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Allan B. Tanner
U. S. Geological Survey
990 National Center
Reston, Virginia 22092, U.S.A.

**UNIVERSITY OF UTAH
RESEARCH INSTITUTE
EARTH SCIENCE LAB.**

U.S. Geological Survey Open-File Report 78-1050

December 1978

This report has been prepared for publication in the proceedings of the Third International Symposium on the Natural Radiation Environment, Houston, Texas, April 23-28, 1978

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ABSTRACT

Since the First International Symposium on the Natural Radiation Environment our understanding of radon migration in the ground has been aided by research in solid-state physics, in occupational health, in uranium exploration, in lunar radioactivity, and in the use of radon-222 for predicting earthquakes. Water is the most important agent in enabling radon isotopes to escape from solid material: water absorbs kinetic energy of the recoil atom of radon; it is an active agent in altering and hydrating mineral surfaces and thus enhancing their emanating power; and it decreases the adsorption of radon on mineral surfaces. Once in rock and soil pores, radon atoms migrate by diffusion and by transport in varying proportions. In diffusion and transport calculations, it is desirable to use the radon concentration in the interstitial fluid as the concentration parameter and to include porosity explicitly. Most values of diffusion coefficient in the literature are for an effective diffusion coefficient, equal to the true diffusion coefficient divided by porosity, and should be corrected before applying boundary conditions. The transport component is important in dry, permeable soils in the upper layers but is much reduced below depths of several tens of meters. Research in disequilibria in the uranium-series and thorium-series radionuclides suggests that much assumed migration of radon-222 is in fact a more general migration of uranium and radium isotopes.

INTRODUCTION

In the proceedings of the First Symposium on the Natural Radiation Environment, I presented a review (Tanner, 1964b) of the processes by which the atoms of the several natural isotopes of radon escape from solid material and migrate in the ground. The present review is intended not to be a revised edition of the earlier review, but to supplement it with information that has resulted from more recent work, that was overlooked, or that appears to need additional treatment or emphasis. This review should serve also as a guide to the literature of recent applications of radon migration to various problems in radiation measurement, lunar studies, uranium prospecting, earthquake prediction, and detection of near-surface geologic features. The review follows topics in the same order as its predecessor; those unfamiliar with the earlier review should find it advantageous to read the two papers sequentially by topic.

MIGRATION OF RADON ISOTOPES

Emanation into Rock Pores

Emanating Power. Before an atom of a radon isotope can migrate, it must escape from the site of its parent radium isotope. Under steady-state conditions, the fraction of the radon atoms formed in a solid that escape from the solid is defined as the emanating power of the solid for the radon isotope in question. In the Soviet literature, the preferred term is coefficient of emanation. "Escape ratio", "Escape-to-production ratio", and "percent emanation" are other equivalent terms used in recent literature. The variability of emanating power among different substances and the effects of heating have been noted since the very early days of radioactivity studies (Rutherford, 1901, and many others). In the 1920's, Otto Hahn and his associates originated the "emanation method" of studying the internal surface and grain size of artificial powders. Flügge and Zimens developed the theory of emanating power in the later 1930's and early 1940's. Wahl (Wahl and Bonner, 1951, Chaps. 9 and 10) comprehensively reviewed emanating-power theory and results to 1950. More recently, Quet et al. (1972, 1975) treated the theory for isolated particles, Andrews and Wood (1972) treated the emanation process with respect to entry into ground water, and Kapustin and Zaborenko (1974a, 1974b) developed the theory for both steady-state and transient conditions and for a non-zero concentration at the pore boundary.

Direct-Recoil Fraction. Upon the decay of an atom of one of the natural isotopes of radium, most of the excess energy is carried off by an alpha particle, and the remainder, 10^4 - 10^5 times greater than typical chemical bond energies, is carried by the resulting radon atom. Figure 1 illustrates the recoil process. Within the mineral grain where the decay takes place, the recoiling radon atom has a range of 20-70 nm (Quet et al., 1975). for minerals of common density. A radon atom that is directed toward the boundary has a chance to escape into the pore. Those radon atoms that terminate their recoil paths in a pore compose the direct-recoil fraction of emanating power. If the pore is filled with gas, the range of the recoiling atom is equal to the fraction of its remaining kinetic energy multiplied by its recoil range in air, which is about 63 μm for Rn-222, 83 μm for Rn-220, or 92 μm for Rn-219 (Flügge and Zimens, 1939).

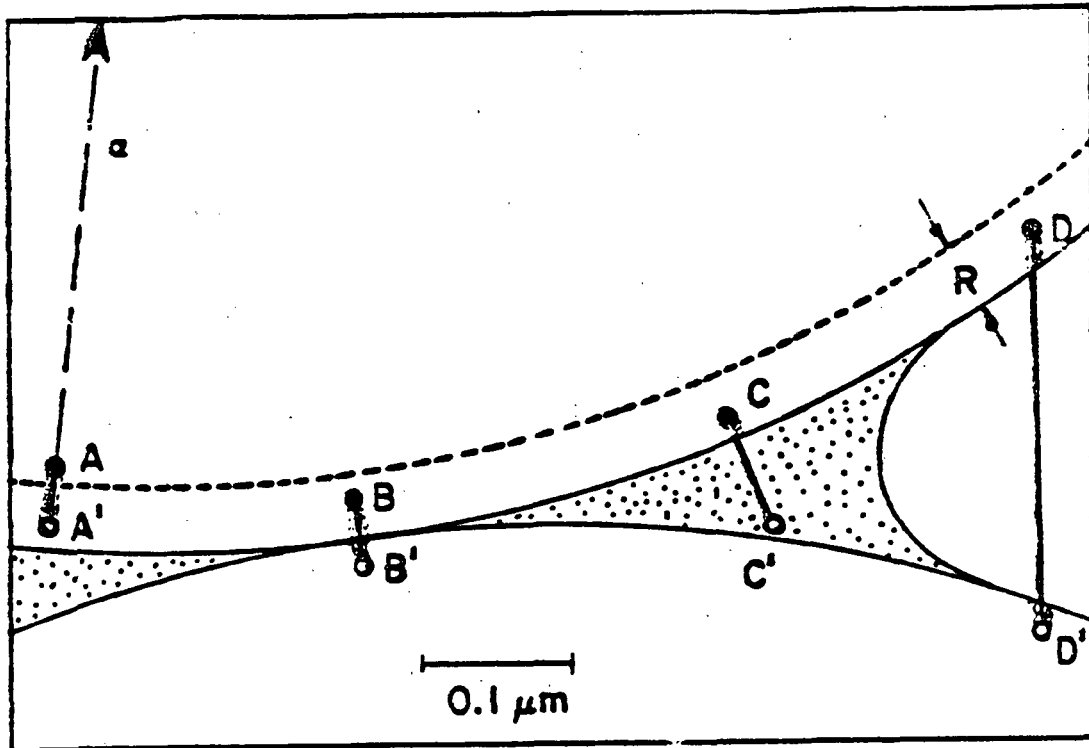


Figure 1. Schematic diagram of emanation processes. Two very fine spherical grains of 2- μm diameter are in contact near B. Water is present in the stippled zone of the pore; the open zone is air filled. Ra-226 atoms (closed circles) decay in the upper grain, each yielding an α particle (as shown at A), and a Rn-222 atom (open circles). Atom A lies at greater depth within the grain than the recoil range, R; recoil Rn-222 atom A' is contained within the grain. Atom B' escapes from the upper grain but buries itself in the lower grain. After escape from the upper grain, atom C' loses the remainder of its recoil energy in the water and is free to diffuse through the pores. Atom D' loses little of its recoil energy in the air and buries itself in the lower grain. Atoms B' and D' may escape from their recoil pockets by diffusion before condensation of the excited atoms of the grain (indirect-recoil effect). Within a mean life of Rn-222 (5.5 days) its diffusion distance in the solid grain is less than the width of any line in the figure. The circles greatly exaggerate the atomic dimensions.

In compacted natural materials either (1) the grain sizes are much larger than the recoil range in the grains, so that few recoil atoms escape from the grains in which they originate; or (2) the pores are much smaller than the recoil range in air, so that the recoil paths do not terminate in the pores if the pores are gas filled. The direct-recoil fraction of emanating power is consequently less than 1% in dry compacted granular materials. If the pores contain water or another liquid, however, the range of the recoil atom is only about 0.1 μm and the probability that it will stop in the pore is greatly increased. The presence of a liquid in the pores thus increases the direct-recoil fraction of emanating power.

