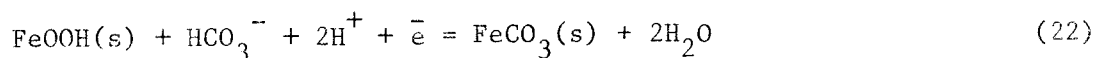
4.2.1. Redox potential measurements

The E_h as well as pH, pS^{2-} , dissolved oxygen and the conductivity were continuously monitored during the sampling procedure, using a flow-through cell where the water passes before coming into contact with the atmosphere. The equipment and the technique used in the field measurements are described elsewhere (15).

The very low E_h -buffering capacity of the water makes the measurements tricky. During the investigation period of two weeks, the measurements were interrupted by calibration procedures with three to four day intervals. Normally a period of minimum three days would be needed before the electrode potentials stabilize after a calibration with the equipment used so far. Hence stable values were not always obtained. The measured potentials were always below zero indicating a reducing environment.

4.2.2. Calculated data

The measured E_h -values should be compared to values calculated for reasonable models including Fe(II)/Fe(III)-equilibria. This kind of calculation was made with models including goethite and free Fe(II), solid Fe(II)-carbonate and Fe(II)-sulfide as redox determining systems according to



Reactions involving other solid phases ($\text{Fe}_2\text{O}_3(\text{s})$, $\text{Fe}_3\text{O}_4(\text{s})$) could be formulated in a similar way. The calculated values are in many cases more negative than the measured values, Fig. 21. The reason for this is likely to be the effect of dissolved oxygen in the system. Improvements of the measuring technique have recently given a better fit between measured and calculated redox potentials. The agreement was very good, however, in laboratory experiments under rigorous control of the oxygen pressure. A good fit was obtained for an Fe(II)/Fe(III)-system in a carbonate water

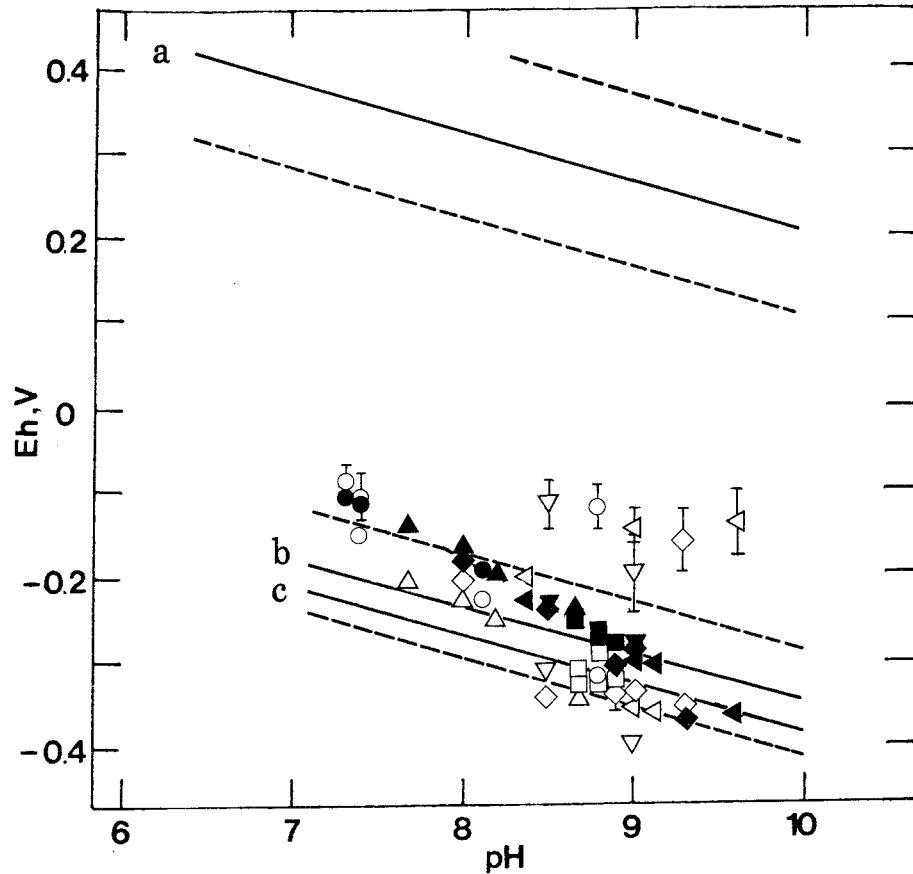


Figure 21 Eh/pH-diagram with measured and calculated redox potentials

- $Eh=0.8-0.06pH\pm 0.1$; equilibrium with air
- $Eh=0.24-0.06pH\pm 0.06$; empirical line covering all the Eh-values calculated from the measured Fe(II)-concentrations
- $Eh=0.21-0.06pH$; equilibrium with $Fe_2O_3(s)/Fe_3O_4(s)$

Symbols as in Fig.3;

Open symbols: Calculated values assuming an Fe(II)/FeOOH(s)-equilibrium;

Filled symbols: Calculated values assuming an Fe(II)/FeOOH(s)- $FeCO_3(s)$ -equilibrium;

Open symbols with bars: Experimental field data with estimated uncertainties.

(e.g. 10 mM) assuming an E_h -control according to reaction (22). (This corresponds to the filled points in Fig. 21).

