The hydrogeochemistry of Norwegian bedrock groundwater – selected parameters (pH, F⁻, Rn, U, Th, B, Na, Ca) in samples from Vestfold and Hordaland, Norway

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Morland, G., Reimann, C., Strand, T., Skarphagen, H., Banks, D., Bjorvatn, K., Hall, G.E.M. & Siewers, U. 1997: The hydrogeochemistry of Norwegian bedrock groundwater - selected parameters (pH, F⁻, Rn, U, Th, B, Na, Ca) in samples from Vestfold and Hordaland, Norway. *Norges geologiske underssøkelse Bulletin 432*, 103-117.

Over 300 samples of bedrock groundwater from Hordaland county and the Oslo rift lithologies of Vestfold county have been collected and analysed for radon and fluoride. A selection of 150 samples have been analysed for a large range of major and trace parameters by ICP-AES and ICP-MS techniques at two different laboratories. 53% of all waters contravene recognised drinking water limits for at least one of the following parameters: pH, U, Rn, F⁻ and Na. The majority of these contraventions are identified in the Hordaland area (64% contravention), with a rate of only 28% contravention in Vestfold. Maximum levels of 2 mg/L, 6840 Bq/L and 9.2 mg/L were recorded for U, Rn and F⁻, respectively. These findings have implications for water resource policy in a country where groundwater is actively being promoted as an alternative to vulnerable surface water sources. Bedrock groundwater may still be considered a viable resource provided that effort is invested in compiling hydrochemical hazard maps for bedrock groundwater and provided that consumers and water suppliers are willing to accept the need for a degree of water treatment in some cases.

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Introduction

In Norway more than 80% of the population receives drinking water from surface water sources (Ellingsen & Banks 1992). In the 1980s the Geological Survey of Norway (NGU) undertook an extensive investigation of the chemical composition of surface and groundwater samples from almost all Norwegian waterworks supplying more than 1000 people with drinking water. This undertaking resulted in data on the chemical composition of water drunk by 70% of the Norwegian population for major and some trace parameters (Flaten 1991). Since that time, increasingly sensitive methods have become available (e.g. inductively coupled plasma mass spectrometry - ICP-MS) for determining very low levels of trace elements. Interest has also increased in the significance of some, previously seldom analysed, parameters in the context of human health and also in the use of surface and groundwaters as a geochemical mapping medium (e.g. the Norwegian survey of 473 lakes; Skjelkvåle et al. 1996).

NGU and the Norwegian Radiation Protection Authority (NRPA) have invested significant effort in recent years in investigating concentrations of radionuclides in groundwater abstracted from Norway's crystalline bedrock (Strand & Lind 1992, Banks et al. 1995a,b). These studies have confirmed that a significant proportion of bedrock groundwaters in some lithologies (particularly granites and gneisses) exhibit radon concentrations of several hundred or even thousand Bq/L. Such concentrations may have a direct health impact via ingestion or an indirect one via degassing inside a house and subsequent inhalation (Swedjemark 1993). The investigations resulted in the NRPA (1995) recommending an action level of 500 Bq/L for Rn in drinking water.

The work of Banks et al. (1995b) and Sæther et al. (1995) revealed that concentrations of other trace elements such as F, Be, Th, U in bedrock may be significant in a health context. In fact, Bjorvatn et al. (1992, 1994) have documented cases of dental fluorosis in southern and western Norway ascribable to the consumption of fluoride-rich bedrock groundwater.

This paper presents selected results from a recent, more extensive survey (Morland et al. 1995, Reimann et al. 1996) of over 300 bedrock boreholes in the Vestfold and Hordaland areas, dominated by granitic and gneissic lithologies. The study uncovered significant levels of Rn (up to 6840 Bq/L) and U (up to 2 mg/L) in bedrock groundwaters. Reports concerning poor water quality have historically always resulted in a certain amount of media hysteria (Ibsen 1882) and the situation is similar today. Unfortunately, some of the media reports have been misleading and may have damaged public confidence in groundwater as a drinking water resource. This paper attempts to discuss the results objectively and to address the issue of whether groundwater can continue to be promoted as the universal, problem-free panacea for municipalities with existing surface water resources of unsatisfactory quality.

Materials and methods

In 1994, groundwater sampling of all known boreholes in bedrock in Hordaland county (ca. 1,000 boreholes) and in three municipalities in Vestfold county (Våle, Svelvik and Holmestrand - 314 boreholes) was undertaken as a collaboration between the health and environment authorities of northern Vestfold and the Department of Dental Research, University of Bergen. The full results of the fluoride analyses of these waters have recently been reported by Bårdsen et al. (1996). By request from NGU additional samples were taken from a selection (all 314 from the Vestfold area, 58 from the Hordaland area) of the drilled wells for more detailed chemical analyses. These were subject to radon analysis at the NRPA and fluoride analysis at the Institute for Dental Research in Bergen. A selection of 150 of the samples was submitted for ICP-MS analysis of trace elements at the laboratories of the Geological Survey of Canada (GSC) and the Federal Institute for Geosciences and Natural Resources (BGR) in Germany. The full results of the study are reported in Morland et al. (1995) and Reimann et al. (1996).

All samples were taken by staff of the local health authorities in the Vestfold area and by the Institute for Dental Research in the Hordaland area. Samples were collected from boreholes which were in regular use (to ensure the collection of «fresh» groundwater). Immediately before sampling, the tap (which may have been at the well-head or indoors) was run for at least five minutes prior to the acquisition of 120 mL aliquots of sample. Temperature was monitored during sampling to ensure fresh groundwater was being sampled. The samples were not filtered (or acidified) in the field. This decision was consciously chosen to meet the objectives of the project as a whole, i.e. to assess the total intake of the analysed elements by people using the groundwater as drinking water. The fact that samples were not filtered may, however, lead to problems in the hydrochemical interpretation of results, as some elements (e.g. Th and the rare earths) are known to be strongly bound to particulate or colloidal matter in the water. No detailed studies of the effects of filtration were carried out during this investigation although such studies are currently underway in a similar national mapping of groundwater quality. Previous studies (Banks et al. 1995b) have indicated that filtration has little effect on determined U and Th concentrations and, by implication, that most groundwaters sampled from long-established, regularly pumped boreholes are relatively free of particulates.

Analytical methods

For sampling of radon, a plastic funnel was inserted below the running water tap, such that the tap mouth was under water and there were no air bubbles in the funnel. Using an adjustable automatic pipette, with disposable tips, a quantum of 10 mL water was taken from the funnel and injected slowly into a 20 mL vial containing 10 mL of prefilled scintillation liquid (Lumagel). The ampoule of scintillation liquid and water was then sealed and shaken. The liquid gels in contact with water, immobilising the radon. Samples were delivered to NRPA within 3 days and analysed using an LKB Wallac 1215 scintillation counter, calibrated using a standard radium solution. Results were corrected for radioactive decay to give a radon concentration in Bq/L at the time of sampling.

The pH of the 58 groundwaters from the Hordaland area and 123 of the groundwaters from the Vestfold area was determined in the field using a portable pH meter.

Afterwards, a 30 mL sample was taken in a polyethylene (PE) bottle for fluoride determination at the local health authorities and at the Institute of Dental Research, University of Bergen, using an ion-sensitive electrode (Orion 960900 combined F electrode).

Finally, two new PE bottles (120 mL) were thoroughly rinsed three times with running tap water and then filled to the top. At NGU the final selection of 150 samples for ICP-MS analysis was made. The samples were sent by courier to the Geological Survey of Canada's (GSC) laboratory

Element	D.L.	Element	D.L.	Element	D.L.
Ag	0.05	Al	2	Ba	0.2
Be	0.005	Cd	0.05	Ce	0.01
Co	0.02	Cr	0.1	Cs	0.01
Cu	0.1	Dy	0.005	Er	0.005
Eu	0.005	Fe	5	Gd	0.005
Ho	0.005	In	0.01	La	0.01
Li	0.005	Lu	0.005	Mn	0.1
Mo	0.05	Nd	0.005	Ni	0.1
Pb	0.1	Pr	0.005	Rb	0.05
Sb	0.01	Sm	0.005	Sr	0.5
Tb	0.005	Tl	0.005	Tm	0.005
U	0.005	V	0.1	Y	0.01
Yb	0.005	Zn	0.5		

Table 1. Elements analysed by ICP-MS at GSC with detection limits in µg/L (D.L.).

where the sample bottles were opened for the first time since sampling, acidified with ultrapure nitric acid (at the rate of 1 mL per 100 mL sample) and shaken for 24 hours. They were then analysed by ICP-MS (VG PlasmaQuad 2+) for the elements listed in Table 1.

Details of the analytical methodology, with associated figure of merit (typical accuracy and precision), can be found in Hall et al. (1996). In addition (detection limits in brackets) Ca (0.2 mg/L), K (0.1 mg/L), Mg (0.2 mg/L) and Na (1 mg/L) were analysed by ICP-AES. International standards as well as GSC-in-house standards and sample blanks were run for quality control purposes. Aliquots of 20 samples (17 samples and 3 standards) were then shipped to BGR's ICP-MS laboratory for analysis. Samples were shipped back to NGU and stored in a refrigerator.

