FACTORS AFFECTING THE MIGRATION OF RADIUS, RADON AND RADON DAUGHTERS IN SOIL SITUATIONS

by

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FACTORs AFFECTING THE MIGRATION OF RADIUM, RADON AND RADON DAUGHTERS IN SOIL SITUATIONS

1. INTRODUCTION

The purpose of this review is to identify the factors determining the distance that radium, radon and decay products migrate in the ground, and also to discuss the implications of radon migration.

Groundwater plays an important role in the migration, dispersion and concentration of radioactive elements in the soil-system. A dynamic equilibrium exists between radioactive elements in the soil and in the water in contact with the soil. This equilibrium undergoes constant disruption during groundwater flow, both as a result of radioactive decay and as a result of the processes of leaching, dissolution, oxidation, adsorption, sedimentation, etc. (Tokarev, 1956). The decay products during the processes of radioactive transformations can also, as a result of radioactive recoil, leave the spheres of the atomic and molecular bonds and be leached out without dissolution of the mineral. Disturbance of the radioactive equilibrium is particularly substantial in natural waters and in the atmosphere (Baranov, 1956).

The following main factors are important in evaluating both the behaviour and fate of radioactivity released by radium and its decay products:

(i) "primary migration" processes: oxidation, leaching, dissolution, diffusion, emanation.

(ii) "secondary migration" processes: deposition, adsorption, ion-exchange, de-emanation.
(iii) hydrogeological and climatic conditions
(iv) physicochemical characteristics of radioactive elements.

2. MIGRATION IN THE GROUND

Tanner (1964) presents an excellent review of many variables significant in the migration of radon isotopes in the ground: the decay rate of the isotope, the diffusion constant for the isotope in the pore-filling fluid, the porosity of the ground, the velocity and composition of the fluid, and the temperature-dependent distribution of the radon isotopes among the phases if the fluid has more than one phase.