Indirect-Recoil Fraction. If a recoiling atom has enough energy to traverse the pore, it penetrates the adjacent solid surface and forms a pocket of depth comparable with the recoil range in the solid material, reduced by the fraction of energy already expended. Flügge and Zimens (1939) estimated the pocket to be about 10 mm in both depth and diameter; Zimen and Mertens (1971) estimated it to be about 10 mm deep and 1 mm in diameter. Recognizing that the energy absorbed along the recoil path was sufficient to melt the solid material in the pocket, Flügge and Zimens considered the possibility of an indirect-recoil fraction of emanating power due to diffusion of the recoil atom out of the pocket while the material was still molten. However, they reasoned that because the thermal conductivity ($10^{-1} \text{ cm}^2/\text{s}$) was several orders of magnitude greater than the diffusion coefficient ($10^{-4} \text{ cm}^2/\text{s}$) in the molten material, the recoil atom would be trapped in the pocket. Since my earlier review, Zimen and Mertens (1971) have reconciled many troublesome experimental observations with the theory by a "knock-out effect": the material of the recoil pocket is briefly transformed to vapor, in which the diffusion coefficient is of the same order of magnitude as the thermal conductivity; the radon-isotope atom then has a significant probability of escaping back into the pore. Carbonates and metal hydroxides may also be explosively dissociated by the knock-out effect, augmenting the indirect-recoil fraction of emanating power.

Diffusion Fraction. Because the direct-recoil fraction of emanating power is very low for sand-sized grains in which the radium-isotope precursor is homogeneously distributed, the much greater emanating power

of many natural materials is often assumed to be due to diffusion of the radon-isotope atom through solid material. My earlier review cited experimentally determined diffusion coefficients for helium, argon, and lead in ionic crystals; it is reasonable to infer diffusion coefficients of 10^{-20} cm²/s or less for the radon isotopes in ionic crystals at room temperature. Such small diffusion coefficients limit movement to only a few lattice constants during the mean life of Rn-222 and indicate little chance of any movement of Rn-220 and Rn-219 in ionic crystals.

The possibility of diffusion of radon-222 through zones of radiation damage has been proposed repeatedly as an explanation of radon loss from radioactive minerals. Recent ingenious experiments by Lambert et al. (1972) and by Lambert and Bristeau (1973) indicate that radiation damage alone does not significantly increase radon escape by diffusion. They exposed a dressed face of a rock or mineral sample to a film of Ra-226 for a period of 1 to 5 weeks, thereby implanting Rn-222 atoms in the sample surface. Each radon atom implanted was then at the end of a disordered recoil pocket leading directly to the surface of the sample. After irradiation, the exposed surface was observed by an α spectrometer for a period of 4 to 5 half lives of Rn-222. The measurement chamber was pumped constantly to remove any radon escaping from the surface; such radon loss would be manifested by a departure in the α counting rate from the decay rate of Rn-222. The samples, experimental temperatures, and apparent radon losses were as follows: (1) NaCl monocrystal, laboratory temperature, $1.7\% \pm 0.7\%$; (2) cleaved mica crystal, laboratory temperature, undetectable; (3) hard fine-grained limestone, 573 K, $3.1\% \pm 2.7\%$; and (4) basalt, 573 K, 0 - 2%. In contrast with these small losses, even where optimum conditions prevailed for diffusion through radiation-damaged zones, emanating powers of α -radioactive zircons have been reported to be as great as 12.1% for Rn-222 (Barretto, 1973, Table 6) and as great as 23% for the much shorter lived isotope, Rn-220 (Starik and Melikova, 1957, Table 6). Zircons, in general, have much lower emanating powers (see, for example, Barretto, 1973, Tables 4 and 5). Most zircons contain water, although it is not an essential constituent (David Gottfried, oral communication, 1978). It is consistent with the above evidence to speculate that the effect of radiation damage is to form a mosaic of channels in which water may be introduced to increase

the direct-recoil fraction of emanating power, or along which the mineral may be altered to increase the indirect-recoil and diffusion fractions.

The experiments by Lambert and his colleagues did not test the possibility of radon losses by indirect recoil during the implantation procedure.

Effect of Moisture and Adsorption on Emanating Power. Water that is present in pores, capillaries, and structural gaps in mineral crystals increases the probability that radon-isotope atoms will terminate their recoil paths in those spaces, thus augmenting the direct-recoil fraction.

Hydrated surfaces tend to have greater emanating power than unhydrated surfaces. Wahl (in Wahl and Bonner, 1951, pp. 289-290) cited a relation found by H. Müller to exist between emanating power and radon adsorption on chabazite, a natural zeolite whose degree of hydration can be altered without changing its crystal structure. In experiments at 293 K on chabazite samples whose degree of hydration ranged from about 7.3 to 0 moles of water per formula weight, the emanating power for Rn-222 ranged from about 63% to about 1%, and the adsorbing power ranged in a nearly complementary manner from 1 to about 73 in relative units. In addition to its tendency to diminish adsorption of radon isotopes, hydration may cause enhanced emanating power through both direct- and indirect-recoil effects.

Adsorption effects on the release of radon isotopes from geologic materials have not been studied sufficiently to determine unambiguously whether they are an important factor. Barrer and Grove (1951) inferred that inert gases were not adsorbed appreciably on beds of small synthetic crystals of analcite, a zeolite. Shashkin and Prutkina (1970) found that 5 to 10 times as much air must be blown through radioactive coal to de-emanate it than through silicate rocks in 5- to 10-g samples. Kelly (1961a, Figure 3) found that Xe-135 frozen on glass at 77 K was 95% desorbed 5 to 6 times faster than if frozen on charcoal; release from both surfaces was nearly complete upon warming up to laboratory temperature. Wertenstein (1935) inferred that the mean residence time of Rn-222 on clean glass surfaces at 90 K was only 4.3 ms. In some unpublished experiments in 1959, I observed that Rn-222 carried in air bubbled through a 1.9- μ Ci Ra-226 solution would pass through a brass cylinder containing copper wool until the temperature in the cylinder was low enough to condense oxygen (90 K), at

which point the Rn-222 was removed from the air stream. If adsorption of radon isotopes is not strong on glass at 90 K, it should be negligible at normal temperatures at the Earth's surface; however, Miller's data on chabazite suggest that adsorption is important to emanating power for some minerals. Adsorption is greater at reduced pressure; the experimental evidence is presented below in connection with emanation from lunar materials.

Effect of Grain Size on Emanating Power. Flugge and Zimens (1939), Quet et al. (1972, 1975) and Kapustin and Zaborenko (1974a) calculated emanating power as a function of grain size. Mercer (1976) calculated the probability of escape of Pb-214 atoms by recoil following decay of Po-218 atoms attached to the surfaces of isolated particles; for spherical, isotropic grains of SiO₂ (and typical common minerals), the probability of escape ranges from about 1.0 for grains of less than 0.1- μ m diameter to about 0.54 for grains of 0.9- μ m diameter.

Andrews and Wood (1972) studied the emanation of Rn-222 into water from various size fractions of crushed and sieved samples of three sedimentary rocks. From assumptions of homogeneous radium distribution, isotropic crystal structure, and grain dimensions much larger than recoil range, they calculated that emanating power resulting from either direct recoil or lattice diffusion would be proportional to (grain diameter)⁻¹. The emanating powers measured for one limestone and one sandstone were, instead, approximately proportional to (grain diameter)^{-1/2}, which was consistent with an isotropic system of internal pathways permitting easy diffusion. Extrapolation of the relation between emanating power and particle diameter gave 100% emanating power for particles less than about 1 μ m in diameter. For another sandstone, the emanating power was nearly independent of size of the sand grains or of aggregates of grains, because the uranium was contained in a cement composed of oxide, carbonate, and silicate. Emanating power could be correlated with iron content.

Megumi and Mamuro (1974) studied the emanation and related characteristics of two soils derived from granite. The concentrations of the precursors of Rn-222 and Rn-220 were progressively greater with decreasing grain size in the range of mean diameter from >2800 to 20 μ m. For one of the soils the emanating power, as measured on the fraction containing the finest grains, was about 25% for Rn-222 and about 10% for Rn-220.

For the other soil, the emanating power for Rn-220 only was measured; it averaged about 13%. The Rn-220 emanating power was nearly independent of grain size with both soils, but most of the Rn-220 was emanating from the finer size fractions because of the greater concentration of the precursors in them. Starik (1959, Table 53) reported that emanating power was similarly not sensitive to grain size, in the range of 50-5000 μm , for all three radon isotopes from radium isotopes adsorbed on barium nitrate crystals.

Emanating Power of Minerals, Rocks, and Soils. Emanating power should be conceived as a property of the material in samples sufficiently small that negligible decay of the radon isotope takes place between the time it becomes available for rapid movement through pores and the time it is collected for measurement. My earlier review cited six publications containing results of determinations of emanating power of mineral and rock samples. Wahl (in Wahl and Bonner, 1951, Table 9A) compiled a table of about 300 determinations of emanating power and of heating experiments with elements and compounds. Iokhel'son (1958) and Shashkin and Prutkina (1970) have described heating experiments with rocks. Barretto (1973, 1975; see also Barretto et al., 1975) determined Rn-222 emanating power of 125 diverse mineral, rock, and soil samples. Austin (1975) reported Rn-222 emanating-power measurements of 760 samples of uranium ores. Shashkin and Prutkina (1970) reported determinations of Rn-222 emanating power of 25 samples of uranium minerals; the Rn-219 emanating power exceeded 10% in three and exceeded 1% in 16 of the 21 samples for which it also was determined.