Several months later, when the results of the initial interlaboratory comparison and the first results from the whole data-set were available, BGR became interested in analysing the whole set of 150 samples. The original samples that had been in Canada and then stored refrigerated at NGU for about 6 months were then shipped to BGR and analysed by ICP-MS (SCIEX ELAN 5000) within two weeks after arrival for the elements listed in Table 2.

Results of interlaboratory comparison and storage effects on water chemistry are reported in Morland et al. (1995) and will be published separately (Hall et al. in prep). Generally, the results obtained from the two laboratories are in excellent agreement, with the exception of Al, Fe, Cr and Ni (see Reimann et al. 1996 for further details). Rare earth element hydrochemistry is discussed by Banks et al. (in prep.).

Element	D.L.	Element	D.L.	Element	D.L.
Ag	0.001	Al	0.05	As	0.025
B	0.01	Ba	0.002	Be	0.002
Bi	0.001	Br	0.1	Ca	10
Cd	0.002	Ce	0.001	Co	0.005
Cr	0.01	Cs	0.001	Cu	0.005
Dy	0.001	Er	0.001	Eu	0.001
Fe	2	Ga	0.001	Gd	0.001
Hf	0.002	Hg	0.005	Ho	0.001
In	0.001	K	10	La	0.001
Li	0.002	Lu	0.001	Mg	10
Mn	0.1	Mo	0.001	Na	10
Nb	0.002	Nd	0.001	Ni	0.002
Total P as PO_4^{3-}	1	Pb	0.002	Pr	0.001
Rb	0.002	Sb	0.002	Sc	0.005
Se	0.01	Sm	0.001	Sn	0.005
Sr	0.01	Ta	0.001	Tb	0.001
Te	0.001	Th	0.001	Ti	0.1
Tl	0.002	Tm	0.001	U	0.001
V	0.01	W	0.002	Y	0.001
Yb	0.001	Zn	0.01	Zr	0.002

Table 2. Elements analysed by ICP-MS at BGR with detection limits in µg/L (D.L.).

Data analysis

All graphics are produced using the DAS program (Dutter et al. 1992), based on exploratory data analysis (EDA) methods (Tukey 1977 and Velleman and Hoaglin 1981). Kürzl (1988), Reimann et al. (1988), Rock (1988) and O'Connor and Reimann (1993) give an introduction to the advantages of using exploratory data analysis methods when dealing with geochemical data.

Analytical results were classified on the basis of the aquifer geology at the borehole location into one of 11 main categories (Tables 3 and 4). During statistical treatment, any data below the detection limits of the technique were set to half the detection limit.

Lithological class	Number of wells	Number of ICP-MS analyses	Rocks
Basa_O	18	4	Permian basalts
Gran_O	44	10	Drammen Granite
Laba_O	10	1	Permian latites and basalts
Lat_O	227	67	Permian latites/rhomb porphyry lavas
Other_O	15	10	Basic igneous rocks, trachyte, syenite, metasandstone

Table 3. The five main lithological classes containing sampled bedrock wells in the Vestfold area.

Lithological class	Number of wells	Rocks
Gran_B	7	Granite and/or granodiorite
Metse_B	4	Metasediments including phyllite and meta-arkose
Miam_B	7	Migmatitic gneisses with amphibolites and metagabbros
Migm_B	7	Migmatites
Mign_B	23	Migmatitic granitic or granodioritic gneisses
Other_B	10	Miscellaneous rock types

Table 4. The six main lithological classes containing sampled bedrock wells in the Hordaland area.

The study areas

Figure 1 shows the general location and size of the study areas, in the southern half of Norway.

Vestfold region

The Vestfold area is dominated by the Permo-Carboniferous Oslo Rift, which comprises Precambrian crystalline basement and Cambro-Silurian sedimentary rocks, including U-rich alum shales near the bottom of the sequence. This sedimentary succession is unconformably overlain by volcanics (basalts, latites and trachytes) and sedimentary rocks of Carboniferous-Permian age. The Precambrian basement complex, as well as the sedimentary and volcanic cover rocks, are intruded by younger plutonic rocks (monzonites, syenites and granites) of mainly Permian age, such as the Drammen Granite. The volcanic rocks in the Oslo Rift are very favourable lithologies for hard rock groundwater boreholes with exceptionally high yields. To the east and west of the Oslo Rift, autochthonous Precambrian basement consists mainly of gneisses, granites and amphibolites dating from the time of the Sveconorwegian orogeny. Boreholes from the following lithologies were investigated in this study: Carboniferous-Permian latites, basalts, trachytes, granites and Precambrian gneisses (Table 3).

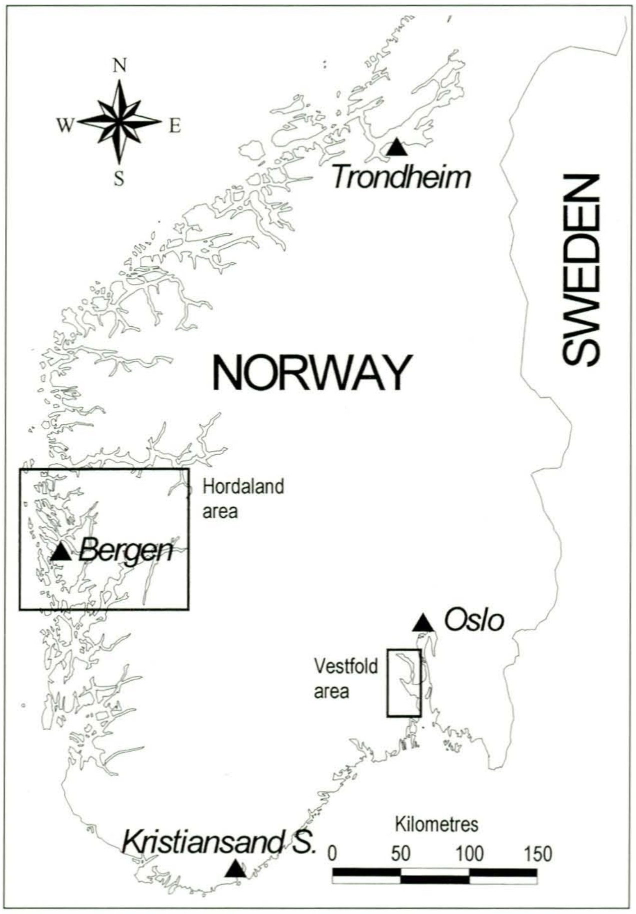

Fig. 1. Map of southern Norway, showing location of the study areas.

Hordaland region

The samples were taken dominantly from the Bergen area of Hordaland. The hard rock lithologies in the surroundings of Bergen are very variable. In the west and north-west, Proterozoic rocks of the Western Gneiss Region (WGR) are present. Migmatitic gneisses and banded gneisses of supracrustal origin are the predominant rocks-types; some gabbros, amphibolites and granites are also present. The rocks of the WGR are overlain by a variety of allochthonous rocks assigned to the Lower, Middle and Upper Allochthons of the Caledonian nappe succession. These rocks are present in the so-called Bergen Arcs, a major convex-towards-the-east, arc-like structure extending notheastwards from Bergen. Within the Caledonian nappes important rock types include diverse Proterozoic granitoid gneisses, a complex of Proterozoic monzonites (mangerites), anorthosites and gabbros, and locally abundant Cambro-Ordovician phyllites. Near Bergen, phyllites, greenstones, amphibolites and metagabbros of Early Palaeozoic age are present within the nappe sequence.

In the southeastern part of the area, southeast of Hardangerfjord, various Proterozoic gneisses and metavolcanic rocks, which are part of the Sveconorwegian basement of southern Norway, are present subjacent to scattered outliers of Caledonian nappe rocks consisting mainly of Cambro-Ordovician phyllites and related rocks.

Groundwater boreholes drilled in these different lithologies tend to display considerably lesser yields than boreholes in the Vestfold area. Boreholes drilled in the following lithologies were sampled in the Hordaland area: granites, migmatitic gneisses, migmatitic amphibolites, migmatites and metasediments (Table 4).

Hydrochemistry

pH

Figure 2 indicates that the distribution of pH is approximately normal (i.e. log-normal distribution of hydrogen ion activity). The median pH of the Vestfold waters was found to be 7.3, all measured samples falling in the range 6.0 to 8.4. The median pH in the Hordaland area is somewhat higher at 7.7, although the range is greater, stretching from 5.8 to 9.1. Compared with hard rock lithologies from other areas, e.g. the Carnmenellis granite (Smedley 1991) or Scilly granite of Cornwall, U.K. (range 5.2 - 6.4; Banks et al. 1997), these pH values are rather high, probably due to longer aquifer residence times and the fact that mafic minerals in Norway's recently glacially scoured bedrock outcrops have not been removed by prolonged subaerial weathering.