<table>
<thead>
<tr>
<th>Diffusing Isotope</th>
<th>Fluid</th>
<th>Medium</th>
<th>D(cm²/sec)</th>
<th>Conditions</th>
<th>Authority</th>
</tr>
</thead>
<tbody>
<tr>
<td>Rn, Tn, An</td>
<td>Air</td>
<td>Continuous</td>
<td>1.0 × 10⁻¹</td>
<td>Various</td>
<td>Various, quoted in Zimens, 1943</td>
</tr>
<tr>
<td></td>
<td>He</td>
<td></td>
<td>2.0 × 10⁻¹</td>
<td>15°C, 76 cm. Hg</td>
<td>Hirst and Harrison, 1939</td>
</tr>
<tr>
<td>Rn</td>
<td>Ne</td>
<td></td>
<td>3.5 × 10⁻¹</td>
<td></td>
<td></td>
</tr>
<tr>
<td></td>
<td>Ne</td>
<td></td>
<td>2.1 × 10⁻¹</td>
<td></td>
<td></td>
</tr>
<tr>
<td></td>
<td>He</td>
<td></td>
<td>9.2 × 10⁻⁴</td>
<td></td>
<td></td>
</tr>
<tr>
<td></td>
<td>Ar</td>
<td></td>
<td>2.69 × 10⁻⁴</td>
<td></td>
<td></td>
</tr>
<tr>
<td></td>
<td>Alcohol</td>
<td></td>
<td>2.67 × 10⁻⁴</td>
<td></td>
<td></td>
</tr>
<tr>
<td></td>
<td>Toluene</td>
<td></td>
<td>2.30 × 10⁻⁴</td>
<td></td>
<td></td>
</tr>
<tr>
<td></td>
<td>Benzene</td>
<td></td>
<td>1.13 × 10⁻⁴</td>
<td></td>
<td></td>
</tr>
<tr>
<td></td>
<td>Water</td>
<td></td>
<td>5.4 × 10⁻⁵</td>
<td></td>
<td></td>
</tr>
<tr>
<td></td>
<td>Air, 4% moisture</td>
<td>Building sand 1.40</td>
<td>1.0 × 10⁻¹</td>
<td>Various</td>
<td>Various, quoted in Zimens, 1943</td>
</tr>
<tr>
<td></td>
<td>Building sand</td>
<td>gm/cm³, 39% porosity</td>
<td>1.2 × 10⁻¹</td>
<td></td>
<td></td>
</tr>
<tr>
<td>Tn</td>
<td>Air, no moisture</td>
<td>Fine sand (mostly</td>
<td>6.8 × 10⁻⁴</td>
<td>Not stated</td>
<td>Grammakov, 1936</td>
</tr>
<tr>
<td></td>
<td>quartz</td>
<td></td>
<td></td>
<td></td>
<td></td>
</tr>
<tr>
<td></td>
<td>Air, 8.1% moisture</td>
<td></td>
<td>5.0 × 10⁻⁴</td>
<td></td>
<td></td>
</tr>
<tr>
<td></td>
<td>Air, 15.2% moisture</td>
<td></td>
<td>1.0 × 10⁻⁴</td>
<td></td>
<td></td>
</tr>
<tr>
<td></td>
<td>Air, 17% moisture</td>
<td>Eluvial-detrital</td>
<td>5.0 × 10⁻⁴</td>
<td></td>
<td></td>
</tr>
<tr>
<td></td>
<td>Sand</td>
<td>deposits of granodiorite</td>
<td>4.5 × 10⁻⁴</td>
<td></td>
<td></td>
</tr>
<tr>
<td></td>
<td>Air, 37.2% moisture</td>
<td>Diluvium of</td>
<td>1.8 × 10⁻⁴</td>
<td>Mean effective</td>
<td>Popretinskiy, 1961</td>
</tr>
<tr>
<td></td>
<td>Diluvium of</td>
<td>metamorphic rocks</td>
<td></td>
<td>value in natural</td>
<td></td>
</tr>
<tr>
<td></td>
<td>Eluvial-detrital</td>
<td>Eluvial-detrital</td>
<td>1.5 × 10⁻⁴</td>
<td>occurrence</td>
<td></td>
</tr>
<tr>
<td></td>
<td>deposits of</td>
<td>deposits of granite</td>
<td></td>
<td></td>
<td></td>
</tr>
<tr>
<td></td>
<td>granite</td>
<td></td>
<td></td>
<td></td>
<td></td>
</tr>
<tr>
<td></td>
<td>Loams</td>
<td></td>
<td>8.0 × 10⁻⁴</td>
<td></td>
<td></td>
</tr>
<tr>
<td></td>
<td>Varved clays</td>
<td></td>
<td>7.0 × 10⁻⁴</td>
<td></td>
<td></td>
</tr>
<tr>
<td></td>
<td>Mud, 1.57 gm/cm³</td>
<td></td>
<td>5.7 × 10⁻⁴</td>
<td>19°-20° C.</td>
<td>Baranov and Novitskaya, 1949</td>
</tr>
<tr>
<td></td>
<td>Mud, 1.02 gm/cm²</td>
<td></td>
<td>2.2 × 10⁻⁴</td>
<td>19°-20° C.</td>
<td></td>
</tr>
<tr>
<td></td>
<td>Barium nitrate</td>
<td></td>
<td>8.0 × 10⁻⁹</td>
<td>Not stated (room</td>
<td>Strassmann, quoted by Flugge and Zimens, 1939</td>
</tr>
<tr>
<td></td>
<td></td>
<td></td>
<td></td>
<td>temperature)</td>
<td></td>
</tr>
</tbody>
</table>
Table 1 presents a summary of coefficients determined experimentally for the diffusion of radon isotopes in various continuous and porous media. A lengthy list of diffusion coefficients for geologic materials of low moisture content may be found in the Soviet Manual for Radiometric Prospecting (Alekseyev, 1957, Table 50). Principal references on the effects of moisture are Grammakov (1936) and Baranov and Novitskaya (1949); the latter also investigated the diffusion rate as a function of temperature.

Several generalisations appear reasonable from the literature:

1. Diffusion coefficients for the several radon isotopes in identical media are practically equal.

2. The diffusion coefficient is not sensitive to the pore diameter in the usual range of geologic materials.

3. Increasing moisture in a porous medium causes reduction of diffusion coefficient greater than can be accounted for by the pore space occupied by water (Grammakov, 1936).

4. The diffusion coefficient is less than the diffusion constant in the presence of a solid matrix. According to Penman (1940, a b as quoted by Baver, 1956 pp213-14), the ratio of the diffusion coefficient for a vapour in a porous medium to the diffusion constant in air is proportional to the pore space, the proportionality constant being 0.66. Penman's relation appears to give fair values for radon diffusion in dry soils.