The recent measurements of emanating power of terrestrial minerals, rocks, and soils agree with measurements reported in my earlier review. Most minerals and nearly all rocks and soils emanate radon isotopes to a far greater degree than can be accounted for by recoil or by diffusion from major-mineral crystals that are structurally intact and in which the radium-isotope precursor is uniformly distributed.

Uniform distribution of the precursor is valid for the radiochemical techniques for which emanating-power theories have been derived, but is not usually valid for rocks and soils. Not compatible with the crystal structure of the major rock-forming minerals, uranium and thorium normally reside in smaller "accessory" minerals, are adsorbed

on clay minerals, or are occluded in fine-grained or amorphous cements and other coatings. The succession of radioactive recoils from uranium and thorium and leaching make the radium isotopes even more available to networks of capillaries and intergranular boundaries (Starik, 1959, pp. 194-196).

Fewer emanating-power measurements of geologic materials have been reported for Rn-219 and Rn-220 than for Rn-222. The published measurements for more than one isotope from a given sample usually do not differ by as much as an order of magnitude. By linear diffusion theory, one expects the differences to be in proportion to the square roots of the half lives; that is, the emanating powers for Rn-219, Rn-220, and Rn-222 should be in the rough proportions, 1.0:3.7:290, if the radium-isotope precursors are distributed similarly. The different geochemical behavior of uranium and of thorium may invalidate conclusions based on comparison of the emanating powers for Rn-222 and Rn-220. However, U-238 and U-235 occur in a nearly constant ratio, and their decay patterns do not differ sufficiently to account for the contrast between the 290-fold difference in emanating power for Rn-222 and Rn-219 expected to result from a diffusion mechanism and the less-than-10-fold differences observed in real samples. The increase of Rn-222 emanating power when samples are immersed in water (Shashkin and Prutkina, 1970) is also inconsistent with a mechanism in which diffusion is the rate-limiting step.

Although the indirect-recoil effect (including the knock-out effect) and adsorption may play a greater role than previously supposed, the conclusions about the emanation process given in my earlier review (Tanner, 1964b, pp. 164-165) may be repeated: "... any appreciable emanations of radon-222, radon-220, or radon-219 atoms come from radium isotopes distributed in secondary crusts or films or in the shallow surface layers (approximately as deep as the recoil range) of intact crystals of the host minerals. Radon isotopes in the deeper regions of the crystals are unavailable without the development of large internal surface, such as may result from chemical corrosion, weathering, or intensive fracturing on a microscopic scale.... The principal mechanism by which the radon isotopes enter the pores, capillaries, or microfractures is radioactive recoil into liquid-containing spaces or diffusion from solid material appreciably smaller than the diffusion length of the most short-lived isotope observed."

Migration in a Porous Medium

Variables that are significant in the migration of radon isotopes in the ground are the decay rate of the isotope, the diffusion constant for the isotope in the pore-filling fluid, the composition of the fluid, the motion of the fluid, and, if the fluid has more than one phase, the distribution of the isotope among the phases.

A liquid (usually water) enhances the emanation process and may tend to prevent adsorption of the radon isotopes. Once a radon-isotope atom has lost its recoil energy, the liquid hinders its migration by lowering the diffusion coefficient and by absorbing it.

Two different mechanisms of migration should be distinguished: diffusion, where the radon isotope moves with respect to the fluid filling the pores of the medium; and transport ("convection" in the Soviet literature), where the fluid itself moves through the porous medium and carries the radon isotope along with it. Either or both mechanisms may be important in a given place.

Distribution of Radon Isotopes Among Fluids. It was found in the early 1900's that a radon isotope distributes itself in definite, temperature-dependent proportions at equilibrium in a system of more than one fluid phase. At temperatures common at the Earth's surface, radon-isotope concentrations are greatest, intermediate, and least, respectively, in the organic-liquid, gas, and water phases of a multiphase system. With increasing temperature, the concentration in the gas phase increases at the expense of the liquid phases. The ratio of Rn-222 concentration in a water phase to that in a gas phase ranges from 0.52 at 273 K to 0.16 at 313 K (Kofler, 1908). Ramstedt (1911) listed the corresponding ratios (Rn in liquid/Rn in gas) for 13 organic liquids, ranging from 4.45 to 55.4 at 273 K and from 3.80 to 23.44 at 291 K. Glycerine is an exception among the organic liquids; it holds a lower concentration of Rn-222 than does water. Bonner (in Wahl and Bonner, 1951, Table 6.15) summarized experimentally determined distribution coefficients. The distribution between gas and water (or aqueous solutions) has been reported by Himstedt (1903), Traubenberg (1904), Hofmann (1905), and Kofler (1908). The distribution between gas and organic liquids has been reported by Traubenberg (1904), Hofmann (1905), Kofler (1908), Ramstedt (1911), and Cherepennikov (1951).

Diffusion of Radon Isotopes in a Porous Medium. Currie (1960a, 1960b, 1961) comprehensively reviewed the diffusion of gases in porous media. His findings that are important to this review are based on hydrogen and may be generalized as follows:

(1) If D is the diffusion coefficient, P is pressure, T is absolute temperature, and if the subscript o indicates standard conditions, then the diffusion coefficient, D_i , in a continuous medium is related to temperature and pressure as follows:

$$D_i/D_o = (T/T_o)^n (P/P_o) \quad (1)$$

where the exponent n is expected to be close to 1.75 for gases and 2.0 for vapors.

(2) Diffusion through dry porous granular materials is influenced by porosity, packing, and particle shape and size, in addition to the composition, temperature, and pressure of the pore-filling fluid. An empirical equation accounting for the solid properties is

$$D/D_i = \gamma \epsilon^\mu \quad (2)$$

Where D_i is the diffusion coefficient in the pore fluid as a continuous medium; ϵ is the porosity; the constant, γ , ranges from 0.8 to 1.0; and μ , a constant ≥ 1 , is probably a measure of pore shape. Equation (2) gives a better fit to experimental data than an often used linear relation of the form

$$D/D_i = a\epsilon + b \quad (3)$$

Various values of the constants a and b have been used in practical applications (Currie, 1960b, p. 318; Kirkham and Powers, 1972, p. 434). Clements (1974, p. 57) found that the values, $a = 0.9$ and $b = 0.12$ gave a diffusion coefficient that was consistent with diffusion in laboratory columns. Doshchechkin and Serdyukova (1969) related the ratio D/D_i to the open, or interconnecting, porosity and the tortuosity (the ratio of the average path length to the straight-line length in the direction of diffusion (ξ)):

$$D/D_i = \epsilon/\xi^2 \quad (4)$$

The heterogeneity of geologic materials is a source of sufficient error to justify using equation (3). Mica and vermiculite, which are flaky minerals, have a shape factor that causes D/D_i to be 1/2 to 1/3 the value expected on the basis of equation (3). Clays and shales contain significant proportions of flaky minerals, usually oriented so as to impede vertical movement; they may be expected to retard diffusion (and transport) to a greater extent than a porous medium having the same porosity but consisting of spherical particles.

(3) Particle shape is less important if the medium is wet. The diffusion coefficient is less for porous media with wet pores than for those that are dry but have the same air-filled porosity; the effect is probably due to blocking of the interpore paths by water. Letting D/D_d be the ratio of the diffusion coefficient for the wet medium to that for the medium when dry, and letting ϵ_a/ϵ_d be the ratio of the air-filled fraction of the porosity of the wet medium to its porosity when dry,

$$D/D_d = (\epsilon_a/\epsilon_d)^\sigma \quad (5)$$

where σ is approximately 4. If the grains themselves are porous, the medium requires too much characterization to generalize here.

Porosity is necessarily a factor in calculations of diffusion and transport in a porous medium, but many of the earlier publications have treated the subject as if the medium were continuous. When the equations published by Grammakov (1936) and cited as equations (1) and (2) in my earlier review are evaluated from experimentally determined concentrations of a radon isotope at different depths in a homogeneous porous medium, the diffusion coefficients obtained are "effective" diffusion coefficients (D^*). The effective diffusion coefficient formally identifies the medium as continuous, rather than porous, and is larger by a factor of (1/porosity) than the "true" diffusion coefficient characteristic of the radon isotope, the interstitial fluid, and the obstructions provided by the solid matrix. Most reports of radon-isotope diffusion in the literature do not enable one to determine which diffusion coefficient is specified, but the manner of measurement, the range of values reported, and the equations used (if given) usually suggest that it is the effective diffusion coefficient.

When the effective diffusion coefficient is used as if it were a constant and is applied to layers of different porosity, or to the interface between soil and the atmosphere, an error can arise because of the change in porosity. Schroeder et al. (1965) discussed the precautions necessary in using Fick's first law to obtain the exhalation of a radon isotope from its concentration gradient in the soil. Bulashevich and Khayritdinov (1959) showed experimentally that the "porous concentration" of Rn-222 was continuous across a sand-air interface; that is, the concentration in the interstitial space and the concentration in the air were equal at the interface. Bulashevich and Khayritdinov have discussed both the concentration parameter and the porosity corrections in diffusion and transport equations and have noted errors in the works of Grammakov and Popretinskiy (1957) and Budde (1960 and earlier papers). Most authors since 1963, notably Schroeder et al. (1965), Doshchechkin and Serdyukova (1969), Robertson (1969), Clements and Wilkening (1974), Culot, Olson, and Schiager (1976), Jeter et al. (1977), Clements et al. (1978), and Jonassen and McLaughlin (1978) have avoided the above errors.

