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THE CONTENT OF SOME NATURAL RADIOACTIVE ELEMENTS, ESPECIALLY ^{222}Rn, IN SOME POTABLE WATERS IN SWEDEN

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The content of ^{222}Rn in drinking water from 118 wells in southeastern Sweden has been measured. The radiation dose from ^{222}Rn is completely dominant in comparison with the dose from other nuclides examined. Only bored wells and especially those in granite bedrock have contents exceeding 20 nCi per l. Dug wells usually have contents less than 2 nCi per l.

As early as 1906 Hj. Sjögren and N. Sahlbom made radon measurements of the water from certain Swedish springs, mainly spas (1). These measurements were continued a few years later and a more systematic check was made of the radon contents in relation to the geological conditions. However, the work was confined to a few geologically limited regions, e.g. the Västgöta Hills, Dalecarlia, Scania and a few places in Norrland (2).

Measurements were made on water from bored wells and springs and the highest activities were reported from Vinterviken, Stockholm (164 M.U.), Bollstanäs (103 M.U.) and Boden Castle (172 M.U.), all these being from deep bored wells with depths of 45, 40 and 193 metres, respectively (1 M.U. = 0.364 nCi per litre). The remaining values were below 100 M.U., although in many cases the water was taken from wells which were deeper than those mentioned above. The conclusion drawn by N. Sahlbom from a geological point of view was that although local variations can occur, the wells bored in acidic rocks such as granites, syenites and pegmatites showed the highest radon contents while others, such as sedimentary rocks, gave water with only small contents of radon except for rocks in the vicinity of alum shales containing radium (Västergötland). The lowest contents were found in limestone and chalk deposits.

More recently, certain more limited areas have been investigated. Armands (3) measured the radon contents of surface waters in peat bog areas around Masugnsbyn, where contents of up to 0.3 μCi/l occur. von Döbeln & Lindell (4) reported results from radon measurements on water from 34 places in Sweden. The highest values in these results were 33nCi/l.

The factors which affect the concentration of radioactivity in water and the occurrence of naturally radioactive water are numerous and complicated. They are the result of interaction between the water and the surrounding rocks, involving leaching, emanation, solution, oxidation, adsorption, precipitation, etc. Furthermore, the meteorological conditions, the season and even the time of day can affect the activity of the water and especially its radon content (5). The hydrological aspects of the radioactivity of water are discussed in detail by Tokarev & Shcherbakov (6).

Most radioactive waters contain such small quantities of radium in relation to their radon contents that most of the radon must originate from other radium in the ground. In granite, which has a normal radium content of 10^{-12}g ^{226}Ra/g, ground water in equilibrium cannot have a radon content of more than approximately 600 pCi/l (7). However, since radon contents approaching 100 nCi/l are not unusual in some water, there must be an appreciably higher concentration of radium in the rock somewhere along the water's way to the spring or the well. The source of such radon has been discussed in detail in a work by K. Kikkawa (7) who points out, inter alia, that due to the low diffusion rate of radon through solid rock, the enrichment may take place in one of the following two ways:

a) water comes into contact with radium-bearing minerals only on the surface of the rock in contact with water,

b) water absorbs radon from radium in the capillaries of the rock, which requires that the rock be porous.

Since radon is a chemically inert gas, the radon content of the water after enrichment will be affected only by radioactive decay, emanation to surrounding air, and mixing with water with low radon content on the way to the well. The emanation and the mixing can be affected by meteorological factors. Water from melting snow and from autumn rains can, for example, cause minima or maxima in the radon contents of the water depending on the infiltration area of the well (8). Radium and other solid substances, on the other hand, may be affected by adsorption and precipitation effects. It is known that radioactive elements in low concentrations are adsorbed by various colloids which may subsequently be precipitated during the passage of the water through the rock – depending on the pH value and various other factors. Against this background

it is not improbable that the decay products formed from radon, RaA, RaB, etc. to RaD (^{210}Pb) and RaF (^{210}Po) suffer the same fate on their way through the rock. Nevertheless, relatively high contents of ^{210}Po have been found in some waters.

At the National Institute of Radiation Protection in Stockholm some measurements have been made on natural radioactivity, especially ^{222}Rn, contained in potable water in central Sweden, mainly from the point of view of radiation protection. The radioactive elements which were investigated were principally ^{222}Rn and, to a lesser extent, ^{226}Ra, ^{210}Pb and ^{210}Po. The results may be of general interest in view of earlier measurements mentioned above. Therefore they are reported more in detail than is motivated purely from the point of view of radiation protection.

The background of the investigations

The immediate cause of these investigations was the interest aroused by the occurrence of ^{222}Rn in certain samples of dairy milk which were collected during measurements of fallout activity. The discovery of ^{222}Rn in dairy milk was particularly interesting since the samples concerned came from a mixture of milk from a large number of farms. Earlier investigations of radon occurrence in milk had indicated that the ^{222}Rn had presumably been conveyed to the milk via radon-rich water drunk by the cows. The relationship between the contents of ^{222}Rn in the milk and in the water is such that the content in the water can be expected to be about 40 times that in the milk.

