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

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Abstract Twenty-eight samples of groundwater from bedrock boreholes in three distinct Norwegian geological provinces have been taken and analyzed for content of Rn, U, and Th, together with a wide variety of minor and major species. Median values of 290 Bq/l, 7.6 μ g/l, and $0.02 \mu g/l$ were obtained for Rn, U, and Th respectively, while maximum values were 8500 Bq/l, $170 \mu g/l$, and 2.2μ g/l. Commonly suggested drinking water limits range from 8 to 1000 Bq/l for radon and 14 to 160 μ g/l for uranium. Radioelement content was closely related to lithology, the lowest concentrations being derived from the largely Caledonian rocks of the Trondelag area, and the highest from the Precambrian Iddefjord Granite of southeast Norway (11 boreholes) where median values of 2500 Bq/l, 15 μ g/l and 0.38 μ g/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.

Key words Groundwater · Radioactivity · Radon · Uranium · Thorium · Norway · Granite · Drinking Water Standards

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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 has been claimed to be responsible for 150-300 lung cancer cases (10-20 percent of the total) annually in Norway (Statens Strålevern 1994), 400-1100 in Sweden (Statens Strålskyddsinstitut 1993), and as many as 5000-20,000 deaths each year in the United States (Zikovsky and Chah 1990). In Scandinavia, radon in houses accounts for about 75 percent 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 several factors, including underlying geology, 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/1 Rn can contribute 0.1 Bq/1 to air concentrations. The USEPA set an action level of 0.15 Bq/1 radon for indoor air (Mose and others 1990a), while Scandinavian lands and the UK use 0.2 Bq/1 (SIFF 1987; DoE 1993). Although direct emanation from the bedrock is the greatest contribution to radon in Norwegian buildings, the groundwater contribution to radon in internal air can be very significant in certain cases (Strand and Lind 1992).

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

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United States 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/1 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 (Jianti and others 1993). The authors have yet to find documented evidence of any desirable effects of radon exposure, however. The consequences of such exposure must continue to be regarded as wholly negative.

Natural occurrence of uranium, thorium, and radon

Geologically, uranium (U), thorium (Th), radium (Ra), and radon (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.

In sedimentary rocks and metamorphic rocks, such radioactive elements can be concentrated: (1) along fault zones or other discontinuities by hydrothermal activity, (2) at palaeo-redox fronts, or (3) where they can bind to organic material in reducing environments, e.g., organicrich 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 $(UO₂)$, which is 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 these minerals often approach a composition symbolized by U_3O_8 . In secondary deposits, coffinite (USiO₄ \cdot nH₂O) 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).

Uranium has three isotopes (Table 1) occurring naturally in significant amounts; but over 99 percent of the global mass consists of ²³⁸U. 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 3–11 times the activity of 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 (micrograms per liter) 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 $1 \mu g = 1.2 \times 10^{-2}$ Bq ²³⁸U, and, assuming equilibrium, a conversion factor of 1μ g = 2.5×10^{-2} Bq $\left[2^{234}U + {}^{238}U\right]$ is thus commonly used (Barnes 1986; Milvy and Cothern 1990).

In terms of mass, naturally occurring thorium consists almost entirely of ²³²Th, and 1 μ g = 4 × 10⁻³ Bq ²³²Th. It decays, via 2^{28} Ra and 2^{28} Ac, to the short-lived 2^{28} Th (half-life 1.91 yr), and the activity (Bq) of the latter radionuclide in groundwater may exceed that due to 232Th. The short-lived ²³⁴Th (half-life 24.1 days) is produced by α -decay of ²³⁸U, itself decaying rapidly to ²³⁴U. ²³⁰Th (half life 77,000 yr) is also part of the 238 U decay series.

Radon is a chemically inert gas that 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

Table 1 Global abundances and half-lives of the commonest U, Th and Rn isotopes, with typical U and Th contents in bulk continental crust, granites and granodiorites (after Killeen and Heier 1975b)

of the uranium-238, thorium-232, and uranium-235 decay series, respectively. As $222Rn$ 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.

