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CHAPTER II.C
TRANSPORT OF RADIONUCLIDES THROUGH SOIL AND GROUND WATER

by

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CHAPTER II.C

TRANSPORT OF RADIONUCLIDES THROUGH SOIL AND IN GROUND WATER

J. Henry Horton, Jr.

I. INTRODUCTION

1. Scope and Aims of This Chapter

Through nuclear waste disposal or accidents, radionuclides come in contact with soil and groundwater. The fate of these radionuclides is of real concern to both the nuclear industry and the general public, and the degree of concern is increasing with the proliferation of nuclear power plants. Man is exposed to radiation as a result of movement (or transport) of the radionuclides into his environment. Water is the principal carrier that induces transport, but chemical characteristics of soil inhibit the transport. Therefore, the purpose of this chapter is to provide a clearer perspective of the impact of radionuclides in soil and groundwater, particularly for those not well-versed in soil science, hydrology, and geology.

Terminology of the scientific disciplines concerned with movement of radionuclides in soil and ground water is not always consistent. For the purpose of this chapter, unconsolidated materials shall be called soil, and consolidated materials shall be called rock. Water in soil above the water table is referred to as soil moisture; below the water table, as ground water.

Soil is a great reservoir from which chemical elements enter all other phases of the environment; therefore, transport of radionuclides through soil is important to many other scientific disciplines concerned with radionuclides in the environment. Plants absorb radionuclides from soil and thus enter into terrestrial ecological systems. Radionuclides in soil can be leached into ground water and eventually emerge into surface water and aquatic ecological systems. If radionuclides adhere to surface soil particles, they can become airborne. Thus, this chapter occasionally overlaps other chapters in this report which concern the environment.

Hundreds of reports have been published on the movement of radionuclides in soils and ground water, and on the basic physics and chemistry which control this movement. Many countries have contributed. Except for transuranic elements, which move so slowly that rates are difficult to measure but are so long-lived that even this movement may be of concern, fundamental knowledge appears adequate. At different locations, there is a great variation between soil and geological strata containing ground water.

Among scientists, disagreements that may exist are likely due to these differences in soil and geological strata rather than to differences of opinion. For the same reason, results obtained at one location can only be applied in a very general manner to any other location. Attempts to define safe or unsafe rates of movement with wide application are not possible. A safe rate is slow enough that radionuclides will not emerge in unacceptable concentrations into an ecological system. This safe rate will be different for each radionuclide and for each site. Therefore, this literature review has been aimed at the basic fundamentals of physics and chemistry which influence the retention or movement of radionuclides in soil and ground water, rather than at attempting to summarize observed rates of movement.

2. Soil and Weather Factors in the United States

Soil is the residue remaining from physical and chemical weathering of rock together with organic matter incorporated into the mineral residue as a result of growth and death of plants and animals in or on the soil.(5) The dominant factors controlling the intrinsic characteristics of soil are climate and parent rock.(45) The many combinations of these two factors have resulted in soils which change greatly from north-to-south and east-to-west within the United States.(68) Superimposed upon these factors are many local factors, such as topography, type of vegetation, cleavage due to subsidence, erosion, and deposition, so that large variations frequently occur within a few feet.

3. The Major Factors Determining Movement of Radionuclides in Soil

The major factors determining movement of radionuclides in soil are soil moisture flow and ion-exchange properties. Moisture flow provides the driving force for movement.(63) Soil retention of radionuclides, by ion-exchange(5,75) or conversion from very soluble forms to a less soluble form, is due to chemical and mineralogical reactions.(1,2,5,9,26,27,28,29,68) The magnitude of these forces is determined by precipitation-evaporation and the mineralogical and chemical characteristics of soil.

In the eastern half of the United States, rainfall decreases from south-to-north; in the western half, rainfall decreases from east-to-west except along the Pacific Ocean where it again becomes high and increases from south-to-north.(69) Annual rates range from a few inches in parts of the west to more than 128 inches in the Pacific Northwest. Throughout the United States, evaporation decreases from south-to-north. Annual rates from an open pan range from greater than 144 inches in the southwest to less than 32 inches in several regions near the Canadian border. In the eastern half of the United States, the precipitation rate is greater than the evaporation rate, therefore some rain flows through the soil to the ground water table. Where this occurs, radionuclides can be leached into ground water and eventually to flowing surface streams. In most of the western half of the United States, the evaporation rate exceeds the precipitation rate to such an extent that ground water is not recharged by local precipitation. Under these conditions, radionuclides in soil could not migrate through soil to ground water except where water is added to soil as with ground disposal of liquid radioactive waste. Under conditions where ground water recharge from local precipitation does not occur, there is always the possibility that precipitation might enter the soil, dissolve radionuclides, and deposit them on the soil surface as evaporation occurs. The radionuclides could then be moved by wind, surface runoff during heavy rains, or other forces.