The highest content in the dairy milk was about 0.5 nCi/l; thus, application of the above factor of 40 should indicate an average content of 20 nCi/l in the drinking water at the farms. Earlier measurements on potable water from different parts of Sweden had shown that such high contents of radon are relatively rare and only to be found in water from deep-bored wells. It was therefore improbable that an average content of 20 nCi/l should have been caused by a widespread radon content of that order of magnitude. There was therefore every reason to suspect that the high average value could originate from a few wells with extremely high radon contents.

The primary objective of this investigation was therefore to determine whether or not there were in fact a few such wells in south-east Sweden with extremely high ^{222}Rn contents and in such case to locate them. The first area selected for investigations was that served by Vimmerby dairy, since one milk sample from that dairy had shown the highest radon content.

A questionnaire was sent to all farms supplying milk to Vimmerby dairy requesting information on the type of well from which they got their water (see Table 1) and on the number of cows. Water samples were then requested

Table 1
Results from the questionnaire on type of water source
(area served by Vimmerby dairy, winter 1966)

Source type	Number	Percentage
Deep-bored wells	41	12
Wells made partly by blasting into the rock	80	23
Wells made by digging	214	61
Natural springs	10	3
Surface water	4	1
Questionnaires not answered or incompletely answered	109	
Total	349 (458)	

from the farms which had deep-bored wells and which therefore had the highest probability of having influenced the average radon content of the dairy milk.

The next step consisted of field measurements in which all the farms in a limited part of the area served by the dairy were visited and the radon content of their water was measured using the Institute's mobile laboratory. When the results of that survey had been analysed, the mobile laboratory was used for an additional survey of the whole of the area served by the dairy but visited only farms which had either deep-bored wells or a large number of cows. On this occasion water samples were also taken for measurements in the laboratories in Stockholm.

During the summer, 65 per cent of the farms had access to surface water for their cattle, and for 35 per cent of all the farms, surface water was the only water which was available. Thirty-five per cent of the farms used subsurface water exclusively.

The final step in February 1970 consisted of a visit to about 20 farms, mostly with deep-bored wells and mostly with radon contents exceeding 10 nCi/l, for the collection of water samples in which the contents of ^{222}Rn, ^{226}Ra, ^{210}Pb and ^{210}Po were subsequently determined.

It will be shown below that the investigation of water supplies in the area served by Vimmerby dairy did not lead to the discovery of any exceptionally high radon contents. However, the highest value (72 nCi/l), found in a deep-bored well (38 m) is more than twice the highest value previously found (von

Döbeln & Lindell (4), measurements in Bergslagen) in measurements carried out at the National Institute of Radiation Protection and somewhat higher than the highest value found by Sahlbom (2) in more systematic measurements of water from wells in different geological regions. However, Sahlbom's measurements did not include Småland. It should also be observed that the average radon content was higher than had been anticipated and this fact, together with the relatively high frequency of deep-bored wells, can help to explain the radon contents found in the dairy milk. A wholly satisfactory solution, however, has still to be found.

Quite apart from the above, however, the results of the investigations will be presented here since knowledge of the contents of natural radioactivity in water is of scientific interest from the point of view of radiation protection. The radiation dose to the stomach, for instance, from radon daughters after the ingestion of water containing radon forms an interesting contribution to the natural radiation dose and this natural dose is one of the most important reference points when the significance of additional sources of radiation is being considered.

Methods of measurement and sources of error

Determination of ^{222}Rn were made both by using a mobile laboratory and by sending samples to the Stockholm laboratory. The field measurements were made with an ionisation chamber instrument (design Sievert and Håkansson) the output of which was fed to a pen-recorder which drew columns at regular intervals (every 6 seconds) on a continuous paper band. The height of the columns was dependent on the ionisation, i.e. on the radioactivity. The number of columns drawn for each sample was about 24. The mean height of these columns was determined and the radon concentration could then be determined with the aid of the calibration constant 0.7 nCi/l per mm net deflection. The total volume of the water sample for these measurements was 4.2 l. In the Stockholm laboratory an NaI crystal (4" × 4" diam.) was used with the sample bottle (about 6 l) standing directly on the crystal. The radon concentration could be determined from calibrations using the same type of bottle containing water with a known radon concentration.

The errors and uncertainties arising from the above measurements are the following:

Laboratory measurements

a) Statistical errors in the counting

Since radon was the only radioactivity (of any significance) present in the water,

the total count was used in the evaluation and the pure statistical errors are therefore small. The mean value of σ is 1.6 % with 0.2 % as the lowest value and 7.5 % as the highest value.

b) Loss of radon during collection of water samples in polythene bottles

The water samples were measured by standing the sampling bottle directly on the crystal and there was therefore no loss other than that which occurred during the filling of the bottle where the sample was taken. This latter loss cannot be determined with any certainty. However, by means of laboratory tests an average loss of 11 % has been estimated. All values from the laboratory measurements have been corrected for this 11 % loss.

