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Radioelement (U,Th,Rn) concentrations in Norwegian bedrock groundwaters.

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RAPPORT

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Tittel:

Radioelement (U,Th,Rn) concentrations in Norwegian bedrock groundwaters.

Konsentrasjoner av radioaktive grunnstoffer (U,Th,Rn) i grunnvann fra fast fjell i Norge.

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Sammendrag:

Twenty-eight samples of groundwater from bedrock boreholes in three Norwegian geological provinces have been taken and analysed for content of ^{222}Rn, U and Th, together with a wide variety of minor and major species. Median values of 290 Bq/l, 7.6 µg/l and 0.02 µg/l were obtained for Rn, U and Th respectively, while maximum values were 8500 Bq/l, 170 µg/l and 2.2 µg/l. Commonly suggested drinking waters limits range from 8 - 1000 Bq/l for radon and 14 to 160 µg/l for uranium. Radioelement content was closely related to lithology, the lowest concentrations being derived from the largely Caledonian rocks of the Trøndelag area, and the highest from the Precambrian Iddefjord Granite of South East Norway (11 boreholes) where median values of 2500 Bq/l, 15 µg/l and 0.38 µg/l respectively were obtained. The Iddefjord Granite is not believed to be unique in Norway in yielding high dissolved radionuclide contents in groundwaters, and several other granitic aquifers warrant further investigation in this respect.

28 grunnvannsprøver fra borehull i fast fjell i tre norske bergrunnsprovinser er analysert for 222-radon, uran og thorium, samt en rekke hoved- og sporelementer. Medianverdier var på 290 Bq/l, 7,6 µg/l og 0,02 µg/l for henholdsvis Rn, U og Th, med maksimumsverdier på 8500 Bq/l, 170 µg/l og 2,2 µg/l. Vanlig foreslåtte drikkevannsgrenser ligger i området 8 - 1000 Bq/l for radon, og 14 - 160 µg/l for uran. Innhold av radioelementer hadde klar sammenheng med litologi, med lavere verdier fra de trønderske kaledonske bergarter og de høyeste fra Iddefjord-granitten på Hvaler, med median-verdier (11 borehull) på 2500 Bq/l, 15 µg/l og 0,38 µg/l for henholdsvis Rn, U og Th. Iddefjord-granitten er sannsynligvis ikke unik med hensyn til radioelementinnhold i grunnvann, og andre granitter (bl.a. Grimstad-, Flå-, Telemarks- og noen Nordlanske granitter) bør prioriteres for videre undersøkelser.

Emneord: Hydrogeologi	Små grunnvannsverk	Grunnvann
Berggrunn	Grunnvannskvalitet	Uran
Thorium	Radon	Fagrapport

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1. INTRODUCTION

Exposure to natural airborne radon (Rn) has been identified as the primary mode of radiation exposure for many populations living in areas underlain by crystalline (particularly acidic igneous) bedrock, or radioelement-rich sedimentary rocks. It is believed to be responsible for 150 to 300 lung cancer cases (10-20 % of the total) annually in Norway, 400 to 1,100 in Sweden and as many as 5,000 to 20,000 deaths each year in the USA (Zikovsky and Chah, 1990). In Scandinavia, radon in houses accounts for about 75% of the total radiation dose (Christensen and others 1990). The problem in Scandinavia is probably particularly acute due to the dominance of crystalline bedrock, the high proportion of time spent indoors and the high degree of house insulation, hindering the escape of radon. The radon dose to a householder thus depends on other factors as well as underlying geology, e.g. construction and insulation of the house, lifestyle, and also water supply. In addition to radon released directly by the underlying ground, it is commonly estimated that the air concentration of radon released by degassing of a domestic water supply is up to 10^{-4} times the radon concentration of the water (Nazaroff and others 1988, Milvy and Cothern 1990); thus use of groundwater containing 1000 Bq/l Rn can contribute 0.1 Bq/l to air concentrations. The USEPA have an action level of 0.15 Bq/l for indoor airborne radon, whilst in Scandinavian a limit of 0.2 Bq/l is commonly used. Although direct exhalation from the bedrock is the greatest contribution to radon in Norwegian buildings, the groundwater contribution to radon in internal air cannot be neglected as insignificant. Indeed, the waterborne radon can be very important in individual cases (Strand and Lind 1992), as is shown in Fig. 1a, illustrating the increase in airborne radon with shower usage in connection with water containing 4300 Bq/l radon.

Several studies (e.g. Mose and others 1990b, Mills 1990) suggest that inhalation of radon is not the only significant pathway for exposure to radon and its daughter isotopes, and that ingestion may also be significant. Mills (1990) estimates that around 5000 cancer deaths in the USA anually may be due to waterborne radon. Of these, over half may be due to ingested radon (e.g. stomach cancer), with around 2000 being lung cancers due to degassing (and subsequent inhalation) of radon released from the water.

It must be noted that disturbingly high concentrations of radioelements are still regarded as being therapeutically desirable in many countries. In the Czech Republic, for example, spas containing up to 8800 Bq/l radon in their groundwater are used for their purported curative properties (Franko and others 1985), and at Fangzi coal field in China, plans are currently underway to exploit radon-containing mine waters in this way (Jianli and others 1993). The authors have yet to find documented evidence of any desirable effects of radon exposure, however, and the consequences of such exposure must continue to be regarded as wholly negative.

2. THE NATURAL OCCURRENCE OF URANIUM, THORIUM AND RADON

Uranium has three isotopes (Table 1) occurring naturally in significant amounts; but over 99 % of the global mass consists of ^{238}U. Although negligible in terms of mass, the shorter lived daughter isotope, ^{234}U is often dominant in groundwater in terms of activity (i.e. Bq) due to preferential alpha recoil (Milvy and Cothern 1990). At Stripa, in Sweden, ^{234}U typically accounted for 3 to 11 times the activity due to ^{238}U in groundwater (Andrews and others 1989), while in bedrock groundwaters around Helsinki, the factor was between 1 and 4 (Asikainen and Kahlos 1979). The activity represented by a given concentration (μg/l) of U in water will depend on the isotopic composition of the dissolved uranium and daughter isotopes, and conversion factors based on equilibrium assumptions can lead to underestimation of activity. It can be shown that 1 μg = 1.2 x 10^{-2} Bq ^{238}U, and, assuming equilibrium, a conversion factor of 1 μg = 2.5 x 10^{-2} Bq [^{234}U + ^{238}U] is thus commonly used (Barnes 1986, Milvy and Cothern 1990).

In terms of mass, naturally occurring thorium consists almost entirely of ^{232}Th, and 1 μg = 4 x 10^{-3} Bq ^{232}Th. It decays, via ^{228}Ra, to the short-lived ^{228}Th (half-life 1.91 yrs), and the activity (Bq) of the latter radionuclide in groundwater may exceed that due to ^{232}Th. The short lived ^{234}Th (half life 24.1 days) is produced by α-decay of ^{238}U, itself decaying rapidly to ^{234}U. ^{230}Th (half life 77,000 yrs) is also part of the ^{238}U decay series.

Radon is a chemically inert gas which occurs in three main forms ^{222}Rn (radon), ^{220}Rn (thoron) and ^{219}Rn (actinon), being the daughter products of ^{226}Ra, $^{228}Ra/^{224}Ra$ and ^{223}Ra respectively. The isotopes are products of the uranium-238, thorium-232 and uranium-235 (U-actinium) decay series, respectively. As ^{222}Rn has a considerably longer half life than the other two isotopes, it is usually the only one significantly present in natural waters, unless unusually high concentrations of thorium are present in the host rock and transport times are very short.

^{222}Rn is derived from the parent radionuclide ^{226}Ra, a long-lived alpha-emitter of half-life 1620 years, and a member of the uranium series. It is thus found in rocks with high uranium content. Radium is an alkaline earth metal (group II), with chemical properties resembling Ca, Ba and Sr. Radon may be produced by *in-situ* radioactive decay of radium in the rock, ejection across the rock-water interface during decay or by decay of dissolved radium. ^{222}Rn, during its half-life of 3.82 days, may travel limited distances (up to tens - hundreds of metres) in the groundwater of a fractured aquifer before decaying to ^{218}Po, ^{214}Pb, ^{214}Bi and ^{214}Po (the so-called radon daughters).

Geologically, U, Th, Ra and Rn tend to occur in many of the same rock types. Both thorium and uranium tend to be concentrated in highly fractionated magmas and hydrothermal solutions and are thus found in acidic igneous rocks (e.g. granites), pegmatites and hydrothermal deposits. Killeen and Heier (1975b) have demonstrated that the central parts of many granite plutons will be more enriched in primary radioelements than the margins. Typical granites will contain 18-20 ppm

thorium and 4-5 ppm uranium in the rockmass, although many granites will be considerably richer (Killeen and Heier 1975b).

In sedimentary rocks and metamorphic rocks, such radioactive elements can be concentrated along fault zones or other discontinuities by hydrothermal activity or where they can bind to organic material in reducing environments, e.g. organic-rich shales, oil reservoirs etc. Another favorable location for uranium enrichment is in the vicinity of unconformities, either within the underlying weathered zone, or in the sediments immediately overlying the peneplain.

The main uranium compound in vein deposits is uranium dioxide (UO_2), being named *uraninite* in its macrocrystalline, and *pitchblende* in its microcrystalline form. Oxidation and uranium loss through decay tend to increase the O/U ratio, so that the minerals often approach a composition symbolised by U_3O_8. In secondary deposits, *coffinite* ($USiO_4.nH_2O$) may also be important. Upon oxidation, e.g. by weathering, somewhat soluble minerals such as *carnotite* (a K-uranyl vanadate), *tyuyamunite* (Ca-uranyl vanadate), *autunite* (Ca-uranyl phosphate) or *rutherfordine* (uranyl carbonate) are formed.

The commonest mineral occurrences of thorium include *monazite* (cerium thorium phosphate), *pilbarite* (thorium lead uranate), *thorite* (thorium silicate) and *thorianite* (thorium uranium oxide).

3. THE HYDROCHEMISTRY OF RADON, URANIUM AND THORIUM

The hydrochemistry of uranium is particularly complex. For a detailed description, the reader is referred to Krauskopf (1979), Garrels and Christ (1965) and Drever (1988). The main features are, however, summarised as follows (from Krauskopf 1979). Uranium can occur in several oxidation states, but only +IV and +VI are important in hydrogeochemistry. The oxidation from uranous state (U^{4+}) to uranyl (UO_2^{2+}) has a redox potential of +0.33 V, placing it in the normal hydrogeochemical range. The uranous ion forms a highly insoluble hydroxide in water, even at low pH values. Upon oxidation, however, the UO_2^{2+} (uranyl) ion (and its hydroxide) is rather soluble. Krauskopf (1979) states that surface waters in contact with uranium minerals will contain a few ppm U, exceptionally up to a few thousand ppm uranium. In summary, therefore, uranium is rather insoluble in reducing environments, but is soluble in oxidising, and particularly acidic, conditions. Uranium (VI) also forms complex ions with hydroxide (in highly alkaline conditions), carbonate, phosphate and maybe even chloride (Nguyen-Trung and others 1991), resulting in elevated solubility (Drever 1988). It may also form soluble complexes with organic/humic species (Higgo and others 1989), and other species such as fluoride and sulphide can be important. Uranium displays considerable chemical similarity to vanadium, forms a number of combined minerals and is insoluble in any water containing >100 μg/l vanadium (Drever 1988). Ingested uranium primarily affects bone and kidney. As the specific radioactivity of uranium is relatively low, it is

thought that the chemical and physiological toxicity of the element may outweigh the radiotoxic effects (Milvy and Cothern 1990)

In contrast to uranium, thorium is highly insoluble under all conditions. It exists almost exclusively in an oxidation state of +IV.

Radon is a chemically inert, but soluble gas. Having such a simple chemistry (in contrast to U and Th) its concentration in groundwater is believed to be directly controlled by (i) hydrodynamic factors and (ii) uranium (or, more precisely, radium) content of rocks in the vicinity of the well (Michel 1990). Like many other solutes, its concentration is commonly found to be lognormally distributed in groundwater (Zikovsky and Chah 1990). Snihs (1973) indicates that groundwater in equilibrium with typical granite with $^{226}Ra = 10^{-12}g/g$ should not exceed c. 22 Bq/l Rn. Levels higher than this are indicative of U or Ra enrichment. Radon concentrations may vary with meteorological factors such as atmospheric pressure, heavy rainfall, snowmelt or frost cover events. Radon concentrations have been used as a diagnostic tool for predicting earthquake events, locating fracture zones and even estimating fracture apertures (Nelson and others 1983).