²²²Rn is derived from the parent radionuclide ²²⁶Ra, a long-lived alpha-emitter of half-life 1620 yr, and a member of the uranium-238 series. It is thus typically found in rocks with high uranium content, although uranium is considerably more mobile than radium and can be weathered out, leaving radium *in situ* in the residual material. 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 d, may travel limited distances (up to tens to hundreds of metres) in the groundwater of a fractured aquifer before decaying to $^{218}P_0$, $^{214}P_0$, $^{214}B_1$ and $^{214}P_0$ (the so-called radon daughters).

Hydrochemistry of uranium, thorium and radon

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, summarized as follows (from Krauskopf 1979). Uranium can occur in several oxidation states, but only $4+$ and $6+$ 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 parts per million and, exceptionally up to a few thousand parts per million uranium. In summary, therefore, uranium is rather insoluble in reducing environments, but is soluble in oxidizing, and particularly acidic, conditions. Uranium(VI) also forms complex ions with hydroxide (in highly alkaline conditions), carbonate, phosphate, and perhaps 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 sulfide can be important. Uranium displays considerable chemical similarity to vanadium, forms a number of combined minerals, and is insoluble in any water containing $> 100 \text{ µg/l}$ 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 $4 +$.

Radon is a chemically inert, but soluble gas. Its concentration in groundwater is believed to be directly controlled by: (1) hydrodynamic factors, (2) uranium (or, more precisely, radium) content of rocks and groundwater in the vicinity of the well (Michel 1990), and (3) the uranium and radium mineralogy within the host rock (Ball and others 1991). Like many other solutes, its concentration is commonly found to be lognormally distributed in groundwater (Zikovsky and Chah 1990). 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).

Radioelements in groundwater

The results of selected surveys of radionuclides in groundwater in Scandinavia and the United States are presented in Tables 2 and 3. Even in relatively shallow granitic groundwaters, concentrations of up to 55,000 Bq/1 radon $(Maine, USA)$ and 14.9 mg/l uranium (Helsinki, Finland) have been recorded. The Finnish Geological Survey has compiled hydrogeochemical maps of radionuclides and statistically analyzed 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).

Previous studies in Norway

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/1 in air (although this is slightly exaggerated due to overrepresentation of houses on the uranium-rich Alun Shale bedrock), representing a dose equivalent of 4 mSv/yr . The highest values appeared to be concentrated in the area around the Oslo graben (Fig. 2 below), 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. In all, 7525 houses, distributed throughout Norway in proportion to population, 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 in Norwegian tap water from drilled

Table 2 Concentrations of Rn, U, and Th in groundwater: Summary of selected previous studies Table 2 Concentrations of Rn , U, and Th in groundwater: Summary of selected previous studies

Table 3 Results of United States survey of radionulides in drinking water (Milvy and Cothern 1990; Cothern 1987; Barnes 1986)

boreholes (70-100 m deep). 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/1 and a range of 130-7000 Bq/1. Other geological formations yielded a range of $\langle 5-1250 \text{ Bg} / 1 \rangle$.

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 μ g/l uranium and 1.2-22 Bq/1 radon. Groundwater from wells, mostly in Quaternary deposits, contained 51-1100 Bq/1 radon.

Arne Gronlie (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-Trondelag. The highest concentration measured was 247 Bq/1 in groundwater from a bedrock borehole believed to be the same as our borehole 22 (Table 4).

Table 4 Details of samples taken during the Norwegian pilot study^a

Samples 1-7 from Oslofjord, 8-19 Hvaler, 20-30 Nord Trøndelag.

Angled borehole. Sample 13 not filtered with $0.45 \mu m$ filter, due to high particulate content

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 yet allowed a strictly geographically or epidemiologically representative sample set to be collected.