2.1 Migration in the Unsaturated, Uppermost Ground Layers

Meteorological factors have a pronounced effect on the transport of radon in the uppermost ground layers. The three most commonly observed effects on radon concentration and exhalation are rainfall
(Wright and Smith, 1915, and many others), freezing and snow cover
(Bender 1934, Zupancic, 1934 and many others). Both transport and
diffusion are affected. With heavy rainfall, the soil gases near
the surface tend to be displaced upward, carrying radon with them
and increasing the exhalation rate temporarily. Thereafter, the
reduced diffusion coefficient and reduced permeability (Alvarez, 1949)
of the wet ground restrict migration by both mechanisms. Jaki (1958)
measured the concentrations of radon, thoron and their decay
products immediately above and below the ground and concluded that
the concentration in the earth's vicinity depends primarily on the
dryness of the ground. Pearson (1967) also studied the influences
of meteorological and soil conditions, time, soil type, and location
upon the release of radon-222 from soils in the local area, Champaign
County, Ill. He concluded that for very low wind speeds, ordinarily
observed at 1.25 cm above the ground, there is no demonstrable
variation of emanation with variation in wind speed. He also
observed that when the soil was wet (air void fraction about 0.17),
the emanation measured was approximately one-third of that observed
when average moisture conditions (air void fraction about 0.40)
were prevalent and that there was a marked decrease of emanation
when the soil was cold or frozen.

2.2 Summary of Migration Distances

Table 2 (Tanner, 1964) is a summary of migration distances for radon
in one-dimensional and cylindrical flow, corresponding respectively to
vertical flow from horizontal sources and to flow into a water well.

TABLE 2 (Tanner, 1964)

SUMMARY OF MIGRATION DISTANCES FOR RADON-222

<table>
<thead>
<tr>
<th>Steady-State Conditions</th>
<th>Approximate Migration Distance (Meters)</th>
</tr>
</thead>
<tbody>
<tr>
<td></td>
<td>Maximum in 5.5 Days (C/C₀ = 1/4)</td>
</tr>
<tr>
<td>One-dimensional diffusion; dry soil</td>
<td>5?</td>
</tr>
<tr>
<td>One-dimensional diffusion; moist soil</td>
<td>0.5</td>
</tr>
<tr>
<td>One-dimensional diffusion; saturated ground</td>
<td>0.02</td>
</tr>
<tr>
<td>One-dimensional transport by ground water</td>
<td>500</td>
</tr>
<tr>
<td>Radial transport by ground water</td>
<td></td>
</tr>
</tbody>
</table>

For the maximum distance at which radon in soil gas would signal a buried deposit of radium-bearing material in a one-dimensional situation, Alekseyev (1957) estimates about 6 metres, Budde (1958b) estimates several metres, Grammakov et al (1958) estimate 13 metres, and Narton and Stegena (1962) estimate 5-10 metres (over petroleum accumulations). Sharkov (1959) states that radon in ground water is removed in tens of metres and, more rarely, in the first hundred metres. Migration of the several most mobile members of the uranium series, uranium-238, uranium-234, radium-226, and radon-222 is probably responsible for many anomalies, particularly those which imply movement through distances of many metres.

2.3 Migration in Waters

It is possible to identify some patterns of radon migration in subterranean waters (Alekseyev, 1957):

1. Radon waters of uranium-radium deposits. The enrichment of water by radon occurs at the expense of radium located in the vicinity
of its parent elements (Thorium-230, U).

2. Radon waters of "dispersed" type. The enrichment comes from radium which occurs in a dispersed state. An important part is played by high emanation and by duration of contact. This is the type of interstitial water characteristic of the zones of weathered crystalline rocks, or the waters in quaternary deposits (with slow, stagnant flow).

3. Radon-radium waters. Radium, upon penetration into liquid medium (as a result of leaching or dissolution), moves with it. In the case of high radium concentrations in water, there may develop a large percentage of radon in water. (For waters containing .4 pCi/L of radon, the concentration of radium must be equal to $4.10 \times 10^{-9}$ g/L, at equilibrium.

4. Depending upon the conditions, the adsorption of radium in the fissure may proceed with totality. Under conditions unfavourable for radon enrichment of water (high velocity of movement, wide fissures etc.) the content of radon in water may be very low. (Contents of radium from $10^{-11}$ to $10^{-9}$ g/L are observed.)