4. RADIOELEMENTS IN GROUNDWATER

Much literature exists on the topic of radioelements in groundwater, both from studies in connection with mobility of radioelements from nuclear waste repositories and from studies examining the health-effects of radionuclides in drinking water. The latter studies have often been performed by non-hydrogeologists and have not tended to emphasise the geological/geochemical controls on radioelement concentrations.

The investigation by Snihs (1973) of the Vimmerby area of Sweden, indicated that only boreholes in granitic bedrock yielded water with over 740 Bq/l, with a maximum of 2660 Bq/l ^{222}Rn. Snihs uses the relationship 3,700 Bq ^{222}Rn/l = maximum permissible concentration for ingestion, which is equivalent to a dose of 0.15 Sv with the gastrointestinal tract as the critical organ. He discovered that the dose from ^{222}Rn itself far exceeds the dose from its daughter radionuclide ^{210}Pb.

The Geological Survey of Sweden has published hydrogeological maps and descriptions of many of the Swedish counties. The typical concentration of ^{222}Rn in crystalline bedrock groundwater ranges from 0 - 500 Bq/l (Pousette and others 1989), but the maximum reported thus far in the published map descriptions is 3400 Bq/l.

Throughout the 1970s and 1980s much mapping of radon and uranium in groundwater has been performed in Finland. A sample of 2065 bedrock boreholes, largely in southern Finland gave a geometric mean (GM) of 240, an arithmetic mean (AM) of 1,020 and a maximum of 77,000 Bq/l ^{222}Rn (Salonen 1988). The corresponding figures for 961 wells and springs in Quaternary deposits

were 31, 92 and 3800 Bq/l. The highest values of U and Rn clearly come from pegmatite-rich granites and migmatites in the south of the country. Lahermo and Juntunen (1991) detail the results of several surveys of uranium in groundwater. In most surveys, the median values are below 1 μg/l, but a study 1388 bedrock boreholes in southern Finland yielded median and AM of 5 and 73 μg/l respectively. Samples with 100 - 1000 μg/l are known from uraniferous granites, while in the Helsinki area, extreme values of up to 14.9 mg/l are recorded (Asikainen and Kahlos 1979). The Finnish Geological Survey have compiled hydrogeochemical maps from the results of these surveys, and statistically analysed the data, finding only a low correlation between ^{222}Rn and uranium in bedrock groundwater, but a moderate correlation between uranium and bicarbonate (Lahermo and Juntunen 1991).

In Denmark, 14 drilled wells in Mesozoic-Quaternary sediments all yielded water with < 1 Bq/l ^{222}Rn. On the Danish island of Bornholm, with more ancient and complex bedrock geology, 49 out of 54 samples gave concentrations over 1 Bq/l, with a maximum of 1070 Bq/l from a granitic lithology (Ulbak and Klinder 1984).

Several detailed and extensive surveys of radionuclides in groundwater have been carried out in USA. The results are summarised in tables 2 and 3, and by Barnes (1986). The granites of Maine are found to be contain among the most radon-rich groundwaters (Table 3 - Michel 1990). Milvy and Cothern (1990) clearly conclude that the threat due to radon heavily outweighs that from U and Ra, being responsible for around 80 % of radionuclide-induced deaths from drinking water. ^{232}Th has been detected in American groundwaters, but rarely exceeds 0.004 Bq/l, with a maximum in drinking water of 0.0004 Bq/l. The uranium duaghter ^{230}Th can be found in higher concentrations (up to 0.015 Bq/l) around uranium mineralisations in New Mexico, with a maximum of 0.0015 Bq/l in drinking water (Barnes 1986).

4.1 Previous studies in Norway

Few studies of radioelements in groundwater have been carried out in Norway. However, two systematic studies of radon in Norwegian houses have been performed (Strand and others 1988, 1991, 1992). In the first study, 1600 houses were monitored, indicating an average radon concentration of 0.1 Bq/l in air (although this may be slightly exaggerated due to overrepresentation of houses on the uranium-rich Alun Shale bedrock), representing a dose equivalent of c. 4mSv/yr. The highest values appeared to be concentrated in the area around the Oslo graben, although this area was also overrepresented in the measurements. The study can also be criticised due to the short integration time of the measurements. In the second study, the defects due to sampling bias and short integration time for measurement were corrected. 7525 houses, distributed throughout Norway in proportion to population (Fig. 1b), were monitored, yielding an annual mean household air concentration of 0.06-0.07 Bq/l.

Strand and Lind (1992) have also carried out a survey of radon and radium Norwegian tap water from drilled deep (70-100 m) deep boreholes. A sample of 229 boreholes yielded a mean ^{222}Rn content of 250 Bq/l. The highest values were obtained from granites, typically in the southeast of the country, with a mean of 1070 Bq/l and a range of 130 to 7000 Bq/l. Other geological formations yielded a range between < 5 to 1250 Bq/l.

A limited survey of uranium content in stream sediments, water and moss, together with radon content in drinking water, was carried out in the vicinity of the Fen carbonatite complex, near Oslo, Norway (Ryghaug 1984). Streamwater often contained over 10 ppb uranium and 1.2 - 22 Bq/l radon. Groundwater from wells, mostly in Quaternary deposits, contained 51 - 1100 Bq/l radon.

Arne Grønlie (1983) and colleagues have investigated the radon content of both groundwater and surface water in the vicinity of the Th- (and to a lesser extent, U-) bearing Leksvik breccia zone in Nord-Trøndelag. The highest concentration measured was 247 Bq/l in a bedrock borehole believed to be the same as our borehole 22 (Table 4). Surface waters contained typically less than 3 Bq/l radon, but ranged up to 20 Bq/l.

5. THE STUDY AREAS

The Geological Survey of Norway has initiated a pilot study to establish whether radon, uranium or thorium levels in groundwater represent a health problem in Norway, and to discover potential correlations between lithology and concentration. The scope of the project has not allowed a strictly geographically or epidemiologically representative sample set to be collected. Such a sample set must await a "phase II" of this project.

Two study areas were chosen, the following criteria being used: (i) ease of accessibility and existing groundwater projects in progress (ii) lithological variation (iii) presence of lithologies suspected to be "high risk". The county north of Trondheim (Nord-Trøndelag) and the area around Oslofjord were chosen.

5.1 Nord-Trøndelag

A simplified geological map of the county of Nord-Trøndelag is shown in Fig. 2. The area can be subdivided very coarsly into three:

1) The Caledonian mountain belt - consisting of a sequence of nappe-piles of gneisses, metasediments and metavolcanics of Precambrian to Lower Palaeozoic age.

2) The Proterozoic (so-called "basement") gneisses occurring west of the Caledonian belt. Although sometimes considered as parautochthonous, these are often also regarded as belonging to the lower allochthons of the nappe sequence. The gneisses north of Namsos are richer in uranium than those to the south (Grønlie and Staw 1987).

3) Windows of Proterozoic "basement" gneisses and metasediments (e.g. the Tømmerås Window)

A dominant tectonic feature is the Møre and Trøndelag fault zone, which largely controls the "grain" of Trondheimsfjord. In several localities in inner Trondheimsfjord, narrow hydrothermal breccia zones containing thorium (and some uranium) occur. The rock-material from the Leksvik breccia zone contains an average 990 ppm Th and 49 ppm U (Grønlie and Staw 1987).

5.2 Oslo Region

The region (Fig. 3) is tectonically dominated by the Oslo rift, of Carboniferous-Permian age. Within the rift can be found sedimentary rocks of Precambrian to Silurian age, including the uranium-rich (10-170 ppm U according to Skjeseth, 1958) Alun Shales. These are overlain by volcanics and sediments of Carboniferous-Permian age, and intruded by igneous rocks of largely Permian age. To the east and the west of the Oslo rift can be found autochthonous Precambrian basement, consisting of gneisses and granites dating from the Sveconorvegian orogeny or earlier. In the extreme southeast of the area is the Precambrian Iddefjord granite, the Norwegian extension of the major Swedish Bohus granite batholith.

The Iddefjord/Bohus granite is one of the "hottest" areas in Scandinavia, with a heat flow density of over 80 mW/m^2 (Čermák and others 1992). Killeen and Heier (1975a) described it as the southern part of a belt of southern Norwegian granites containing elevated contents of uranium and thorium. In the Iddefjord granite, they recorded average contents of 9.9 ppm U and 50 ppm Th, with maxima exceeding 30 and 70 ppm respectively. It appears to be particularly enriched in uranium on its eastern side. Radioelements in groundwater in this area may be derived from radioactive elements dispersed in the granite's groundmass, or may be derived from occurrences of specific minerals in pegmatite dykes. Several occurrences of uranium and thorium minerals in pegmatites in the Iddefjord granite are reported by Bjørlykke (1939) including uranium (IV) oxide, thorite, samarskite (up to 15 % uranium oxides), monazite (up to 19 % thorium oxide) and xenotime. The hydrogeology and hydrochemistry of the Iddefjord granite are relatively well known from the Hvaler group of islands and are described in papers by Banks and others (1993a,b). Samarskite, monazite and xenotime are specifically reported from Hvaler (Bjørlykke 1939). For these reasons, the Hvaler islands were focussed on during the sampling programme.

6. METHODS AND EQUIPMENT

Bedrock boreholes or wells were chosen (Table 4) with emphasis being given to the following criteria:

(i) borehole should be in regular use or should be naturally overflowing, such that "fresh" groundwater is sampled.

(ii) borehole should not be newly drilled. Investigations (Banks and others 1993b) have indicated that newly exposed rock surfaces and drilling cuttings can substantially affect water chemistry.

(iii) borehole should give low possibility for degassing, i.e. sampling points at borehole head, or sampling points which are part of a closed system (e.g. pressure tank) were preferred.

(iv) the water should not contain particulate matter or humus.

In practice, however, some boreholes did not satisfy all criteria (i.e. minor infringements of (iii) and (iv)). In particular sample 13 came from a relatively newly drilled (7 month old) borehole which still contained a sufficient particulate load to prevent filtering and field acidification. All results quoted below for this borehole refer thus to the unfiltered sample. Sample 12 came from a borehole with a permanent problem of particulate and humic matter in the water, but which was able to be filtered in the field.

Sampling took place in autumn 1992 and winter 1992-93. Prior to sampling, the tap was allowed to run for at least 5 minutes. All polythene flasks were rinsed thoroughly three times with groundwater, and twice with filtered (0.45 μm Millipore filter) water before sampling.

The following samples were then taken in polythene bottles with screw caps.
(a) 2 x 100 ml unfiltered, unacidified
(b) 2 x 100 ml filtered (0.45 μm Millipore filter and polythene syringe) and acidified (10 drops 65% Ultrapur nitric acid) in the field.
(c) 1 x 500 ml unfiltered, unacidified

One quantum of sample (a) was analysed at the Geological Survey of Norway (NGU) for 7 anions (F^-, PO_4^{3-}, Br^-, Cl^-, $SO_4^{=}$, NO_3^- and NO_2^-) by ion chromatography. One quantum of sample (b) was analysed by inductively coupled plasma emission spectroscopy at NGU for Si, Al, Fe, Ti, Mg, Ca, Na, K, Mn, P, Cu, Zn, Pb, Ni, Co, V, Mo, Cd, Cr, Ba, Sr, Zr, Ag, B, Be, Li, Sc, Ce, La and Y (although many of these have inappropriately high detection limits).

The second quanta of samples (a) and (b) were analysed at the Norwegian Institute for Air Research (NILU) by ICP Mass Spectrometry for Pb, Cd, Cu, Zn, Cr, Ni, Co, Fe, Mn, V, As, Ba, Sr, Al,

Sb, Bi, Tl, U, Th, Be, Li, Rb, Cs, Mg, Ca, Mo, Y and La. The standard method used at NILU was employed - 10 ml of each sample was taken, and 100µl 1% HNO_3 was added. 50 µg/l Sc, Re and In were also added to the sample as an internal standard. The ICPMS instrument is calibrated against reference standards provided by SPEX Industries, and calibration allows a maximum of 10% deviation at a concentration of 10 µg/l. Brackish/saline water samples can lead to greater errors, and Cr, V, Fe and Ni are amongst the most problematic elements in such a situation.