Two study areas were chosen, the following criteria being used: (1) ease of accessibility and existing groundwater projects in progress, (2) lithological variation, and (3) presence of lithologies suspected to be "high risk." The county north of Trondheim (Nord-Trondelag) and the area around Oslofjord were chosen (Figs. 1 and 2).

Nord-Trendelag

A simplified geological map of the county of Nord-Trondelag is shown in Fig. 1. The area can be subdivided very coarsly into three units: (1) the Caledonian mountain belt-consisting of a sequence of nappe-piles of gneisses, metasediments and metavolcanics of Precambrian to

Fig. 1 Simplified geological map of the Trøndelag area, showing location of sampled boreholes. Inset shows map of Norway with study areas outlined

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 (Gronlie and Staw 1987); (3) windows of Proterozoic "basement" gneisses and metasediments (e.g., the Tommerås Window). A dominant tectonic feature is the Møre and Trondelag 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 rockmaterial from the Leksvik breccia zone contains an average 990 ppm Th and 49 ppm U (Gronlie and Staw 1987).

Oslo region

The region (Fig 2) is tectonically dominated by the Oslo rift, of Carboniferous-Permian age. Sedimentary rocks of Precambrian to Silurian age occur within the rift, 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,

Fig. 2 Simplified geological map of the Oslofjord area, showing location of sampled boreholes

and intruded by igneous rocks, largely of Permian age. Autochthonous Precambrian basement occurs to the east and the west of the Oslo rift, consisting of gneisses and granites dating from the Sveconorvegian orogeny or earlier. In the extreme southeast of the area is the Precambrian Iddetjord 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/m² (Cermák and others 1992). In the Iddefjord granite, Killeen and Heier (1975a) 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 derive from radioactive elements dispersed in the granite's groundmass or from occurrences of specific minerals in pegmatite dykes. Several occurrences of uranium and thorium minerals in pegmatites in the Iddefjord granite are reported by Bjorlykke (1939), including uranium(IV) oxide, thorite, samarskite (up to 15 percent uranium oxides), monazite (up to 19 percent thorium oxide), and xenotime. The hydrogeology and hydrochemistry of the Iddetjord 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 (Bjorlykke 1939). For these reasons, the sampling program was focused on the Hvaler islands.

Methods and equipment

Bedrock boreholes or wells were chosen (Table 4) with emphasis on the following criteria: (1) the borehole should be in regular use or should be naturally overflowing, such that "fresh" groundwater is sampled; (2) the 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; (3) the borehole should have a low possibility for degassing, i.e., sampling points at borehole head or sampling points that are part of a closed system (e.g. pressure tank) were preferred; (4) the water should not contain particulate matter or humus.

In practice, however, some boreholes did not satisfy all criteria (i.e., minor infringements of items 3 and 4). In particular, sample 13 came from a relatively newly drilled (7 months 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- 1993. Prior to sampling, the tap was allowed to run for at least 5 mins. All polythene flasks were rinsed thoroughly three times with groundwater and twice with filtered $(0.45 \text{-} \mu \text{m}$ Millipore filter) water before sampling. The following samples were then taken in polythene bottles with screw caps: (1) 2×100 ml unfiltered, unacidified; (2) $2 \times$ 100 ml filtered $(0.45 \text{-} \mu \text{m})$ Millipore filter and polythene syringe) and acidified (10 drops 65 percent Ultrapur nitric acid) in the field; and (3) 1×500 ml unfiltered, unacidified.

One aliquot of sample (1) was analyzed at the Geological Survey of Norway (NGU) for seven anions (F⁻, $\overline{PO_4}^{3-}$, Br⁻, Cl⁻, SO₄²⁻, NO₃⁻, and NO₂⁻) by ion chromatography. One aliquot of sample (2) was analyzed by inductively coupled plasma emission spectroscopy at NGU for Si, A1, 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 aliquats of samples (1) and (2) were analyzed at the Norwegian Institute for Air Research (NILU) by ICP mass spectrometry (ICPMS) for Pb, Cd, Cu, Zn, Cr, Ni, Co, Fe, Mn, V, As, Ba, Sr, A1, Sb, Bi, T1, 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 100 μ l 1 percent HNO₃ was added; 50 μ g/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 percent deviation at a concentration of 10 μ g/l. Brackish/saline water samples can lead to greater errors, and Cr, V, Fe, *Ni,* and possibly As are among the most problematic elements in such a situation.