In conclusion, it should be noted that, in natural radon waters, the radioactive equilibrium between radon and the parent elements is usually sharply shifted towards the enrichment of waters with radon. There is no interdependence between the concentrations of radon, radium, and uranium in waters. Consequently, the geochemistry of radon in waters is not related to the geochemistry of uranium and may also be unrelated to that of radium. (Alekseyev, 1957).
3. IMPLICATIONS OF RADON MIGRATION AT PORT HOPE: A DISCUSSION OF POSSIBLE PHYSICAL AND CHEMICAL CONTROL MECHANISMS.

In view of the fact that chemical and radioactivity data on the behaviour of radium-226 and radon-222 in the Port Hope environment are not available, any conclusions reached here must be considered tentative. Nevertheless, it is necessary to suggest various mechanisms that affect and control in ground water the behaviour of radium-226 and radon-222.

3.1 Radon-Sources in Underground Waters

The half-life of radon-222 is 3.8 days. The presence of radon in ground-water at a substantial distance (>10 m) from its parent requires either that its initial concentration be quite high or that it move rapidly. Neither condition is likely to prevail for the following reasons:

1. In the saturated sediments the diffusion coefficient of radon-222 can be expected not to exceed its diffusion coefficient in water, which is reported by Zimens (Table 1) to be $1.13 \times 10^{-15}$ cm$^2$/sec at 18°C. One-dimensional diffusion at this rate results in a hundred-fold diminution of concentration over a distance of about 0.1 m. Therefore, transport, rather than diffusion must be the dominant mechanism if there is to be significant migration of radon. But transport by water moving nearly horizontally at a typical rate of 0.3 m/day or less would take radon only a short distance in the 25 days required for hundred-fold diminution by radioactive decay (Table 2).

2. It is conceivable that paths of high permeability exist along which radon is transported quickly enough that much of it is undecayed
after the passage. It is geologically reasonable that such high-speed conduits can exist, but direct evidence for or against the existence of such channels in the Port Hope area needs to be established. Weighing against such a possibility of conduits is the consideration that continuously high permeability would not be maintained over a path length of about 100m. (Thomas and Nelson, 1948). Also, even a short section of low permeability in an otherwise highly permeable chain would retard water movement long enough to permit substantial decay of radon-222 (Tanner, 1964).

Surface runoff would, however, constitute a fast and therefore important water vector. Surface runoff may be important in the Pidgeon Hill area.

Vertical transport of anomalous amounts of radon-222 from bedrock is improbable and hydrologically unreasonable (Rogers, 1954). It is therefore necessary to look backward along the migration path for an adequate supply of the radium-226 parent. The probable sources of the natural sediments noted by Rogers (1954) suggest that the radium-226 source of the radon-222 is a dispersed one rather than a concentrated one. He also observed that most of the radon measured in a water sample comes from radium-226 that is quite close to the well bore and is immobile enough that it is not present in the water sample.

3.2 Physical Mechanisms

The radon concentration in the interstitial water is dependent not only on the source density but also on the ease with which radon escapes from the sites of disintegration of radium-226 atoms into
the interstices of the sediments, and on the porosity of the sediments. These latter factors represent physical controls on the radon distribution and should be independent of the source density because of the extreme dispersion under natural conditions of the radium-226 on an absolute basis (approx. \(10^{-13}\) grams of radium per gram of sediment) (Tanner, 1964).

It will be argued, in the following considerations of possible physical controls, that none of the physical factors are of any great significance or responsible for patterns of radon distribution in ground water. The physical controls could, of course, combine to yield an observed pattern but a systematic study would have to be conducted to reinforce such observations.

The ease with which radon can get into the interstitial water depends on the material in or on which radium is immobilized. Flugge and Zimens (1939) suggest that since the range of radon-ion recoiling from a disintegrating radium atom and the diffusion rate of the radon atom thereafter are so small, that the contribution of radon from within sediment grains to interstitial waters is likely to be small.

The contribution from exterior sources and microfissures may be appreciable if weathering or chemical corrosion has formed coatings or a crust in which radon may diffuse much more rapidly than in a crystal (Hayase and Tsutsumi, 1958). Films of hydrous of gelatinous iron oxide, such as might be deposited on the grain surface, will also yield a large fraction of the radon emanating from radium trapped in them (Hahn, 1936). In addition, radon should be readily available
from radium-226 adsorbed on clay particles or on the exterior surfaces of clay lenses.