The 500 ml sample (c) was used for laboratory determinations at NGU of pH, electrical conductivity and alkalinity. Standard methods employed at NGU are described by Ødegård and Andreassen (1987).

For sampling of radon, a plastic funnel was inserted below the running sampling 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 ampule of scintillation liquid was then sealed and shaken. The liquid gelifies on contact with water, immobilising the radon. Flasks were delivered to the Radiation Protection Authority (NRPA) within 3 days and analysed using an LKB Wallac 1215 scintillation counter, calibrated using a standard radium solution. Results were back adjusted for radioactive decay to give a radon concentation in Bq/l at time of sampling. The overall uncertainty in the method is estimated to be around 20 % at the 95 % confidence level, and the lower limit of detection is 1 Bq/l.

7. RESULTS

The duplicate analyses made on many elements at NGU and NILU, and the analyses at NILU on field-acidified and field-unacidified samples allowed independent control of many parameters. In the case of most parameters (including U, Th), field acidification did not dramatically affect the analysis outcome (Fig. 4), indicating that unacidified, unfiltered samples can give satisfactory analystical results for U and Th, at least for relatively "clear" groundwater samples. Some discrepancies were discovered between NGU's and NILU's analytical results, particularly a few elements which are rather sensitive to pH and/or redox conditions (and thus to filtering and storage), such as Fe and Al, and a few elements which are known to be problematic for ICP-MS techniques in rather brackish groundwaters, such as Cr, Fe and V. Further discussion of analystical comparisons can be found in Banks and other (1993c).

In the following analysis, NGU's values for major elements (Na, K, Ca, Mg and elements not analysed by NILU, such as Zr) are used, while NILU's results for field acidified samples are used for trace elements (including U and Th). Analysis results below the detection limit were set to half the detection limit for the purposes of statistical analysis.

The results of the U, Rn, Th, Cl, and Na analyses for each borehole are given in Table 4. Cl can be regarded as an indicator of marine influence, and the parameter Na - Cl as a coarse indicator of water-rock interaction (Banks and others 1993b).

8. CORRELATION STUDIES

In order to interpret the data, the wells have been divided into 8 lithological groups as follows:

1 = Quaternary sediments
2 = rhomb porphyry basalts
3 = metadiorite
4 = metasediments (schists, meta-arkose)
5 = granodiorite gneiss
6 = Precambrian gneiss (Trøndelag)
7 = Precambrian gneiss (Oslo)
8 = Iddefjord granite.

The lithologies are chosen such that they exhibit a general increase in acidity and thus in expected incompatible element content. The wells are also divided up into three geographical groups: (a) = Hvaler, (b) = Oslo region (excluding Hvaler) and (c) = Trøndelag. All major and trace elements have been examined using box plots (Fig. 5) for each of the three geographical groups, and it is revealed that many elements, including U, Th and Rn, show considerably higher concentrations in the Hvaler area than otherwise in the Oslo area, with the Trøndelag area showing the lowest concentrations (Table 5). Other parameters which follow this pattern are Si, Al, Fe, Na, Cd, Cu, Zn, Cr, V, F, Cl, $SO_4^{=}$, Y, Be and Mo. Parameters such as Bi, La, E.C., Tl, Zr, Co, Pb and B show elevated levels for the Hvaler area, but little significant difference between Oslo and Trøndelag. Elements which show the opposite trend, with highest concentrations in the apparently more calcareous geological environment of Trøndelag, include Ca, Mg, Sr, Rb, Cs, pH and alkalinity. Plots of element concentrations versus lithology (Fig. 6) tend to confirm these trends with, for example, higher concentrations of many trace metals in the Iddefjord granite. Details of the interpretation of trace metal chemistry are published in a separate report (Banks and others 1993c), while the analytical data are documented in Banks (1993).

What such analyses of lithological and geographical trends do not reveal, however, are the hydrogeological reasons for elevated concentrations of many elements in the Hvaler/Iddefjord granite area. Although none of the geographical areas are located far from the coast, the Trøndelag wells are least marine dominated and Hvaler (being an island group) is most coastal in character. Chloride concentrations, for example, are not related to lithology (as demonstrated in Banks and others 1993b) but are marine-related. Species such as sulphate will be largely marine-related, but also be influenced by elevated levels of anthropogenic fallout in southeast Norway as compared to

Trøndelag. There are thus at least three separate controlling trends from Trøndelag through Oslofjord region to Hvaler:

(i) Increasing bedrock "acidity" i.e. geological occurrence of incompatible elements
(ii) Increasing marine influence
(iii) Increasing fallout from atmospheric contamination

This paper is primarily concerned with identifying the causes of elevated thorium, uranium and radon levels in groundwater. Any correlation between these radioelements and other elements or lithology may be:

(i) purely coincidental (i.e. non-geological), such as that between chloride and lithology
(ii) reflecting a covariation in element concentrations with respect to geological environment - covariations between "incompatible" late-melt elements in acidic rocks.
(iii) reflect a direct causitive relationship, e.g. expected relationships between U and Rn (the one being derived from the other) or between HCO_3^- and U (if the one complexes with and mobilises the other).

Correlation matrices have been produced between all elements, for the entire data set (N = 30) and for purely the Iddefjord granite lithology (N = 11), for both untransformed and log-transformed data. Correlation coefficients over 0.5 were obtained as shown in Table 6. Generally speaking, U, Th and Rn showed only rather weak correlations with other elements. The strongest, most persistent correlations were found between Th and Bi, La and Y (the latter two being chemically analogous to the actinides). Weaker, but persistent, correlations were found between Rn and F, Zr and B. Uranium showed moderate correlations with Mo, As and Sb. These correlations probably reflect covarying degrees of enrichment in the host rocks (i.e. type ii above). No significant or persistent correlations were found between the uranium and major parameters which might be expected to affect mobility, such as alkalinity, Cl, V or pH (i.e. type iii above).

Surprisingly, only rather weak correlations between uranium, radon and thorium themselves were found (in accordance with findings of Lahermo and Juntunen 1991), and these correlations only occurred within the entire data set, reflecting the coarse-scale covariation in host rock contents of these elements (Fig. 7a). Within a single lithology (the Iddefjord Granite), these correlations disappear (Fig. 7b, Table 7), indicating that the hydrochemistry is dominated by hydrodynamic factors, redox- and pH conditions and interaction between various dissolved species, rather than purely by lithological content of uranium and thorium. Asikainen and Kahlos (1979) also note a similar local lack of correlation between U and Rn in bedrock groundwater.

9. DISCUSSION

Maximum concentration levels (MCL) for U, Th and Rn drinking water have not been agreed upon in most countries. Suggestion and tentative limits are summarised in Table 8.

As regards radon, and taking the Swedish levels of 100 Bq/l in groundwater for possible adverse effects, and 1000 Bq/l as the level for recommended remedial action (Table 8), the following comments can be made. The two Quaternary wells sampled give no grounds for concern over radon levels. Of the ten bedrock boreholes in Nord-Trøndelag, six exceed the 100 Bq/l boundary, although there is no clear lithological correlation. The highest value is 240 Bq/l, and is derived from a borehole believed to be associated with a Th- (and U-) enriched breccia zone. Previous measurements, using different sampling techniques (degassing of water) on this borehole yielded similar results (247 Bq/l - Grønlie 1983). The bedrock of Nord-Trøndelag thus appears to yield groundwaters with inferior Rn levels compared with the Oslofjord region. This probably reflects the rather calcareous and uranium-poor nature of the Caledonian metasediments, and the uranium-poor nature of the basement gneisses in the area south of Namsos (Grønlie and Staw 1987).

Boreholes in the gneissic and rhomb-porphyry rocks of Oslofjord gave radon levels all exceeding 100 Bq/l, and approaching 1000 Bq/l in some cases. The gneisses typically yielded higher concentrations of both radon and uranium than the rhomb-porphyries. No samples were, however, taken from either the Palaeozoic metasediments (including the Alun Shales) or from Permian intrusives (such as the Drammen granite). This must be regarded as a weakness of the current pilot study. In the Iddefjord granite of the Hvaler area, the majority of the samples exceeded 1000 Bq/l radon, reaching a maximum of 8500 Bq/l. These values must be regarded as high, in the context of proposed domestic water limits/action levels ranging from the USEPA's 11 Bq/l to the Swedish 100 Bq/l (Table 8). On Hvaler itself, many, but not all, of the users are holidaymakers, using the supplies for only a few weeks per year, thus lessening any health impact. On the mainland area of the Granite, both in Sweden and Norway, many permanent inhabitants obtain groundwater from bedrock boreholes, and the consequences of the use of such boreholes should be evaluated. Across the border, in Sweden, 50 % of all boreholes in Bohuslän are reported to yield groundwater with $>$ 1000 Bq/l radon, with a maximum of $>$ 30,000 Bq/l in a borehole penetrating a uranium-rich pegmatite (Gustav Åkerblom, SSI, Sweden, pers. comm. to Knut Ellingsen, 1993).

The health related impact of uranium and thorium in the groundwaters is far more difficult to judge. According to some tentative standards, levels of uranium higher than between 14 and 160 μg/l in drinking water can exceed acceptable limits of intake, and certainly the two maximum levels of 150 and 170 μg/l, from boreholes in the Iddefjord granite, are regarded as being undesirable. The remainder of the samples lie under c.40 μg/l. As regards thorium, four boreholes on Hvaler yield water exceeding 1 μg/l. As Th is generally regarded as being more radiotoxic than uranium, such concentrations cannot necessarily be disregarded as negligible in a health-related context. Although the distinctions between the Oslo rift and Trøndelag regions are less clear for uranium

and thorium than for radon, the groundwaters from Hvaler clearly exhibit the highest levels of both uranium and thorium.

It has long been known that the Iddefjord/Bohus granite contains anomalously high concentrations of uranium and thorium, but it is interesting to note that Kileen and Heier (1975a,b) regard it as only one of several anomalously radioactive granites of similar age in southern Norway (e.g. the Telemark suites, some of which contain up to 60 ppm Th and 13 ppm U, and the Flå granite) which might warrant further investigation as regards groundwater. Lindahl (1983) regards the southern Norwegian granites as inferior in U and Th content to several granites in the northern county of Nordland (up to 50 ppm U). It would appear that a wider survey of the possible health implications of radioelements in groundwater from Norwegian granites is long overdue. This survey should be based upon already-existing geochemical and radiometrical studies of Norwegian bedrock.

10. TREATMENT OF WATER FOR RADIONUCLIDES

Although the results of this survey do indicate that Rn and U occur in unacceptably high concentrations in some Norwegian bedrock groundwaters, this should not necessarily be seen as a major drawback to the exploitation of this resource. Investigations have indicated that several methods exist which can effectively treat small scale water supply sources for these parameters. For radon, the following three methods (together with capital costs in the USA in 1988) are among the most appropriate, all giving removal rates of above 95 % (Nazaroff and others 1988, Kinner and others 1990):

(i) Granular activated carbon adsorption ($ 850 pr. household). The Swedes have rather more negative results using this method, namely c.50 % removal of radon, up to 1000 Bq/l (Gustav Åkerblom, Statens Strålskyddsinstitut - pers. comm. to K.Ellingsen, 1993).

(ii) Diffused bubble aeration ($ 2000 pr. household)

(iii) Packed tower aeration

Other methods could include:

(iv) Modification of external storage reservoir and distribution system to give increased degassing area and residence time.

(v) *In-situ* aeration in the borehole, although Swedish experiences have proved rather negative (Gustav Åkerblom, SSI, pers. comm. to K. Ellingsen, 1993).

Of these, methods (iii) and (iv) are probably more appropriate for somewhat larger waterworks. Users of high-Rn groundwater should also be encouraged to ensure effective ventilation of rooms where water is used.

As regards uranium, methods which have been shown to be effective to over 90% removal include coagulation/filtration, lime addition, anion exchange, reverse osmosis and, under certain circumstances (low pH) cation exchange. Activated carbon adsorption may also represent a possible treatment.

12. CONCLUSIONS

Parallel sampling (with and without field filtration and acidification) indicates that field filtration and acidification are not critical for obtaining reproducable results when analysing uranium and thorium in "clean" (i.e. non-humic, non-turbid) groundwater by ICPMS. Particularly reproducible results are obtained for uranium.