The retention capacity of soils for radionuclides depends on size, mineralogical composition, and organic content of the soil particles.(5,58) Soils are composed of particles ranging in size from colloidal to sand. The smallest particles (<0.002 mm) are referred to as clay.(11) Silt ranges in size from 0.002 to 0.02 mm, and sand is >0.02 mm. These various size particles may be in a state of complete dispersion, but are usually aggregated.(7) Clays are the most active fraction of soil because their surface area per unit volume is large. Soils include minerals such as quartz, feldspars, micas, amphiboles, pyroxenes, olivine, carbonates, gypsum, and pyrite that are residues from the parent rock. Soils also include various clay minerals (kaolinite, illite, vermiculite, montmorillonite, and chlorite), iron oxides, and titanium oxides that are either residues from the parent rock or are formed during weathering of parent rock.(5) The cation exchange capacities of kaolinite and and related minerals range from 3 to 15 milliequivalents per 100 grams (meq/100 g). For montmorillonite and related minerals, the cation exchange capacities range from 80 to 150 meq/100 g. The cation exchange capacity of organic matter depends on its stage of decomposition, but can be as high as 400 meq/100 g. Thus, soils high in organic matter and/or montmorillonite have high cation exchange capacities. These characteristics exist in the western half of the country.(45) In the eastern half of the country, kaolinite is the predominant clay mineral, and organic content of soils decreases from north-to-south. Thus, the cation exchange capacities of soils in the southeast are relatively low.

Anion exchange does not occur in western soils, but has been observed in eastern soils.(13) The exact mechanism is not known,(58) and the capacity for anion exchange is small compared to the capacity for cation exchange.(13)

Several reactions occur in soils which convert certain radionuclides from soluble and readily ion exchangeable forms into relatively insoluble and not readily ion exchangeable forms. These reactions are commonly called "fixation" processes. Examples of these processes include precipitation of soluble phosphorous compounds by reaction with iron, aluminum, or calcium compounds;(5) precipitation of soluble strontium by reaction with calcium carbonates, calcium phosphates, or iron and aluminum hydroxides;(1,2,28,68) and entrapment of cesium within minerals such as illite.(9,26,27,29,44) The latter reaction is dependent upon the ionic radius of cesium, which is very similar to the ionic radius of potassium that has been removed from the original mineral (illite) during the weathering process. When radionuclides are accumulated in soil by ion exchange, they remain readily available to plants growing on the soil; but if removed from solution by one of the "fixation" processes, they are only slowly available to plants growing on the soil.(5,12,16)

II. TRANSPORT IN SOILS (ABOVE THE WATER TABLE)

1. Soil Moisture Flow

Mathematical equations for calculating water flow in unsaturated soil above the ground water table have received much attention for several decades.(41,60,66) This flow obeys the same general equation as water flow in the saturated zone below the water table. This equation is commonly referred to as Darcy's law and according to it:

$$Q = -KA\,(h_2 - h_1)/\,(Z_2 - Z_1) \qquad (1)$$

where:

Q = quantity of water flow per unit time

K = hydraulic conductivity

A = cross-sectional area through which flow occurs

h_1 and h_2 = hydrostatic heads at locations 1 and 2

Z_1 and Z_2 = distance from some reference point to locations 1 and 2.

Use of this equation to calculate ground water flow (saturated flow below the water table) is relatively simple. However, calculating flow in the unsaturated zone above the water table is mathematically very complicated because the value for K is dependent upon the water content of the soil and varies as flow occurs. Likewise, values for h_1 and h_2 (these pressures are less than atmospheric) are also dependent upon the water content of the soil and also vary as flow occurs. Yet through many years of effort, satisfactory means of calculating flow for many conditions have been developed.(41,66) Although these equations are useful in calculating flow, they do not describe some characteristics of soil moisture flow that are most useful in considering movement of radionuclides in soil, especially those moved only by infiltrating rain.