The sources of major radon contribution described above, however, are those already exposed to interstitial water. Experiments by Starik and Melikov (1957, Table 13) have revealed that the amount of radon made available to passing fluids may be the same or less from rock after it is crushed than before. They also observed (Table 14) the temperature dependency of emanation of radon from uranium ore at 20°C and concluded that the increase was approximately 1 per cent/°C.

The effect of temperature on enrichment of ground water with radon is shown in Table 3. (below). Since radon is a gas, an increase in the temperature results in enhanced degassing of the water, due to which the coefficient of distribution of radon between water and air (coefficient of solubility) is decreased.

Table 3.

<table>
<thead>
<tr>
<th>Temperature of the water in °C</th>
<th>α</th>
<th>Temperature of the water in °C</th>
<th>α</th>
<th>Temperature of the water in °C</th>
<th>α</th>
</tr>
</thead>
<tbody>
<tr>
<td>0</td>
<td>0.510</td>
<td>30</td>
<td>0.200</td>
<td>70</td>
<td>0.118</td>
</tr>
<tr>
<td>5</td>
<td>0.420</td>
<td>40</td>
<td>0.160</td>
<td>80</td>
<td>0.112</td>
</tr>
<tr>
<td>10</td>
<td>0.350</td>
<td>50</td>
<td>0.140</td>
<td>90</td>
<td>0.109</td>
</tr>
<tr>
<td>20</td>
<td>0.225</td>
<td>60</td>
<td>0.127</td>
<td>100</td>
<td>0.107</td>
</tr>
</tbody>
</table>
Numerous investigations have shown that the theoretical values of the coefficients \( a \) are fairly close to those observed in nature. (Tokarev, 1956). (See Table 4).

Table 4. (Tokarev, 1956)

<table>
<thead>
<tr>
<th>Spring</th>
<th>Temp. of Water in °C</th>
<th>Rn cont. in Mc. of water</th>
<th>Rn cont. in Mc. of gas</th>
<th>Value of actual theor.</th>
</tr>
</thead>
<tbody>
<tr>
<td>Shiviya (Transbaikal)</td>
<td>0,1</td>
<td>16,2</td>
<td>39,6</td>
<td>0,40</td>
</tr>
<tr>
<td>Archan-Tunkinskii</td>
<td>8,8</td>
<td>2,4</td>
<td>3,3</td>
<td>0,73</td>
</tr>
<tr>
<td>Kislovodsk</td>
<td>13,6</td>
<td>4,7</td>
<td>7,0</td>
<td>0,67</td>
</tr>
<tr>
<td>Uch-Kainarskii (Northern Kirgiziya)</td>
<td>21,0</td>
<td>5,0</td>
<td>18,5</td>
<td>0,29</td>
</tr>
<tr>
<td>Agura, well No. 6, 400 m deep</td>
<td>29,6</td>
<td>3,5</td>
<td>10,2</td>
<td>0,34</td>
</tr>
<tr>
<td>Novaya Matsesta, well No. 9, 100 m deep</td>
<td>30,9</td>
<td>0,7</td>
<td>2,4</td>
<td>0,29</td>
</tr>
<tr>
<td>Tekhaltuba</td>
<td>34,0</td>
<td>4,3</td>
<td>13,6</td>
<td>0,23</td>
</tr>
</tbody>
</table>

As a result of the foregoing considerations, physical disturbances as such would not cause marked changes in the availability of radon from radium present in the sediments.

For a given amount of radon emanation into the interstices, the radon concentration will be greatest in sediments of lower porosity. Substantial porosity differences may be found among unconsolidated sediments (Manger, 1962). However, among heterogeneous sediments, the porosity range is smaller. In the Port Hope area, it is assumed that it is unlikely that porosity differences greater than twofold exist among zones from which radon is gathered. If this assumption
proves not valid, because of physical dumping, then it must be
given due consideration. The effect of strata variations on porosity
is difficult to assess. If a twofold or greater difference in average
porosity of the aquifer material between two points is observed,
then the variations should be taken into account.

Tokarev (1956, pp44) points out that radon is not adsorbed on rocks.
It can be assumed that differences in radon adsorption are negligible.

Hence, if the interpretation that physical factors are not responsible
for radon distribution is correct, then the distribution in waters
must be determined by the distribution of radium-226 in the sediment.

3.3 Chemical Mechanisms

From the foregoing, it is inferred that the radon concentrations in
waters would be roughly proportional to the concentrations of im-
mobilized radium in the sediments. The logical question to ask is:
What mechanism controls the small fraction of radium that could be
present in the sediments?