This study indicates that concentrations of the radioelements uranium, thorium and radon in bedrock groundwater can be correlated with geological provinces and with lithology. The Sveconorwegian Iddefjord granite yields groundwater with considerably higher concentrations of all these elements than the nearby Precambrian gneisses and Permian lavas of Oslofjord, which in turn are enriched as compared with the rocks of the Caledonian orogenic belt in Nord-Trøndelag. Two "control" samples of groundwater from Quaternary deposits in these areas yielded very modest radioelement concentrations.

Weaknesses in the pilot study can be summarised as follows:

(i) the sampling programme was not adequately large or well-designed to allow epidemiological analysis
(ii) the sampling programme was insufficiently large to be representative for some lithologies (e.g. Quaternary, Precambrian gneisses in Trøndelag)
(iii) Several important lithologies were not represented in the Oslo area, notably the Permian intrusives and the Oslo graben sedimentary sequence.
(iv) Radium content was not analysed.

Concentrations of radon in at least some groundwaters from all three bedrock provinces exceed 100 Bq/l, the Swedish lower regulatory limit. Thus, it appears that groundwater from most bedrock lithologies can represent a potential health problem under adverse circumstances (groundwater not aerated prior to use, poor ventilation in house etc.). In groundwater from the Iddefjord Granite, radon exceeded 1000 Bq/l (the Swedish limit for remedial action) in 8 of 11 samples. A similar pattern is observed for the various geological provinces and lithologies as regards uranium and

thorium, the highest values clearly being observed in the Iddefjord granite. The two highest concentrations of uranium exceed most of the commonly proposed MCLs. Surprisingly, however, the correlations between U, Th and Rn themselves are rather weak, particularly within one lithology, indicating that hydrodynamic factors, complexing, pH- and redox-conditions and solution/recoil phenomena are the major controlling factors for radioelement concentration, often masking the effect of mere radioelement concentrations in the bedrock.

The groundwaters observed to have elevated concentrations of U and Rn should be treatable using "domestic" technologies such as aeration, ion exchange or active carbon adsorption.

The Iddefjord Granite is by no means alone in being enriched in U and Th in Norway; other granites which may be enriched in radioelements include many of the Telemark suite, the Flå granite, the Grimstad granite and certain older granites in Nordland county. The pilot study clearly demonstrates the need for a wider survey of radioelements in bedrock groundwater. Such a study should be designed so as to allow an epidemiological analysis of cancer occurrence, and possible correlations with:

(i) Groundwater usage in the home
(ii) Radioelement concentrations in groundwater
(iii) Geology and tectonic situation

13. TECHNICAL NOTE - TERMINOLOGY AND UNITS

The terminology and units used in the study of radioactivity can be confusing. There are three main ways of quantifying radioactivity and its effect on human beings:

♦ Activity: the number of disintegrations per second (i.e. the "amount" of radioactivity) is best measured by the unit Bequerel (Bq), and depends upon the type of radionuclide and the quantity in question. Older units are the Curie (Ci) and the Mache Unit. 1 Bq = 1 disintegration per second. 1 Ci = 3.7×10^{10} Bq. 1 Mache Unit = 3.6×10^{-10} Ci/l = 13.3 Bq/l.

♦ Radiation dose: radiation energy absorbed per unit mass is measured in Grays (Gy). This will depend on the type of radiation and its energy (and thus the radionuclide), distance from source and properties of the absorbing material. 1 Gy = 1 J/Kg = 100 rad.

♦ Effective dose depends on the radionuclide (i.e. the type and energy of radiation), the exposure pathway (direct, breathing, ingestion etc.) and the organ of the body in question. The unit of effective dose is the Sievert (Sv). 1 Sv = 100 rem.

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Figures

Black triangles show detection limits, where relevant, on Figs. 4 - 8.

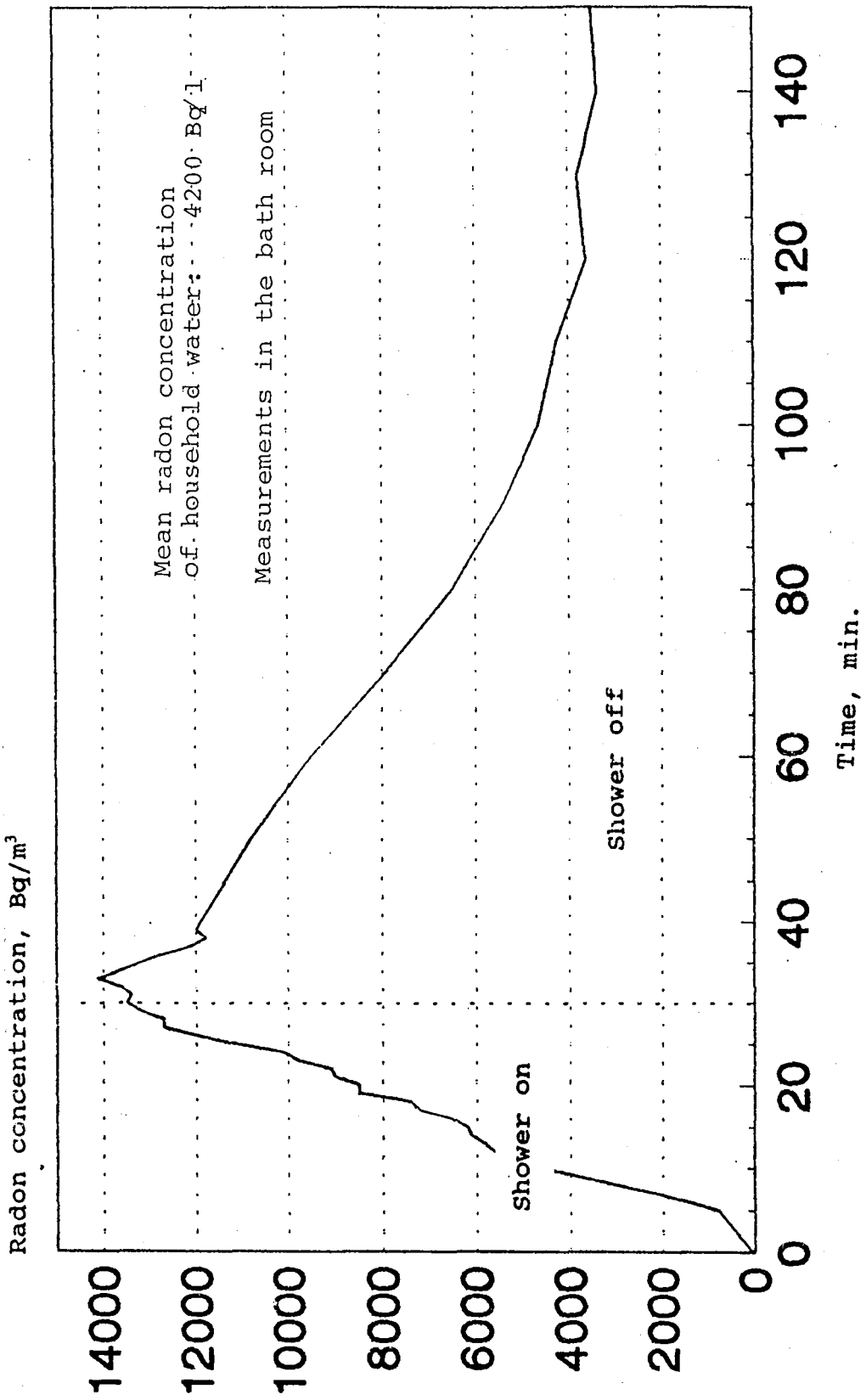


Fig. 1a. Effect of using a shower (water containing 4300 Bq/l) on airborne concentrations of radon in the bathroom.

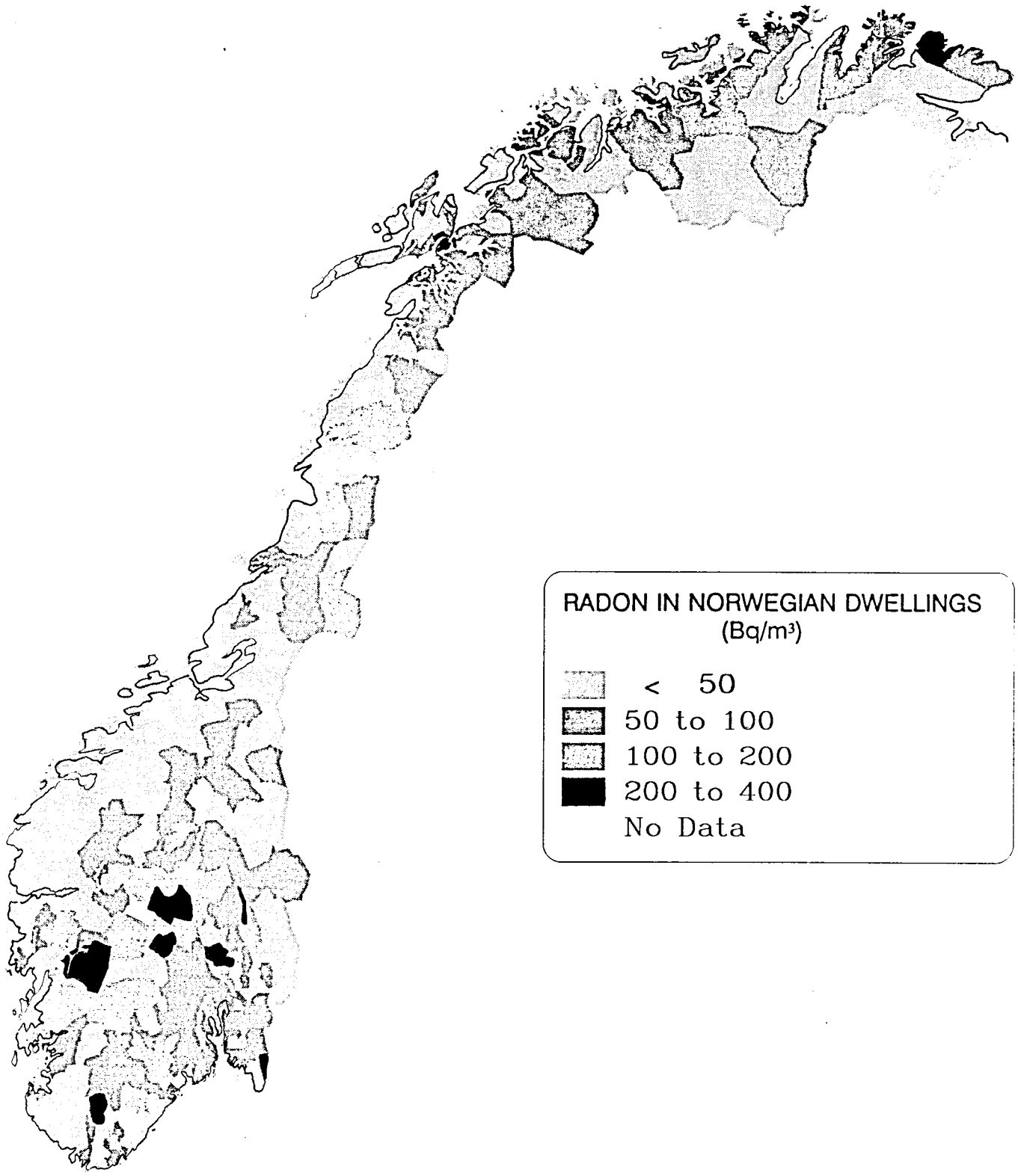


Fig. 1b. National survey of radon in household air, showing geographical distribution of average radon concentration.

