INGESTED RADON AS A SOURCE OF HUMAN RADIATION EXPOSURE

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Abstract—^{222}Rn is usually mentioned as a source of radiation exposure of man in connection with inhalation rather than ingestion. However, since radon is a natural contaminant of ground water, it is of interest to study also the doses caused by intake with water. For ingestion of water containing radon, the stomach is the critical organ. The radon retention in the body is mainly determined by the transport from stomach to blood, since radon is rapidly eliminated from blood. After 4 hr only a few per cent of the ingested amount remains in the body, but is then likely to be eliminated at a lower rate, probably because of the high solubility in adipose tissues. The dose equivalent per 1 μCi radon ingested is estimated at 200 mrem, corresponding to $(MPC)_w = 10^{-4}$ μCi/ml (100 nCi/l.). Concentrations of up to 5000 nCi/l. have been reported in the literature. A frequent value for tap water from ground water supplies is 1 nCi/l. in many countries, while waters from deep bored wells frequently show concentrations of the order of 10 nCi/l. It is interesting to note that radon concentrations of up to 1 nCi/l. have been observed in some dairy milk.

INTRODUCTION

Although there are numerous reports on the content of ^{222}Rn in drinking waters (distributed in time from 1904, when one paper was published by Mme Curie(1) up to the present no value for the maximum permissible concentration of this nuclide has been proposed until recently (Bernard,(2) Andersson and Nilsson,(3) von Döbeln and Lindell,(4) and Hursh *et al.*(5)), with the exception of a value given in 1953 by the United States National Committee on Radiation Protection in NBS Handbook 52(6) but not repeated in later NCRP reports. The International Commission on Radiological Protection does not recommend any value for $(MPC)_w$.

There are several obvious reasons for this, one being that radon in water rarely constitutes an occupational risk since it is usually naturally occurring. Another reason is that the radon does not remain in the water very long, and that it stays an even shorter time in the body once ingested. Already in the 1910's the apparent mean residence time was found to be less than one hour, and in the 1930's detailed studies of the radon retention was published, e.g. by Stefen Meyer(7) and by Fernau and Smereker.(8)

With the present attitude towards radiation protection, however, even radon in water has become of some interest from the radiation protection point of view. Protection efforts are not only directed towards the radiation worker; ICRP has given "dose limits" also for the general public, "intended to provide standards for the design and operation of radiation sources so that it is unlikely that individuals in the public will receive more than a specified dose".

These dose limits have not been designed to apply to naturally occurring radiation sources or, in fact, to any *occurring* situation. For such situations it is countermeasures rather than planning that are required. However, the $(MPC)_w$-values are based also upon the doses per unit intake of activity, and the results of such calculations are also needed in the assessment of the possible risk from a given source. Therefore, the doses due to intake of radon with drinking water justify some investigation. The doses may not always be entirely insignificant: it has been shown that there exist in nature drinking waters that may give local tissue doses as high as 1 rem/day at a water consumption of 1 l./day, i.e. 1 rem per liter consumed.

LEVELS OF ^{222}Rn IN WATER

A number of relatively recent reviews (Federov and Baranov,[9] Hursh *et al.*,[5] Muth *et al.*,[10] Kiefer and Maushart,[11] Turner *et al.*,[12] Smith *et al.*,[13] von Döbeln and Lindell[4]) and old reports (Sahlbom,[14] Stephan,[15] Engelmann,[16] Ludewig,[17,18] Genser[19,20]); mostly on spa waters, relatively portray the frequency of various ^{222}Rn levels in ground water.