5. IDENTIFIED MINERAL PHASES

Fissure filling materials in contact with groundwater have been sampled and identified from all the test sites (18,19,22,23). Data from only two water sampling levels in one hole are available from Kamlunge so far. The minerals identified in this drilling core will not be discussed, since further analysis, both of mineral phases and groundwaters, at larger depths are in progress.

5.1. Fjällveden

Kaolinite, calcite, chlorite, illite, laumontite, pyrite, smectite, and serpentine have all been identified within fissures in the drilling cores. Most common fissure filling minerals are kaolinite, calcite, and to some extent chlorite and illite. Both kaolinite and laumontite would be stable phases in contact with the analysed waters according to Fig. 16. Illite and chlorite are likely to be unstable under the same conditions.

Plagioclase, which is the main constituent of the bedrock, is likely to be unstable and would be transformed into kaolinite. The occurrence of calcite at the deepest level would be expected, according to Fig. 13.

5.2. Gideå

Laumontite, pyrite, chlorite, calcite, smectite and stilbite are the most important fissure filling minerals identified within the drilling cores. Smectite and stilbite are likely to have a preference to fissures within the dolerite dykes. From Fig. 13 and 16 it is suggested that calcite, laumontite and probably also montmorillonite (smectite) would be stable phases. However, chlorite which is a mineral usually formed during hydrothermal conditions, is likely to be unstable in most of the analysed waters. Fluorite, which would be expected in Gi4 at the largest depth according to Fig. 19, was not found.

5.3. Svartboberget

Some of the most important fissure filling minerals identified at Svartboberget are calcite, chlorite, illite, pyrite, epidote and some zeolite minerals (mostly laumontite). Laumontite as well as montmorillonite would be stable phases during present conditions according to Fig. 16. It is also likely that chlorite would be stable in some waters as well as illite. Kaolinite has not been identified as a filling mineral although it is likely to constitute a stable phase. Also calcite would be expected, Fig. 13.

6. ISOTOPE ANALYSIS

6.1. Stable isotopes, ^{18}O and deuterium

The variation of $\delta^{18}\text{O}$ with depth and TDS is illustrated in Fig. 22 and Fig. 23, respectively. It is evident, that $\delta^{18}\text{O}$ is fairly constant within each test area, regardless both of the depth and largely also the sampling hole, except for the saline waters which exhibit considerably higher values. Generally there is a tendency for decreasing $\delta^{18}\text{O}$ with increasing TDS. The low $\delta^{18}\text{O}$ -values indicate, that the salinity probably does not origin from a marine water. However, the differences between the test areas are significant. This can possibly be explained by latitudal differences.

A linear relation is obtained between D and $\delta^{18}\text{O}$, Fig. 24, represented by the equation

$$D = 8.1\delta^{18}\text{O} + 10.7 \quad (24)$$

This local meteoric water line is similar to previous observations reported in the literature (31). The d-value (10.7) corresponds to what could be expected from present oceanic precipitation (32). This indicates, that the water has not been subjected to any major exchange processes with the surroundings.

6.2. Tritium and ^{14}C

Several ^{14}C -determinations have been carried out on waters from Fjällveden, Gideå and Svartboberget. Many of these waters are supersaturated with