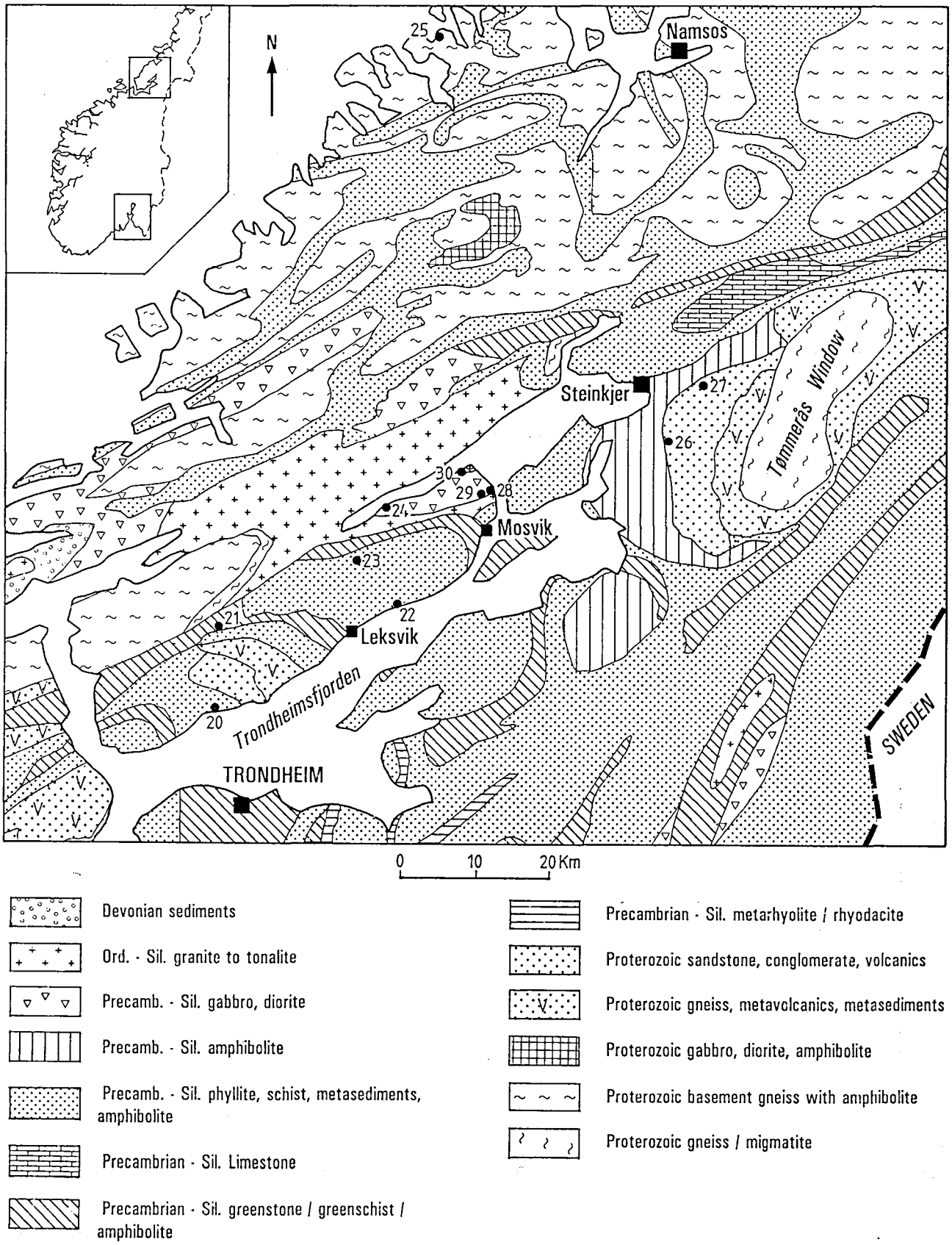
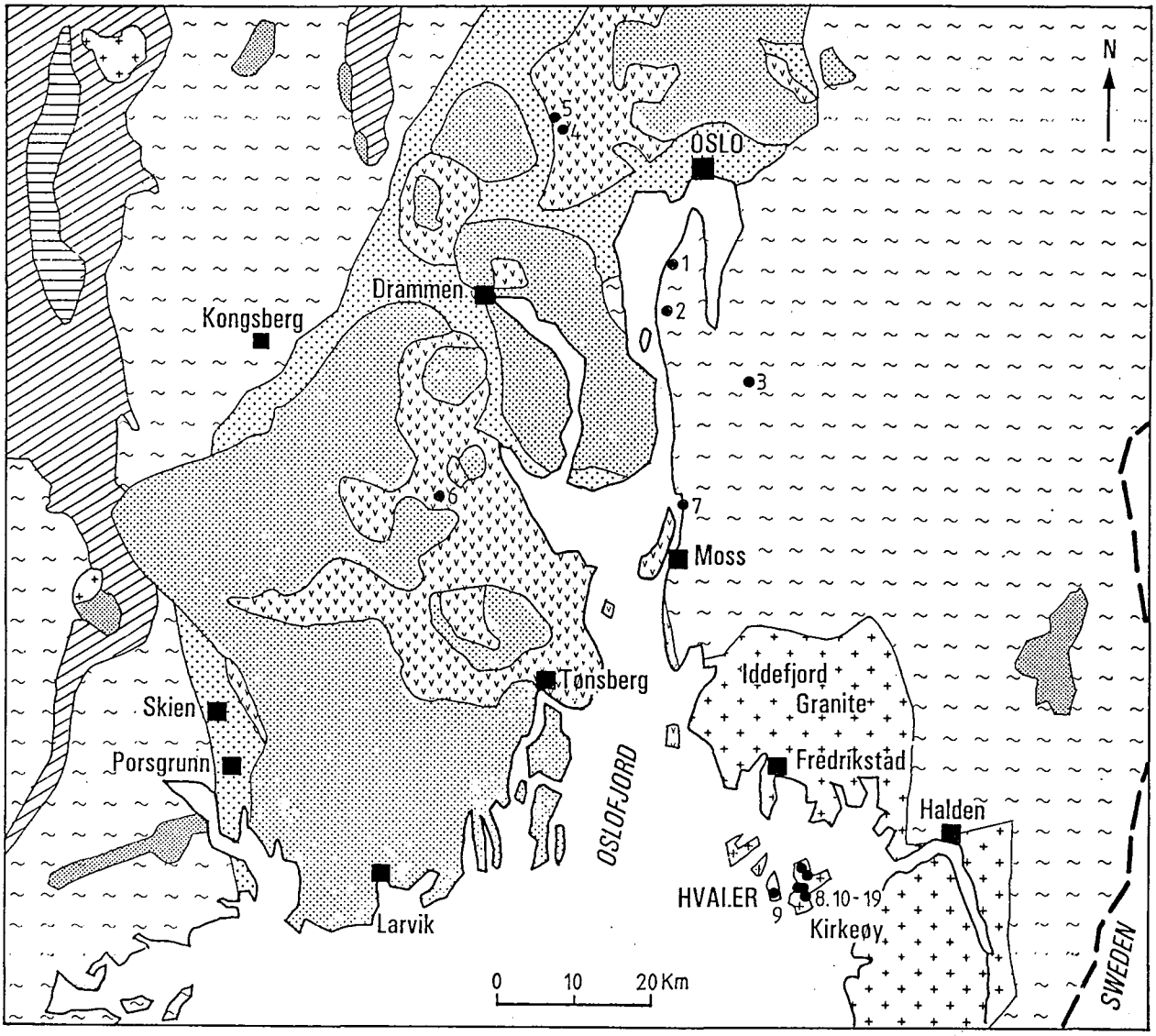


Fig. 2. Simplified geological map of the Trøndelag area showing sample locations (numbers refer to Table 4). Inset shows location of Figs. 2 and 3 on map of Southern Norway.



Permian intrusives

Carb. - Perm. volcanics with sediments

Precambrian - Silurian metasediments

Proterozoic granite / tonalite

Proterozoic mafic / ultramafic intrusives

Proterozoic metasediments / volcanics (sandstone, mica schist, gneiss)

Proterozoic metarhyolite / rhyodacite

Proterozoic gneiss

Fig. 3. Simplified geological map of the Oslofjord area showing sample locations (numbers refer to Table 4).

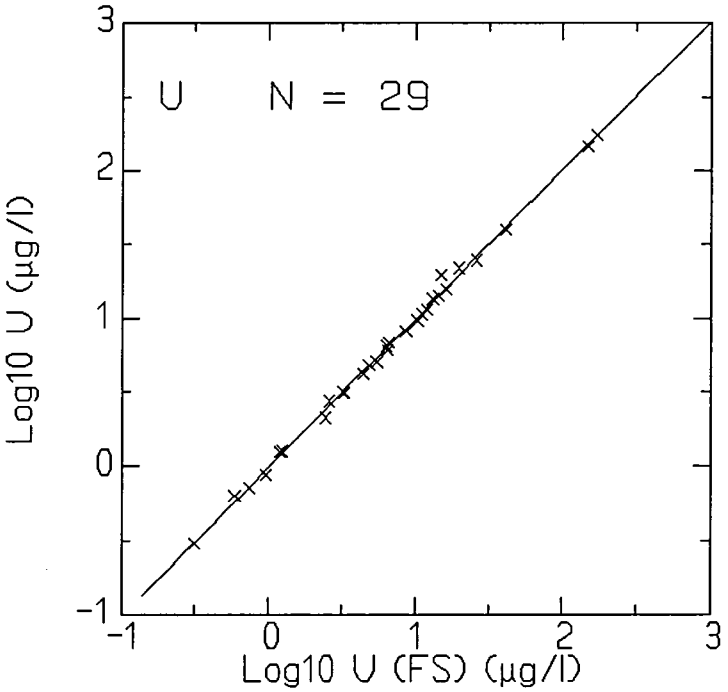


Fig. 4(a). Correlation between analytical results for field filtered and acidified (FS) samples with field unfiltered/acidified samples for uranium; $r^2 = 0.997$. Note $\log_{10}$ scales. Sample 13 excluded, N = 29.

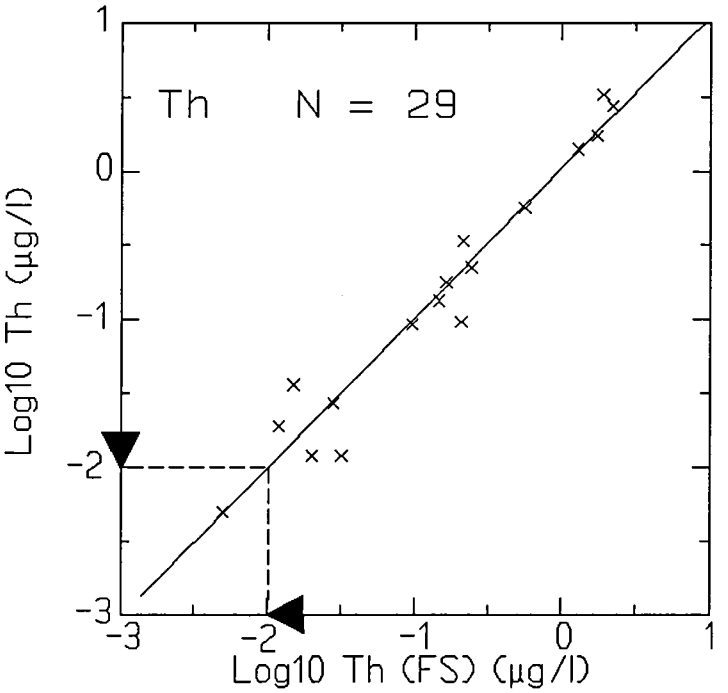


Fig. 4(b). Correlation between analytical results for field filtered and acidified (FS) samples with field unfiltered/acidified samples for thorium; $r^2 = 0.977$. Note $\log_{10}$ scales. Sample 13 excluded, N = 29.

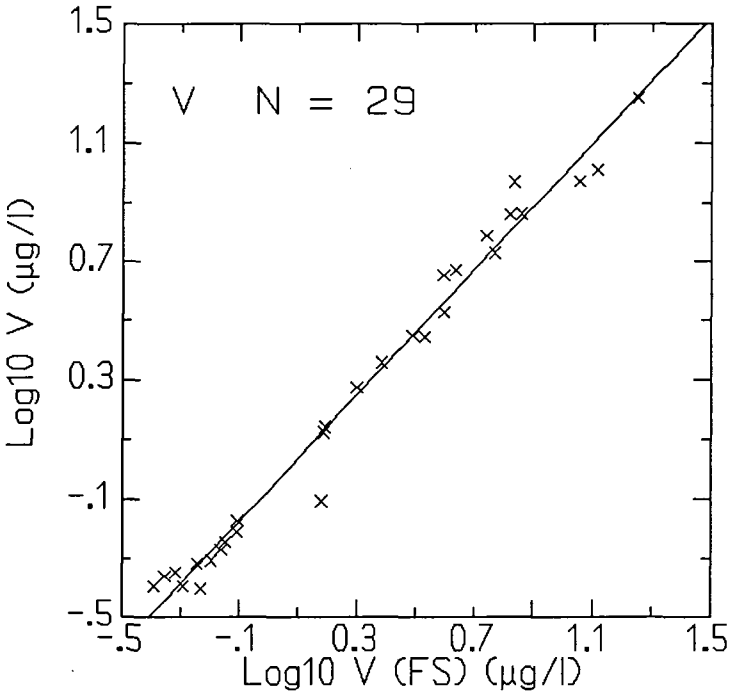


Fig. 4c. Correlation between analytical results for field filtered and acidified (FS) samples with field unfiltered/acidified samples for vanadium, $r^2 = 0.982$. Note $\log_{10}$ scales. Sample 13 excluded, N = 29.