The correlation of pH with aquifer lithology is not immediately clear, although it will be noted that two acidic lithologies (Vestfold granite and Hordaland migmatites) have the lowest pH values. The migmatitic gneisses, amphibolites and metagabbros of the Hordaland area are the groups with the highest pH. The majority of groundwaters fall well within the

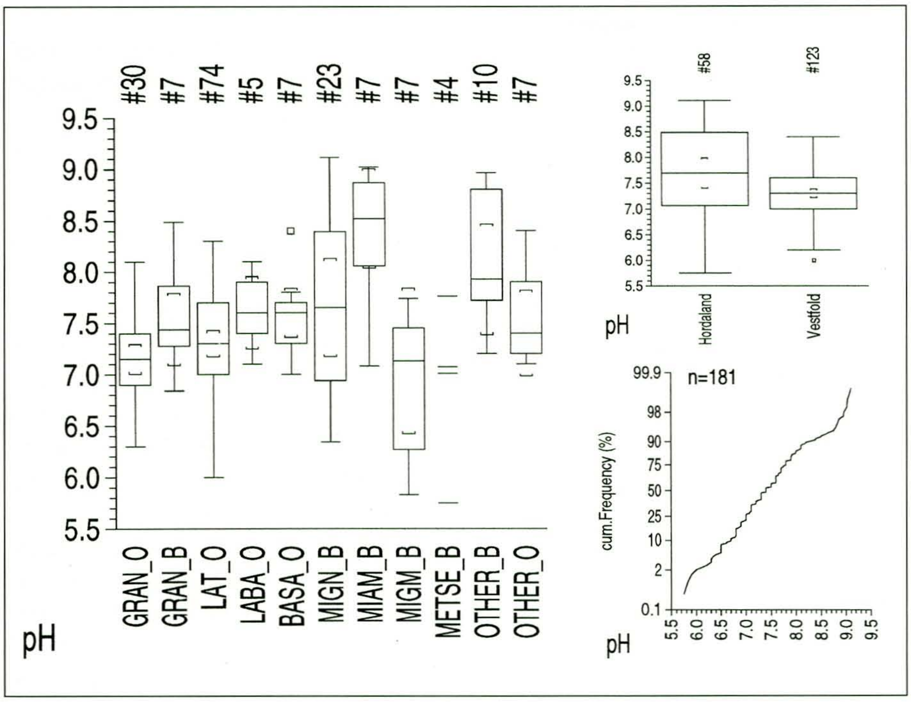

Fig. 2. Diagram showing: left, boxplots for distribution of pH in bedrock groundwaters, according to lithological class (see text for explanation): top right, boxplots for distribution of pH in the Vestfold and Hordaland areas: bottom right, cumulative probability distribution plot of pH for the entire data set (n = 181). # = total number of samples in boxplot group.

Parameter	Norm	Source	Hordaland	Vestfold	Total
			failures/ total	failures/ total	failures/ total
pH	<8.5	1	14/58	0/123	14/181
	>6.5	1	4/58	5/123	9/181
U_BGR	<20 µg/L	3	11/58	9/92	20/150
U_GSC	<20 µg/L	3	10/58	9/92	19/150
Rn	<100 Bq/L	4	41/56	211/265	252/321
	<500 Bq/L	6	15/56	40/265	55/321
	<1000Bq/L	5	4/56	24/265	28/321
F^-	<1.5 mg/L	1	20/58	40/313	60/371
Na	<20 mg/L	2	34/58	36/92	70/150
	<150mg/L	1	4/58	2/92	6/150
Combined norms for pH, U, Rn (500 Bq/L), F and Na (150 mg/L)	Compliance Failure		20/56 36/56	18/25 7/25	38/81 43/81
Ca	< 25 mg/L	2	16/58	61/92	77/150
	> 15 mg/L	2	29/58	16/92	45/150
K	< 12 mg/L	1	0/58	3/92	3/150
Mg	< 20 mg/L	1	0/58	7/92	7/150
Ba	< 100 µg/L	2	2/58	15/92	17/150

1: Norwegian maximum (or minimum) permitted concentration, Norway (Sosial- og helsedepartementet 1995)
2: Norwegian guideline value, Norway (Sosial- og helsedepartementet 1995)
3: Canadian drinking water limit (Barnes 1986)
4: Swedish 'concern' level (SIFF 1987)
5: Swedish maximum level (SIFF 1987)
6: Norwegian recommended maximum (NRPA 1995)

Table 5. Summary table showing proportion of analysed bedrock groundwater samples failing with respect to cited drinking water norms.

limits allowed by the Norwegian drinking water standards (Sosial- og helsedepartementet 1995), namely 6.5 < pH < 8.5, although a minority of samples (13%) fall both above and below these limits (Table 5).

Fluoride

Water may be a significant source of fluoride in the diet. It is known that fluoride is required in certain quantities for normal tooth and bone development. Deficiency can result in malformation of bones and decreased resistance to dental caries. Several studies (e.g. Rock et al. 1981) seem to demonstrate that artificial fluoridation of naturally fluoride-poor drinking water can decrease incidence of caries.

Many children are indeed recommended to take fluoride supplement tablets in Norway today. Nevertheless, excessive fluoride may cause negative health effects, referred to as fluorosis. Dental fluorosis results in chalkiness and mottling of the teeth, while osteofluorosis results in rough, thickened and chalky bones, particularly in the jaws, fingers and ribs. Fluorosis may also cause lameness and limb stiffness in cattle and sheep (it was first observed in volcanic terrain in Iceland in c. 1000 AD; Shupe et al. 1979). Cases of dental fluorosis have been identified by Bjorvatn et al. (1992, 1994) in some parts of western Norway, which appear to be linked to consumption of fluoride-rich bedrock groundwater, in a region where levels may exceed 9 mg/L (Bjorvatn 1996, Figure 3).

Previous studies of the regional distribution of fluoride in bedrock groundwater in cool temperate areas include that of Corbett & Manner (1984) in Ohio, U.S.A., where levels of up to 5.9 mg/L were recorded. In the current study, the highest concentrations were recorded from the Hordaland area (Fig. 3), where around one third (20 of 58) of all samples

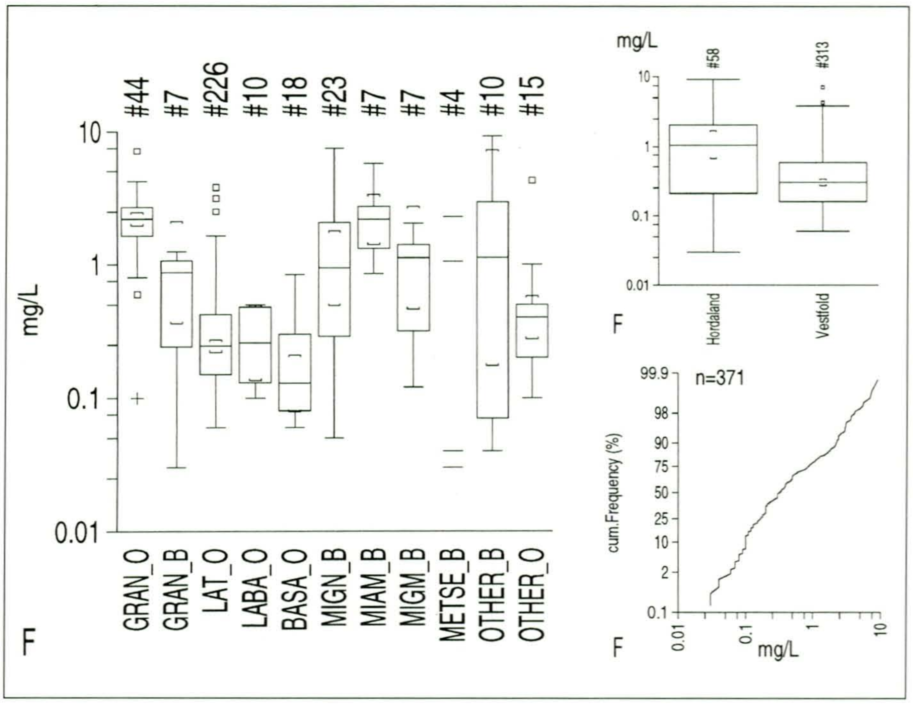

Fig. 3. Diagram showing (note the log scales): left, boxplots for distribution of fluoride (mg/L) in bedrock groundwaters, according to lithological class (see text for explanation): top right, boxplots for distribution of fluoride in the Vestfold and Hordaland areas: bottom right, cumulative probability distribution plot of fluoride for the entire data set (n = 371). # = total number of samples in boxplot group.

exceeded the Norwegian drinking water limit of 1.5 mg/L (Sosial- og helsedepartementet 1995) and a maximum concentration of 9.2 mg/L was recorded (Table 5). These high levels are typically derived from the gneissic lithologies and especially from the lithological group MIAM_B, consisting of migmatitic gneisses, metagabbros and amphibolites. In the Vestfold area, levels were generally lower, although a significant number of samples (40 out of 313 samples), mostly from the Drammen Granite and some from the latites, exceeded the 1.5 mg/L limit (Table 5).

Figure 4c reveals that in the Hordaland samples of elevated pH (>8.5) there are considerably elevated concentrations of fluoride. These high pH values are typically related to the migmatitic gneisses with intercalated bodies of metagabbro and amphibolite and it is likely that the fluoride is being derived by anion exchange for hydroxide ions on sites on amphiboles or sheet silicates. Positive correlations of fluoride with pH have been observed several times previously in Norwegian bedrock groundwaters, with a similar explanation (Banks et al. 1993, Englund & Myhrstad 1980).