c) Volume dependence

The water samples measured did not always have the same volume. The mean value of the volumes was 5.54 l with a standard deviation in the series of about 7 %. The calibration constant has dimensions pCi/l per count per unit time, i.e. the concentration is given and the error in the concentration corresponding to the above 7 % is less than 2.5 %. Earlier experiments using so-called "Marinelli bottles" containing solutions of ^{137}Cs have shown that changes of $\pm$ 6.6 % in the volume (about 5 l) resulted in variations of 2.5 % in the values for the concentration. In the radon measurements, with the bottle standing on the crystal, the error would be smaller.

d) Correction for decay

For the cases where water samples were supplied by post, the correction for decay was made using the time difference from the date of postmark to the time of measurement. Measurements were made between 10.00 hours and 16.00 hours and the decay time t was calculated in units of 24 hours back to the postmark date. On the reasonable assumption that the water sample had been taken on the same day or at the earliest on the day before the postmark date, we have $t - 6 \text{ hours} < t < t + 30$ hours, i.e. the "correct" value R (from this point of view) in relation to the given value A must lie within the limits $0.96\ A < R < 1.25\ A$.

The field measurements

a) Errors in the readings from the pen-recorder

When using the ionisation chamber, the radon content was determined from the mean height of about 24 recorded "columns" in relation to the height recorded for a given background. The position of the mean height was determined by eye and drawn in with a ruler. The error in the reading therefore consists of two errors, one in the visual determination of the mean height and the other in the measurement of the net height. These errors have been investigated in a series of particularly exact measurements and together with other uncertainties

(background variations, etc.) have resulted in every calculated value of the radon content having an uncertainty varying between 1 and 2 nCi/l. This means that individual values of < 2 nCi/l are very uncertain. However, the uncertainty is somewhat less for the mean value of groups of values of, say, < 2 nCi/l, since the random variation of the individual errors between positive and negative values means that they cancel one another out statistically.

b) Losses while filling the sampling bottles

As in the case of the measurements in Stockholm, there is an uncertainty here due to the radon loss during filling. The values have been corrected for an estimated loss of 11 %.

c) Volume dependence

This was not a source of error since all samples had the same volume.

d) Correction for decay

No correction for decay was necessary since the samples were measured on the spot. However, it should be noted that the samples were generally measured before gamma equilibrium was obtained in order to save time. Since in many cases parallel measurements were made in the Stockholm laboratory, corrections could be made for this. The correction factors which were determined in this way and then applied to samples which were only measured in the field imply a degree of uncertainty particularly in the low values. This source of error, however, is not normally dominant.

Determination of ^{226}Ra, ^{210}Pb and ^{210}Po.

In the last sampling programme, not only the radon contents but also the contents of ^{226}Ra, ^{210}Pb and ^{210}Po in water from 19 wells were measured, the wells being selected because of their high radon contents in earlier sampling programmes. Most of these wells were deep boreholes.

The water samples were obtained during visits to the places concerned. In order to avoid water which had been standing still in the distribution system, the water was allowed to run for some time before the sample was taken. For the ^{210}Pb and ^{210}Po analysis, 1.2 l of water were taken (0.5 M due to the addition of 50 ml conc. HCl). The water sample was immediately boiled for 10 minutes, after which 140 ml distilled water was added. This procedure eliminated more than 95 % of the radon. For the radium and radon determination, a 5 l sample was taken (0.5 M due to addition of 240 ml conc. HCl). The addition of acid is essential in this connection in order to prevent adsorption on the walls of the bottle, particularly of ^{210}Po. The chemical analyses and measurements were made in the laboratory.

The ^{210}Pb analysis was carried out as follows:

A calcium carrier (60 mg Ca) was added to a 400 ml water sample and then precipitated together with lead as a phosphate. The precipitate was dissolved and boiled in concentrated nitric acid in order to destroy organic material (9). A lead carrier (20 mg Pb) was added and after a complete chemical separation with lead as the only remaining metallic ion, the lead was precipitated as chromate (10).

After weighing and computation of the chemical yield (50–85 %), the precipitate was moved to a standard planchette and covered with plastic foil (6 mg/cm^2). With a covering foil of this thickness, the β-irradiation from ^{210}Pb (85 % E_{max} = 0.015 MeV, 15 % E_{max} = 0.061 MeV) and the α-irradiation from growing ^{210}Po (5.3 MeV) are both absorbed. The ^{210}Bi (E_{max} = 1.16 MeV) can then be measured and its growth can be followed until equilibrium with ^{210}Pb is reached after about 25 days. The counting efficiency for ^{210}Bi under these conditions is 39 % in a low-β counter of Intertechnique type RA-12.

Analysis of the ^{210}Po was carried out in accordance with Black's method (11): ^{210}Po was precipitated self-electrolytically from 200 ml samples (0.5 M due to addition of HCl) on silver foil with a diameter of 30 mm. The yield of ^{210}Po in all experiments in which known quantities of ^{210}Po were added was > 95 %. The counting efficiency with a ZnS detector was 35 %.