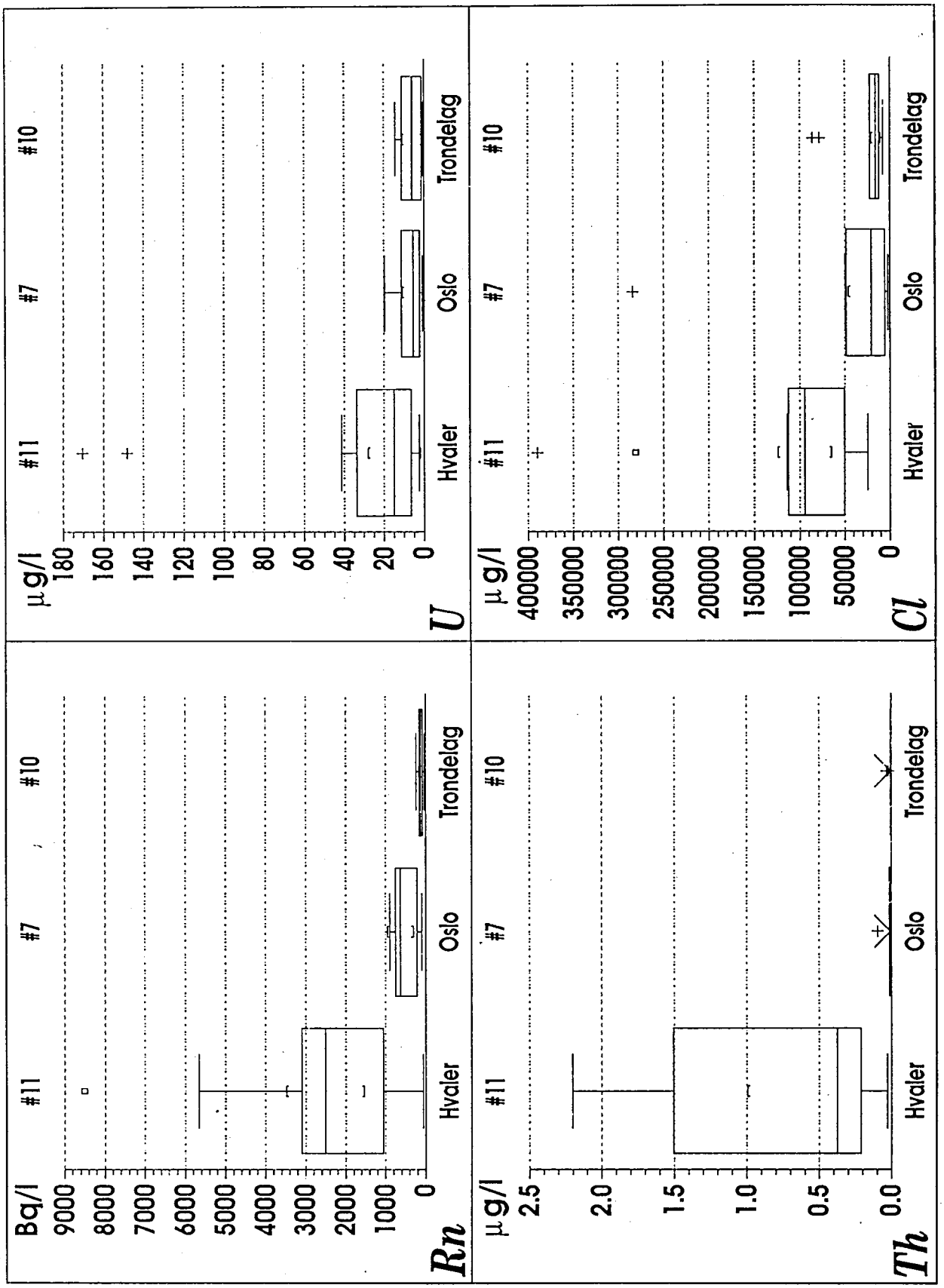


Fig. 5. Box plots showing concentrations of Rn, U, Th and Cl in bedrock groundwater from the three geographical sub-areas (N_{total} = 28). Box shows interquartile range, horizontal line = median, whiskers show range excluding outliers. Outliers shown by crosses and squares.

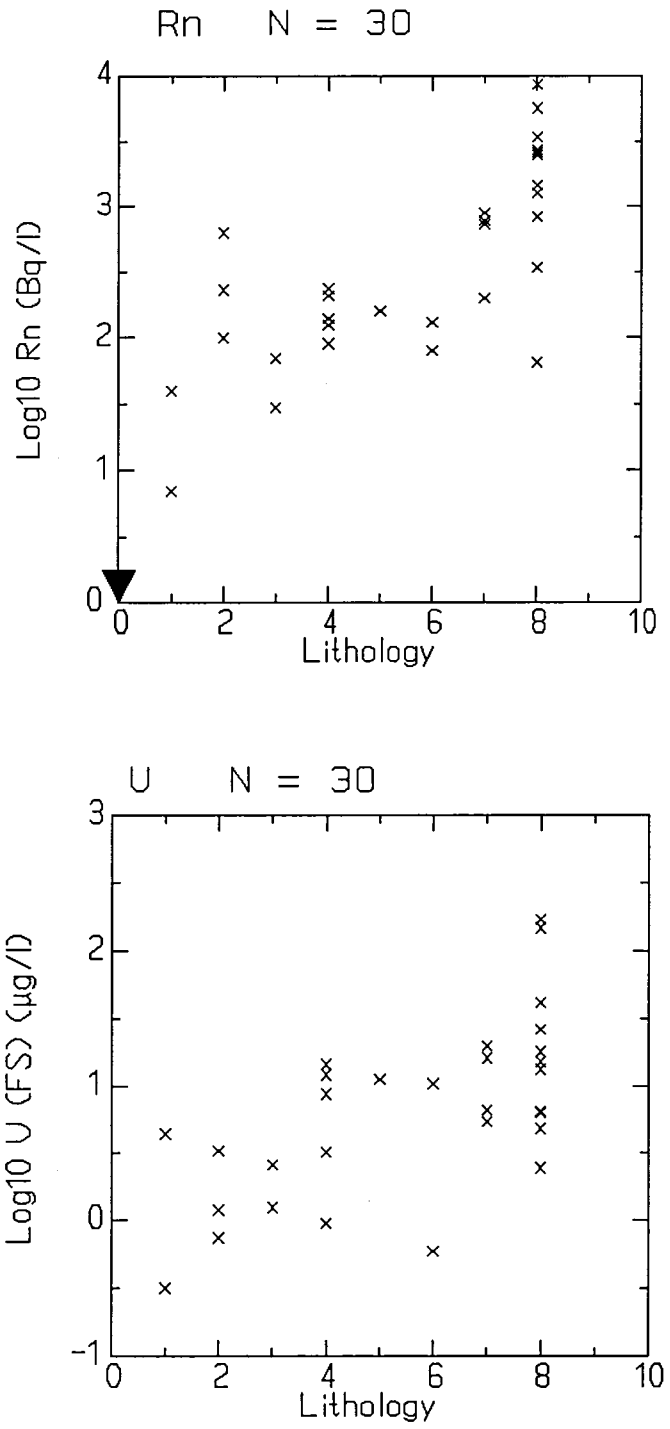
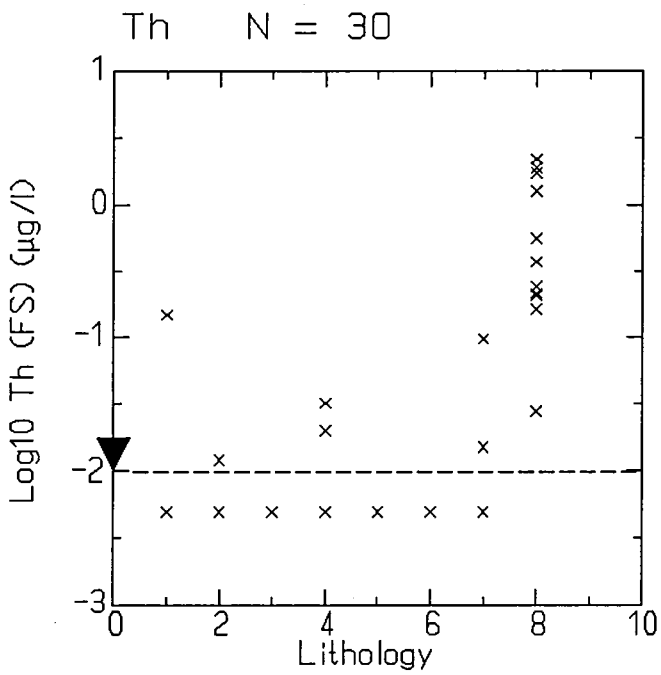


Fig. 6(a,b). Plots of $\log_{10}$ concentrations of Rn and U against lithological grouping (1 = Quaternary, 2 = rhomb-porphyry, 3 = metadiorite, 4 = schists, metaarkose, 5 = granodioritic gneiss, 6 = Precambrian gneiss, Trøndelag, 7 = Precambrian gneiss, Oslofjord, 8 = Iddefjord granite).



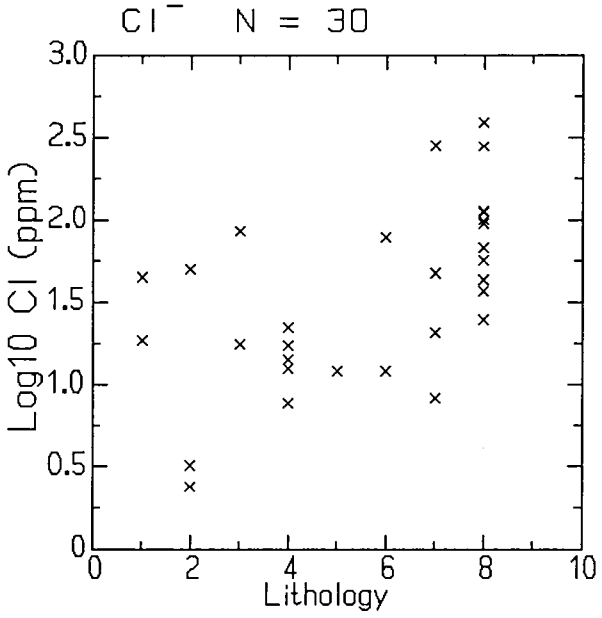


Fig. 6(c,d). Plots of $\log_{10}$ concentrations of Th and Cl against lithological grouping (1 = Quaternary, 2 = rhomb-porphyry, 3 = metadiorite, 4 = schists, metaarkose, 5 = granodioritic gneiss, 6 = Precambrian gneiss, Trøndelag, 7 = Precambrian gneiss, Oslofjord, 8 = Iddefjord granite).