Bulashevich and Kartashov (1967) and Bulashevich et al. (1970) reported measuring the diffusion coefficient of rocks in place by introducing an artificial pulse of radon at a point. Their method gave reasonable values of $D^*=2.9 \times 10^{-2}$ and $3.8 \times 10^{-2} \text{ cm}^2/\text{s}$ for rocks in two locations.

Ghosh and Sheikh (1976) measured diffusion of Rn-222 through non-radioactive sections of rock, obtaining $D^* (\text{cm}^2/\text{s})=1.436 \times 10^{-5}$ for muscovite chlorite schist, 1.15×10^{-6} for epidiorite, and 0.79×10^{-6} for quartzite.

Porstendörfer (1968) measured the diffusion coefficients for Rn-222 and Rn-220, obtaining values of $D^* (\text{cm}^2/\text{s})=3.2 \times 10^{-6}$ in bacon, 1.3×10^{-6} in paraffin, and 5.7×10^{-4} in polystyrene foam.

Migration in Unsaturated, Undisturbed Rock and Soil

Mechanisms of Movement. Both diffusion and transport by moving fluids are significant in moving radon isotopes in the ground. In general, diffusion is the dominant mechanism in the intergranular channels, capillaries, and smaller pores; in the larger pores, transport may become important or even dominant.

The diffusion processes and sources of pressure gradients that cause gas flow are detailed by Wilkening (1978). Knudsen diffusion is applicable where the diffusing atom's mean free path (0.1 μm in air at STP) is much greater than the capillary diameters, and the gas atoms collide mainly with the solid walls; within leached zones of radiation damage, fine capillaries, and cements, Knudsen diffusion should prevail. In the larger capillaries and in the pores, the gas atoms collide mainly with other atoms of the fluid, resulting in molecular diffusion.

The relative contributions of diffusion and transport to migration of radon isotopes and other gases are quite difficult to determine in practice. In my earlier review (Tanner, 1964b, p. 169-170), I showed that one-dimensional migration of a radon isotope in a homogeneous porous medium results in exponentially decreasing concentration of the isotope in the direction of migration. The exponential argument includes both diffusion and transport parameters. Unless the diffusion coefficient or the transport velocity is accurately known, any combination of the two components will satisfy the simple diffusion model. Robertson (1969) found that field studies based on injections of Kr-85 closely matched calculations based on a diffusion model, using an effective diffusion coefficient of about 0.02 cm^2/s . Schroeder *et al.* (1965) found average concentrations of Rn-222 at 0.87-, 1.50-, and 2.86-m depths in soil at the Nevada Test Site to fit a diffusion model using $D^* = 0.036 \text{ cm}^2/\text{s}$; within the upper few decimeters of the soil, however, transport in addition to diffusion caused the effective diffusion coefficient to be 0.1 cm^2/s -- nearly that for Rn-222 in open air. Israëli (1962 and earlier publications) has shown that typical concentrations of Rn-222 in the atmosphere can be balanced by a deficit in the uppermost layer of soil of typical uranium-series activity, and that typical values of Rn-222 exhalation are obtained, if a diffusion model is used with $D^* = 0.05 \text{ cm}^2/\text{s}$.

Jonassen and McLaughlin (1978) have described use of a diffusion model and measurements of Rn-222 buildup from samples held at various pressures in a pressure vessel to obtain the rate of exhalation of Rn-222 from building materials and walls. The effect of pressure decreases on Rn-222 exhalation from walls was evaluated by McLaughlin and Jonassen (1978).

Although not directly applicable to diffusion of radioactive nuclides, the extensive work by Newton and Round (1961) on the diffusion of helium through sedimentary rocks is noteworthy because it illustrates some of the considerations necessary for multilayer modeling.

Meteorological Effects. Since my earlier review was compiled, relatively few general studies have been reported on the effects of meteorological variables on the concentrations of radon isotopes in soil gas and their exhalation into the atmosphere. Pearson and Jones (1965, 1966) observed depression of the rate of exhalation of Rn-222 during periods when the soils were cold or frozen; during a winter thaw the exhalation rate rose by a factor of two or more. Wind speed at 0.5-m height and the fraction of soil space occupied by air were also monitored but gave no obvious correlations with exhalation. Megumi and Mamuro (1973) measured both Rn-222 and Rn-220 exhalation at Osaka; they found little seasonal variation in Rn-222 exhalation, but the summer exhalation rates for Rn-220 were 2 to 3 times greater than those in winter because they were strongly affected by the soil moisture content. Bakulin (1969) found seasonal changes for exhalation of Rn-222 to be not more than 10%; the maximum exhalation was in the summer. Gabelman (1972) found the α activity of soil gas to correlate directly with temperature and inversely with atmospheric pressure and rainfall. A heavy rain sealed the surface and caused an increase of α activity in the soil gas during the following 12-hr period; as the rain percolated downward through the measurement zone to the water table, the α activity decreased severalfold. Guedalia *et al.* (1970) found no direct influence of the variations in atmospheric pressure on the exhalation of Rn-220. Mageru and Rezlescu (1965) inferred that temperature, pressure, and wind speed were the principal meteorological factors in Rn-222 variation in the atmosphere near the surface.

Decreasing atmospheric pressure causes an increase in exhalation of Rn-222 and an upward shift of its concentration profile in soil gas, as noted in my earlier review. No general analytical treatment of the combined effects of diffusion and a varying transport component, induced by atmospheric pressure changes, appeared until that of Clements and Wilkening (1974). They found that the differential equation combining diffusion and transport components may be solved by numerical methods

using the soil parameters of porosity and permeability, the interstitial-fluid parameters of density and dynamic viscosity, the Rn-222 parameters of decay constant and production rate, the appropriate true diffusion coefficient, and an analytic expression for the velocity field resulting from a small pressure change. Experimental measurements of Rn-222 exhalation best fit their model using maximum transport velocities of about 4×10^{-6} m/s upward or downward, depending upon the direction and magnitude of the pressure change. Because the pressure changes are propagated progressively more slowly with distance below the surface, day-to-day cycles of atmospheric pressure result in significant transport components at shallow depths only. On the basis of a sinusoidal variation of pressure and soil parameters typical of his area of investigation, Clements (written communication, 1977) calculated that the velocity would be diminished by a factor of 1/e at depths of 16.5 m and 28.5 m, respectively, for sinusoids of 1- and 3-day periods. Numerical results and curves showing the pressure and velocity variations at various depths as results of linear and sinusoidal pressure changes are also given in a progress report of modeling studies by Jeter *et al.* (1977).

Migration in Unsaturated, Fractured Rock, Disturbed Soil, and Underground Cavities. Migration through fractures, vents, and underground cavities is not amenable to general mathematical analysis because the open-space system is unique at each location. However, migration of the radon isotopes can be described in a qualitative way for some systems, such as volcanic vents and caves. Iwasaki *et al.* (1968) concluded that the source area for Rn-222 and Rn-220 should be near the ground surface at a volcanic vent they investigated. Wilkening (1977, 1978; see also Wilkening and Watkins, 1976) has reported the variation of Rn-222 in a horizontal tunnel, under the influence of atmospheric pressure changes, and in Carlsbad Caverns, under the influence of seasonal changes in the convective flow of air between the cave and the atmosphere.

Migration in Saturated Rock and Soil. As noted in my earlier review, migration of radon isotopes through significant distances in saturated rock and soil requires transport, because diffusion coefficients are very small, and, in many instances, the flow pattern cannot be adequately defined for mathematical analysis. A graph showing relative Rn-222 concentration in water as a function of distance from an emanating rock

surface, for diffusion plus various transport velocities, was given by Andrews and Wood (1972); a transport velocity of only 10^{-7} m/s is more effective than diffusion in moving Rn-222 away from the rock surface. Korshunkov (1970, Fig. 2) found that the Rn-222, Ra-226, and uranium concentration in water withdrawn from a borehole had maximum values at a certain withdrawal rate; at higher rates the decline in concentration of Rn-222 exceeded those of Ra-226 and uranium. Kimura and Komae (1978) have shown various applications based on the migration of Rn-222-bearing ground water into surface waters, wells, and gas spouts. Hammond et al. (1977) found that about 40% of the Rn-222 in waters of the Hudson River Estuary entered by molecular diffusion from sediments; stirred-up and suspended sediments and dissolved Ra-226 accounted for the remainder.