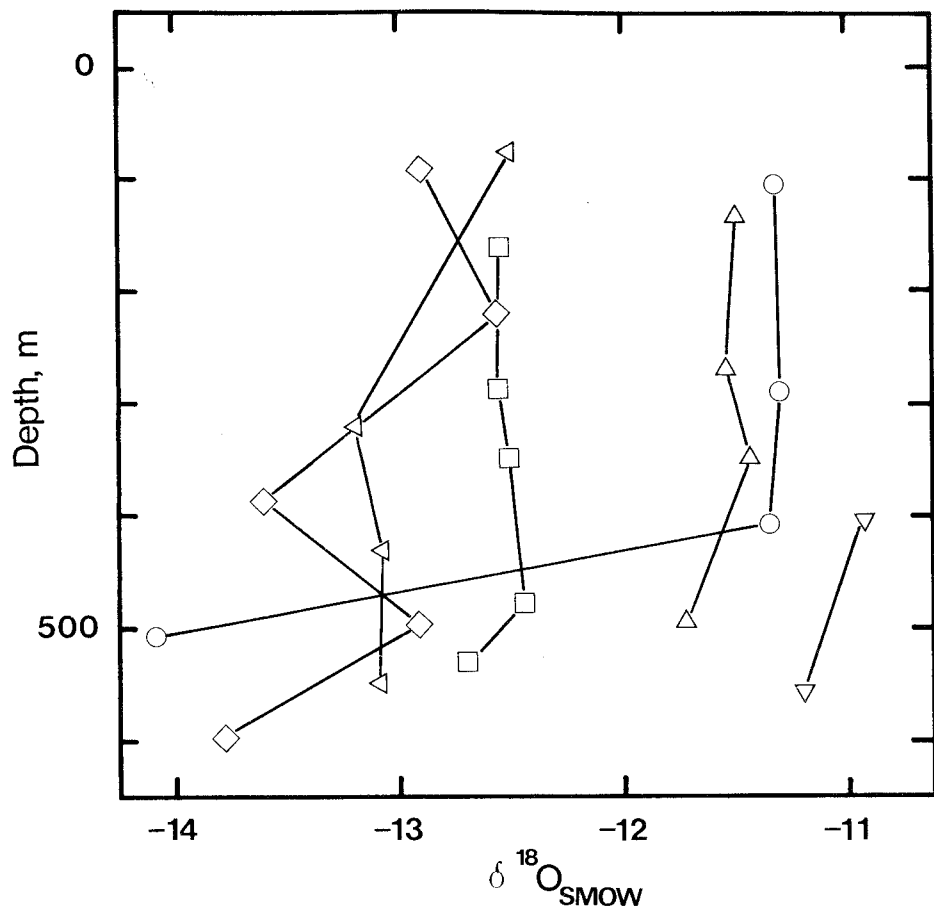


Figure 22 $\delta^{18}\text{O}$ in the sampled groundwaters
 Symbols as in Fig. 3.

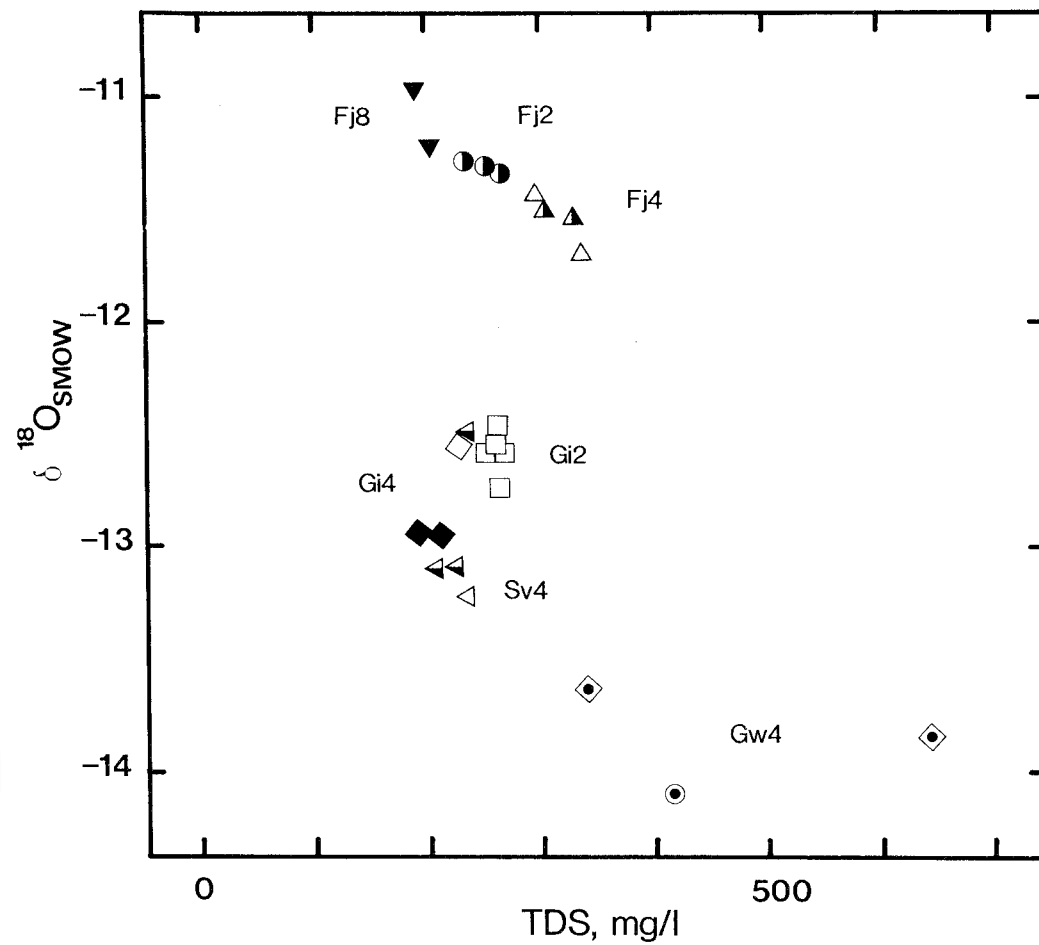


Figure 23 $\delta^{18}\text{O}$ vs TDS
 Symbols as in Fig. 13.