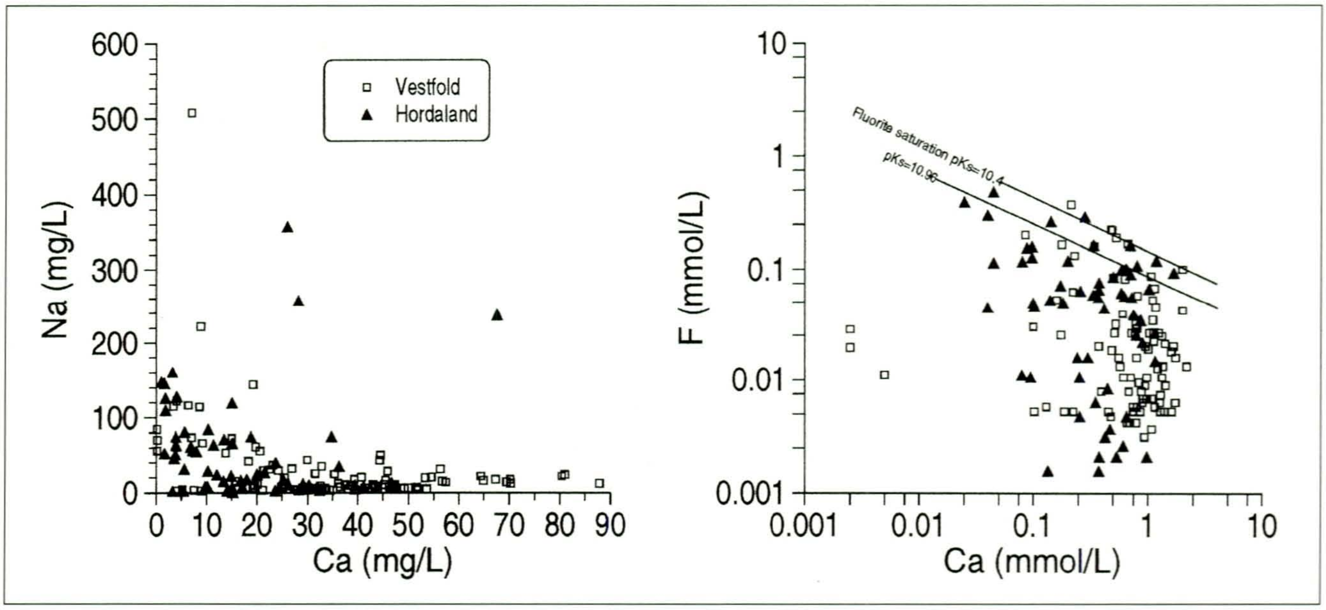

Fig. 5. X-Y graphs showing (note the log scales) (left) the relationship between calcium and sodium concentrations (mg/L) in the groundwaters from the Vestfold and Hordaland areas and (right) the relationship between molar concentrations of calcium and fluoride in the same areas. The upper line shows saturation with respect to fluorite according to Krauskopf (1979)'s value of pK_s = 10.4 while the lower line uses Nordstrom & Jenne (1977)'s value of 10.96, both at 25°C. Both lines are adjusted to T = 6°C using the Van't Hoff isotherm. Corrections for activity have not been made.

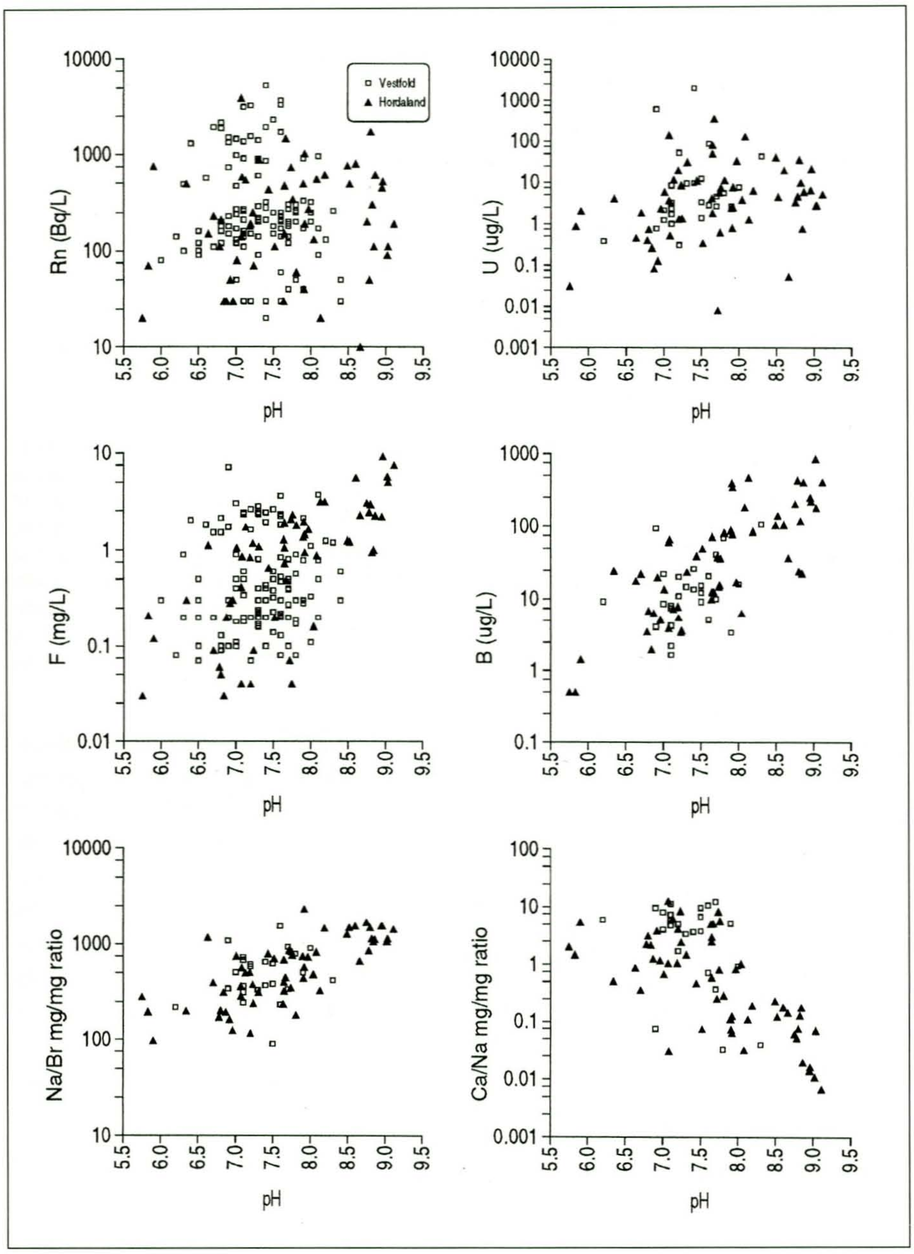

Fig. 4. XY graphs (note the log scales) showing the relations of (a) Rn, (b) U (Canadian values), (c) F-, (d) B, (e) Na/Br ratio and (f) Ca/Na versus pH for the datasets from the Vestfold and Hordaland areas.

Figure 5 reveals no clear correlation of F^- with Ca, although it will be noted that the most fluoride- and calcium-rich waters impinge upon the line defining fluorite saturation. A plausible model for fluoride evolution in these waters thus involves the derivation of fluoride by anion exchange on amphiboles, sheet silicates and, possibly, apatite, at elevated pH, with an upper limit being defined by the locus of fluorite saturation. Another possible source of fluoride may be in fluorite within the rock matrix or as a fracture mineralisation.

Radon

Apparent correlations have emerged between the incidence of lung cancer and the concentration of radon in household air. Hitherto, some 20 epidemiological studies have been carried out in mines (including uranium mines) and some 30 studies in residential environments. The most important of these are described by ICRP (1993), UNSCEAR (1994), Lubin et al. (1994), WHO (1996) and Lubin & Boice (1997). Although radon-containing water was once thought to have positive health-effects (Albu et al. 1997), there now exist epidemiological studies which appear to link radon concentration in water with incidence of gastric cancer (Mose et al. 1990). Water with elevated radon concentrations can, according to recent research, result in significant radiation doses, particularly for young children (UNSCEAR 1993, Swedjemark 1993).

Hitherto, the highest radon activity (8,500 Bq/L) documented from Norwegian groundwaters was from the Precambrian Iddefjord Granite of south-eastern Norway (Banks et al. 1995b). In recent months, the NRPA have also analysed a groundwater sample from the same lithology, near the town of Fredrikstad, with an activity of 19,900 Bq/L

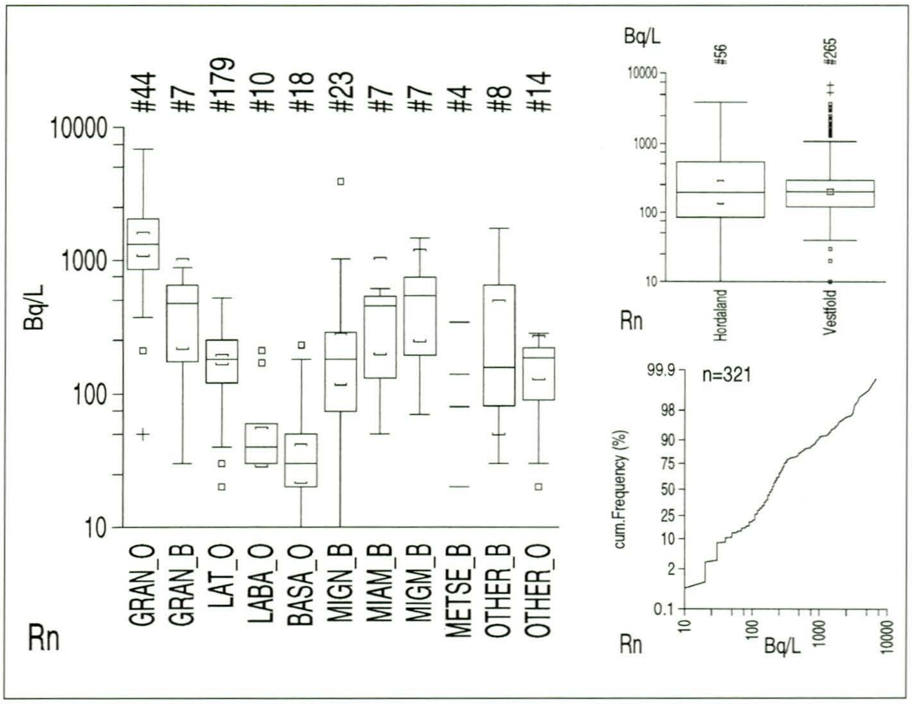

Fig. 6. Diagram showing (note the log scales): left, boxplots for distribution of Rn (Bq/L) in bedrock groundwaters, according to lithological class (see text for explanation): top right, boxplots for distribution of Rn in the Vestfold and Hordaland areas: bottom right, cumulative probability distribution plot of radon for the entire data set (n = 321). # = total number of samples in boxplot group.