RADON MIGRATION IN THE LUNAR SOIL

Studies of radon migration in the lunar soil and of α radioactivity of the lunar surface were provoked by speculation of Kraner et al. (1966) that Rn-222 might diffuse from the lunar soil, form a rarified atmosphere, and implant Po-218 in the lunar surface upon its decay. The speculation was based on the typical emanating power of terrestrial soils and a diffusion coefficient comparable with those observed on Earth. Some variation was subsequently found in the rate of α -particle emission from the lunar surface, but the rate was several orders of magnitude lower than that expected on the basis of the assumptions by Kraner et al. (Turkevich et al., 1970; Gorenstein and Bjorkholm, 1972; and others). The radioactivity of lunar samples returned by the Apollo missions has been examined by several groups (Lindstrom et al., 1971; Adams et al., 1971; Grjebine et al., 1972; Stoenner et al., 1972; Yaniv and Heymann, 1972; and subsequent papers by the same groups). A moderate enrichment of Po-210 has been found on the surfaces of some samples of lunar fines, indicating implantation of Pb-210 and its precursors from Rn-222 in the interstices.

Both reduced emanating power of the lunar soil and retarded diffusion cause the exhalation of Rn-222 to be much less from the lunar surface than from the Earth's surface. Yaniv and Heymann (1972), Adams et al. (1973), Lambert et al. (1973), and Stoenner et al. (1972, 1974) have made emanating-power measurements of the fine fractions of various lunar samples under simulated lunar conditions of low pressure. Room-temperature values of emanating power were less than 1%. At temperatures below 243 K

the emanation of Rn-222 is decreased, but it has not been determined whether the decrease is due to lessened emanating power or to lessened ability of the Rn-222 atoms to diffuse from the sample (Lambert et al., 1973). Adams et al. (1973) found that the emanating power of a standard terrestrial granite at 1 atm was 11% - 9% in the range 530-300 K, about 8% at 253 K, and about 3% at 193 K; adsorption was inferred to cause the marked reduction of emanating power at low temperature.

Adams et al. (1971) de-emanated a 473-mg fraction of dust (<60 μm) from Apollo 12 soil sample 12070 at room temperature and 1 atm pressure. Their result for the rate of emanation of Rn-222, when combined with the precursor activity in a different split of the sample, measured by other investigators, yielded a Rn-222 emanating power of 48%, within estimated accuracy limits of $< \pm 50\%$ of the result. This anomalously high emanating power may have resulted from using a value of precursor activity that was not representative of the sample that was de-emanated, or it may be due to a combination of very fine grain size with great specific surface and adsorption of moisture during preparation; the result is otherwise not consistent with the emanating-power mechanisms acknowledged in these reviews. Adams et al. (1973) later reported emanating power $(0.25 \pm 0.15)\%$ for a split of the same sample; the measurement was also made at room temperature and 1 atm pressure. Yaniv and Heymann (1972) obtained values of $<0.73\%$ and $<0.57\%$ on 887 mg of another split at room temperature and $\sim 10^{-5}$ atm pressure. Experiments with an Apollo 14 sample showed up to severalfold increases in apparent emanating power when the pressure was increased to 1 atm, but the higher pressure alone could not account for a two-order-of magnitude increase.

Because there is practically no water in the lunar soil, few atoms terminate their recoil paths in intergranular spaces; the direct-recoil fraction of emanating power is consequently low. There is insufficient water and oxygen to permit formation of hydrated or corroded surfaces, such as are found to correlate well with highly emanating terrestrial soils; thus, the indirect-recoil and the diffusion fractions of emanating power are also low. In addition, adsorption plays a much more important role on the Moon than on Earth because (1) the adsorption sites on the lunar grains must be sparsely occupied, (2) the enthalpy of adsorption

is much greater in the lunar vacuum, (3) the gas molecules in lunar soil nearly always collide with solid surfaces, and (4) the average soil temperature on the Moon is lower than that on Earth.

Stoenner et al. (1972, p. 1710) summarized evidence that the mean grain size of at least some lunar soil is very small, on the order of micrometers, and that the specific surface area for adsorption is large.

Kinoshita (1908) found that the "condensation" temperature of Rn-219 and of Rn-220 not only is spread out over several tens of degrees but also is strongly dependent on pressure. Wertenstein (1935) presented strong arguments that (even with the large amounts of radon isotopes used by the early experiments) the radon-isotope atoms are not plentiful enough to form a continuous layer on a cold surface, and that the "condensation" and "vaporization" phenomena referred to in even the modern literature are in fact effects of trapping by condensing gases. On the basis of experiments with glassware outgassed at 693 K, Wertenstein calculated a mean residence time of 4.3 ms for the true adsorption of Rn-222 atoms on glass at 90 K and 1 atm pressure. The enthalpy of adsorption, ΔH_a , may be calculated from the well-known equation,

$$\tau = \tau_0 \exp (\Delta H_a / RT) \quad (6)$$

where τ is the mean residence time, τ_0 is a constant $\sim 10^{-13}$ s, R is the gas constant, and T is the absolute temperature. For $\tau = 4.3$ ms, $\tau_0 = 10^{-13}$ s, $R = 8.3$ J/mol-K, and $T = 90$ K, the value of $\Delta H_a = 18$ kJ/mol (4.4 kcal/mol). Stoenner et al. (1972) cite 4.3 kcal/mol (18 kJ/mol) as the heat of vaporization of liquid Rn(-222). From diffusion of Rn-222 through columns of fine particulate materials at pressures of 2×10^{-2} to 20×10^{-2} torr (2.7 to 27 Pa), Friesen and Adams (1976) obtained $\Delta H_a = 8-9$ kcal/mol (34-38 kJ/mol). Chackett and Tuck (1957) inferred from measured values of ΔH_a of neon, argon, krypton, and xenon on charcoal in the pressure range 10^{-2} to 10^{-4} mm (10^0 to 10^{-2} Pa) that ΔH_a for Rn(-222) should be 13.5 kcal/mol (56.6 kJ/mol). These widely ranging values of the enthalpy of adsorption of Rn-222, probably equally applicable to the other radon isotopes, demonstrate the strong dependence of adsorption on pressure and imply that the radon-isotope atoms stick to surfaces for long periods in the vacuum and at the average temperatures prevailing on the Moon.

ENVIRONMENTAL RADIOACTIVITY

Effects of Radon-Isotope Migration on the Gamma-Ray Field

The short-lived decay products of radon isotopes, particularly Pb-214 and Bi-214 (from Rn-222) and Tl-208 (from Rn-220) are the most important gamma-ray emitters of the uranium and thorium series. Rn-222 and Rn-220 migrations, if they take place, are consequently significant to measurements of gamma radiation fields.

Beck (1974) calculated that the gamma-ray flux from Rn-222 decay products in the atmosphere is about 2% of the gamma-ray flux from Rn-222 decay products in the soil during normal turbulence but can rise to about 20% during atmospheric inversions. Tyukhtin (1974) inferred that seasonal variations in gamma-spectrometric analyses of rocks in situ were due to variations in exhalation of Rn-222 and Rn-220. Errors in gamma assay and corrections based on emanation of Rn-222 from natural materials were discussed by Bolotnikov (1961, 1964, 1965), Artsybashev and Bolotnikov (1961), and Filimonov (1974). Novikov (1966) treated the same problem for Rn-220 emanation from thorium ores. Putikov (1963) noted substantial errors in borehole gamma-ray measurements resulting from emanation. Løvborg *et al.* (1978) reported values ranging from 153 to 242 ppm for the apparent uranium concentration in a uranium-loaded concrete calibration pad; the variation was inferred to be due to Rn-222 migration from the pad and could be correlated with the degree of sealing of the concrete by ice or moisture. Culot *et al.* (1976) have applied linear diffusion theory to multilayered porous media to estimate the gamma-ray field over a radon barrier.

Natural soil radioactivity has been used as the gamma-ray source for determination of the water equivalent of snow cover by gamma-ray absorption (Verzhinina and Dimakhsyan, 1969; Bissell and Peck, 1973). Garrigue (1939) measured Rn-222 concentrations as great as 1 nCi/l under the snow layer in the Pyrenees. Precipitation was shown by Bissell and Peck to cause an increase in the gamma-ray intensity above snow; it was not determined whether precipitation was causing displacement of Rn-222 upward to the soil surface or whether some other mechanism was responsible.

Radon in Mines

As was found for cellar air by Schmid (1932), Rn-222 concentrations in mine atmospheres are greatest during periods of decreasing atmospheric

pressure (Schroeder, 1964). Serdyukova et al. (1965) devised methods of calculating the average diffusion coefficient for Rn-222 in the walls of mine workings from its concentration in mine air. Pohl-Rilling and Pohl (1969) showed methods of calculating supply and removal of Rn-222 as a function of pressure changes. Bykhovskiy et al. (1969) have published a monograph on the engineering of mines to minimize exposure to Rn-222 and its decay products. Thompkins and Cheng (1969) measured exhalation from the rock walls of a Canadian uranium mine; they have described the technique and calculations. Franklin and Marquardt (1976) operated continuous Rn-222 monitors at five locations in an inactive uranium mine and observed the same inverse correlation with atmospheric pressure reported by other workers.