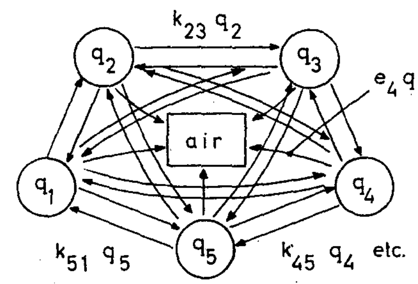
Sahlbom found already in 1915 that typical Swedish values for the radon concentration in ground water in sedimentary rocks ranged between 0.7 and 1.1 nCi/l., while wells in igneous rocks usually covered the range 2–20 nCi/l. Kiefer and Maushart found levels between 0.1 and 2.5 nCi/l. in tap waters from a number of German towns which use ground water or springs. Surface waters usually show less than 0.05 nCi/l. Turner *et al.* found an average of 1.1 nCi/l. in Cornish waters, while they assumed that the average for the whole of Britain is only 0.002 nCi/l. Smith *et al.* found average concentrations of 16 and 30 nCi/l. in wells in Maine and New Hampshire, respectively, with maximum values as high as 200 nCi/l. The highest levels found in Sweden are near the same level (Armands[21]).

Spa waters in Oberschlema in Germany have been reported by Kuroda[22] and by Genser[20] to hold activities ranging from 100 to 5000 nCi/l.

The radon content of the drinking water has been suggested by Allen-Price[23] as a possible cause of an alleged higher than normal cancer frequency in certain parts of West Devon, but these levels as reported by Abbatt *et al.*[24], were not found to be higher than 13 nCi/l.

THE FATE OF THE INGESTED RADON

In 1933, Fernau and Smereker[8] gave an exponential expression for the radon expiration rate after ingestion with water. Their expression contained two exponential terms and represented a rapid increase, with a time constant $k_A = 0.149$ min^{-1}, followed by a more slow decrease, corresponding to a time constant $k_B = 0.023$ min^{-1}. Fernau and Smereker correlated the time constants to the elimination from the gastrointestinal tract and the blood, i.e. from two compartments. A more general multi-compartment analysis will be needed to illustrate the phenomenon over a longer time period. The general circuit graph of a five compartment system would be as follows



The *amount* of radon transferred from compartment (m) to compartment (n) per unit time can be written as

$$R_{mn} = k_{mn}q_m - k_{nm}q_n$$

where the q's denote the quantity of radon in the various compartments.

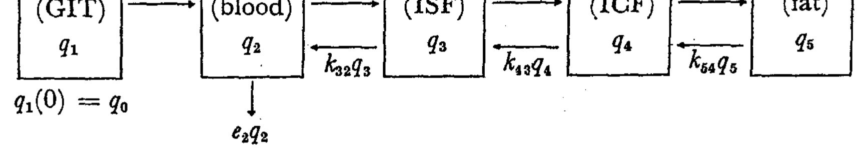
The radon transport will be governed by five differential equations, namely

$$\frac{dq_1}{dt} = -(k_{12} + k_{13} + k_{14} + k_{15} + e_1)q_1 + k_{21}q_2 + k_{31}q_3 + k_{41}q_4 + k_{51}q_5;$$

$$\frac{dq_2}{dt} = +k_{12}q_1 - (k_{21} + k_{23} + k_{24} + k_{25} + e_2)q_2 + k_{32}q_3 + k_{42}q_4 + k_{52}q_5;$$

. . . etc.

where the e's denote elimination constants. A detailed analysis of this system has been reported by von Döbeln and Lindell.[4] A rough approximation of the circuit graph can be made as follows:



The analysis yields curves of the type shown in Fig. 1. In addition to the total retention curve, the retention is also shown in the following five compartments: blood (B), stomach (S), extracellular fluids (ECF), intracellular fluids (ICF) and fat (F). The broken curve represents the retention in fat calculated on the basis of experimental data presented by Hursh *et al.*(5) and Harley *et al.*(25) The long-term retention is entirely governed by the fat content.