The radon contents of the water samples were determined by means of gamma-spectrometry directly on the sampling bottles. The radium determination was carried out using a selective adsorption method. This method is based on

Table 2.
^{210}Pb and ^{210}Po in H_3PO_4 of different makes

Manufacturer	Activity		
Merck, Germany	0.8 – 1.1	pCi	^{210}Po/ml
Pro analysi	–"–		^{210}Pb/ml
Riedel–De Haën A. C., Germany	1.5	pCi	^{210}Po/ml
Pro analysi	"		^{210}Pb/ml
Fisher, USA	0.5	pCi	^{210}Po/ml
	"		^{210}Pb/ml
La Fonte, Switzerland	< 0.003	pCi	^{210}Po/gram
Puriss, crystallized	< 0.01	pCi	^{210}Pb/gram
	0.5	pCi	^{212}Pb/gram

Table 3.
Distributions of radon contents in wells of different types

^{222}Rn content (nCi/l)	Number of wells in group		
	Bored	Partly blasted	Dug
$\leqq 2$	9	9	47
2 – 10	16	6	19
10 – 20	6	0	0
20 – 30	3	0	0
30 – 50	2	0	0
> 50	1	0	0
Total	37	15	66

the selective adsorption of radium ions from solutions to a barium sulphate precipitate which is fixed in a gelatine layer on a glass plate (12). The α-irradiation from the adsorbed radium is then measured with a ZnS detector with a low background.

The following source of error is relevant in addition to the adsorption effect for ^{210}Po already mentioned. About 4 ml of phosphoric acid (H_3PO_4) was used in the determination of ^{210}Pb. Activity analyses of phosphoric acid of different makes showed that in certain cases they contained about 1 pCi ^{210}Pb/ml. In one case ^{212}Pb was present. The activity contents of phosphoric acid of different makes is shown in Table 2.

When making low-activity measurements of ^{210}Pb it is therefore most important to make note of this source of error and to use suitable phosphoric acid. Phosphoric acid is made from fluoride apatite, $Ca_5(F_1Cl_1OH)\ (PO_4)_3$, originating in Algeria, Tunisia and the U.S.A. Much of the radioactivity in igneous rocks is known to be concentrated in the mildly radioactive accessories, zircon, sphene and apatite (13).

The results of the radon determinations

The wells investigated have been subdivided into three groups: dug wells, wells partly formed by blasting, and bored wells. Of these types, dug wells in particular could be expected to differ from bored wells in the areas from which they collected water, and a comparison between them should be of interest from

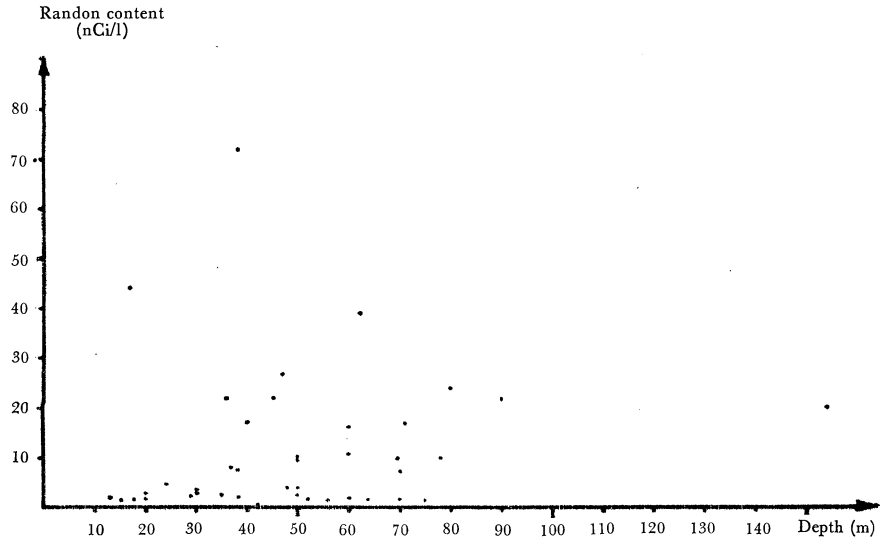

Fig. 1.
Radon contents of bored wells vs. the depth of the well.

that point of view. The mean depth of dug wells was 4 metres (1–11 m), for partly blasted wells it was 7 metres (3–11 m), and for bored wells 50 metres (13–154 m).

Table 3 shows how the radon contents were distributed for the various types of wells.

The table shows that radon contents of $>$ 10 nCi/l occur only in deep-bored wells. The highest value found was 72 nCi/l; the depth of the well concerned was 38 metres.

In Fig. 1 the radon contents of bored wells and the depth of the well have been plotted. It can be seen that there is no correlation between the radon content and the well depth ($r = 0.1$).

In order to determine whether there is a geological relationship in the radon contents, the geological maps have been studied and the wells have been further divided according to the geology of the infiltration area of the wells. In Fig. 2a-c the wells are divided according to the type of bedrock without regard to the type of soil. In Fig. 2d the dug wells over granitic bedrock have been further divided according to the type of soil.

The following conclusion can be drawn from Fig. 2.