It is suggested, with respect to the Port Hope situation, that radium
in waters would either have been leached from the waste material or
introduced from radium sources (parent materials) outside the area
(i.e. dumping operations).

3.4 Leaching of Radium

Radium may be leached from the sediments under the following conditions:

1. pH of water is low enough to dissolve the alkaline earth carbonates.
2. if chelating agents are available to remove the alkaline earth or iron cations from precipitates in which radium is trapped.

3. or if other ions are present in sufficient concentration to displace radium.

Tokarev and Shcherbakov (1956, pp63) and Mazor (1962) have noted the tendency of ground waters high in chloride to be enriched in radium or have greater power to leach radium from rock (see Table 5).

Table 5 (Tokarev, 1956)

<table>
<thead>
<tr>
<th>Medium</th>
<th>% of leaching</th>
<th>Medium</th>
<th>% of leaching</th>
</tr>
</thead>
<tbody>
<tr>
<td>H₂O</td>
<td>0.012</td>
<td>0.0001-н. BaCl₂</td>
<td>0.040</td>
</tr>
<tr>
<td>0.001-н. NaCl</td>
<td>0.016</td>
<td>0.0001-н. BaCl₂</td>
<td>0.080</td>
</tr>
<tr>
<td>0.001-н. CaCl₂</td>
<td>0.032</td>
<td>0.001-н. BaCl₂</td>
<td>0.15</td>
</tr>
<tr>
<td>0.001-н. SrCl₂</td>
<td>0.036</td>
<td>0.01-н. BaCl₂</td>
<td>0.15</td>
</tr>
<tr>
<td>0.001-п. PbCl₂</td>
<td>0.088</td>
<td></td>
<td></td>
</tr>
</tbody>
</table>

Thus, on the basis of the effect which they produce on leaching of radium, the cations can be arranged in the following series:

\[ \text{Ba}^{++} > \text{Pb}^{++} > \text{Sr}^{++} > \text{Ca}^{++} > \text{K}^{+} > \text{Na}^{+} \]

The importance of water that is high in chloride is its complement of positive ions, which tend to compete for adsorption sites with radium and other alkaline earth ions, and its tendency to be concentrated enough to lower the activity coefficients of the constituents, thereby reducing their tendency to precipitate.
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An index of the tendency of water to promote desorption of the alkaline earth ions in favour of sodium ion is the sodium adsorption ratio (SAR) (Hem, 1959. pp 148). Tanner (1964) had observed high SAR values for waters containing more than 200 ppm chloride and concluded that high chloride waters should therefore have a greater ability to extract adsorbed radium and other alkaline earth ions. He also calculated the activity coefficients of selected ions in water samples using graphs given by Hem (1961 a). The activity coefficients for calcium and bicarbonate ions were 1.5 times lower in the high chloride waters than in the low chloride waters. (Table 6)

<table>
<thead>
<tr>
<th>Well Designation</th>
<th>Cl⁻ (p.p.m.)</th>
<th>Activity Coefficient</th>
<th>Calculated pH</th>
<th>Measured pH</th>
</tr>
</thead>
<tbody>
<tr>
<td></td>
<td></td>
<td>Fe⁺⁺</td>
<td>Ca⁺⁺</td>
<td>HCO₃⁻</td>
</tr>
<tr>
<td><strong>Burmester Area, Tooele County, Utah</strong></td>
<td></td>
<td></td>
<td></td>
<td></td>
</tr>
<tr>
<td>(C-2-5)bd-1.....</td>
<td>47</td>
<td>0.51</td>
<td>0.73</td>
<td>0.92</td>
</tr>
<tr>
<td><strong>Woods Cross Area, Bountiful District, Davis County, Utah</strong></td>
<td></td>
<td></td>
<td></td>
<td></td>
</tr>
<tr>
<td>(H 2-1)26dca-3</td>
<td>46</td>
<td>0.52</td>
<td>0.74</td>
<td>0.93</td>
</tr>
<tr>
<td>(H 2-1)26dcd-1</td>
<td>62</td>
<td>0.45</td>
<td>0.69</td>
<td>0.91</td>
</tr>
<tr>
<td>(H 2-1)26cdd.....</td>
<td>985</td>
<td>0.21</td>
<td>0.46</td>
<td>0.80</td>
</tr>
</tbody>
</table>

Table 6 (Tanner, 1964)
Hence the solubilities of the alkaline earth compounds in the high-chloride waters are thus effectively increased, and there is a greater tendency to retain them in solution. As was shown by the experiments of Starik (1957), the solubility of uranium and radium increases on increased acidity or alkalinity (Tables 7 and 8).