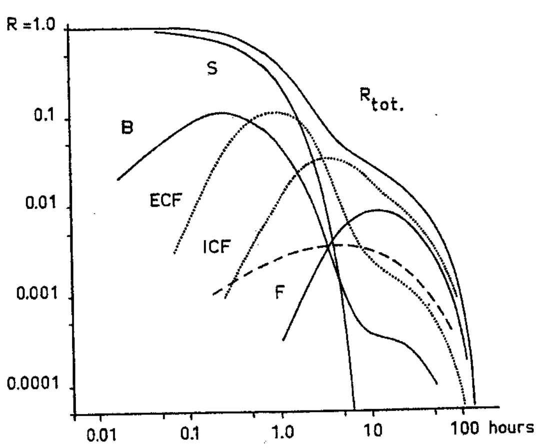


FIG. 1.

The mathematical model, used here only for illustrational purposes, reflects facts that were reported and well understood already by Meyer in 1937.(7) The radon is transferred from the stomach to the blood at a rate which varies depending upon whether the water was ingested on a full or on an empty stomach. Fat in the stomach will decrease the rate of transfer because radon is more soluble in the fat which is also more slowly absorbed than the water. An equilibrium between the radon in the fat and in the intestinal fluids will probably not be attained until the fat is in emulsion after the influence of bile and other intestinal fluids.

Once the radon has reached the blood, it is eliminated very rapidly through the lungs, with an effective half-time of only about one minute. While in the blood, the radon is retained possibly by adsorption and absorption processes.

After four hours, only a few per cent of the ingested amount remains in the body. The remaining radon is mainly located in the adipose tissues, because of the higher solubility in fat. This has been demonstrated on the rat by Nussbaum and Hursh,(26) in a study of the distribution between different organs under equilibrium conditions.

Figure 2 shows some examples of retention curves for ^{222}Rn after a single ingestion, as suggested by various authors. Curve 1 represents a single exponential function assumed by Andersson and Nilsson.(3) Curve 2 illustrates the double exponential expression suggested by Fernau and Smereker.(8) Curves 3 and 4 correspond to data obtained by Hursh *et al.*(5) for an empty and a full stomach respectively. Curve 5 can be obtained from the compartment analysis discussed above, on the basis of experimental data presented by Harley *et al.*(25)

Retention measurements by von Döbeln and Lindell(4) on subjects who had ingested radon with water on an empty stomach yielded the data shown in Fig. 3. The technique did not permit measurements for more than about five hours. The diagram actually shows the body content of the daughter product ^{214}Bi (RaC) as measured by whole-body counting. The full

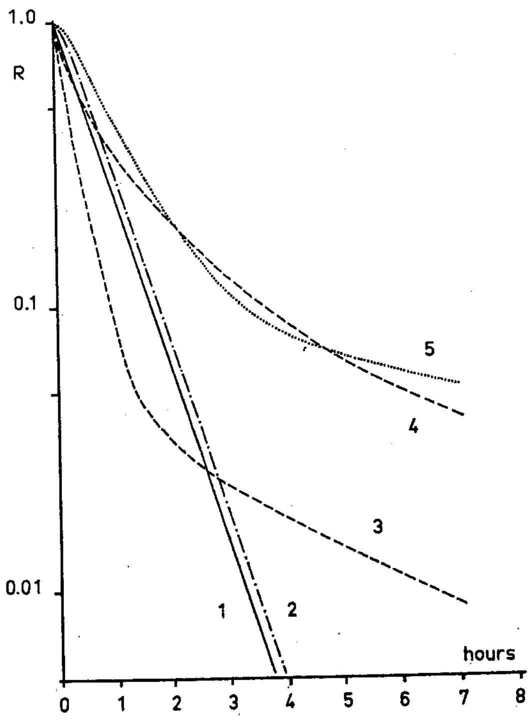


FIG. 2.