1) Amongst the bored wells, only those with a granite bedrock have radon

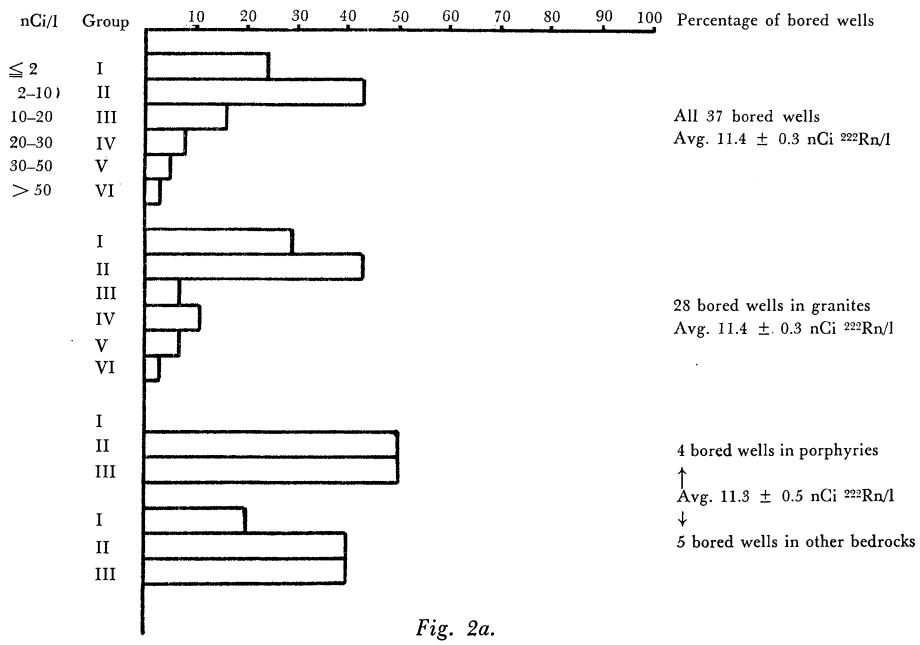

Fig. 2a.
^{222}Rn in bored wells.

contents exceeding > 20 nCi/l. Sub-division according to the type of soil would be meaningless since the great majority of these wells were in morainic soil (of the three wells in gravel, however, there was one with a radon content of > 20 nCi/l). It is not probable that the type of soil had any relevance in the case of bored wells.

2) Blasted wells form an intermediate class from the point of view of the collecting area. Both the type of bedrock and the type of soil cover can be expected to be significant. For these wells there was also a tendency for the average radon content to be higher when the bedrock was granite than for the others, although none of the wells had a radon content exceeding 20 nCi/l.

3) Even for dug wells a tendency was found for the average radon content to be higher when the bedrock was granite than for other types of rock. The relative difference between the number of wells in groups I and II (see Fig. 2) is, however, larger for the dug wells than for the blasted ones. For the latter type the number in group II is about the same as in group I.

4) From Fig. 2 d it can be seen that for dug wells over a granite bedrock those in till (morainic soil) have a higher proportion in group II than those in gravel, i.e. the average radon contents of their water is higher.

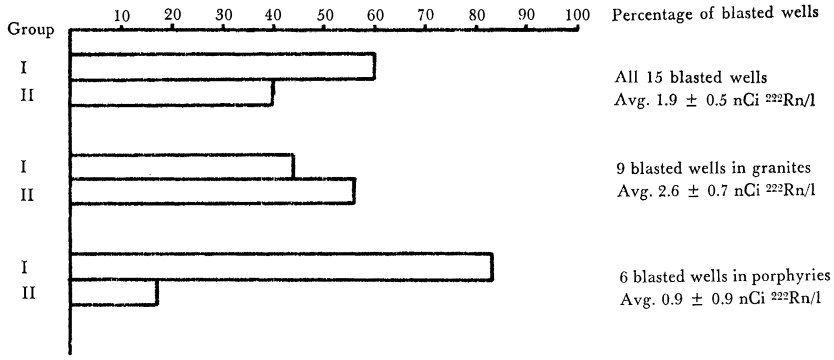

Fig. 2b.
^{222}Rn in blasted wells.

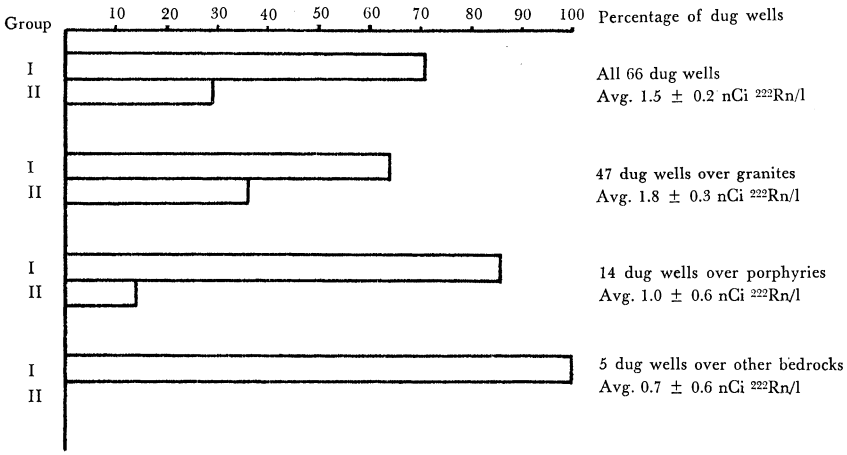

Fig. 2c.
^{222}Rn in dug wells.