Table 7 (Starik, 1957)

<table>
<thead>
<tr>
<th>Medium</th>
<th>Content of Ra in solution in % of weighed sample</th>
<th>Content of U in solution in % of weighed sample</th>
<th>Ra/U</th>
</tr>
</thead>
<tbody>
<tr>
<td>H₂O</td>
<td>0.018</td>
<td>?</td>
<td>?</td>
</tr>
<tr>
<td>0.001-N. HCl</td>
<td>0.28</td>
<td>0.05</td>
<td>5.6</td>
</tr>
<tr>
<td>0.01-N. HCl</td>
<td>0.49</td>
<td>0.30</td>
<td>2.0</td>
</tr>
<tr>
<td>0.1-N. HCl</td>
<td>1.50</td>
<td>1.18</td>
<td>1.3</td>
</tr>
<tr>
<td>1.0-N. HCl</td>
<td>4.19</td>
<td>4.16</td>
<td>1.0</td>
</tr>
</tbody>
</table>

Table 8 (Starik, 1957)

<table>
<thead>
<tr>
<th>Medium</th>
<th>Content of Ra in solution in % of weighed sample</th>
<th>Content of U in solution in % of weighed sample</th>
<th>Ra/U</th>
</tr>
</thead>
<tbody>
<tr>
<td>H₂O</td>
<td>0.81</td>
<td>?</td>
<td>?</td>
</tr>
<tr>
<td>0.01-N. NaHCO₃</td>
<td>0.90</td>
<td>7.8</td>
<td>0.12</td>
</tr>
<tr>
<td>0.1-N. NaHCO₃</td>
<td>1.23</td>
<td>15.4</td>
<td>0.08</td>
</tr>
<tr>
<td>0.5-N. NaHCO₃</td>
<td>7.32</td>
<td>48.0</td>
<td>0.15</td>
</tr>
</tbody>
</table>
The transfer of radium from the solid matrix (soil and rock) into water is influenced by the chemical composition and acidity of the solution, amount of free oxygen and temperature, which in general affects the solubility.

The process of leaching can be considered as consisting of three stages (Tokarev, 1956):

i) As a result of reactive recoil, the radium and its isotopes accumulate in the water-filled "capillaries" of soils;

ii) An adsorption equilibrium is established between radium in solution and radium at the walls of "capillaries";

iii) As a result of diffusion the following equilibrium is established:

Ra in "capillary" water $\rightleftharpoons$ Ra in gravitational water

The diffusion occurs very slowly; even after one year the diffusion equilibrium does not have time to become established (Table 9).

Table 9 (Tokarev, 1956 pp 31)

<table>
<thead>
<tr>
<th>Consecutive Number</th>
<th>Time Interval Between Repetition of Experiment</th>
<th>Amount of Leached-out Radium, in %</th>
</tr>
</thead>
<tbody>
<tr>
<td>1</td>
<td>At the start of experiment</td>
<td>0.0150</td>
</tr>
<tr>
<td>2</td>
<td>After 13 days</td>
<td>0.0088</td>
</tr>
<tr>
<td>3</td>
<td>After 26 days</td>
<td>0.0040</td>
</tr>
<tr>
<td>4</td>
<td>After 15 months</td>
<td>0.0073</td>
</tr>
</tbody>
</table>
The rate of diffusion of radium from the "capillaries" into gravitational water is greatly dependent on the specific surface of the particles. The more finely dispersed the particles, the higher is the rate of diffusion of radium from solids into solution (Table 10).

Table 10 (Tokarev, 1956 pp 31)

Correlation Between Leachability of Radium and Dispersion-State of the Rock

| Specific surface of particles, in cm²/gram | 0.54 | 0.96 | 1.33 | 2.73 |
| Amount of leached-out radium, in % | 0.0025 | 0.0037 | 0.0080 | 0.0150 |
remedies, is available. (Environmental Radon and Radon Daughter Measurements at Port Hope, by C. R. Phillips and H. L. Pai).
adsorbed, at any dilution, by the precipitate, if the surface charge is opposite in sign to the charge of the ion being adsorbed and if the adsorbed compound is difficulty soluble in the given solvent. Radium in waters is readily adsorbed by clayey and by organic substances. This is evidenced by the great enrichment with radium of precipitates deposited by a number of radioactive springs (Tokarev, 1956).