The 500-ml sample (3) 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, 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 vial of scintillation liquid was then sealed and shaken. The liquid gels on contact with water, immobilizing the radon. Vials were delivered to the Radiation Protection Authority (NRPA) within three days and analyzed using an LKB Wallac 1215 scintillation counter, calibrated using a standard radium solution. Results were back-adjusted for radioac-

Fig. 3 Correlation between analytical results for field filtered and acidified (FS) samples with field unfiltered/unacidified samples for (a) uranium; $r^2 = 0.997$ and (b) thorium; $r^2 = 0.977$. Note \log_{10} scales. Sample 13 excluded, $n = 29$. Black triangles show detection limits for Th

tive decay to give a radon concentation in becquerels per liter at time of sampling. As 222 Rn is the only isotope with a significant half-life, the radon concentrations quoted in this paper, determined by this method, represent concentrations of 222 Rn. The overall uncertainty in the method is estimated to be around 20 percent at the 95 percent confidence level, and the lower limit of detection is 1 Bq/1.

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 verification of many parameters. In the case of most parame-

Fig. 4 Box plots of analytical results for Rn, U, Th, and C1 for bedrock boreholes in each of the three geographical subareas. Boxes contain the middle 50 percent of the data, whiskers show range of non-outlying data, crosses and small squares show extreme and nearer outliers

ters (including U, Th, and V), field acidification did not dramatically affect the analysis outcome (Fig. 3), indicating that unacidified, unfiltered samples can give satisfactory analytical results for U and Th, at least for relatively "clear" groundwater samples. Some discrepancies were discovered between the NGU and NILU analytical results, particularly for a few elements that are rather sensitive to pH and/or redox conditions (and thus to filtering and storage), such as Fe and A1, and a few elements that are known to be problematic for ICP-MS techniques in rather brackish groundwaters, such as Cr, Fe, and V. Further discussion of analytical comparisons can be found in Banks and others (1995).

In the following assessment, the NGU values for major elements (Na, K, Ca, Mg, and elements not analyzed by NILU, such as Zr) are used, while the NILU 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 purposes of statistical evaluation.

The results of the U, Rn, Th, C1, and Na analyses for each borehole are given in Table 4. Cl can be regarded as an indicator of marine influence, and the difference Na -C1 as a coarse indicator of water-rock interaction (Banks and others 1993b).

Correlation studies

In order to interpret the data, the wells have been divided into eight 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), and $8 =$ Iddefjord granite. The lithologies were chosen in order to 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 (Banks and others 1995) (Fig. 4) 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 in the Oslo area, with the Trondelag area showing the lowest concentrations (Table 5). Plots of element concentration versus lithology (Fig. 5) tend to confirm these trends, with the highest concentrations being recorded in the Iddefjord granite.

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, as reflected in chloride concentrations (see Figs. 4 and 5). Banks and others (1993b) have demonstrated that chloride concentrations at Hvaler are not derived in significant quantities from the granite, 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 Trondelag through Oslofjord region to Hvaler: (1) increasing bedrock "acidity," i.e., geological occurrence of incompatible elements; (2) increasing marine influence; and (3) increasing fallout from atmospheric contamination.

Any correlation of groundwater concentrations of radioelements with other elements or lithology may be: (1) purely coincidental (i.e., nongeological), such as that between chloride and lithology; (2) reflecting a covariation in element concentrations with respect to geological environment, e.g., covariations between "incompatible" latemelt elements in acidic rocks, or (3) reflecting a direct causative 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 mobilizes 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.