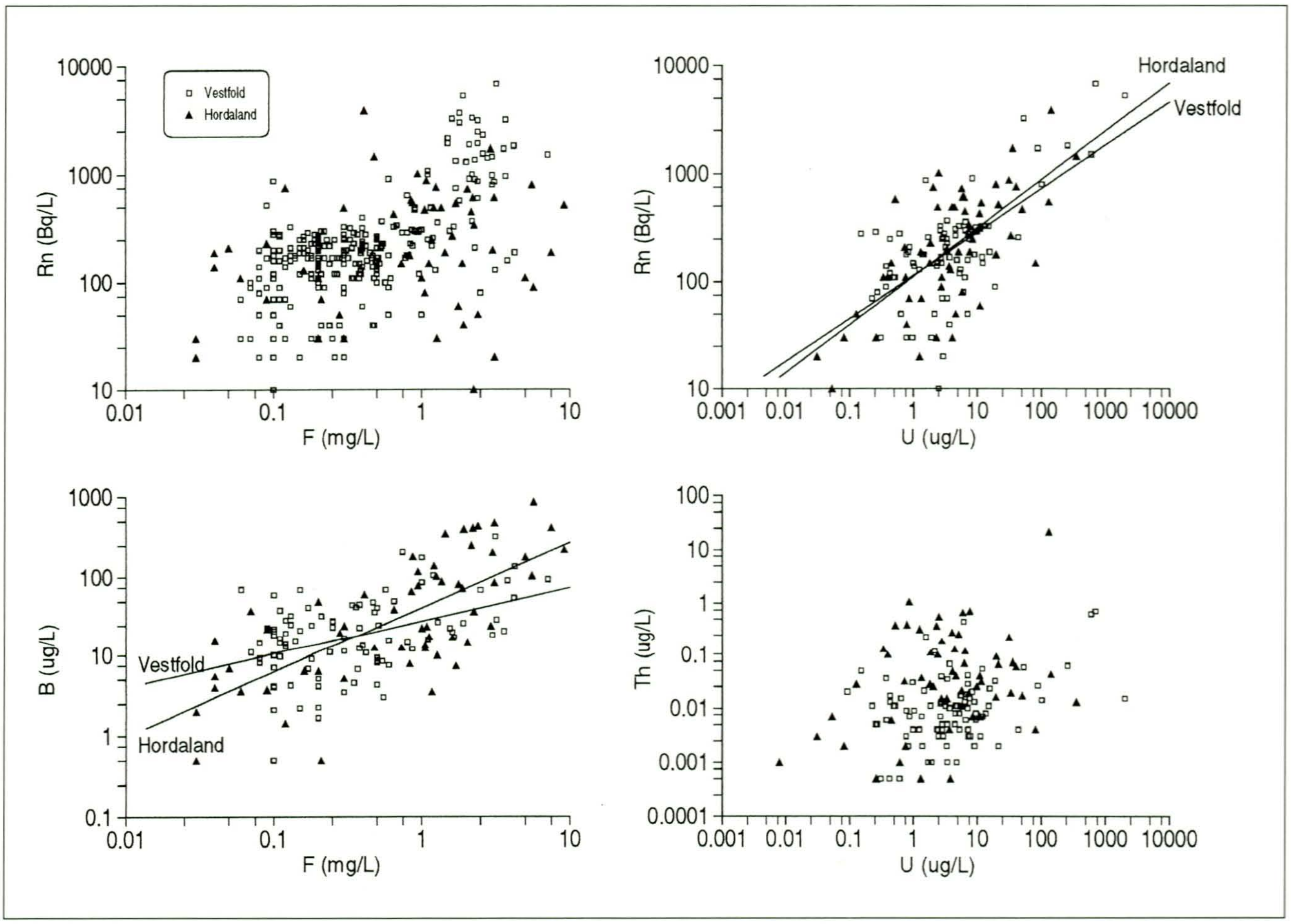

Fig. 7. X-Y graphs (note the log scales) showing covariation (a) of F- and Rn, (b) of U and Rn, (c) of F and B and (d) of U and Th in the waters from the Vestfold and Hordaland areas.

(Lind 1997). Almost 80% of samples taken from this lithology exceeded 1,000 Bq/L according to the data of Banks et al. (1995b). These values are modest, however, compared with the maxima recorded in drinking water in Sweden (57,000 Bq/L - Åkerblom & Lindgren 1996) and Finland (77,500 Bq/L - Salonen 1994).

Sweden operates with a «concern» level of 100 Bq/L for municipal drinking water, above which it is recommended that the possible effects of the radon on water users should be considered. For groundwater extracted from a private borehole to use in a single household, the similar limit is 500 Bq/L. Sweden has a maximum limit of 1,000 Bq/L, above which action to limit the dose from radon is recommended (Statens strålskyddsinstitut 1996). Norway has recently introduced a recommended maximum level of 500 Bq/L (NRPA 1995). These levels are based on radiation doses to children and adults due to ingestion and inhalation of degassed radon (Strand & Lind 1992). For children, ingestion results in the most significant dose; for adults, inhalation is the critical pathway.

On the basis of 56 boreholes in the Hordaland area and 265 boreholes in the Vestfold area (Fig. 6), around 80% of boreholes (252 out of 321) had a radon activity of more than 100 Bq/L, while some 17% (55 out of 321) had a concentration above 500 Bq/L. The highest recorded level was 6840 Bq/L. The most problematic lithological group from the point of view of radon are the Vestfold granites, dominated by the Permian Drammen granite, where almost 90% of all boreholes exceeded 500 Bq/L and over 50% of all boreholes exceeded 1,000 Bq/L. The lowest radon activities are related to the basalts of the Vestfold area. The latites and rhomb porphyries which, with their relatively high permeability, form the dominant bedrock groundwater supply around Oslofjord, exhibit radon concentrations typically in the range 100-500 Bq/L.

Figure 7a suggests that there are a significant number of cases, especially in the Vestfold area where high Rn is accompanied by high F^- concentrations. Positive correlation between Rn and F^- has been noted by Banks et al. (1995a,b). A possible coexistence of fluoride and uranium in various mineral assemblages (e.g. apatite, sheet silicates such as biotite) is well known. No radium analyses have, as yet, been performed on the waters, but Strand & Lind (1992) note that there is generally no clear correlation between radon and radium in Norwegian groundwater.

Uranium and Thorium

Previously, the highest reported uranium concentration in Norwegian bedrock groundwater was from the Iddefjord granite (170 µg/L - Banks et al. 1995b), while levels of over 14 mg/L have been reported from granites near Helsinki in Finland (Asikainen & Kahlos 1979). For thorium, Banks et al. (1995b) reported a maximum of 2 µg/L. No Norwegian drinking water limit exists for uranium or thorium whereas Canada uses a maximum of 20 µg/L for uranium (Barnes 1986).

In this survey, uranium was determined by ICP-MS both in Canada and Germany. Below 100 µg/L, the inter-laboratory calibration is good. Above this level, the Canadian results exceed the BGR results to a degree which increases with concentration. The highest concentration was found in a sample returning a value of some 2,020 µg/L from Canada and 890 µg/L from Germany. In the diagrams and in the subsequent discussion, the Canadian values for uranium are cited as these are the values measured shortly after sampling.