Pearson (1967) examined soil samples collected from a wide area of the United States for "emanating radium 226" content. Clay type soils contained from \((0.342 \text{ to } 0.690) \times 10^{-12}\) grams of emanating radium 226" per gram of dry soil while sandy soils contained from \((0.147 \text{ to } 0.244) \times 10^{-12}\) g/g. Materials removed from an area where mining for uranium 238 or radium 226 had been practiced contained from \((0.45 \text{ to } 14.3) \times 10^{-12}\) grams per grain of dry soil.

Adsorption processes are akin to the processes of coprecipitation, which are characteristic of elements having similar ionic radii. Precipitation as radium sulphate is one of the processes that might remove radium from water. Boltzman (1963) conducted a literature search on the solubility of radium sulphate in water and concluded that the accepted value is Nikitin and Tomatschoff's value of \(2.1 \times 10^{-6}\) g/l, at 25°C.

It is well known that radium ions are efficiently removed from solution by coprecipitation with barium as the sulphate (Gordon and Rowley, 1957). When barium ions are present in waters with
sulphate ions, the formation of insoluble barium sulphate leads to the result that when this barium sulphate separates as a precipitate it entrains a major portion of the radium, since barium is the nearest chemical analogue of radium. This permits an important conclusion to be reached that on precipitation of barium compounds (in the form of barium sulphate) from underground waters, there takes place a depletion of the waters in radium; the radium is deposited upon the surface of water-conducting fissures (Tokarev, 1956).

Another common mechanism of radium immobilization is coprecipitation of radium 226 with calcium carbonate deposits as water bearing calcium bicarbonate evolves carbon dioxide to the atmosphere. (Miholic, 1958). Coprecipitation of radium with calcium carbonate is possible in all waters where conditions of calcium carbonate saturation are possible.
4. CONCLUSIONS

Various mechanisms that control radon and radium migration in soil are suggested. The following tentative conclusions can be drawn:

1. The three most significant meteorological factors that affect radon concentration and exhalation in the uppermost ground layers are rainfall, freezing and snow cover.

2. Radon concentration in water is mainly dependent on radium concentration in the sediments.

3. Surface run-off is a likely mechanism of dispersion of radon from a soil matrix. Underground percolation is likely to be so slow that decay intervenes.

4. Radium in waters is readily adsorbed by clayey and organic substances.

5. The transfer of radium from the solid matrix (soils and rock) into water is influenced by the chemical composition and acidity of the solution, amount of free oxygen and the temperature, which in general affects the solubility.

6. Precipitation is one of the processes which might remove radium from water. Upon precipitation of barium compounds (in the form of barium sulphate) from underground waters, there takes place a depletion of the of the waters in radium with the result that it is deposited upon the surface of water-conducting fissures.

7. Radium is also immobilized by coprecipitation with calcium carbonate in waters where conditions of calcium carbonate solution are available. Coprecipitation with ferric hydroxide is likely at interfaces between iron-bearing low-oxygen water or oxygen in rich water.
8. Surface run-off is a likely mechanism of dispersion of radium (via leaching) from a soil matrix.

5. **RECOMMENDATIONS**

1. The processes of adsorption and coprecipitation of radioactive elements in low concentrations in water that take place with active participation of hydroxides of iron and aluminum, silica gel, clayey, colloidal particles, peat and so on are not understood very well at all, and warrant further study.

2. No observations of dynamics of such processes in material conditions are known to have been carried out yet. Such observations are required.

3. No clear cut correlation has been obtained between the observed increases in radioactivity in the fine colloidal fractions of soils ( < 0.02 mm) and the content in them of organic substances. The effect of colloids and organic substances on radioactive elements should be investigated.

4. A systematic study of the hydrogeological and hydrochemical conditions in the area should be conducted to determine causes and probable pathways of observed anomalous radon concentrations. Without such understanding there can be no guarantee that soil removal would result in any more than a temporary reduction in radon and gamma levels, especially in the Pidgeon Hill area.

A report of field measurements of radon and radon daughters in buildings at Port Hope, together with a discussion of possible causes and
remedies, is available. (Environmental Radon and Radon Daughter Measurements at Port Hope, by C. R. Phillips and H. L. Pai).
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