When water enters the soil surface, it is most likely to enter the largest pores and flow from these into smaller pores.(37) If the smaller pores contain water supported by capillary forces, this water is displaced downward thus forcing water just above the water table into the water table. Because of this displacement mechanism, the flow of soil moisture is very slow. In central South Carolina, soil typically retains about 20 volume percent water by capillary forces. Annual ground water recharge is equivalent to about 15 inches of rain. Soil moisture flows downward at a rate of about 7 feet per year.(34) The flow rate can be estimated for other areas if the rate of ground water recharge is known, because displacement of water held by capillary forces is almost complete. With these very slow flow rates, leaching of radionuclides in soils is slow and especially so if radionuclides are adsorbed by ion-exchange or retained by any one of the fixation methods.

The nature of soil moisture flow is of particular interest in leaching of buried solid waste. If a burial site is carefully selected to obtain uniform soil permeabilities, and if burial trenches are designed to prevent horizontal water flow into or within trenches, then waste should seldom if ever be in contact with water-saturated soil. Natural precipitation would be the only source of water. If buried waste were always in unsaturated soil, leaching of radionuclides from the waste would be eliminated or greatly reduced. This is particularly true when radionuclides are on the interior of pipes or vessels or any other container which forms a cavity in the soil; for in unsaturated soil, water will not flow into cavities in the soil.(15)

2. Ion-Exchange and other Mechanisims that Retard Movement of Radionuclides in Soil

Adsorption of radionuclides by soil due to ion-exchange obeys the same basic rules as adsorption of ions by ion-exchange resins.(36) When an ion in solution is adsorbed, it displaces an equivalent amount of another ion from the adsorbing surface. The reaction obeys the laws of mass action; at equilibrium, a distribution coefficient can be determined. For soils, it is commonly expressed as:(52)

$$K_d = (F_s/F_l) \cdot (V/M)$$

K_d = distribution coefficient

F_s = fraction of radionuclide adsorbed on soil

F_l = fraction of radionuclide in water

V = volume of water

M = mass of soil

If the K_d for a radionuclide in a particular soil and the rate of soil moisture flow are known, then the rate of movement of the radionuclide can be calculated by the following equation:(65)

$$\frac{V_a}{V_w} = \frac{1}{1 + (K_d)\cdot(R)} \qquad (2)$$

where:

V_a = average velocity of radionuclide

V_w = average velocity of water

K_d = distribution coefficient for adsorption of radionuclide by soil

R = ratio of weight of soil to volume of water per unit volume of soil.

If a uniform concentration of a radionuclide is continually added to a column of soil, as could occur with ground disposal of liquid waste, the effluent concentration curve will be the typical

S-shaped curve observed with ion exchange resins,(36) and the above equations can be used to calculate the volume of flow which will have occurred when the effluent concentration is 0.5 of the influent concentration. If a quantity of a radionuclide is added to soil and then leached (as would occur when precipitation is the only source of water added to soil), the effluent concentration curve will be the typical bell-shaped curve observed in ion-exchange chromatography. If dispersion coefficients have been determined, the shape of these two types of concentration curves can be calculated. By using these basic fundamentals and more complex mathematics, satisfactory methods have been developed for describing the movement of radionuclides in the ground.(4,21,32,33,43,63)

Ion-exchange adsorption selectivity of radionuclides by soils is based upon the same general principles as adsorption of stable isotopes by ion-exchange resins.(36) The greater the valence and the less the degree of hydration, the more readily the ion will be adsorbed. The selectivity sequence for some common divalent cations is Ba > Pb > Sr > Ca > Ni > Cd > Cu > Cs > Zn > Mg > UO_2; and for monovalent cations, the sequence is Tl > Ag > Cs > Rb > K > NH_4 > Na > Li. The relative adsorption of radionuclides from alkaline solutions by Hanford soils was determined to be Pu > rare earths > Sr > Cs > Ru.(10) Ruthenium may exist as a cation, an anion, or a neutral molecule. Except for cesium, the adsorption of radionuclides by Oak Ridge soils followed the lyotropic series; so the relative order of adsorption by a hydrogen-ion saturated soil was: Nb = Zr > Cs > Ru > Y > Sr.(67)