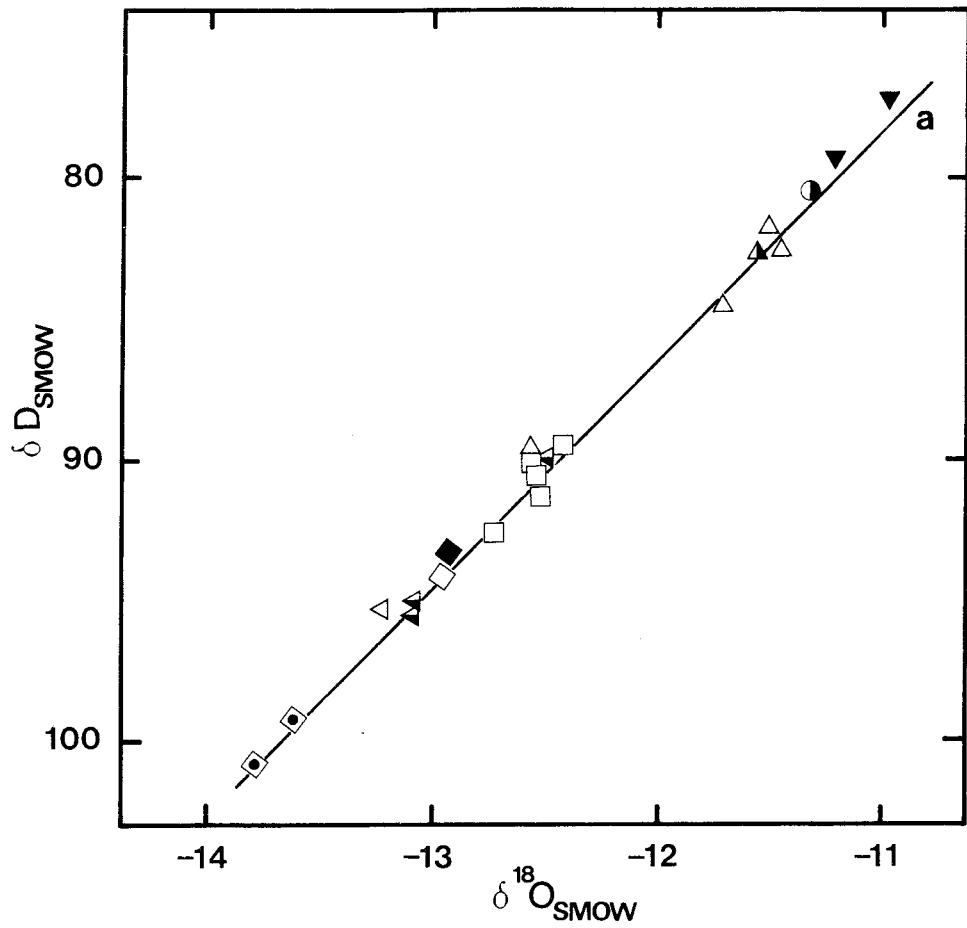


Figure 24 D vs $\delta^{18}O$
 a. $D=8.1\delta^{18}O+10.7$; local meteoric water line
 Symbols as in Fig. 13.

respect to calcite (see 3.2.1.). This means that ^{14}C -enriched calcite can be deposited on fissure walls, thus depleting the water with respect to ^{14}C . The interaction with carbonate-rich solid phases in terms of sorption and exchange reactions is not negligible and difficult to correct for in the calculations of apparent ^{14}C -ages. Thus, high ^{14}C -ages should be considered as qualitative indications of a long residence time rather than exact age determinations.

Some of the sampled waters were analysed for ^3H (tritium). Fig. 25 shows the tritium content (TU) vs the ^{14}C -ages. Presence of tritium in the water would generally indicate contributions of surface water components of recent origin (younger than 30 years).

The variations in both ^{14}C -ages and tritium contents are large for the waters from Fjällveden. An exception is Fj2:596, which has a high ^{14}C -age and no detectable tritium, indicating that this water is old and without young components. Fj2:596 is one of the saline waters (GW4).

Some of the water samples from Gi4 have large tritium contents, e.g. Gi4:498. Thus, it is likely, that these waters would include a young component. This would in fact explain the observed variations in salinity with depth (cf. Fig. 11) at Gi4. In contrast to Gi4, the waters from Gi2 show no detectable contents of tritium and only small variations in apparent ^{14}C -ages. This is in accordance with the observations that the variations in chemical composition with depth is minor. It should be pointed out that Gi4 is located in an in-flow area in contrast to Gi2, which is located in an out-flow area.

The highest ^{14}C -ages were generally observed for the samples from Svartboberget. The tritium concentrations were here below the detection limit.

The high tritium concentrations in certain samples indicate the presence of either drilling water, surface water introduced during the sampling procedure or contamination during the sample handling. The existence of a fraction of surface water in the deep groundwater even under natural undisturbed conditions does not seem probable, considering the high ^{14}C -ages, but can not a priori be excluded. No evident correlation between the tritium content and the estimated amount of drilling water in the sampled waters could be observed.