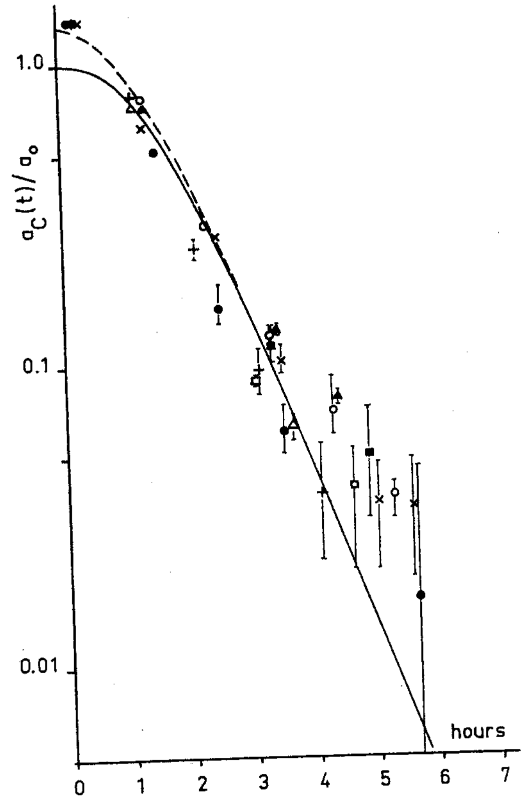


FIG. 3.

curve represents the body content of ^{214}Bi calculated on the assumption of a single exponential retention of ^{222}Rn ingested in initial equilibrium with ^{214}Bi, while the broken curve is based upon the assumption that 25% of the radon gas is lost initially. The experimental points have been normalized to make the average retention between 3 and 4 hr fall upon the calculated curve.

The long-term retention is only vaguely indicated by these data, but experiments aimed at measuring the retention at more than five hours after ingestion will be carried out.

MAXIMUM PERMISSIBLE CONCENTRATIONS

On the basis of the information referred to above, the radiation doses to various organs and hence also values for $(MPC)_w$ can be calculated. The results derived by von Döbeln and Lindell are shown in Table 1:

Table 1. *Suggested* $(MPC)_w$ *Values for* Rn^{222}

Organ of reference	Dose equivalent per 1 μCi radon ingested (mrem)	$(MPC)_w$ (μCi/ml)
Critical organ		
GIT (stomach)	200	10^{-4}
Other organs		
Liver	7	3.10^{-3}
Fat	6	3.10^{-3}
Whole body	2	3.10^{-3}
Lung	2	10^{-2}
Kidney	1	2.10^{-2}

The stomach is the critical organ with a dose equivalent to the stomach wall of 200 mrem per μCi radon ingested and $(MPC)_w = 10^{-4}$μCi/ml. This value of $(MPC)_w$ is 50 times as high as the value once suggested in NBS Handbook 52, but it is in accordance with the estimate by Hursh *et al.*[5] It is 1/5 of the value proposed by Bernard[2] who, however, suggested fat as critical organ, and 1/9 of the value proposed by Andersson and Nilsson.[3] Both the latter values seem to have been derived on the unrealistic assumption that all energy delivered within the body is dissipated in living tissues. In fact, 50–75% of the energy is delivered in the stomach content and in the mucus, even if the energy from the α-emitting daughters which fail to penetrate the mucus is completely neglected.

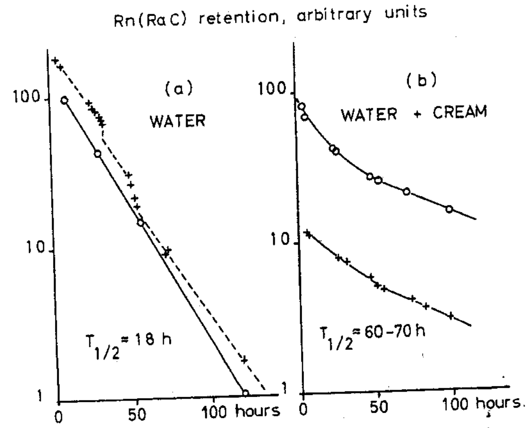


FIG. 4. Rn(RaC) retention, arbitary units.