The distribution of uranium seems to follow that of ra-

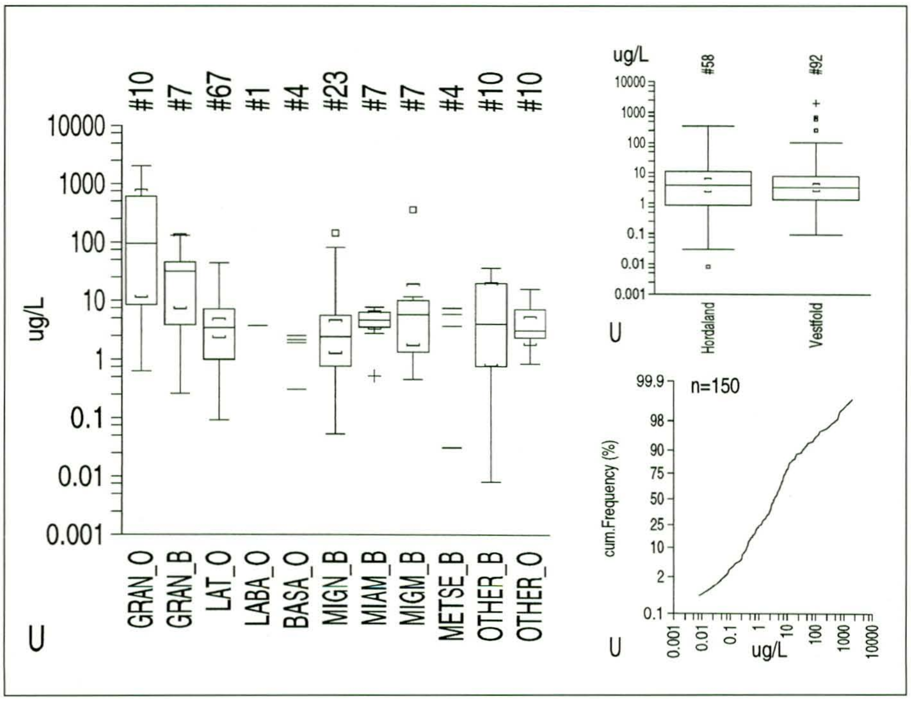

Fig. 8. Diagram showing (note the log scales): left, boxplots for distribution of U (µg/L) in bedrock groundwaters, according to lithological class (see text for explanation): top right, boxplots for distribution of U in the Vestfold and Hordaland areas: bottom right, cumulative probability distribution plot of uranium for the entire data set (n = 150). # = total number of samples in boxplot group. In all cases, the figures refer to uranium analyses carried out by GSC in Canada.

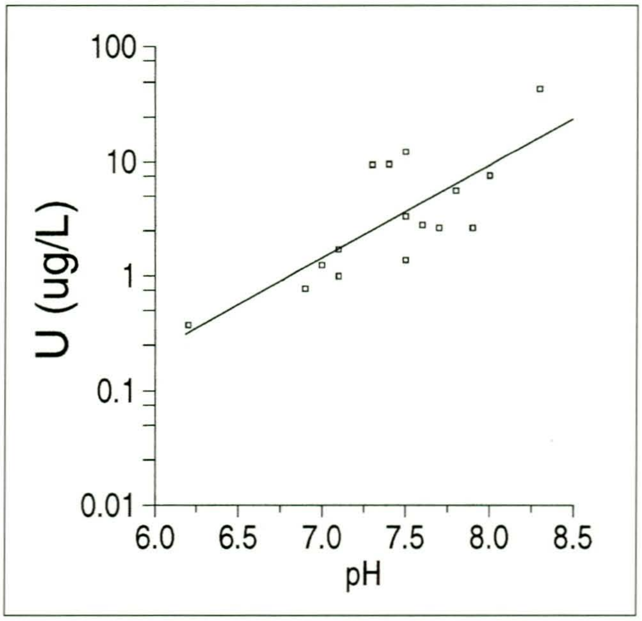

Fig. 9. XY-diagram showing (note the log scale) the correlation between U and pH in the Permian latites/rhomb porphyry lavas from Vestfold.

don quite closely (Fig. 7b). Figure 8 shows that the highest concentrations (with a median of some 100 µg/L and a maximum of 2 mg/L) are from the Vestfold granites. The next highest levels are from the Hordaland granites. Some 13% (19 out of 150 samples) of all groundwaters exceed the Canadian limit of 20 µg/L and these are dominantly from the granite lithologies. Although there exists a clear correlation between Rn and U for the total data set (Fig. 7b), this is mainly due to the large differences in Rn and U groundwater contents between the different lithologies (Figs 6 and 8), rather than to correlation within a given lithology. This is in accordance with Banks et al. (1995 a,b, 1997), and indicates that, although U content in aquifer material may act as a coarse control on both dissolved Rn and U contents in groundwater, other factors such as residence time, fracture aperture, redox conditions or weathering history control the distribution of these parameters within a given aquifer type. There may be a weak positive correlation of U with pH (Fig. 4b). The correlation is considerably stronger in the Permian latites and rhomb porphyry lavas from the Vestfold area (Fig. 9).

Thorium in groundwater seems to exhibit a very weak positive correlation with uranium (Fig. 7d), probably reflecting a coarse co-variation in Th and U content in host rocks. Thorium is generally rather insoluble, particularly so in reducing conditions, hence redox potential is likely to be a major factor in controlling thorium distribution in groundwater. In this survey almost all samples contain thorium at less than 1 µg/L, although a single sample from the Hordaland granites returned a value of some 20 µg/L (Fig. 10). Again the highest concentrations are derived from the acidic lithologies (gneisses and granites). The low solubility of Th probably implies that it poses less of a concern in the context of human health than U or Rn, but given that Th is significantly more toxic than either U or Rn, the health impact of µg/L-levels of Th cannot necessarily be disregarded as insignificant.

Boron

Boron is an essential element for plants but becomes phytotoxic in high concentrations. Concentrations of boron may thus be of concern regarding the use of groundwater for irrigation. Its essentiality for human health has not been clearly demonstrated, but its utility as a tracer of sewage leakage or of leachate from waste disposal sites renders knowledge of background concentrations in groundwater of considerable

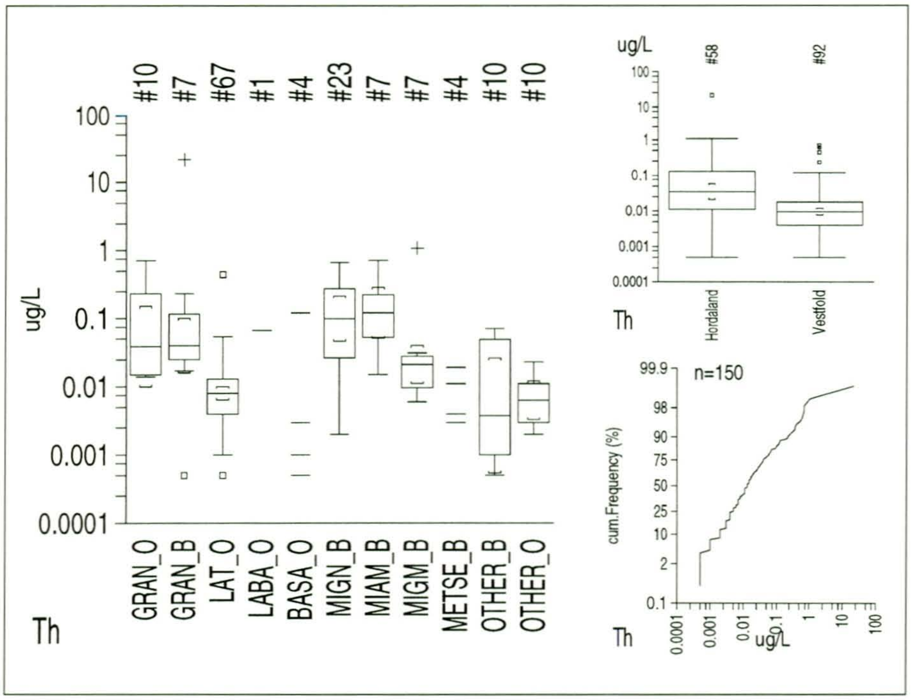

Fig. 10. Diagram showing (note the log scales): left, boxplots for distribution of Th (µg/L) in bedrock groundwaters, according to lithological class (see text for explanation): top right, boxplots for distribution of Th in the Vestfold and Hordaland areas: bottom right, cumulative probability distribution plot of thorium for the entire data set (n = 150). # = total number of samples in boxplot group. In all cases, the figures refer to thorium analyses carried out by BGR in Germany.

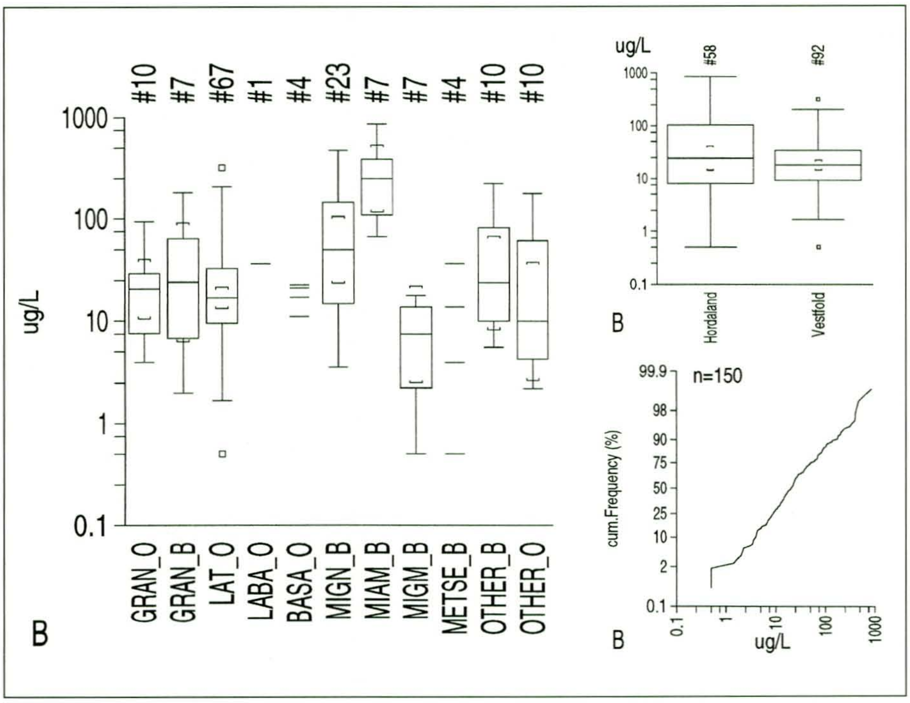

Fig. 11. Diagram showing (note the log scales): left, boxplots for distribution of B (µg/L) in bedrock groundwaters, according to lithological class (see text for explanation): top right, boxplots for distribution of B in the Vestfold and Hordaland areas: bottom right, cumulative probability distribution plot of boron for the entire data set (n = 150). # = total number of samples in boxplot group. In all cases, the figures refer to boron analyses carried out by BGR in Germany.