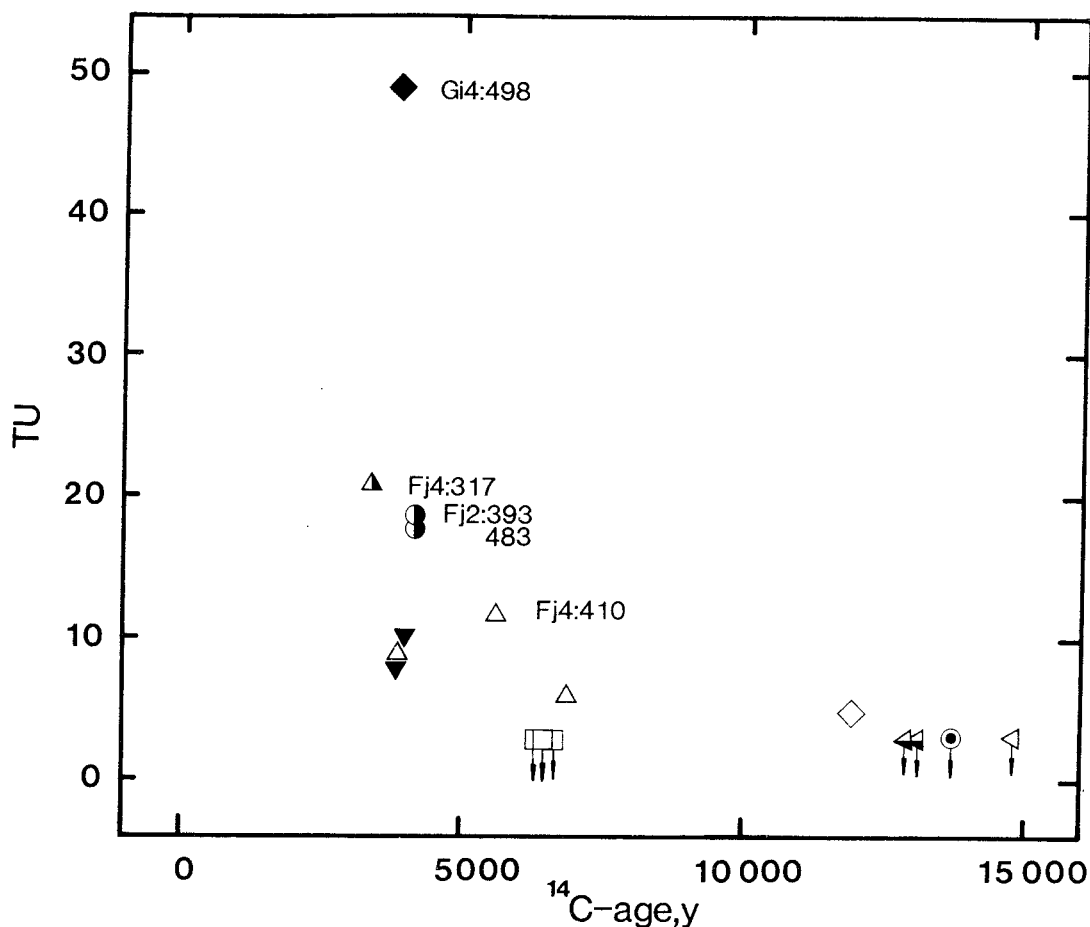


Figure 25 Tritium content (TU) vs apparent ^{14}C -age

Symbols as in Fig. 13; values below the detection limit are indicated by ↓

An intrusion of surface water caused by the drilling operation, e.g. through water-bearing fracture systems in contact both with the sampled fracture zone within packers and the over-lying parts of the drilling hole could be possible. The disturbance of the original conditions is very significant also considering the amounts of water pumped from each sampling level (several cubic meters) during a short time (less than two weeks).

7. CONCLUSIONS

The sampled groundwaters from Fjällveden, Gideå, Svartboberget and Kamlunge are generally $\text{Na}^+ - \text{Ca}^{2+} - \text{HCO}_3^-$ -dominated waters of low salinity (100-300 mg/l TDS) and high pH (8-9) and with total carbonate in the range 100-200 mg/l.

The waters are generally saturated or even oversaturated with respect to calcite, at least at the deepest sampling levels, but not saturated with respect to fluorite and gypsum.

Calcite is, as expected, a common fracture filling product.

The contents of organic material is generally considerable (1-5 mg/l), largely consisting of fulvic acids of intermediate molecular weight (<1000).

The contents of natural particulate matter (silica?) appears to be below 0.3 mg/l in undisturbed systems.

Studies of ^{18}O and deuterium indicate, that the water probably have a non-marine origin, and that the exchanges with the surroundings have been minor.

Determination of ^{14}C -ages and tritium indicate, that most of the waters appear to be old (long residence times), but also that young water components occasionally are present in the samples. It is probable, although not proven, that these young contributions are introduced due to disturbances of the natural conditions in the drilling and sampling procedure.

Intrusions of old saline waters (up to ca 650 mg/l TDS) are observed at Fjällveden (Fj2,4) and Gideå (Gi4) at the deepest sampling levels. These waters, which are $\text{Na}^+\text{-Cl}^-$ -dominated and with a low total carbonate content and high pH, possibly origin from relict seawater or relict Baltic Seawater.

The conditions in the waters are generally strongly reducing due to the presence of Fe(II)/Fe(III)-buffering systems. Pyrite is fairly abundant in fractures.

The presence of fracture filling materials like kaolinite, smectites and zeolites (e.g. laumontite) appears to have a large influence on the water composition (cation ratios).

8. ACKNOWLEDGEMENTS

Valuable comments and advices from Dr. K. Nordstrom, Prof. I. Grenthe, Ms. K. Axelsson, Dr. F. Karlsson and Dr. O. Brotzen are gratefully acknowledged.

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SALINE WATERS

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SALINE WATERS

1. INTRODUCTION

It is quite common for deep subsurface groundwaters to have increased salinities compared to shallow groundwaters, regardless of rock type (White, et al., 1963). Subsurface saline waters may contain from hundreds to thousands of milligrams per liter of chloride. The salinity content is related to the hydrogeologic origin and therefore the groundwater age as well as the geochemical processes occurring during its evolution. The groundwater age is essential for determining the safe storage of radioactive waste over long periods of time. In addition, the salinity may affect the corrosion potential of a canister, the ion exchange properties of a bentonite back-fill and the mobility of radionuclides. This report summarizes data on saline groundwaters in Sweden including data from the Finnsjön study site.