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Table

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Fig. 5a-d Plots of log_{10} concentrations of Rn, U, Th, and CI 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). $FS =$ field filtered and acidified. Black triangles show detection limits for Rn and Th

Generally speaking, U, Th, and Rn showed only rather weak correlations with other elements. The strongest, most persistent correlations were found for Th with Bi, La and Y (the latter two being chemically analogous to the actinides). Weaker, but persistent, correlations were found

Table 6 Correlation coefficients (r) between radioactive elements and other dissolved species^a

 $^{\circ}$ 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. Cr and V are known to suffer significant interference with C1 in the ICPMS determination and correlations involving these parameters may be subject to error (see Banks and others 1995)

Fig. 6 Correlation of radon (crosses) and thorium (squares) with uranium for (a) the entire data set ($n = 30$) and (b) the Hvaler data set ($n = 11$). Note log_{10} scales. Correlation coefficients: Rn vs U; $r = 0.53$ (whole set), $r = -0.11$ (Hyaler): Th vs U; $r = 0.32$ (whole set), $r = -0.46$ (Hvaler). Black triangles show detection limits for Rn and Th

for Rn with 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 2 above). No significant or persistent correlations were found between uranium and major parameters that might be expected to affect mobility, such as alkalinity, C1, or pH (i.e., type 3 above).

Surprisingly, only rather weak correlations between uranium, radon, and thorium themselves were found [in accordance with findings of Lahermo and Juntunen (1991)1. These correlations only occurred within the entire data set, reflecting the coarse-scale covariation in host rock contents of these elements (Fig. 6a). Within a single lithology (the Iddefjord Granite), these correlations disappear (Fig. 6b, 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.

Doses and drinking water limits

Maximum concentration levels (MCL) for U, Th, and Rn in drinking water have not been agreed upon in most countries. Suggestion and tentative limits are summarized in Table 8. Swedjemark (1993) cites radon dose-equivalents (Table 9), which suggest that, for young infants, the dose from ingestion of radon could outweigh the dose from inhalation of degassed radon. She implies that the threat from the ingested parent nuclide 222 Rn far outweighs the dose derived from daughters. The Finnish Strålsäkerhet-

Table 7 Correlation coefficient between three radioactive species and F, C1, pH, and alkalinity, for entire data set $(n = 30)$ and Hvaler bedrock subset^a

 $^{\circ}$ Correlation coefficients (r) greater than 0.5 are shown in bold type

Table 8 Existing and proposed drinking water standards for Rn, U, Ra and gross radioactivity

^b Based on assumption 1 μ g \geq 0.025 Bq

scentralen (1993) is in rough agreement with Swedjemark, suggesting that an ingested concentration of 300 Bq/1 is equivalent to the maximum permissible effective dose from radionuclides in ingested drinking water of 0.5 mSv. Milvy and Cothern (1990), considering water resources generally, conclude that the threat due to radon heavily outweighs that from uranium and radium, being responsible for around 80 percent of radionuclide-induced deaths from drinking water.

Discussion

The following comments can be made regarding radon, taking the Swedish levels of 100 Bq/1 in groundwater for possible adverse effects, and 1000 Bq/1 as the level for

recommended remedial action (Table 8). The two Quaternary wells sampled give no grounds for concern over radon levels. Of the ten bedrock boreholes in Nord-Trondelag, six exceed the 100 Bq/1 threshold, although there is no clear lithological correlation. The highest value is 240 Bq/1, from a borehole believed to be associated with a Th- (and U-) enriched breccia zone. The bedrock of Nord-Trondelag thus appears to yield groundwaters with inferior Rn levels compared with the Oslofjord region. This probably reflects the rather calcareous and uraniumpoor nature of the Caledonian metasediments and the uranium-poor nature of the basement gneisses in the area south of Namsos (Gronlie and Staw 1987).