interest. Studies of a single lithology, the Iddefjord Granite, by Banks et al. (1993) indicate that boron in groundwater in coastal regions is partly derived from marine salts in recharge water and partly from water-rock interaction. Boron concentrations recorded in this survey vary between approximately 1 µg/L and 850 µg/L (Fig. 11). Boron shows a positive co-variation with pH (Fig. 4d) and also with fluoride (Fig. 7c). As with fluoride, the highest boron concentrations are clearly derived from the lithological group containing migmatitic gneisses, amphibolites and metagabbros from the Hordaland area.

Sodium and Calcium

Both sodium and calcium have been regarded as problematic parameters in the context of Norwegian groundwater resources for some time. The former Norwegian guideline level for sodium (SIFF 1987) was set at a level of only 20 mg/L, which was frequently exceeded in the bedrock groundwater of a country with a high degree of marine influence (airborne and relict marine-derived salts in recharge water) and an abundance of rocks containing sodic plagioclase. The maximum concentration for sodium has been recently defined at the more realistic level of 150 mg/L, although the guideline value of 20 mg/L is retained (Hellesnes 1995, Sosial- og helsedepartementet 1995).

Calcium has also long been regarded as undesirable from an aesthetic point of view in Norway, due to scaling of kettles, and problems with foaming of soap. Former guidelines (SIFF 1987) stipulated maximum limits for calcium in, despite the fact that calcium is not regarded as toxic in drinking water. This despite the fact that several other European nations have set minimum levels for hardness in drinking water, a concept now accepted in Norway (see below). These minimum levels have been ostensibly promoted by the observation that the incidence of heart disease in several countries, such as the UK (Crawford et al. 1971, Lacey 1981) and Norway (Glattre et al. 1977), is inversely correlated with water hardness. The reasons for this remain unclear, although several possible explanatory models have been proposed:

- Calcium-rich waters are often poor in sodium, a high dietary content of which is known to exacerbate hypertensive disorders.
- A high water hardness hinders the solubility of many toxic metals, such as lead, which otherwise may be solubilised from geological materials or from the distribution network (Crawford & Clayton 1973).
- Heavy industry, particularly in the UK, has grown up in areas with soft water supplies suitable for textiles manufacture and boiler feed water (i.e. it is the industrial environment rather than the water per se which leads to the disease).

Currently, Norway advises a guideline range of 15-25 mg/L for calcium, yet (slightly perversely) sets a minimum level of 60 mg/L calcium-equivalents (Ca or Mg) for artificially softened water (Sosial- og helsedepartementet 1995).

Figure 12 displays the sodium concentrations recorded during this study. It will be noted that around 50% of waters (70 out of 150) exceed the Norwegian guideline of 20 mg/L and 4% (6 out of 150) exceed the limit of 150 mg/L, while the maximum observed concentration is 508 mg/L. Banks et al. (1993) demonstrated that sodium in groundwater in a coastal granite lithology was partly derived from marine salts (from precipitation or direct intrusion) but was in many cases largely derived from silicate weathering or possibly ion

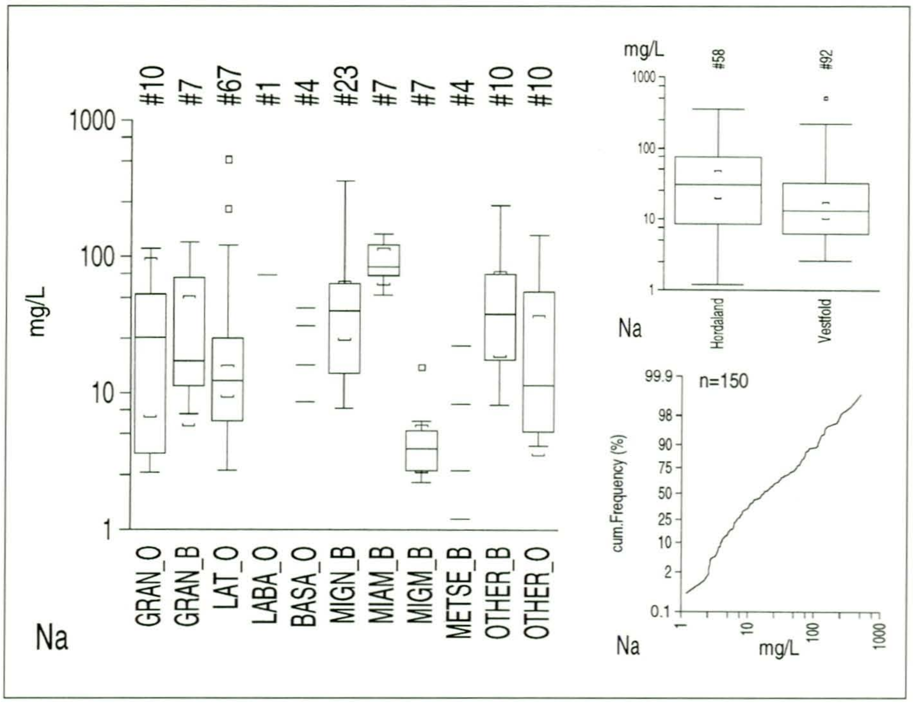

Fig. 12. Diagram showing (note the log scales): left, boxplots for distribution of Na (mg/L) in bedrock groundwaters, according to lithological class (see text for explanation): top right, boxplots for distribution of Na in the Vestfold and Hordaland areas: bottom right, cumulative probability distribution plot of sodium for the entire data set (n = 150). # = total number of samples in boxplot group.

exchange.

Figure 13 shows the distribution of bromide, which element is assumed to be a good indicator of sea salts in groundwater. It will be noted that there is a considerable degree of correlation between Na and Br, indicating the effect of marine influence on Na concentrations. The groups with a (presumably) lithologically-derived sodium-excess are also clearly identified on Fig. 14, which shows the Na/Br mass ratios. This ratio has a value of c. 160 in ideal sea water (Rösler & Lange 1975, Open University 1989).

The range of calcium concentrations extends to higher values in the Vestfold area than in the Hordaland area (Fig. 5). There appears to be a general inverse relation between Na and Ca concentrations. Samples containing more than 60 mg/L Na generally contain less than 30 mg/L Ca. This relationship is further elaborated by the correlation of the Ca/Na ratio with pH (Figs. 4e and 4f). While it will be noted that Ca is dominant over Na in many samples of pH below 8, above pH 8, calcium falls to very low values and the Na/Br ratio increases dramatically (i.e. a lithological source of Na). It will be noted that almost all of the samples of pH greater than 8 are derived from Hordaland, dominantly from the lithological

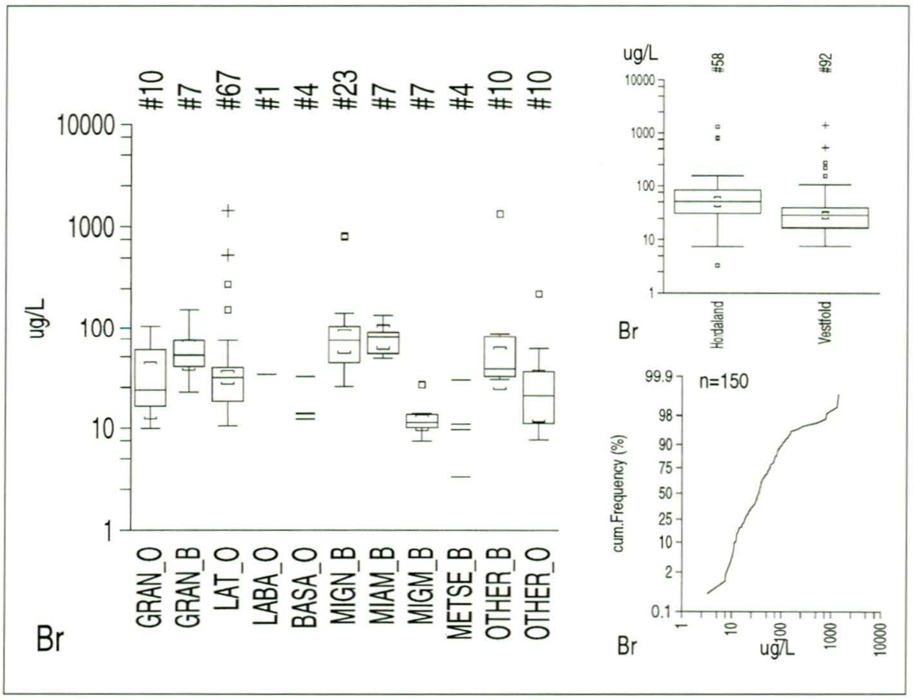

Fig. 13. Diagram showing (note the log scales): left, boxplots for distribution of Br (µg/L) in bedrock groundwaters, according to lithological class (see text for explanation): top right, boxplots for distribution of Br in the Vestfold and Hordaland areas: bottom right, cumulative probability distribution plot of bromine for the entire data set (n = 150). # = total number of samples in boxplot group. In all cases, the figures refer to bromine analyses carried out by BGR in Germany.