Table 2. Bunsen and Ostwald Solubility Coefficients for Inert Gases in Water and Olive Oil at 37°C (*Ref. Lawrence et al.*, 1946; *last entry: Nussbaum*, 1957.)[29]

Gas	Molecular weight	Water	Olive oil	Oil/Water ratio
		Bunsen absorption coefficients (α)		
Helium	4	0.0085	0.015	1.7
Neon	20.2	0.0097	—	—
Nitrogen	28	0.013	0.067	5.2
Argon	39.9	0.026	0.14	5.3
Krypton	83.7	0.045	0.43	9.6
Xenon	131.3	0.085	1.7	20.0
Radon	222	0.15	19.0	125.0
		Ostwald absorption coefficients (αI)		
Radon (from above):		0.17	21	125
Radon (Nussbaum):		0.165	6.25	≈40

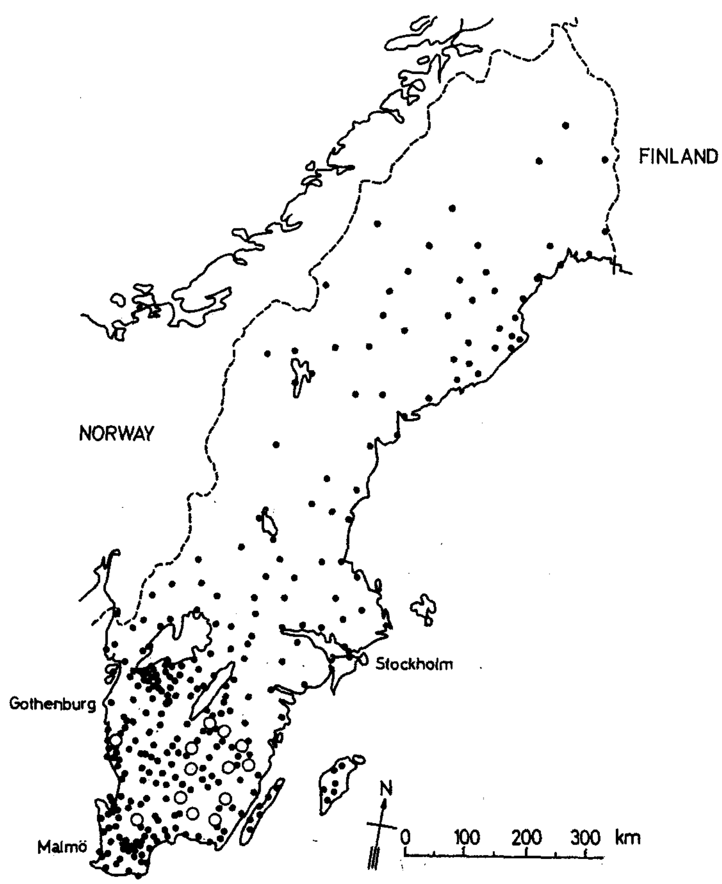


FIG. 5.

On the basis of $(MPC)_w = 10^{-4}$ μCi/ml, or 100 nCi/l., some natural waters exceed the maximum permissible occupational levels. In risk assessments, however, it must be borne in mind that the ICRP method of calculating $(MPC)_w$ is based on the assumption that the total water intake is ingested as drinking water and, furthermore, that radon is likely to disappear from the water unless it is ingested immediately after tapping.

RADON IN MILK

If water containing radon is kept in an open container of normal shape, the radon is eliminated with an apparent half-time of about 20 hr (cf. Fig. 4(a)). If the water is stirred the loss is greater, and if the water is poured from one container to another, about 10–15% of the radon is lost each time (the broken curve in Fig. 4(a)). If some cream is added to the water, the retention time will increase (Fig. 4(b)) and approach the physical life of ^{222}Rn (91.8 hr half-life). Again, the explanation is the higher solubility in fat, which is also illustrated by the Ostwald solubility coefficients for inert gases in water and oil as shown in Table 2.