2. OCCURRENCE OF SALINE GROUNDWATERS IN SWEDEN

Saline groundwaters in Sweden may originate from five possible sources: (1) seawater, (2) Baltic seawater, (3) "relict Baltic", (4) "ancient seawater" and (5) residual igneous/metamorphic fluids. Mixtures of these may also occur and this possibility can make any interpretation extremely difficult.

2.1 SEAWATER

The surface water of the world's oceans has a very constant composition for all of the major constituents and many of the minor and trace constituents.

The concentrations of the major ions in seawater are shown in Table 1. Note that Na > K and Mg >> Ca. Several element ratios are shown in Table 2 for the purposes of comparison with the other types of saline waters. The occurrence of seawater in Swedish groundwaters is very limited and would be found along the west and south coast, near highly populated areas such as Göteborg and Skåne, where high pumpage of coastal groundwater supplies encourages the intrusion of seawater (Nordberg, 1981).

2.2 BALTIC SEA WATER

The Baltic sea is a mixture of seawater diluted with fresh water from rivers entering the Baltic. The dilution factor is roughly 6.5 for surface Baltic water, but this factor can be considerably less at depth, because the denser seawater enters the Öresund (the Denmark-Sweden sound) along the sea bottom. Since the Baltic can be thought of as a simple mixture, the element ratios should be the same. Extensive investigations by Kremling (1969, 1970, 1972) at 21 different stations in the Baltic from the Öresund to the Bothnian Bay and the Gulf of Finland show that even with considerable dilution the element ratios deviate only slightly from seawater (see Table 2). Coastal Baltic seawater intrusion also occurs in some coastal aquifers along Sweden's east coast (e.g. Sund and Bergman, 1981).

2.3 "RELICT BALTIC" SEAWATER

During the post-glacial period of the last 10,000 years there have been two major occurrences of seawater intrusion into the Baltic area. One occurrence is the Yoldia sea invasion of approximately 7,000-8,000 years ago and the Littorina sea invasion of

approximately 3,500 - 5,500 years ago. The end of the Littorina period is quite gradual and concurrent with the isostatic uplift of the Fenno-Scandian shield. These sea invasions transgressed directly across central Sweden leaving behind marine clays and saline groundwaters, which presently contaminate hundreds of wells (Nordberg, 1981; Lindewald, 1981; Engqvist, 1981). These waters are characterized by chloride concentrations in the range of 500-8,000 mg/L and their occurrence in shallow wells (typically 100 m in depth). They occur in Quaternary deposits, sedimentary rocks and in crystalline, pre-Cambrian bedrock. Examples are reported by Engqvist (1969), Moller, et al. (1974) and Möller, et al. (1981). These waters have chemical compositions which have been modified from the original seawater concentrations by clay formation, ion-exchange processes and mineral precipitation by mixing with freshwaters and non-saline sediments and rocks (Jacks, 1973, 1978; Agerstrand, et al., 1981). Element ratios for these saline waters are shown in Table 2. Geochemical processes have caused quite a range in values, but a consistent feature is seen in the increase in Ca/Cl and Ca/Mg compared to seawater. These ranges of element ratios reflect the removal of some Mg during smectite clay formation and dissolution of calcite and ion exchange to give increased Ca concentrations. Br/Cl ratio has not yet been analyzed for these waters, but it should be virtually the same as seawater.

2.4

"ANCIENT SEAWATER"

Deep subsurface brines have been found in Skåne at depths of over 1 km in Cretaceous and Jurassic sedimentary rocks (Brotzen and Assarsson, 1951). Chloride concentrations can be as high as 154,000 mg/L and the general characteristics are similar to other brines in deep sedimentary basins of the world.

By comparing the elements ratios of ancient seawater, believed to have originated with the depositional environment of 70-200 million years ago, with seawater, several major changes are apparent. The K/Cl has sharply dropped due to clay formation, e.g. illite. The Ca/Cl has markedly increased, while the Mg/Cl has markedly decreased due to clay formation. The Br/Cl ratio has increased by nearly a factor of 2 and the I/Cl ratio has increased by a factor of 15.

2.5

RESIDUAL IGNEOUS/METAMORPHIC FLUIDS

Recent investigations at the Stripa Test Site have indicated that deep saline groundwaters in the Stripa granite may be related to the inherent fluids of the igneous and metamorphic rocks found there (Nordstrom, 1983). Several anomalies of the element ratios in Table 2 suggest that the saline source cannot be ascribed to any of the four previously described origins. Notably, the K/Cl and Mg/Cl ratios are much lower and the Ca/Mg, Br/Cl and I/Cl ratios are much higher. The only type of natural water that gives such a high Ca/Mg ratio is a geothermal water of 200-350°C (e.g. Ellis and Mahon, 1977). The Br/Cl ratio indicates a non-marine origin and leaching of fluid inclusions from the Stripa granite give identical Br/Cl ratios as those in the groundwater. These lines of evidence strongly favor fluid inclusions as the source of salinity. Recent data on deep brines in the Canadian Shield (Fritz and Frape, 1982) show that they have similar element ratios to those at Stripa. These brines are also likely to be remnants of residual metamorphic fluids undiluted by younger meteoric water.