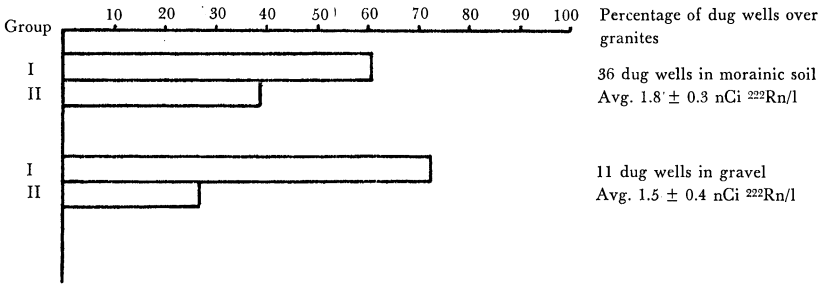

Fig. 2d.
^{222}Rn in dug wells in different soils over granites.

These observations are substantially the same as those found by Knutsson (8) in his measurements made in an adjacent area. The last observation, no. 4, requires a comment. It might be thought that the type of bedrock was of no direct significance for dug wells. However, till, in particular, contains mostly particles of the local bedrock. Gravel soils of this area have often been transported by glacial streams and the particles may therefore originate from rocks a considerable distance away which are not necessarily granite. An alternative explanation might be that till material emanates radon to the surrounding water more effectively, since the average size of its particles is smaller than that of gravel. Till is also less permeable and this results in a smaller loss of radon to the air.

This has practical significance from the point of view of radiation protection. The overwhelming majority of communal water supplies in Sweden originate from surface water or from wells dug in gravel. These observations and conclusions thus imply that radon contents of the communal water supplies of the surrounding areas can be expected not to show especially high radon contents ($>$ 10 nCi/l) but rather to have lower contents than dug wells in general.

When determinations of ^{226}Ra, ^{210}Pb and ^{210}Po were being made for water from bored wells in February 1970, the radon contents also were measured. From Table 4 below it can be seen that the radon contents were in general very similar to those found from the previous sampling during September–October 1966.

It also appears that the fact that the average temperature of the samples in February 1970 was 3^{0} C lower than that in September–October 1966 had no significance either. In dug wells, on the other hand, the radon contents may show a seasonal variation depending on the precipitation, the melting of the snow cover and the ground temperature (8).

The results of the determinations of ^{226}Ra, ^{210}Pb and ^{210}Po

The results from these determinations are shown in Table 4.

The conclusions which can be drawn are as follows:

a) The radium content is low compared with the radon content. There is no simple relationship between the radium content and the radon content. The radium content of a water sample therefore seems to be of no significance to its radon content, an observation which is not new (14). The mean value of the radium content in these radon-rich waters is 0.56 pCi/l, i.e. a value which lies within the normal values which would apply to drinking water reported in (15). As a comparison, it may be mentioned that drinking water in Stockholm has a radium content of 0.1 pCi/l.

b) The content of ^{210}Pb and especially of ^{210}Po is also low compared to the radon content. This either means that the age of the water from the time of its radon enrichment is low or that lead and polonium have been precipitated on their way to the well. That latter possibility is not improbable, particularly in the case of polonium. The actual content of ^{210}Pb, however, is higher than is usual in drinking water. The values given in (15) are below 0.1 pCi ^{210}Pb/l. At present there are no representative values for normal ^{210}Pb content in drinking water in Sweden. In a small number of measurements recently made on the drinking water in the Stockholm area and its surroundings (within 100 km), the values given in Table 5 were obtained.

Table 4.
^{222}Rn, ^{226}Ra, ^{210}Pb and ^{210}Po in some deep-bored wells in south-east Sweden

Well no.	Depth (m)	^{222}Rn Sept.–Oct. 1966 (nCi/l)	^{222}Rn Feb. 1970 (nCi/l)	^{226}Ra (pCi/l)	^{210}Po (pCi/l)	^{210}Pb (pCi/l)	Po/Pb
15	62	39	38	0.16	< 0.2	1.3	< 0.16
47	47	27	26	0.56	< 0.2	1.6	< 0.13
38/40	154	20	24	0.3	0.6	2.3	0.26
63	80	24	42	0.59	< 0.3	1.3	< 0.23
187	20	–	15	0.8	–	–	–
216	50	10	13	0.58	2.0	2.6	0.78
217	50	10	10	0.34	0.2	< 0.2	–
230*)	36	22	< 0.3	0.18	< 0.2	< 0.2	–
238**)	6	5	4	0.25	< 0.2	0.5	< 0.4
283	45	22	22	0.62	< 0.2	1.3	< 0.16
289	37	8	6	0.16	< 0.2	< 0.2	–
290	71	17	15	0.14	< 0.2	0.9	< 0.23
293	90	22	22	0.22	< 0.1	< 0.3	–
364***)	5	7	11	0.75	0.6	2.0	0.3
368*)	40	17	16	1.3	1.1	11.6	0.1
381	38	72	80	2.5	< 0.1	4.0	< 0.03
518	60	16	15	0.64	4.1	0.8	5
617	17	44	43	0.29	< 0.2	< 0.2	–
623	78	10	13	0.18	< 0.3	< 0.3	–

*) Different wells were used in the two sampling programmes. However, the two wells involved were of the same depth and were close together.
**) A blasted well.
***) A dug well.