Because of its higher solubility in fat, radon is also retained in milk. This has been shown to have some interesting consequences. It has been demonstrated by von Döbeln and Lindell[4] that ^{222}Rn can be found in cow's milk if the animals have had access to radon-rich water. The ratio of the concentrations (nCi/l.) found in milk and in water at the same farm was less than 0.04, with 0.025 as a representative value. This means that if milk is found to contain ^{222}Rn, the water supply is likely to hold the 40-fold concentration. This opens a possibility to trace high radon concentrations in ground water through milk.

Magi and Lindell[27] have recently reported on a survey (for ^{137}Cs) covering milk from all Swedish dairies (about 300), which revealed the presence of radon in samples from 13 dairies located in a relatively limited part of Sweden, mostly in Småland. The location of the dairies is indicated in Fig. 5, where the dairies with radon are represented by circles.

The highest concentration was 0.6 nCi/l. which would suggest that the *average* concentration for about 400 farms delivering milk to this dairy would be at least 25 nCi/l., or that a few farms have water supplies with much higher concentrations. This is being further investigated.

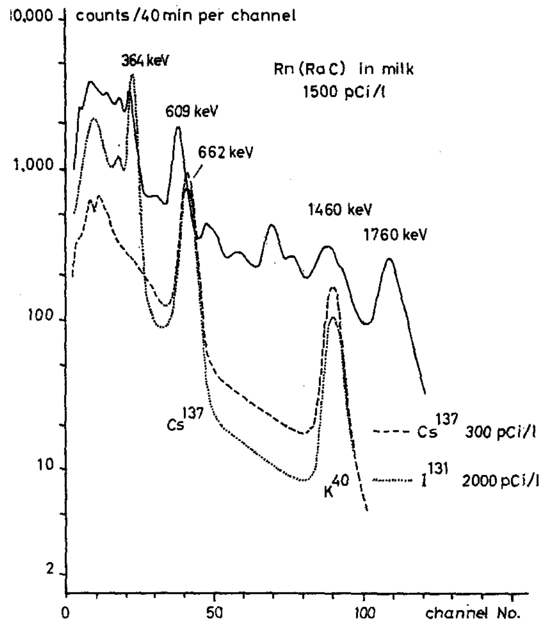


FIG. 6. γ-spectra illustrating some of the most active milk samples found in the Swedish sampling programme of 1962; cf. discussion in the text.

Even though the concentrations of ^{222}Rn in milk are much lower than in water, they are sometimes sufficiently high to dominate the γ-spectrum completely, even at relatively high levels of artificial contamination, as can be seen from Fig. 6. Not even the highest ^{131}I concentration measured in Swedish milk (in 1962) did give such a high counting rate as the highest ^{222}Rn concentration found in milk. The naturally occurring contamination can, indeed, be as intriguing as the far more investigated contamination caused by nuclear debris.

REFERENCES

1. P. CURIE and E. LABORDE. Sur la réactivité des gas qui se degagent de l'eau des sources thermales. *Compt. rend. Acad. d. Sc. Paris* **138,** 1150–1153 (1904).