Boreholes in the gneissic and rhomb-porphyry rocks of Oslofjord yield radon levels all exceeding 100 Bq/1; some approach 1000 Bq/1. The gneisses typically exhibited 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). In the Iddefjord granite of the Hvaler area, the majority of the samples exceeded 1000 Bq/1 radon, reaching a maximum of 8500 Bq/1. These values must be regarded as high, in the context of proposed domestic water limits/action levels ranging from the USEPA's 11 Bq/1 to the Swedish 100 Bq/1 (Table 8). On Hvaler itself, many, but not all, of the users are vacationers, 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 borehotes should be evaluated.

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 $14-160 \mu g/l$ in drinking water can exceed acceptable limits of intake, and certainly the two maximum levels of 150 and 170 μ g/l, from boreholes in the Iddefjord granite, are regarded as being undesirable. The remainder of the samples lie under 41 μ g/l. Four boreholes on Hvaler yield water exceeding 1 μ g/l thorium. As thorium is generally regarded as being more radiotoxic than uranium, such concentrations cannot necessarily be disregarded in a health-related context. Although the distinctions between the Oslo rift and Trondelag 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 unusually high concentrations of uranium and thorium. It is interesting to note, however, that Killeen 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) that might warrant further investigation of 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). A wider survey of the possible health implications of radioelements in groundwater from Norwegian granites is long overdue. This survey should be based upon alreadyexisting geochemical and radiometrical studies of Norwegian bedrock.

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. Several methods exist that can effectively treat small-scale water-supply sources for these parameters. For radon, the following three methods (together with capital costs in the US dollars in 1988) are among the most appropriate; all are reported to give removal rates of above 95 percent (Nazaroff and others 1988; Kinner and others 1990): (1) granular activated carbon adsorption (\$850/household), (2) diffused bubble aeration (\$2000/ household), and (3) packed tower aeration. Other methods could include: (4) modification of external storage reservoir and distribution system to give increased degassing area and residence time, and (5) *in situ* aeration in the borehole, although Swedish experiences have proved rather negative (Gustav Åkerblom, SSI, personal communication).

Of these, methods 3 and 4 are probably more appropriate for somewhat larger waterworks. Users of high-radon groundwater should also be encouraged to ensure effective ventilation of rooms where water is used.

Methods that effectively remove up to 90 percent uranium 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.

Conclusions

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

The concentrations of uranium, thorium, and radon in bedrock groundwater can be correlated with geological province and 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 in comparison to the groundwaters of the Caledonian orogenic belt in Nord-Trondelag. Two "control" samples of groundwater from Quaternary deposits in these areas yielded low radioelement concentrations.

Weaknesses in the pilot study can be summarized as follows: (1) the sampling program was not adequately large or well-designed to allow epidemiological analysis; (2) the sampling program was insufficiently large to be representative for some lithologies (e.g., Quaternary, Precambrian gneisses in Trondelag); (3) several important lithologies were not represented in the Oslo area, notably the Permian intrusives and the Oslo graben sedimentary sequence; and (4) radium content was not analyzed.

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

the house, etc.). In groundwater from the Iddefjord granite, radon exceeded 1000 Bq/1 (the Swedish limit for remedial action) in eight of 11 samples. A similar pattern is observed for the various geological provinces and lithologies in respect to 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 with excessive concentrations of U and Rn should be treated using appropriate "domestic" technologies, which may include aeration, ion exchange, or active carbon adsorption.

The lddefjord granite is not the only bedrock substantially enriched in uranium and thorium in Norway; other granites that may be enriched in radioelements include members 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 allow an epidemiological analysis of cancer occurrence and possible correlations with: (1) groundwater usage in the home, (2) radioelement concentrations in groundwater, and (3) geology and tectonic situation.

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Appendix: 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 \times 10¹⁰ Bq. 1 mache unit = 3.6×10^{-10} Ci $\hat{A} = 13.3$ 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). $1 \text{ Sv} = 100 \text{ rem.}$

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