Table 5.
^{222}Rn, ^{210}Pb and ^{210}Po in drinking water in the Stockholm region

Place	^{222}Rn content (pCi/l)	^{210}Pb (pCi/l)	^{210}Po (pCi/l)
Märsta	400	< 0.3*)	< 0.06*)
Stockholm	< 60*)	< 0.3	< 0.2*)
Svartsjölandet	< 60	< 0.3	0.2
Sigtuna	< 60	< 0.3	< 0.2
Uppsala	1000	< 0.3	< 0.2

*) Below detection limits.

From Table 5 it can be seen that the "normal values" for ^{210}Pb and ^{210}Po in ordinary water can be expected to lie below the detection limits.

c) The values for the concentration of ^{210}Pb show considerable spread but also a positive correlation to those for the ^{226}Ra concentration. However, the correlation is only moderate ($r = 0.6$). This does not correspond with the results reported by Holtzman (16) who showed a negative correlation in a material which also had considerable spread in the values. He also showed a negative correlation with the depth of the well. In the material presented in this report there is no correlation whatever ($r < 0.1$) with the well depth.

d) The ratio of ^{210}Po to ^{210}Pb is as a rule less than 1. In one case, however, (well No. 518), a surprisingly high ratio of 5 was obtained. Further samples will be taken from that well for detailed studies.

The radiation dose

In the samples of drinking water investigated, which had radon contents between 4 and 80 nCi/l (and one < 0.3 nCi/l), the ^{210}Pb content was measurable in 12 cases (of 18), being between 0.5 and 12 pCi/l with a mean value of 2.5 pCi/l. The ^{210}Po content was measurable in only six cases. From the point of view of radiation dose this means that the intake of ^{210}Pb via water could be up to 100 times greater than normal. The consequences of this from the point of view of dose cannot be determined with any great accuracy due to a number of uncertain factors concerning, inter alia, the metabolism of lead in the human body. For the sake of comparison, however, one can apply a treatment which is analogous to that used by Holtzman (17). He considers the contributions of va-

rious sources to the ^{210}Pb content of the human skeleton and finds that consumption of 1.5 litres of water containing 0.4 pCi ^{210}Pb/l will contribute about 12 % of the ^{210}Pb content of the human skeleton. Using 10 pCi ^{210}Pb/l in drinking water as a starting point, the assumptions imply that drinking water accounts for nearly 80 % of the resulting dose of approximately 100 mrem/year (instead of the corresponding 30 mrem/year according to Holtzman).

It is of interest to consider the magnitude of these radiation doses compared to the radiation dose contribution due to radon in water. In this case the critical organ is the gastro-intestinal tract. According to von Döbeln & Lindell (4), 100 nCi ^{222}Rn/l corresponds to one MPC with the ICRP assumption regarding the intake of 2200 cc of water per day, etc. Since the greatest part of this water is ingested in food, coffee, etc., a large proportion of the radon will have left the water before ingestion. The amount of water drunk directly can be estimated to about one-tenth of the total water intake, i.e. about 200 cc per day. This leads to a radiation dose of about 1500 mrem/year in the gastro-intestinal tract. The highest radon content in the drinking waters investigated was 80 nCi/l; this would result in about 1000 mrem/year. The radiation dose due to radon is thus still the dominating one.

As radon measurements are much easier to perform than measurements on ^{210}Pb and ^{210}Po, it should be of interest, still from the point of view of radiation dose, to be able to predict the expected maximum levels of these nuclides from the result of radon measurements, or at least appraise the probability that the concentration of ^{210}Pb (and ^{210}Po) should exceed some settled value. If it is

Table 6.

Radon content in the well water (nCi/l)	Required residence time (days)
430	0.25
210	0.5
100	1
45	2
27	3
12	5
4	10
1.4	15
0.6	20

assumed that ^{210}Pb and ^{210}Po contents occur due to the decay of radon in the water, it is clear that high primary radon contents are required to make the ^{210}Pb and ^{210}Po contents significant. Total decay of 1 nCi ^{222}Rn/l produces 0.5 pCi ^{210}Pb/l. This activity is produced in 1.3 hours in water with an initial content of 100 nCi ^{222}Rn/l, in 14 hours in water with a content of 10 nCi ^{222}Rn/l, and so on. If the content of ^{210}Pb must exceed 10 pCi/l before it is significant from the point of view of radiation dose, it follows that at least 20 nCi ^{222}Rn/l must have decayed completely in the water. With the assumption that the water receives its radon content within a locally limited region on its way through the rock (14) and that the radon content then decreases due to radioactive decay on the way to the well, the following relationship can be established between the required residence time in the rock and the radon content in the out-going water (see Table 6) if the condition that 20 nCi ^{222}Rn/l shall have decayed is to be fulfilled.