2. S. R. Bernard. An $(MPC)_W$ for Rn^{222}. Paper presented at the Eighth Annual Meeting of the Health Physics Society, New York, 1963.
3. I. O. Andersson and I. Nilsson. Exposure following ingestion of water containing radon-222. Paper presented at the (IAEA) Symposium on Assessment of Radioactive Body Burdens in Man, Heidelberg, 1964.
4. W. von Döbeln and B. Lindell. Some aspects of radon contamination following ingestion. *Arkiv för fysik (Stockholm)* **27,** 531–572 (1965).
5. J. B. Hursh, D. A. Morken, Th. Davis and A. Lovaas. The fate of radon ingested by man. Univ. of Rochester Atomic Energy Project Report No. UR-648 (1964), also *Health Physics* **11,** 463–476 (1965).
6. National Committee on Radiation Protection (U.S.), Maximum permissible amounts of radioisotopes in the human body and maximum permissible concentrations in air and water. National Bureau of Standards, Handbook 52. Washington, 1953.
7. S. Meyer. Physikalische Grundlagen von Emanationskuren. Strahlentherapie **58,** 656–663 (1937).
8. A. Fernau and H. Smereker. Über des Verbleiben radioaktiver Substanz im Organismus bei Radium-Emanationstrinkkuren. *Strahlentherapie* **46,** 365–373 (1933).
9. E. K. Federov and V. I. Baranov. Content of natural radioactive substances in the atmosphere and in natural waters in the territory of the USSR. Academy of Sciences, Moscow (1956).
10. H. Muth *et al.* The normal radium content and the Ra^{226}/Ca ratio of various foods, drinking water and different organs and tissues of the human body. *Health Physics* **2,** 239–245 (1960).
11. H. Kiefer and R. Maushart. Die natürliche Radioaktivität im Wasser. *Nukleonik* **1,** 22-27 (1958).
12. R. C. Turner, J. M. Radley and W. V. Mayneord. Naturally occurring alpha-activity of drinking water. *Nature* **189,** 348–352 (1961).
13. B. M. Smith, W. N. Grune, F. B. Higgins and J. G. Terrill. Natural radioactivity in ground water supplies in Maine and New Hampshire. *Journal Am. Water Works Ass.* **53,** 75 (1961).
14. Naima Sahlbom. Om radioaktiviteten hos svenska källvatten och dess samband med de geologiska förhållandena. *Arkiv för Kemi, Mineralogi och Geologi (Stockholm)* **6,** No. 3, 1–52 (1915).
15. I. Stephan. Der Emanationsgehalt ausserdeutscher radioaktiver Quellen nach französischen und amerikanischen Messungen aus den Jahren 1918–1923. *Strahlentherapie* **16,** 560–574 (1923).
16. W. Engelmann. Die Radioaktivität der Heilquellen im deutschen Sprachgebiet und ihr Anteil am deren therapeutischer Wirkung, Strahlentherapie **18,** 686–709 (1924).
17. P. Ludewig. Der Emanationsgehalt der radioaktiven Quellen in Brambach und Oberschlema. *Strahlentherapie* **18,** 718–720 (1924).
18. P. Ludewig. Der Emanationsgehalt der Quellen in den wichtigsten radioaktiven Quellengebieten. *Strahlentherapie* **19,** 170–171 (1925).
19. C. Genser. Über die Entstchung und die Natur radioaktiver Quellen. *Geologische Rundschau* **23,** 188–238 (1932).
20. C. Genser. Radioaktive Heilquellen in Deutschland. *Z. der Deutschen Geologischen Gesellschaft* **85,** 482–495 (1934).
21. G. Armands. Geochemical prospecting of a uraniferous bog deposit at Masugnsbyn, Northern Sweden. Swedish Atomic Energy Company Report AE-36, Stockholm (1961).
22. P. K. Kuroda. Radioactivity tables of some natural waters. University of Arkansas (1953).
23. E. D. Allen-Price. Uneven distribution of cancer in West Devon. *Lancet* **II,** 1235–1238 (1960).
24. J. D. Abbatt, J. R. A. Lakey and D. J. Mathias. Natural radioactivity in West Devon water supplies. *Lancet* **II,** 1272–1274 (1960).
25. J. H. Harley, E. Jetter and N. Nelson. Elimination of radon from the body. New York's Operations Office Report No. 3, United States Atomic Energy Commission (1951).
26. E. Nussbaum and J. B. Hursh. Radon solubility in rat tissues. *Science* **125,** 552–553 (1957).
27. A. Magi and B. Lindell. Rn^{222} in milk. *Proc. International Symposium on Radioecological Concentration Process*, Stockholm, 25–29 April 1966. Pergamon Press, London.
28. J. H. Lawrence *et al.* Preliminary observations on the narcotic effect of xenon with a review of values for solubility of gases in water and oils. *J. Physiol.* **105,** 197–204 (1946).
29. E. Nussbaum. Radon solubility in body tissues and in fatty acids. Univ. of Rochester Atomic Energy Project Report No. UR-503 (1957).