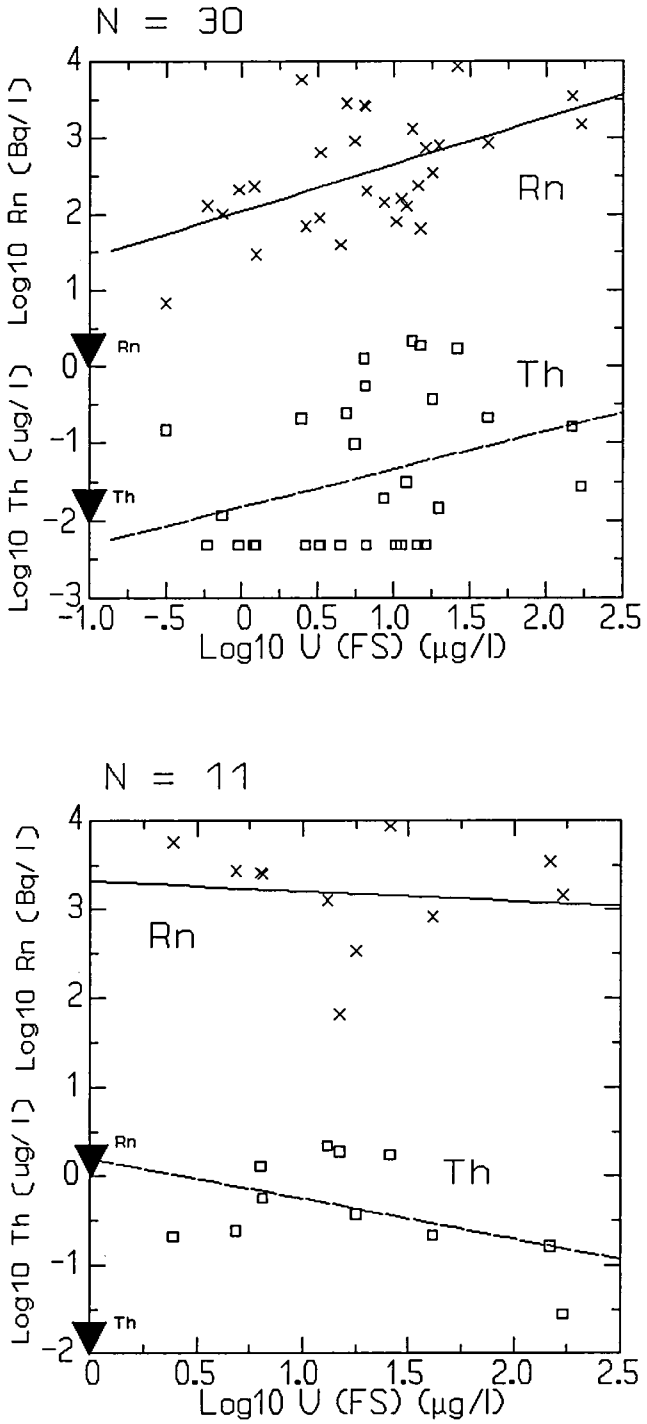


Fig. 7. Correlation of radon (crosses) and thorium (squares) with uranium for (a) the entire data set (N=30) and the Hvaler data set (N=11). Note $\log_{10}$ scales. Correlation coefficients: Rn vs. U; r= 0.53 (whole set), r = -0.11 (Hvaler): Th vs. U; r = 0.32 (whole set), r = -0.46 (Hvaler).

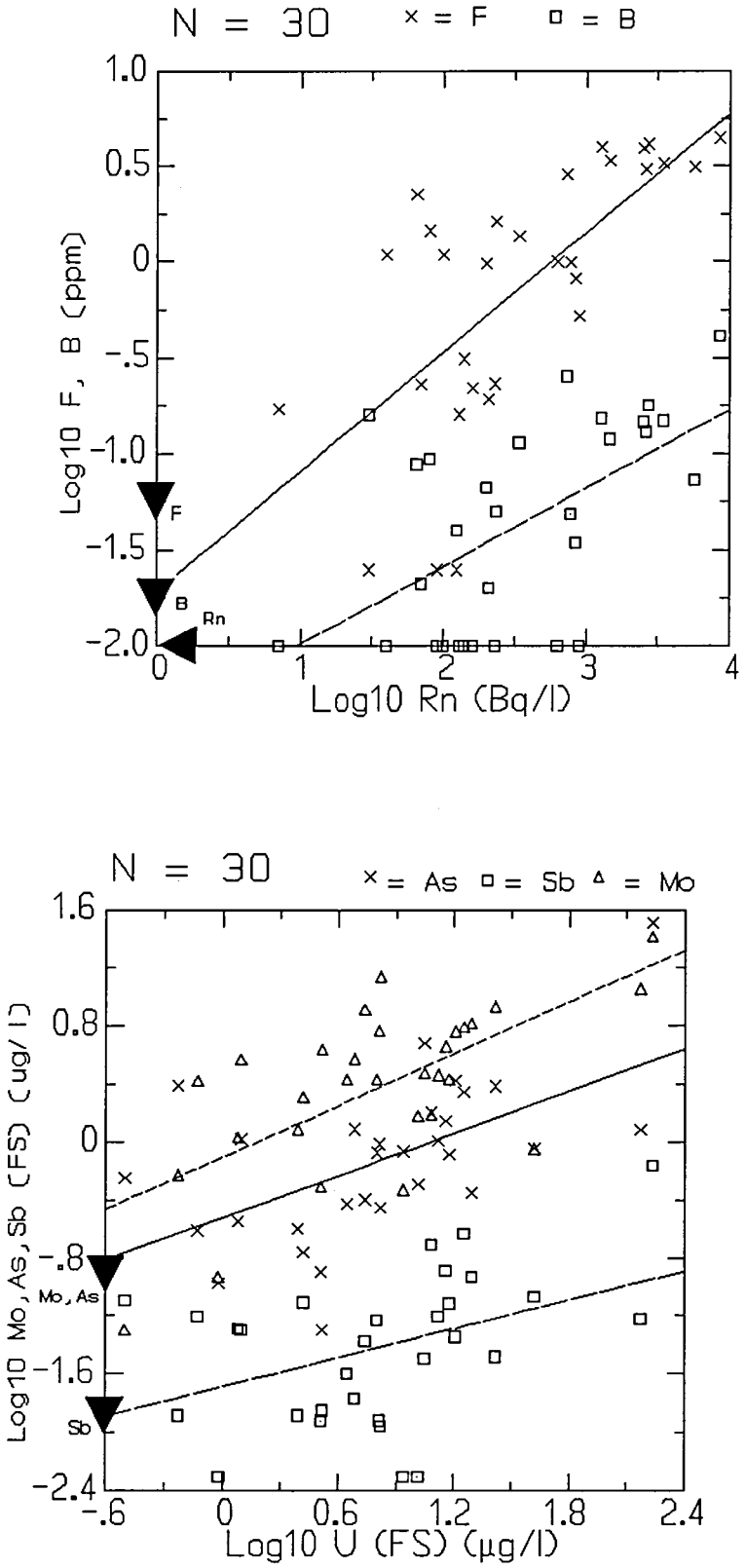


Fig. 8a,b. Correlation of (a) $Log_{10}Rn$ with $Log_{10}F$ (r = 0.67) and B (r = 0.56) and of (b) $Log_{10}U$ with $Log_{10}Mo$ (r = 0.65), As (r = 0.56) and Sb (r = 0.41).

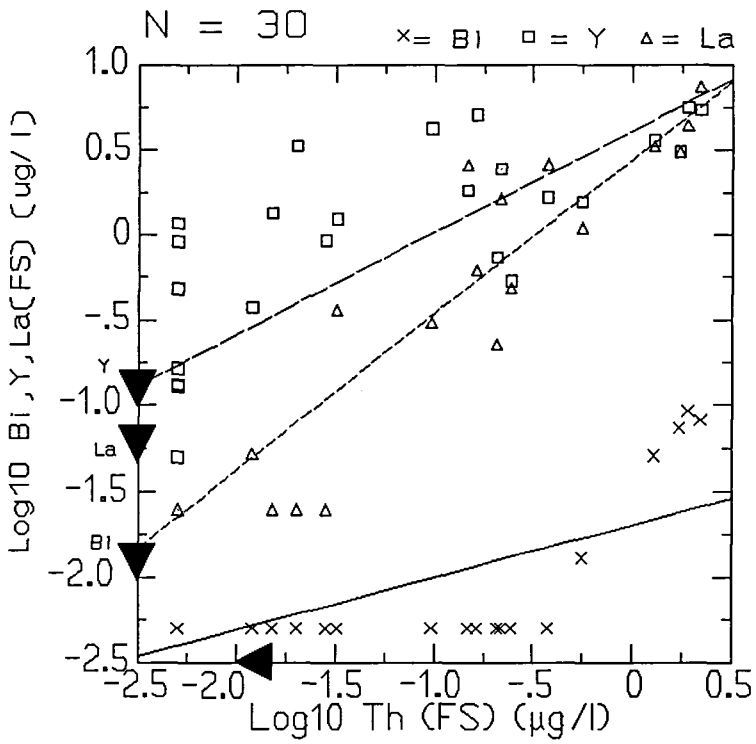


Fig. 8c. Correlation of $Log_{10}Th$ with $Log_{10}Bi$ (r = 0.73), La (r = 0.95) and Y (r = 0.78).

Radionuclide	Abundance	Half life (yr)
U-234	0.0056 %	2.5×10^5 y
U-235	0.720 %	7.0×10^8 y
U-238	99.276 %	4.5×10^9 y
Th-228	Trace	1.91 y
Th-230	Trace	7.7×10^4 y
Th-232	100%	1.4×10^{10} y
Th-234	Trace	24.1 days
Rn-219	-	3.92 sec
Rn-220	-	54.5 sec
Rn-222	-	3.8 days

Table 1: Global abundances and half-lives of the commonest U, Th and Rn isotopes.

Radionuclide	Average population weighted concentration in U.S. ground-water supplies (Bq/l)	Average population weighted conc-entration in U.S. water supplies (surface and ground) (Bq/l)
Rn-222	22	1.9 - 11
Ra-228	0.026	0.015 - 0.037
Ra-226	0.015	0.011 - 0.03
U	0.07	0.011 - 0.074
Po-210		< 0.005
Pb-210		< 0.004
Th-230		< 0.0015
Th-232		< 0.0004

Table 2: Results of United States survey of radionulides in drinking water (Milvy and Cothern 1990; Cothern 1987; Barnes 1986).

Area	Aquifer(s)	Reference	Radon (Bq/l)	Uranium (ppb)	Thorium (ppb)
"Standard" Seawater	Seawater	Horne 1969	0.6 x 10^{-12} ppb	3	0.05
Streamwater, Nordagutu, Norway	Surface water	Ryghaug 19	1.2 - 22	often > 10	
Ten Japanese rivers	Surface water	Miyake and others 1964		0.34 - 1.23	0.0087 - 0.048
Peat bog surface waters, Sweden	Surface water	Snihs 1973	up to 11,000		
Groundwater, Virginia and Maryland, USA	Not specified	Mose and others 1990a	4 - 300		
Stripa, Sweden, borehole M3 borehole 410	Quartz monzonite	Nelson and others 1983	48,000 - 70,000 18,000 - 21,000		
Stripa, Sweden Shallow groundwater Deep groundwater	Quartz monzonite	Andrews and others 1989	190-710 @ 100-74,400 @	0.8 - 90 0.02 - 35	
Leakage of ground-water, Olav's mine, Røros, Norway	Pyrrhotite mine	Myran 1973	170		
Wakulla Spring, Florida	Sedimentary rocks	Osmond 1964		0.5	nd
Leon well, Florida	Sedimentary rocks	Osmond 1964		nd	0.1
Martins thermal wells, Florida	Sedimentary rocks (Hawthrone phosphatic beds)	Osmond 1964		nd - 4	0.1 - 2
Well water, Utah, USA	Quaternary sediments	Tanner 1964	15 - 67		
Groundwater, Kalmar County, Sweden	Bedrock	Pousette and others 1981	Up to 3758		
Groundwater, Blekinge County, Sweden	Bedrock Quaternary	Pousette and others 1983	Median 161 Max. 2316 Median 33 Max. 874		
Groundwater, Stockholm County, Sweden	Bedrock Quaternary	Engqvist and Fogdestam 1984	Median 70 Max. 1166 Median 24 Max. 553		
Groundwater, Södermanlands County, Sweden	Bedrock Quaternary	Pousette and others 1984	Median 92 Max. 3398 Median 37 Max. 733		
Groundwater, Halland County, Sweden	Bedrock Quaternary	Karlqvist and others 1985	Median 23 Max. 231 Median 10 Max. 72		
Groundwater, Skaraborg County, Sweden	Bedrock Quaternary	Wikner and others 1991	Median 39 Max. 265 Median 5 Max. 106		
Groundwater, Kronoberg County, Sweden	Bedrock Quaternary	Söderholm and others 1987	Median 126 Max. 1204 Median 18 Max. 154		
Groundwater, Jönköping County, Sweden	Bedrock Quaternary	Pousette and others 1989	Median 95 Max. 1189 Median 25 Max. 401		