Radon-222 Exhalation from Mill Tailings

Uranium milling results in waste ("tailings") that contains Ra-226 of moderate-to-high emanating power. Raghavayya and Khan (1975) estimated that the amount of Rn-222 emanating from tailings used to fill exhausted parts of mines would be much greater than that emanating from the ore itself. The release of Rn-222 to the atmosphere from waste dumps was measured by Bernhardt et al. (1975). Clements et al. (1978) found that a two-layer diffusion model of exhalation fit measurements when used with a true diffusion coefficient of $0.004 \text{ cm}^2/\text{s}$ ($D^* = 0.01 \text{ cm}^2/\text{s}$). Because of the strong affinity of radon isotopes for organic liquids, I speculate that petroleum-based sealants should be more effective than concrete for reducing exhalation from tailings piles.

Radon in Natural Gas and Petroleum

The presence of Rn-222 in some natural gases and petroleum has been known since the early 1900's (Burton, 1904). The possible health hazard from Rn-222 in natural gas was suggested by Bunce and Sattler (1966) and evaluated by Gesell (1974, 1975) and by Barton et al. (1975). The radioactivity of Rn-222 in natural gas, as delivered to consumers, appears to be a minor source of radiation exposure.

The migrations of the various nuclides of the uranium series in the natural-gas reservoir of the Panhandle Gas Field of Texas were studied in an extensive investigation reported by Pierce et al. (1964). Within the interstices of carbonate reservoir rocks, the uranium isotopes reside

generally in organic solids and petroleum. Ra-226 tends to be leached into coexisting brines; upon decay of the Ra-226, the product Rn-222 migrates back into the organic liquid and natural gas phases.

Martelly (1950) reported radioactivity corresponding to that of 10.6-hr Pb-212, a decay product of Rn-220, in gas spouts on the Santa Elena Peninsula of Ecuador.

Luca Muro (1947) found a sharp increase of α activity in the soil gas at a depth of 70 cm over a shallow deposit of asphaltite.

GEOPHYSICAL APPLICATIONS

Location of Buried Faults and Geological Structures

Zones of anomalously high surface gamma-ray activity and Rn-222 concentration in soil gas have long been associated with buried faults or dislocations. Schoeneich (1960) located faults overlain by thin sediments. Vogler (1960) presented several arguments to prove that uranium and Ra-226 migration from faults and accumulation by means of ion exchange with clays in the soil is the principal cause of emanometric anomalies over the faults; he showed examples of correlation between emanometric anomalies over several faults and the radioactivity of the A₂, B₁, and B₂ soil horizons at the same respective locations. Budde (1961) considered that faulting would create greater surface area of the material in the fault zone and consequently greater emanation release. Jirkovský (1962) presented examples of the use of emanometry to locate faults and geologic contacts buried by alluvium as an aid to geological mapping and engineering. I (Tanner, 1964a) found that Rn-222 anomalies in ground water in the vicinity of a fault scarp were probably due to coprecipitation of Ra-226 at the locus of interaction between water rising along the fault and normally flowing ground water. Israël and Björnsson (1967) measured Rn-222 and Rn-220 in soil gas in traverses over five faults. The isotopes showed similar trends for some traverses, suggesting that the radon isotopes were derived from Ra-226 and Ra-224 at the sampling points. The authors caution that the several instances of anomalies in Rn-222 only could be due to the disparate geochemical behavior of the uranium and the thorium series, as well as to migration of Rn-222 from greater depth than the migration distance of Rn-220 (roughly 0.1 m). An anomaly at Bad Nauheim appeared to be due in part to migration of Rn-222 from below

3 m. Lauterbach (1968) related anomalies in profiles of gamma radiation from Bi-214 to the patterns of dispersion of Ra-226 rising with water along fault planes. Gabelman (1972) inferred that Rn-222 in near-surface soil gas had migrated along faults from relatively deep sources at a salt dome. Bulashevich et al. (1976) and Chalov et al. (1976) have studied methods of identification of waters from faults by means of the ratios of uranium-series nuclides.

Use of Rn-222 and gamma-ray measurements in prospecting for petroleum was discussed in my earlier review (Tanner, 1964b, pp. 176-177). Additional references are Kellogg (1957), Liu et al. (1959), Walker and Litzenberg (1959), Sikka (1963), Alekseyev and Gottikh (1965), and Foote (1969). Armstrong and Heemstra have discussed case histories (1972a, 1972b) and provided an extensive review (1973) of the use of radiometric methods in connection with petroleum exploration. Gates and McEldowney (1977) showed hypothetical routes by which Rn-222 would leak from hydrocarbon reservoirs via faults to the surface, but presented no supporting argument other than the existence of anomalous concentrations of Rn-222 in soil gas over uranium ore bodies, which subject is discussed below.

Exploration for Uranium

For many decades, the measurement of Rn-222 in soil gas has been used as a method of greater sensitivity than measurement of surface gamma-ray activity for locating deposits of radium or uranium. Many investigations have assessed the depth of penetration of the method and the effects of meteorological factors. The most definitive field study yet noted was the first, reported by Bogoslovskaya et al. in 1932. By means of ionization chambers and electrometers, they measured the Rn-222 concentration in soil gas samples brought up by tubes from 0.5- and 1-m depths at various points in a 10-m-square plot of ground, and from depths of 0.5, 1, 2, 3, 4, 5, and 6 m in the center. Lateral differences at 1-m depth were about 10-20% of the mean value upon stabilization after emplacement of the sampling tubes. Little change of concentration was found below 3 m. After the background measurements had been made, porous uraniferous rock was buried at 5-m depth. The effects of the disturbance caused by emplacement of the source rock and of the soil-gas sampling tubes were negligible after about one mean life

of Rn-222 (5.5 days). Table 1 gives results selected from daily measurements during a 30-day period at points above and in the uraniferous source rock. These results illustrate (1) the very marked reduction of Rn-222 concentration with distance above the source layer and (2) the strong influence of variations in atmospheric pressure. If one were to assume that all the Rn-222 migration from the source layer were due to diffusion, the calculated diffusion coefficients would range over an order of magnitude, depending upon the atmospheric-pressure trend of the previous several days. The source layer was detectable, with a gross-signal-to-background ratio of about 2:1, at 1-m depth, 4 m above the source layer. At distances of more than 2 m to the side of the source layer the Rn-222 concentration at 1-m depth was not significantly greater than background.

Pradel et al. (1963) conducted tests of Rn-222 as a prospecting method, finding that fluctuations as great as 100-fold were observed in the Rn-222 concentrations in single small samples of soil air taken on different days, but that 20- ℓ samples gave results that were reproducible within a factor of 1.5. Although Rn-220 and wet soils also presented problems, the method was concluded to be useful in exploration for uranium.

Case Histories. Some of the numerous papers describing use of radon methods of prospecting through 1961 were cited in my earlier review (Tanner, 1964b, p. 177-178). Below are summarized those case histories in which anomalously greater-than-background concentrations of Rn-222 in soil gas ("anomalies") were inferred to be related to uranium mineralization at known minimum depths.

Bowie et al. (1971, Figure 5) have shown the results of traverses over a uranium-bearing vein under about 1 m of glacial drift. During dry weather, the vein was detected by a 2- to 3-fold anomaly in Rn-222 in soil gas but not by surface gamma radiation; following a heavy rain, the soil-gas anomaly increased about 4-fold and a surface gamma-ray anomaly, about twice background, appeared.

Dyck (1969a) measured a peak-to-background ratio of about 15 for Rn-222 in soil gas in gravel and glacial till overlying a uraniferous stratum; the depth to the source was estimated at 1.5 to 4.5 m. Surface gamma radiation was similarly anomalous, but exceeded background by a factor of only about 3.

TABLE I

RADON-222 IN SOIL GAS ABOVE URANIFEROUS
LAYER AT 5-m DEPTH (from Bogoslovskaya *et al.*, 1932)

Radon-222 Concentrations in Relative Units

Conditions	Depth (m)					
	0.5	1	2	3	4	5
Barometer stable 5 days preceding	2.82	4.08	7.63	21.7	74.8	436
Barometer rising 2 days preceding	2.0	3.39	5.12	10.45	38.0	436
Barometer falling 2 days preceding	3.24	5.76	9.74	28.2	126.5	447
Mean	3.41	4.45	8.25	22.9	82.0	437
Mean for overburden	-	2.6	5.0	5.6	6.2	6.6
Mean effect due to buried layer	-	2.25	3.25	17.3	75.8	430.6

Skvarla (1964) described the use of borehole probes utilizing inflatable packers, adsorption of Rn-222 from the borehole air on charcoal, and measurement of gamma rays from the collected decay products from exploration boreholes in patterns having spacing as great as 1/2 mile (800 m). He reported discovery of uranium ore by the method in the Maybell area of the Colorado Plateau. In the Uravan area of the Colorado Plateau, anomalous Rn-222 was detected more than 100 m from the nearest known uranium mineralization.