2.6

SALINE GROUNDWATERS AT FINNSJÖN

Groundwaters in FI5, -FI8 show high salinities below 140 m, with chloride concentrations as high as 5800 mg/L (Laurent, 1982). The general chemical characteristics are very similar to the Stripa groundwaters: high Ca/Mg and Br/Cl ratios and low Mg/Cl ratio.

The most likely origin of these waters seems to be a residual igneous/metamorphic fluid, which means that the groundwater has a different age and a different origin than the salinity. However, such an origin will not be typical of all deep saline groundwaters in crystalline bedrock. For example results from the Loviiso site in Finland show saline waters at depths of >75 m in crystalline rock (Snellman, 1982). These waters have chloride concentrations of about 5,000 mg/l and are very similar in composition to the relict Baltic waters found in shallow wells in Sweden. Element ratios such as Ca/Mg and Mg/Cl compare very well to the relict Baltic ratios in Table 2, but Br/Cl ratios will provide the most definitive evidence for a marine origin. Finally, it should be anticipated that mixtures of residual igneous/metamorphic fluids and Yoldia/Littorina seawater might occur at some sites.

3.

CONCLUSIONS

Saline groundwaters in crystalline bedrock of Sweden are most likely to have originated from either relict Baltic seawater (3,500-8,000 year old water) or residual igneous/metamorphic fluids or mixtures of these. These two types can be distinguished on the basis of Ca/Mg, Mg/Cl and Br/Cl ratios. If a study site contains relict Baltic seawater then the water at that depth is in the order of 3,000-8,000 years old. If a study site contains water of the residual igneous/meta-

morphic fluid type, it is likely to be older than the relict Baltic type, otherwise it would have been diluted out by fresh groundwaters. By implication then they should be considerably greater than 5,000 years and probably greater than 10,000 years.

Table 1. Composition of seawater (mg/kg from Drever, 1982)

Na	10,760
K	399
Ca	411
Mg	1,290
Cl	19,350
SO ₄	2,710
HCO ₃	142
Br	67
I	0,060

Table 2. ELEMENT RATIO CHARACTERISTICS OF SWEDISH
SALINE WATERS (BY WEIGHT)

	<u>SEAWATER</u>	<u>BALTIC</u>	<u>"RELICT BALTIC"</u>	<u>"ANCIENT SEAWATER"</u>	<u>STRIPA</u>
Na/Cl	.55	.55	.29-.90	.39-.63	.44
K/Cl	.0206	.0205	.003-.066	.009-.013	.003
Ca/Cl	.021	.020-.023	.040-.54	.18-.21	.26
Mg/Cl	.067	.067	.049-.079	.008-.016	.0007
Ca/Mg	.32	.30-.34	1.4-57	20-24	350
Br/Cl	.00347	.00336-.00348		.005-.006	.0103
I/Clx10 ⁶	3.1			40, 53	350

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INVESTIGATION OF THE CORRELATION BETWEEN
PARTICLE SIZE AS MEASURED BY HIAC AND TURBIDITY

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1983

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INVESTIGATION OF THE CORRELATION BETWEEN PARTICLE
SIZE AS MEASURED BY HIAC AND TURBIDITY

Kenneth Rosenquist

1983-03-02

Calculations of HIAC-curves

The HIAC-curves give the percentage distribution of the number of particles in the interval 2-80 μm , the absolute number of particles per ml and the percentage particle volume distribution in the above interval. The number of particles in each size class has been calculated from the percentage distribution of the number of particles. This calculation has been performed in a special way because the number of particles bigger than 13 μm are in general < 1% of the total number of particles. For this interval it is not possible to read the distribution of the number of particles in the diagram. In this interval about 50% of the total particle volume is found which has been recalculated to absolute volume assuming a mean size of 25 μm . The amount of material (mass) in each size class has been calculated assuming a density of 2.7 g/cm^3 . The turbidities have been calculated according to

$$\tau = \frac{3 \cdot \theta}{4 \cdot r} Q$$

θ = relative volume

τ = turbidity

r = particle radius (cm)

Q = "efficiency factor"

Q is a complicated function but in a first approximation it can be assumed to be 2 if the particle size is above 1 μm . The obtained τ has been related to turbidity measured as NTU through literature data on chlorobenzene, which for the instrument HACH in question is used as a calibration substance. It was thereby obtained $\tau = 2 \cdot 10^3$ NTU. The interesting parameters of the samples such as calculated number of particles, calculated mass and calculated turbidity have been listed in the table.

Discussion

According to the formula used the turbidity is decreasing with increasing particle radius. This implies that

correlation between measured turbidity and mass-concentration should only be expected if the size distribution is unchanged. The table of results shows that this cannot be the case for the samples in the table.

When turbidity is calculated as the sum of the turbidities of the included fractions a relatively good correlation with measured turbidity is obtained. These values have been plotted in figure 1. Most of the samples have measured turbidities below 3.5 NTU (9 out of 15 points). In this area it is often difficult to perform reproducible measurements. In many cases the particle frequency is also very low (< 500 particles/ml). In very particle-rich samples there is no correlation between measured and calculated turbidity.