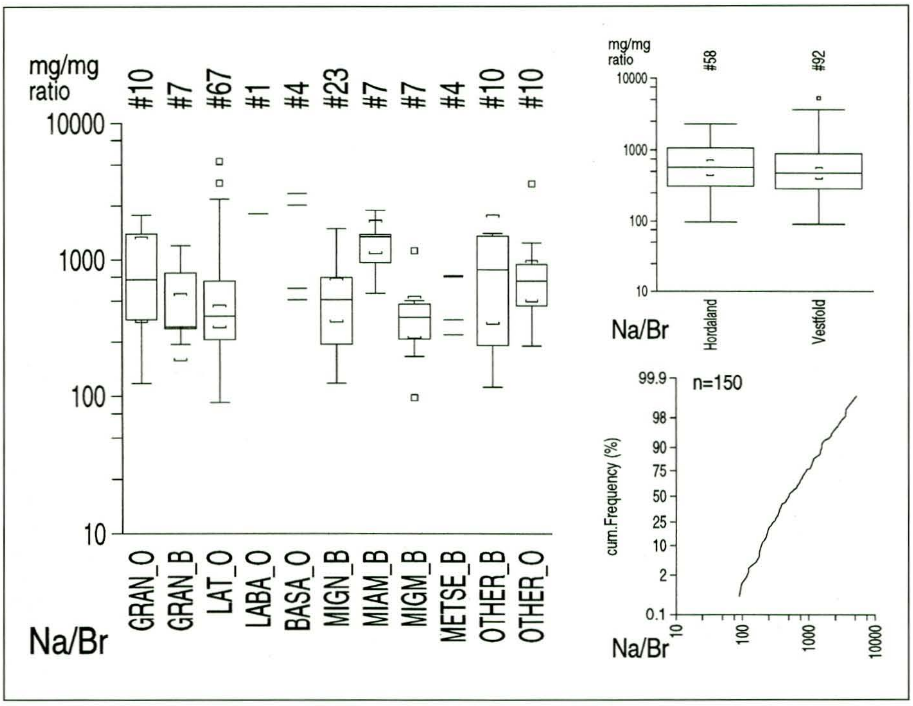

Fig. 14. Diagram showing (note the log scales): left, boxplots for distribution of Na/Br (mg/mg) in bedrock groundwaters, according to lithological class (see text for explanation): top right, boxplots for distribution of Na/Br in the Vestfold and Hordaland areas: bottom right, cumulative probability distribution plot of the sodium to bromine ratio for the entire data set (n = 150). # = total number of samples in boxplot group. In seawater the ratio Na/Br = 161-162 (Rösler & Lange 1975, Open University 1989).

group of «migmatitic gneiss/metagabbro/amphibolite». One might argue that these features merely reflect the petrochemical composition of this lithology, but a low Ca/Na ratio is not directly consistent with such basic lithologies and suggests somewhat more complex processes than congruent weathering reactions, such as ion exchange. The features observed here are reminiscent of those described by, for example, Edmunds (1981) and Banks (in press) in classic sedimentary aquifers of the UK (the Lincolnshire Limestone and the Coal Measures). In these cases, at a given stage in groundwater evolution, reducing conditions commence with a decline in Eh, reduction of sulphate, generation of alkalinity and a slight rise in pH. Commensurate with this is the exchange of Ca for Na, which is possibly related to the presence of reduced sulphide-phase exchange surfaces in the aquifer. The removal of Ca from the aquifer raises the fluorite saturation «ceiling» permitting increased solubility of fluoride. The data from the Hordaland and Vestfold areas are consistent with a similar step in groundwater evolution, although in Norwegian crystalline rocks one cannot relate such hydrochemical evolution to clear flow pathways, as is possible in the well-defined stratabound Lincolnshire Limestone. In the absence of redox indicators (Eh, dissolved oxygen, sulphur and nitrogen species) and alkalinity, however, the model must remain speculative in this case. This should underline the necessity of future groundwater chemical surveys to include analysis of anions and alkalinity in the analytical programme, despite the costs associated with these analyses, if a full understanding of groundwater genesis is to be achieved.

Discussion and conclusion

By examining the contents of several potentially undesirable parameters in Norwegian drinking water derived from bedrock groundwater from the Vestfold and the Hordaland areas, it has been demonstrated that 53% of all waters contravene recognised drinking water limits for one or more of the following parameters: pH, U, Rn, F^- and Na (Table 5). The majority of these contraventions were identified in the Hordaland area (64% contravention), with a rate of only 28% contravention in the Vestfold area.

It is clear from the results of this survey that there is cause for concern over the suitability of some untreated groundwaters from bedrock aquifers as drinking water resources. Concentrations of Rn, U and F^- have been presented which exceed accepted drinking water norms, in some cases very significantly. Other elements with possible health (Be, Al) or aesthetic (Fe, Mn) implications, which occur in concentrations exceeding accepted norms in these waters, are presented and briefly discussed by Reimann et al. (1996). Groundwater, particularly from hard rock aquifers, should not be presented as a universal panacea for municipalities with existing poor quality surface water-based water supplies, as has been the tendency in some quarters. Alternative resources to vulnerable Norwegian surface water intakes should be assessed, including:

a) High altitude lakes
b) Lower altitude deep lakes with well-defined thermoclines and long residence time
c) Bedrock and drift groundwater

A broader survey of groundwater chemistry in bedrock and drift deposits is required in order to satisfactorily assess the last option (c). Nevertheless, it would be wrong to place too much emphasis on the negative aspects of groundwater quality. Studies by Sæther et al. (1995) and Morland et al. (1996) have demonstrated that water quality problems associated with Rn and F^- (and presumably U) are not normally encountered in Quaternary sand and gravel aquifers in Norway, which form the basis for most large-scale waterworks. Morland et al. (1996) demonstrate that the radon activity in groundwater from Quaternary aquifers from 31 large Norwegian waterworks does not exceed 80 Bq/L.

In bedrock groundwater, many geochemical parameters exhibit a heavily skewed, quasi-log-normal distribution. This implies that the health-related parameters tend to only exceed drinking water norms in a minority of groundwaters, which are typically abstracted from relatively well-defined lithologies. In the case of Rn, F^- and U, for example, granitic groundwater can be predicted to carry a greater risk of limit-exceedance than basaltic lithologies. From Table 5, it can be seen that from the areas of Hordaland and Vestfold studied, 53% of boreholes yielded water which did not satisfy at least one of the following criteria: Norwegian drinking water limits for pH, Na, Rn and F^- or the Canadian limit for U. It is almost certainly the case that these results are not representative of Norwegian bedrock groundwater as a whole, as the areas chosen do seem to contain a high proportion of «high-risk» lithologies. For example, a recent study of groundwater from 26 bedrock boreholes in Caledonian metasediments, metavolcanics and Precambrian gneisses in the counties of Nord- and Sør-Trøndelag did not record any radon concentration exceeding the recommended maximum of 500 Bq/L (Doherty 1996).

Finally, it should be remembered that most «problem-parameters» can be treated. For radon, treatment is available for domestic water supplies using several methods, including simple aeration and carbon adsorption. Various methods are assessed by Nazaroff et al. (1988), Kinner et al. (1990) and Boox (1995a,b). Fluoride is more problematic to treat, the most effective, although expensive, technologies being anion exchange and reverse osmosis.

In summary, groundwater in bedrock can continue to be regarded as a potentially attractive alternative water resource in many areas. The risk due to elevated concentrations of health-related parameters can be reduced by the following measures:

- access to a comprehensive database on the occurrence of hydrochemical parameters in bedrock groundwaters of differing lithologies in order to identify «risk-lithologies».
- the application of analytical programs which include the more important health-related parameters (F, Rn, U and Be) that are often not analysed as «standard components» by conventional analytical laboratories.
- the willingness to accept and invest in cost-effective water treatment facilities for «problematic» parameters, rather than to expect the «perfect» resource, where no form of treatment is necessary.

Acknowledgements

The base of this investigation was carried out as a joint project between Norges geologiske undersøkelse, Statens strålevern, Institutt for odontologisk forskning, Geological Survey of Canada and Bundesanstalt für Geowissenschaften und Rohstoffe. We would like to thank A. Bårdsen of the Universtity of Bergen, H. Dalsbø Antonsen of «Næringsmiddeltilsynet i Nordre Vestfold», T.E. Næss of «Miljørettet helsevern for nordre Vestfold», and A.K. Kolstad and B. Lind of Statens strålevern, for their co-operation in this project.

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Manuscript received February 1997; revised manuscript accepted May 1997.