Stevens et al. (1971) observed diverse results in a study of three areas in the western United States: in one, the Rn-222 content of soil gas correlated with uranium in the soil but not with buried uranium deposits; in another, impermeable strata prevented detection of uranium deposits by emanation methods; in another, Rn-222 in soil gas gave an 8-fold anomaly over a uranium-bearing sandstone buried at a depth of 0.3 to 9 m in a sandstone stream-channel sequence.

Miller and Ostle (1973) described the successful tracing of a uranium-mineralized fault, covered by as much as 2 m of glacial drift, by measurement of Rn-222 in soil gas.

Caneer and Saum (1974) described a soil-gas Rn-222 anomaly that correlated closely with the vertical projection of uranium ore at a depth of 110-120 m.

Gingrich (1975) and Gingrich and Fisher (1976b) have reported results of Rn-222 measurements of soil gas by means of sensitized cellulose nitrate films positioned so as to detect only airborne emitters in soil gas. Gingrich and Fisher have cited six instances where anomalous soil-gas α measurements have been correlated with significant uranium deposits at depths of 25, 75, 100, about 150, about 150, and 185 m, respectively. The 75-m- and 185-m-deep deposits were below the respective water tables, at 3-24 m and 30 m. The 100-m-deep deposit was displaced down the bedding plane of its host rock, so that its vertical projection to the surface was displaced from the location of the soil-gas anomaly. Numerous other associations of anomalies with uranium mineralization at unspecified depths have also been cited. Because the film measurements are usually made for 30-day periods, they have the advantage of averaging out daily (but not seasonal) fluctuations. By means of a permeable barrier, Rn-220 can be delayed so that it and its decay products cannot reach the sensitive film (Ward et al., 1977).

Morse (1976), using an α -scintillation chamber, found that measurements of Rn-222 in soil gas in podzolic soils and glacial till could locate radioactive pegmatite buried by about 3 m of overburden. Fluctuations in Rn-222 concentration were not significant.

Warren (1977) reported tests of the film method and of cups containing electronic semiconductor α detectors, which permit readings to be made without disturbance of the sample point. Little significant difference was found among results using counting periods of 3 days and longer. Both types of apparatus showed strongly anomalous α activity in soil gas over uranium mineralization located at depths of less than 20 m.

Causes of Radon-222 Anomalies. Enough case histories have correlated buried uranium deposits with Rn-222 soil-gas anomalies to suggest strongly that they are causally related. The relation is sometimes stated and more often is implied or inferred to be one of direct migration of Rn-222 from the uranium deposit to the near-surface point of measurement. Migration through the greater of the distances cited in the case histories above requires such great attenuation of the Rn-222 flux density that the remainder should be masked by a larger background Rn-222 concentration generated by Ra-226 in the soil. It is easily derived that the background concentration, B, is related to the concentration of Ra-226 as follows:

$$B \text{ (nCi/l)} = (\rho \cdot K \cdot e^{226}\text{Ra})/3\epsilon \quad (7)$$

where ρ is the soil density in g/cm^3 ; K is the fractional emanating power; ϵ is the soil porosity; and $e^{226}\text{Ra}$ is the U-238 equivalent at equilibrium, in parts per million, of the activity of Ra-226 present. Selecting reasonable values of 0.2 for emanating power, 1.5 for density, and 0.2 for porosity, we obtain the result that the background concentration of Rn-222 in soil gas should be roughly 0.5 nCi/l-ppm U-238 if equilibrium prevails through Ra-226. Considering that soils typically contain 1 ppm of U-238 (Baltakmens, 1976), and that both emanating power and porosity tend to be greater for fine soils and less for coarse soils, so that the ratio, K/ϵ , is not greatly variable, we may conclude that the undepleted concentration of Rn-222 in typical soil gas should be in the range 0.1-1 nCi/liter of soil gas. Depletion of the Rn-222 by

exhalation would affect both the signal and the background Rn-222. The progress report of Jeter et al. (1977, Figure 9) shows attenuation curves for a 1-m-thick layer of 0.6% U_3O_8 of 20% emanating power at a depth of 60 m in a homogeneous inactive overburden having an associated true diffusion coefficient of $0.02 \text{ cm}^2/\text{s}$ ($D^* = 0.13 \text{ cm}^2/\text{s}$). The Rn-222 concentration is equal to the nominal background level of 0.1 nCi/l ($10^{-3.25} \text{ d/s-cm}^3$ of soil) at a depth of about 35 m in the absence of a transport component. In the presence of a $1 \times 10^{-4} \text{ cm/s}$ constant upward transport component, the background level is reached at about 17-m depth. Figure 17 of their work shows that even relatively long-period (7-day) atmospheric pumping has less than a 3% effect at 20 m; thus, the simple diffusion model is appropriate for deeply buried ore. The concentration-cell mechanism proposed by Mogro-Campero and Fleischer (1977) for generating constant upward transport velocities substantially greater than the $1 \times 10^{-4} \text{ cm/s}$ used by Jeter et al. could cause Rn-222 migration through longer distances, but the hypothesis remains to be proved.

Where a uranium deposit lies more than a few meters below the water table, the diffusion process is not adequate to result in significant soil-gas anomalies from migrating Rn-222; a transport mechanism is required. The radon isotopes are too dispersed to associate and form bubbles that could move because of their buoyancy. They can be carried by major gases, as is observed at some natural springs and in volcanic gases. The possibility of emanation of major gases from a distributed uranium deposit (in contrast with mineralization along a fracture), so as to be useful in prospecting, does not appear to have been tested. To the extent that upward-moving water might transport radon isotopes, it should be even more effective in creating Rn-222 anomalies by transporting Ra-226, U-234, and U-238 upward from a deposit and leaving a "halo" of activity nearer the surface. Classic examples of such anomalies are the highly emanating hot-spring deposits, in which Ra-226 is coprecipitated with calcium and magnesium carbonates (see, for example, Wollenberg, 1974). Fluctuations of the water table during a period commensurate with the half lives of Ra-226 (1.6×10^3 years) and U-234 (2.45×10^5 years) should be geologically reasonable and adequate to have resulted in many, if not all, Rn-222 soil-gas anomalies. Since my earlier review, research has

shown that U-234 in particular is preferentially leached from uranium-bearing rock and migrates in ground waters (Granger, 1963; Rosholt et al., 1966; Osmond and Cowart, 1976).

Soil-gas Rn-222 anomalies that might well have been inferred as rising from a roll-front uranium deposit were found by Martin and Bergquist (1977, Figure III-10) to be closely correlated with the gamma-ray analyses of the soil samples removed from the holes bored for the soil-gas analyses; the gamma-ray analyses were dependent upon the concentrations of the short-lived Rn-222 decay products, Pb-214 and Bi-214. Schutz (1978) found, from measurements of Ra-226 and Pb-210 from holes 3 to 60 m deep over a uranium deposit at about 130-m depth, that the Rn-222 measured in the soil gas could be accounted for by local sources.

Lack of correspondence between surface gamma-ray anomalies and those of Rn-222 in soil gas is sometimes proposed as evidence for migration of Rn-222 from depth. Such a conclusion is quite unnecessary. Gamma radiation measured at the surface by total-count instruments includes cosmic radiation and radiation from K-40, from the thorium series, and sometimes from fallout, as well as that from the uranium series. The gamma-ray component from the uranium series is depleted further by exhalation of Rn-222 from the upper soil layers. The insensitivity of surface gamma radioactivity to buried uranium in comparison with Rn-222 in soil gas is consequently an effect of a poorer signal-to-background ratio. Sensitivity can be improved by making the gamma-ray measurement in the ground, where the cosmic and fallout radiations are shielded to some extent and where the concentrations of Rn-222 and its decay products are much greater; use of gamma-ray spectrometry makes the measurement specific to the uranium series. Michie et al. (1973) have described a thorough examination of radiometric anomalies in northern Scotland, where surface gamma-ray, gamma-probe, and soil-gas Rn-222 measurements and analyses of soil and water samples were all used to study the dispersion of uranium-series nuclides from shallow bodies of uranium-bearing rock. Uranium, copper, lead, zinc, and all the radiometric anomalies were found to be displaced by as much as 100 m downslope in overburden comprising peat, soil, glacial till, and residual soil; in some places, the gamma-probe and Rn-222 anomalies largely coincided.

Bhatnagar (1973) and Tewari and Bhalla (1976) have found that Rn-222 concentrations in soil gas at a given place fluctuate so that repeated measurements give a lognormal distribution in ground containing only local sources of Rn-222 and a compound lognormal distribution if Rn-222 is derived from both local and distant sources.

An excellent review of the use of Rn-222 for prospecting for uranium and descriptions of the origins of various types of anomalies have been presented by A.Y. Smith et al. (1976).