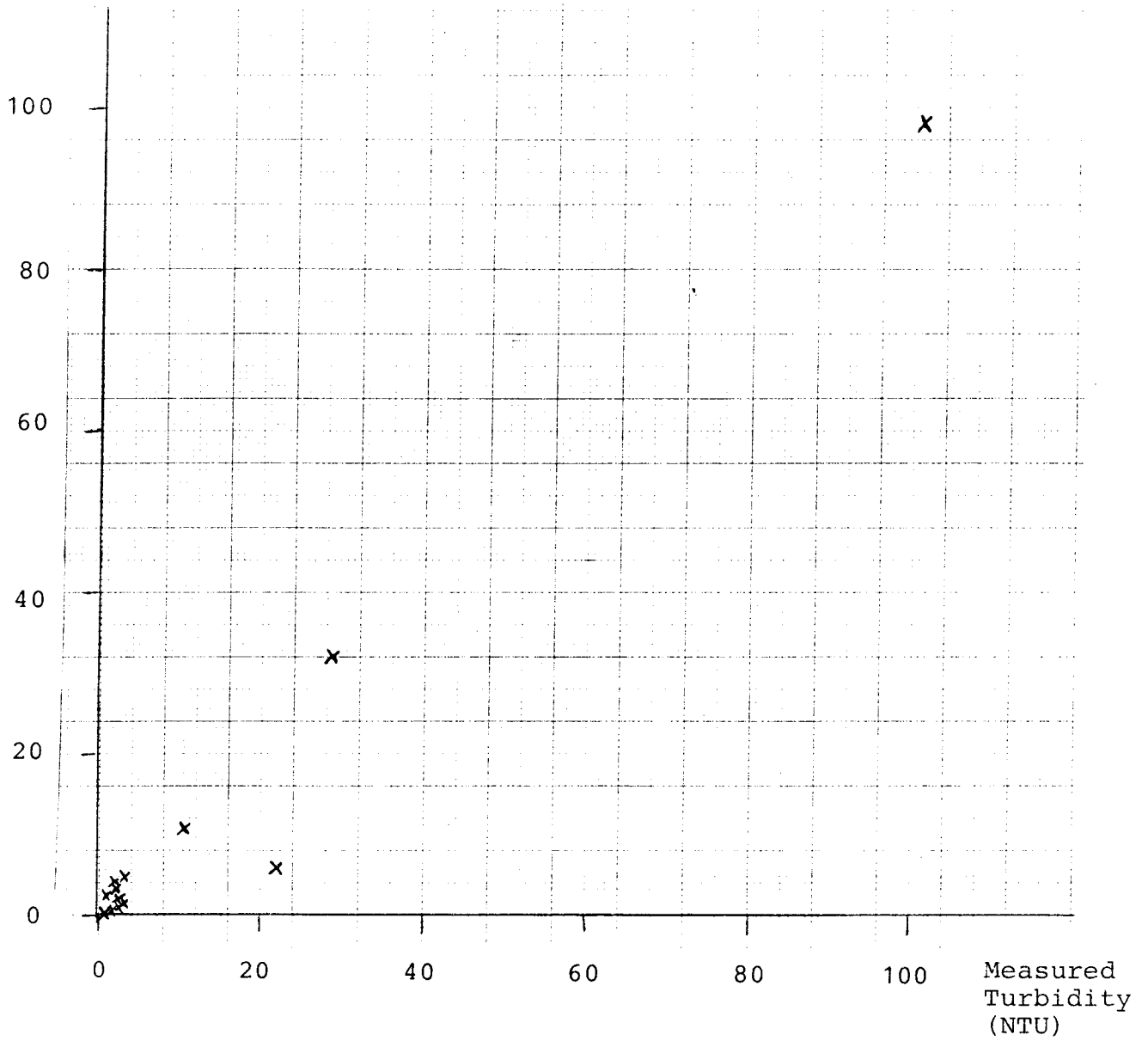
The intermediary interval only includes a few points which, however, seem to be lineary correlated. To be confident, more points are of course necessary.

Finally, it should be pointed out that the calculations of expected turbidities are very approximate and include a number of assumptions and generalizations.

Table

Sample	Part Conc (part/ml)	Calc Mass Conc ($\mu\text{g/ml}$)	Meas Turb (NTU)	Calc Turb (NTU)
KM3 445	3820	1.3	-	11.7
GI4 512	37700	11.4	101.0	98.1
GI4 404	40	0.02	1.1	0.1
GI2 602	1400	0.3	2.0	3.8
GI2 400	75	0.12	1.2	0.6
GI4 616	2860	1.44	-	10.4
FJ2 123	76	0.1	-	0.4
FJ2 342	226	0.14	2.4	0.8
FJ2 483	3550	2.07	-	11.3
FJ2 605	302	0.54	0.7	2.6
FJ4 151	553	0.12	3.3	1.2
FJ4 317	4560	0.44	2.2	4.2
FJ4 410	593	0.18	2.4	1.9
FJ4 496	351	0.17	-	1.3
FJ8 470	4820	0.33	22.0	5.8
FJ8 666	8250	0.57	11.0	10.6
SV5 160	2960	0.36	3.5	5.1
SV5 357	23400	2.19	29.0	32.8
SV5 615	65700	111.1	25.0	1434
SV5 719	430000	64.9	52.0	809
GI2 544	374		2.2	
GI2 544	-		-	

Calculated
Turbidity
(NTU)



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