Groundwater, Stockholm, Boden, Bollstanäs; Sweden	Bedrock	Snihs 1973	1390 - 2320		
Groundwater, Vimmerby, Sweden	Bedrock	Snihs 1973	< 70 - 2660		
Groundwater, (largely southern) Finland	Quaternary deposits	Salonen 1988	GM 31 AM 92 Max. 3800	Median 1.5 Max. 10	
Groundwater, (largely southern) Finland	Bedrock	Salonen 1988 Asikainen and Kahlos 1979	GM 240 AM 1,020 Max. 77,000	Median 16 Max. 14,900	
Groundwater, Denmark	Mainland - Mesozoic to Quaternary Bornholm - older bedrock	Ulbak and Klinder 1984	Max. < 1 Max. 1070		
70 - 100 m deep drilled (mostly bedrock) wells, Norway	Granitic areas	Strand and Lind 1992	Mean 1070 Range 130-7000		
Maximum concentrations reported from the U.S.		Milvy and Cothern 1990 Michel 1990 Barnes 1986	55,000 (granite, Maine)	255 Bq/l = < 10 mg/l* (Denver, Co.)	^{232}Th 0.004 Bq/l = 1 µg/l ^{230}Th 0.015 Bq/l
Average conc., granite groundwater, Maine, U.S.A.	Granite	Michel 1990	8200		
990 public groundwater sources in the U.S. (finished water in distribution)	Various	Longtin 1990	Max. 951 Mean 24	Max. 88.2 Ave. 1.85	
Groundwater, Skalná spring, Czech Republic	Palaeozoic granite	Franko and others 1985	3100-7800		
Groundwater, Jáchymov borehole, Czech Republic	Eocambrian - Palaeozoic granitoids, schists (?) and phyllites	Franko and others 1985	8800		
Groundwater, Mladkov spring, Moravia, Czech Republic	Proterozoic Ortogneiss and migmatites	Franko and others 1985	1170		
Groundwater, Velká úpa spring, Czech Republic	Proterozoic muscovite albite schist (?) and phyllite	Franko and others 1985	2880		
Oberschlema spring		Hevesy and Paneth 1938	39,900		
Joachimstal mine leakage, Germany		Hevesy and Paneth 1938	26,600		
Ischia, Old Roman Spring		Hevesy and Paneth 1938	4921		
Alum Spring, Aix-les-Bains, France		Hevesy and Paneth 1938	745		
Groundwater, Nordagutu, Norway	Mostly Quaternary	Ryghaug 1984	51 - 1100		
Groundwater, Leksvik, Norway	In vicinity of Th-breccia	Grønlie 1983	3 - 247		

Table 3: Literature survey of concentrations of Rn, U and Th in natural waters. @ = conc. in Bq/kg. * using conversion factor of 1 µg ≥ 0.025 x 10^{-2} Bq. AM = arithmetic mean, GM = geometric mean. nd = not detectable.

Sample nr.	Lithology (Class)	Depth (m)	Rn (Bq/l)	U (µg/l)	Th (µg/l)	Na (meq/l)	Cl (meq/l)
1	Precambrian gneiss	60 m	890	5.5	0.097	0.57	0.23
2	Precambrian gneiss	90 m	780	20	0.015	1.8	1.3
3	Precambrian gneiss		200	6.6	< 0.01	3.7	0.58
4	Permian rhomb porphyry	60 m	100	0.74	0.012	0.13	0.07
5	Permian rhomb porphyry	25 m	230	1.2	< 0.01	0.93	1.4
6	Permian rhomb porphyry		630	3.3	< 0.01	0.14	0.09
7	Precambrian gneiss	65-100 m (2 wells)	730	16	< 0.01	12	8.0
8	Iddefjord granite		5700	2.4	0.21	3.1	1.0
9	Iddefjord granite		1470	170	0.028	2.6	0.69
10	Iddefjord granite	c.70 m	2600	6.3	1.3	4.1	1.9
11	Iddefjord granite	80 m	8500	26	1.7	14	11
12	Iddefjord granite	80 m	65	15	1.9	3.3	2.8
13 @	Iddefjord granite	80 m *	340	18	0.38	7.3	7.9
14	Iddefjord Granite	101 m	2500	6.5	0.56	5.1	2.7
15	Iddefjord granite	70 m	840	41	0.22	1.2	1.2
16	Iddefjord granite	c. 45 m	1280	13	2.2	4.8	1.6
17	Iddefjord granite	60 m	2800	4.8	0.24	7.4	3.2
18	Iddefjord granite	c. 80 m	3500	150	0.16	5.3	3.1
19	Quaternary sand.	2.5 m	7	0.32	0.15	0.82	1.3
20	Precambrian-cambrian garnet-mica schist	52 m	90	3.2	< 0.01	1.0	0.40
21	Precambrian-cambrian garnet-mica schist	120 m	210	0.96	< 0.01	2.3	0.36
22	Precambrian-cambrian quartz and garnet-mica schist	25.5 m	240	14	< 0.01	1.8	0.49
23	Precambrian-cambrian mica schist	75 m	125	12	0.032	0.43	0.63
24	Ordovician(?) metadiorite	80 m	30	1.2	< 0.01	5.4	2.4
25	Precambrian gneiss	80 m	80	10	< 0.01	3.3	2.2
26	Late Pre€-palæozoic metaarkose	120 m	140	8.7	0.020	0.45	0.22
27	Quaternary sediments	1 m	40	4.4	< 0.01	0.37	0.52
28	Ordovician(?) metadiorite	119 m	70	2.6	< 0.01	1.4	0.50
29	Pre€.-€. granodioritic gneiss	100 m	160	11	< 0.01	0.86	0.34
30	Precambrian granitic gneiss	71 m	130	0.59	< 0.01	0.67	0.34

Table 4. Details of samples taken during the Norwegian pilot study. Samples 1 - 7 from Oslofjord, 8 - 19 Hvaler, 20 - 30 Nord Trøndelag. * = angled borehole.
@ = Sample 13 was not filtered using a 0.45 µm filter, due to particulate content.

	Rn (Bq/l)	U (μg/l)	Th (μg/l)	Rn (Bq/l)	U (μg/l)	Th (μg/l)
	Whole data set			Oslo Rift		
N =	28	28	28	7	7	7
Maximum	8500	170	2.2	890	20	0.10
Minimum	30	0.59	< 0.01	100	0.74	< 0.01
Arithmetic	1230	20	0.33	510	7.6	0.02
Geometric	430	7.3	0.04	390	4.4	0.01
Median	290	7.6	0.02	630	5.5	< 0.01
Standard dev.	1950	40	0.63	320	7.4	0.03
	Iddefjord granite			Trøndelag		
N =	11	11	11	10	10	10
Maximum	8500	170	2.2	240	14	0.03
Minimum	65	2.4	0.03	30	0.6	< 0.01
Arithmetic	2700	41	0.81	127	6.5	0.009
Geometric	1530	17	0.43	111	4.0	0.007
Median	2500	15	0.38	128	5.9	< 0.01
Standard dev.	2500	60	0.81	63	5.3	0.009

Table 5. Statistical analysis of the Norwegian pilot study data set (excluding Quaternary wells).

Radon	Untransformed variables	Whole set (N=30)	Zr (0.83), F, B, Tl, Na, Be, Cl, EC, V (0.5)
		Hvaler (N=11)	Zr (0.77), B, Alk, Na, Co, K, F, EC, Br, Al, Tl (0.51)
	Log10	Whole set (N=30)	F (0.67), Zr, B, U, Th, Mo (0.52)
		Hvaler (N=11)	Co (-0.78), Alk, K, Al, Zr, Cs, F (0.55)
Uranium	Untransformed	Whole set (N=30)	Mo (0.77), As, Sb, Li (0.52)
		Hvaler (N=11)	Mo (0.85), Li, As, Sb, Rb, Si (-0.50)
	Log10	Whole set (N=30)	Mo (0.65), As, B, Rn, EC (0.50)
		Hvaler (N=11)	Sb (0.72), As, Rb, Si, Mo, Cd, Ni (0.53)
Thorium	Untransformed	Whole set (N=30)	Bi (0.98), La, Y, Pb, Tl, Zr, F, B (0.53)
		Hvaler (N=11)	Bi (0.98), La, Y, Pb, Al (0.54)
	Log10	Whole set (N=30)	La (0.95), Y, Be, Bi, Cd, Ca, V, Pb, Sr, Tl, Zr, Cr, Mg, F, B, Cl, Rn, Si, Al (0.50)
		Hvaler (N=11)	La (0.92), Bi, Tl, Rb, Y, Br, Ca, Sr, Pb, As (-0.51)

Table 6. Correlation coefficients (r) between radioactive elements and other parameters. Values of r in excess of 0.5 are given in descending order. Underlined values indicate negative correlations; maximum and minimum values of r are given in parentheses.

			Rn	U	Th	F	Cl	pH	Alk
Radon	Untransformed variables	Whole set (N=30)	*****	0.23	0.43	**0.72**	**0.55**	0.14	0.17
		Hvaler (N=11)	*****	-0.07	0.06	**0.58**	0.47	0.41	**0.69**
	Log10	Whole set (N=30)	*****	**0.53**	**0.52**	**0.67**	0.34	0.25	0.06
		Hvaler (N=11)	*****	-0.11	-0.18	**0.55**	0.01	0.49	**0.75**
Uranium	Untransformed	Whole set (N=30)	0.23	*****	-0.02	0.37	0.09	0.16	0.04
		Hvaler (N=11)	-0.07	*****	-0.43	0.01	-0.19	0.34	0.12
	Log10	Whole set (N=30)	**0.53**	*****	0.32	0.49	0.39	0.25	0.06
		Hvaler (N=11)	-0.11	*****	-0.46	-0.19	-0.06	0.12	-0.08
Thorium	Untransformed	Whole set (N=30)	0.43	-0.02	*****	**0.59**	0.41	-0.10	-0.12
		Hvaler (N=11)	0.06	-0.43	*****	0.27	0.25	-0.07	0.00
	Log10	Whole set (N=30)	**0.52**	0.32	*****	**0.53**	**0.53**	-0.35	-0.27
		Hvaler (N=11)	-0.18	-0.46	*****	0.15	0.48	-0.31	-0.11

Table 7. Correlation coefficients between the three radioactive species and Cl, F, pH and alkalinity, for the entire data set (N = 30) and the three geographically defined bedrock subsets. Correlation coefficients (r) greater than 0.5 are shown in bold type.

Organisation	Rn	U	Ra
U.S.EPA recommended limit for Rn in air (Mose and others 1990a)	4 pCi/l = 0.15 Bq/l		
Max.recommended concentration in drinking water (Snihs 1973)	100 nCi/l = 3,700 Bq/l		
Max. permitted total radium in water, U.S.E-PA, and equivalent concentrations of U and Rn giving lifetime risk of 4 x 10^{-5} (Milvy and Cothern 1990)	2.2 Bq/l	0.74 Bq/l = < 30 μg/l^{+}	5 pCi/l * = 0.185 Bq/l
USEPA proposal for Rn in water (alternative levels suggested by AWWA) (AWWA 1993)	11 (37-185) Bq/l		
Range of suggested MCLs (Milvy and Cothern 1990)	22 - 74 Bq/l	0.7 - 4 Bq/l = <30 - <160 μg/l^{+}	
Range of suggested MCLs (Kinner and others 1990, Sorg 1990)	8 - 370 Bq/l	0.37 - 1.85 Bq/l = <14 - <75 μg/l^{+}	
Radon in drinking water in Sweden (based on possibility for degassing), SIFF (1987) Action required Possible action No action required	 > 1000 Bq/l 100 - 1000 Bq/l < 100 Bq/l		
Canadian MCL for uranium in drinking water (Lahermo and Juntunen 1991, Barnes 1986)		20 μg/l	
Gross α-activity (excluding U and Rn). U.S.-EPA (Milvy and Cothern 1990)	15 pCi/l = 0.555 Bq/l.		
Gross α-activity WHO (1984), SIFF (1987)	3 pCi/l = 0.1 Bq/l (based on ^{226}Ra, i.e. excluding radon)		

Table 8. Existing drinking water standards for Rn, U, Ra and gross radioactivity. * = fixed USEPA standard for ^{226}Ra + ^{228}Ra. $^{+}$ = based on assumption 1 μg $\geq$ 0.025 Bq.