Earthquake Prediction

Annual measurements of Rn-222 concentration in hot mineral water from an aquifer 1300 to 2400 m deep in the Tashkent Basin, Uzbek S.S.R., yielded consistent but slowly rising values of 0.55 to 0.60 nCi/l during the years 1956-1959. When measurements were resumed in 1965, the values had risen to nearly 1.3 nCi/l and continued to rise at a rapid rate, reaching 1.5 nCi/l on April 20, 1966. Following a magnitude-5.3 earthquake near the water well on April 26 and the aftershocks, the Rn-222 concentrations had decreased to 0.75 nCi/l and continued to decrease to about 0.5 nCi/l. These findings, published by Ulomov and Mavashev (1967), provoked great interest in the use of changes in the Rn-222 concentrations in ground water and in soil gas to warn of impending earthquakes. In May 1976, the Rn-222 concentration in thermal mineral water in the same basin rose from a normal level to one about four times higher in about a day's time, oscillated for 4 days until the magnitude-7.3 Gazli earthquake struck about 20 km away, and then declined to the normal level in another week (Sultankhodzhayev et al., 1976). Instances of Rn-222 anomalies in ground water preceding earthquakes have been reported by the Red Mountain Observatory, Kunming, China (Sykes and Raleigh, 1975). The Seismological Brigade of Hebei (Hopeh) Province, China (1975), has reported ground-water Rn-222 anomalies preceding seven earthquakes from 1969 to 1974, ranging in magnitude from 4.3 to 7.9; they imply that anomalies have been noted with many other earthquakes. Accurate predictions were issued for the magnitude-4.3 earthquake at Sin Tang on August 5, 1971, and for the magnitude-4.9 earthquake at Hew-Jin on June 6, 1974. Although Rn-222 anomalies in ground water in connection with land subsidence and slides in Japan have been

observed by Kimura and Komae (1978), they have not observed premonitory effects such as those mentioned above. Studies of the use of Rn-222 to predict volcanic eruptions in Kamchatka have been reported by Chirkov (1973) and by Sugrobov and Chirkov (1974).

Measurements of Rn-222 in ground water and in soil air near potential earthquake locations in California are being conducted by several groups, represented in the Seismicity and Radioactivity session of this symposium and by publications by Teng et al. (1975), King (1976a, 1976b, 1977b, 1977c, 1977d, 1978), King and Teng (1976), A.R. Smith, Bowman et al. (1976), A.R. Smith, Wollenberg, and Mosier (1976, 1977), Birchard and Libby (1976, 1977, 1978), Shapiro et al. (1977), and Wollenberg et al. (1977). King (1976b, 1978) observed two large coherent maxima of radon-isotope concentration in soil gas from 20 stations along fault segments of about 60-km length in central California; the maxima correlate with two moderate earthquakes, with earthquake energy release, and with the winter seasons of 1975-1976 and 1976-1977. He believes the seasonal correlation to be fortuitous because of the absence of similar anomalies at stations in the same climatic zone but removed from the earthquake region. Birchard and Libby (1976, 1978) noted both positive and negative anomalies in soil gas preceding several moderate earthquakes in southern California, but question the negative anomalies. Shapiro et al. (1977) reported a decrease in the Rn-222 concentration in ground-level air before a magnitude-3.2 earthquake near Newhall, California, in early 1977. Cathey (1977) and Moore et al. (1977) reported decreases in Rn-222 concentration in ground water preceding a magnitude-2.3 earthquake in South Carolina on February 23, 1977.

Methods of continuous monitoring of Rn-222 in ground water have been described by Cathey (1977) and by Noguchi and Wakita (1977).

Neither the details of the behavior of rock and interstitial water in the zone of an impending earthquake nor the mechanisms causing the anomalous Rn-222 concentrations have yet been established. Two principal models of the phenomena occurring in an earthquake zone and their implications with respect to various parameters of potential value in prediction have been discussed by Mjachkin et al. (1975). Substantial fluid migration is permitted by one of the models and is

required by the other. Mavlyanov et al. (1971) suggested that ultrasonic vibrations in the deep zones promote release of U-234 and Rn-222. Gorbushina et al. (1972) noted that the concentration of helium and the ratio, $^{234}\text{U}/^{238}\text{U}$, were anomalously great in some ground waters near Tashkent at the time of the 1966 earthquake and also proposed that ultrasonic vibrations could cause release of the helium and U-234 and could accelerate their diffusion. Sultankhodzhayev et al. (1974) noted that the Rn-222 concentration in ground water is controlled by the rate of flow of the water through radioactive layers of the ground and reported that the emanating powers of some rock samples were increased at high pressure.

Intuition tells us that pre-earthquake fracturing, if it takes place, should increase the rock surface exposed to circulating fluids and consequently should increase Rn-222 release to the fluids, resulting in a positive anomaly. However, the effect of deformation is probably not so simple, owing to sometimes counteracting factors: emanating-power change, transient release of local concentrations of Rn-222, permeability change, and change in the rates of flow of fluids through zones of greater and lesser concentrations of emanating Ra-226. Emanating power is directly related to the total surface area communicating with rock pores; if an extensive network of internal capillaries in the grains of a rock is responsible for most of its emanating power, as must often be true, simple shearing of the rock grains should increase the total surface and the emanating power only slightly. Starik and Melikova (1957, pp. 225-226 of translation) concluded that pulverization of the mineral khlopinite did not increase its emanating power. Starik and Lazarev (1960) stated that the emanation ability of minerals depends only slightly on the change in specific [outer] surface; they point out that the internal surface is due to a network of capillaries that is little affected by crushing. The Seismological Brigade of Peking (1977) measured the quantity of Rn-222 that could be flushed from Ra-226-loaded porous cement cubes before and during uniaxial compression. Following a 3-year storage period, each of three cubes was flushed with air, the effluent being continuously monitored by a scintillation counter. Uniaxial compression was applied until rupture occurred. The concentration of Rn-222 collected increased at the time of rupture by 22% to 107%.

Similar effects were said to be observed in experiments with natural rock samples. Because the experiment periods (about 1 hr) were brief compared with the half life of Rn-222, it is open to question whether the observed increases would persist for a long enough time to result in significant anomalies. Fracturing is an unlikely explanation for Rn-222 anomalies that have been observed as much as 300 km from the epicenters.

The most comprehensive explanation of Rn-222 behavior noted in connection with deep earthquakes is that of Sultankhodzhayev et al. (1977). They considered that the emanating power of the reservoir rocks may be changed as the microcapillary system in the rocks is changed by deformation; that the rate of movement of ground water from Ra-226-bearing layers to inactive layers should influence Rn-222 concentration in the water; and that electrical changes accompanying deformation of the rock may cause changes in its adsorption capacity and consequently in the retention of Rn-222 on the rock surfaces. Sultankhodzhayev et al. (1977) believe that it is possible to predict the location of the focus of an earthquake by means of continuous measurement of Rn-222 in ground water from points along radii from the epicentral zone. Whatever mechanisms may be suggested, they need to account for the observations that Rn-222 concentrations in ground water may be either anomalously high or anomalously low prior to an earthquake, may be observably anomalous far away from the zone of inelastic deformation, and return to the normal level later.

It is reasonable that squeezing in zones of compression causes a net upward and outward flow of soil gas, thereby shifting upward the profile of Rn-222 concentration near the surface. Measurements of Rn-222 in soil gas at a standard depth in the soil should then show positive anomalies near zones of compression (and, conversely, negative anomalies near zones of dilatation, where intergranular spaces are enlarging). This mechanism has been proposed by King (1976b, 1977b) and by Birchard and Libby (1976, 1978) to explain soil-gas Rn-222 anomalies observed respectively in central and southern California.

RECOMMENDATIONS FOR RESEARCH

The importance of adsorption in retarding the movement of radon isotopes and holding them on the surfaces of common minerals, rocks, and soils under typical conditions needs to be studied. The effect of water on adsorption should be determined.

Radiation-damage experiments of the type described by Lambert and Bristeau (1973) should be performed on a greater variety of minerals and rocks. The effect of varying the absolute pressure during the α -counting period and the indirect-recoil mechanism should be studied concurrently.

Treatments of radon-isotope diffusion have tacitly assumed that gravitational effects are negligible. Although the assumption is reasonable, it appears not to have been verified. It could be tested simply by performing diffusion measurements in horizontal and vertical orientations.

The long-distance migration of Rn-222 needs to be tested rigorously at places where it is thought to occur. One approach would be to measure the profiles of Rn-222 concentration in interstitial fluids at different depths to sufficient depth to establish whether the profile followed the required approximately exponential form. A second approach, reported by Schutz (1978), would be to measure the Pb-210 concentration profile, which should indicate the average Rn-222 profile for the preceding several decades. The Ra-226 concentration at each point should be determined in either of the profile measurements. A third, but less satisfactory, approach would be to compare measured Rn-222 concentrations at a standard depth in the horizontal plane with the Rn-222 production rates at the respective sites. For all three approaches, corrections would have to be made for the disturbance caused by the measurements and for the effects of variable porosity, saturation, temperature, and emanating power; and measurements should be sufficiently standardized to permit computation of the balance between sources and losses of Rn-222.

ACKNOWLEDGEMENTS

I thank my colleague, Frank E. Senftle, for calling attention to many pertinent abstracts. Numerous authors have favored me with much-appreciated reprints of their publications.

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