The implication of the figures in Table 6 is that if, for instance, the measured radon content is 100 nCi/l, the water must have an "age" of at least one day, from the time of radon enrichment, if the content of ^{210}Pb in the water is to exceed 10 pCi/l. This, however, is purely a physical-mathematical analysis of an idealized condition. The true situation is certainly much more complicated; for instance, precipitation of activity reduces the content of ^{210}Pb. The quantitative occurrence of possible precipitation in certain waters has been shown by experience in direct measurement of radon contents in the field. In some cases the radon daughters were not in equilibrium with the radon immediately after the sample was taken. This implies either a very rapid replacement of the water or a rapid precipitation of the radon daughters.

The values of required residence time given in Table 6 are, however, minimum values. Since smaller radon contents require longer residence times in the rock if the same ^{210}Pb content is to be obtained, the probability of relatively higher precipitations of ^{210}Pb and ^{210}Po is also increased. It is thus reasonable to conclude that radon concentrations below about 100 nCi/l will hardly involve radiation protection problems with regard to ^{210}Pb and ^{210}Po.

SUMMARY AND CONCLUSIONS

The content of ^{222}Rn in drinking water from 118 wells has been measured and in water from 19 of these wells the content of ^{226}Ra, ^{210}Pb and ^{210}Po has also been examined. The conclusions are

1) Only bored wells and especially those in granite bedrock have radon contents which exceed 20 nCi/l.

2) Dug wells usually have low radon contents (< 2 nCi/l). Wells on granite bedrock and in till have higher average radon contents than do others.

3) Communal wells (each supplying water to more than 200 people) are most often dug in sand and gravel and can therefore be expected to contain low radon concentrations.

4) The contents of ^{226}Ra, ^{210}Pb and ^{210}Po in water from bored wells are low compared to ^{222}Rn. There are no significant correlations between the contents of these nuclides or between their contents and the depths of the wells.

5) The radiation dose from ^{222}Rn is completely dominant in comparison with the radiation dose caused by the other nuclides examined.

6) There is reason to believe that water with a radon content lower than about 100 nCi/l does not contain ^{210}Pb and ^{210}Po of any significance as regards radiation dose.

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REFERENCES

1. Sjögren, Hj. & Sahlbom, N. (1907) Arkiv för kemi, mineralogi och geology, Bd. 3, 1907.
2. Sahlbom, N. (1916/17) Om radioaktiviteten hos svenska källvatten och dess samband med de geologiska förhållandena. *Arkiv för kemi, mineralogi och geologi,* Bd. 6.
3. Armands, G. (1961) Geochemical prospecting of a uraniferous bog deposit at Masugnsbyn, Northern Sweden. Atomic Energy Company Report, AE-36, 1961.
4. von Döbeln, W. & Lindell, B. (1964) Some aspects of radon contamination following ingestion. *Arkiv för Fysik,* Bd. 27, Nr. 32.

5. Kikkawa, K. (1954) Study on Radioactive Springs. *Jap. J. Geophysics 1* (1).
6. Tokarev, A. N. & Shcherbakov, A. V. (1960) *Radiohydrogeology, Moscow, 1956.* Translation Series AEC-tr-4100.
7. Kikkawa, K. The Sources of Radon to the Radioactive Springs. Geophysical Institute, Kyoto University.
8. Knutsson, G. (1970) Personal communication.
9. Harley, J. Manual of standard procedures. NYO-4700 (2nd ed.) U.S. Atomic Commission (1967). Procedure E-Pb-01-01.
10. The radiochemistry of lead. NAS–NS 3040. U. S. Atomic Energy Commission (1961). Procedure No. 11.
11. Black, S. C. (1956) Low level Polonium determination of tissue and urine. Univ. of Rochester. Atomic Energy Project. Report UR 463. Rochester N. Y. October 1956.
12. Kulich, J.: Unpublished data.
13. Nuclear Geology (1954) A Symposium on Nuclear Phenomena in the Earth Sciences. Edited by Henry Faul.
14. Tanner, A. B. (1964) Physical and Chemical Controls on Distribution of Radium-226 and Radon-222 in Ground Water near Great Salt Lake, Utah. In "The Natural Radation Environment" edited by John A. S. Adams and Wayne M. Lowder.
15. Report of the United Nations Scientific Committee on the Effects of Atomic Radiation. UN New York 1966.
16. Holtzman, R. B. (1964) Lead-210 (RaD) and Polonium-210 (RaF) in Potable Waters in Illinois. In "The Natural Radiation Environment" edited by John A. S. Adams and Wayne M. Lowder.
17. Holtzman, R. B. (1963) Measurement of the Natural Contents of RaD (Pb-210) and RaF (Po-210) in Human Bone – Estimates of Whole-Body Burdens. Health Physics. Vol. 9, No. 4, April, 1963.

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