3. Soil Chemistry of Radionuclides

The chemistry of radionuclides in soils has been reviewed by Schulz.(64) Over 275 fission products have been identified. Most of these are of little concern in soil because of their relatively short half-lives or low abundance. Those fission products of interest include ^{90}Sr - ^{90}Y, ^{89}Sr, ^{137}Cs - ^{137}Ba, ^{106}Ru - ^{106}Rh, ^{103}Ru - ^{103}Rh, ^{140}Ba - ^{140}La, ^{144}Ce - ^{144}Pr, ^{141}Ce, ^{95}Zr - ^{95}Nb, ^{91}Y, ^{147}Pm, ^{143}Pr, and ^{151}Sm. Those created by neutron activation that are of interest in soil include ^{57,58,60}Co, ^{54}Mn, ^{51}Cr, ^{55,59}Fe, and ^{65}Zn. Some of these nuclides are very immobile. They include the rare earths (^{140}La, ^{144}Ce - ^{144}Pr, ^{141}Ce, ^{147}Pm, ^{143}Pr, and ^{151}Sm) and ^{90,91}Y, which form polyvalent ions in acid solution and precipitate as hydroxides or carbonates in neutral or alkaline media. In acid soils, polyvalent cations will be strongly bound to soil by ion-exchange or as precipitates. In addition to the rare earths, ^{95}Zr - ^{95}Nb ions are sorbed by soil in some undefined but very immobile form.

Radionuclides which have some degree of mobility include ^{54}Mn,^{57,58,60}Co, ^{65}Zn, ^{55,59}Fe, ^{51}Cr, ^{103,106}Ru, ^{89,90}Sr, ^{137,140}Ba, and ^{137}Cs. Manganese, cobalt, zinc, chromium, and ruthenium tend to react in soils in a similar manner. They are most soluble under acid conditions and precipitate as hydroxides as the pH increases. All except zinc have more than one valence state and become less mobile when oxidized to higher valences. Ruthenium can form complexes which are either uncharged or anionic, and these tend to be very mobile. Strontium is relatively mobile except when precipitated with calcium carbonates, calcium phosphates, or with iron and aluminum hydroxides.(1,2,28,68) Cesium is generally not very mobile because of its fixation by mica-like minerals.(9,26,27,29,44) Tritium is a special case; it will likely be present in soil as water and will move with no retention by soil except that due to capillary forces.(13,14)

Movement of transuranic radionuclides in soil needs much more research.(51) These nuclides are usually very insoluble in soil, and their mobility is so low that they have not received much attention until very recently. Because of the long half-life of some of them, especially ^{239}Pu (24,000 yrs), very small continual releases to soil could eventually lead to a high concentration at the point of release into soil. Movement into soils has been attributed to movement of particles and/or chelation of insoluble compounds by organic materials in soil. Colloidal plutonium with a valence of 4, e.g., $Pu(OH)_4$, is the form most likely to exist. These colloids are positively charged up to about pH 8 and negatively charged at higher pH's. This condition explains the extreme immobility which has been observed in soils between pH 2 and 8, because the positively charged colloidal plutonium is strongly adsorbed by the negatively charged soil. Some data on comparative uptake of transuranic elements by vegetation suggest that the relative mobility of these ions in soils is Cm > Am > Pu = Np; but other data suggest that the order is Np > Cm $\overline{>}$ Am > Pu.

III. CONDITIONS UNDER WHICH GROUND WATER EXISTS AND THE NATURE OF ITS FLOW

The same chemical and mineralogical reactions which retard movement of radionuclides in soil are applicable to movement of radionuclides in water-saturated earth materials below the water table. Each radioactive element moves at its own unique rate dependent upon its chemistry, the chemical characteristics of the ground water, and upon the geological formation containing the ground water. In contrast, flow of ground water and conditions under which it exists are considerably different from soil moisture. Therefore, emphasis will be placed upon the widely variable conditions under which ground water exists and the nature of its flow.

Ground water may be found in one continuous body or in several separate strata.(19,31,39,75) The thickness of the water-saturated zone varies from a few feet to many hundreds of feet. The upper surface of ground water (the water table) in general tends to follow the shape of the land surface and fluctuates according to the rate of infiltration of precipitation. In some cases, a local zone of saturation may exist at some level above the main water table due to a stratum of low permeability in the unsaturated zone. This condition is referred to as perched ground water. When ground water is present in a permeable stratum, it is called an aquifer. When an aquifer is between two impermeable strata, it is said to be confined and is not open to atmospheric pressure. Confined aquifers are called artesian aquifers; when a well is drilled into them, water rises in the well to some level above the top of the aquifer.

Flowing ground water eventually emerges into surface waters, such as springs, streams, lakes, and oceans.(39) Conditions that cause perched ground water merely delay flow of soil moisture to the water table, because perched ground water either flows slowly through the supporting strata or off the end of it. Water in unconfined aquifers generally emerges into the nearest surface stream and generally does not flow underground from one watershed to another, whereas water in confined (artesian) aquifers may flow long distances underground, passing beneath unconfined aquifers feeding surface streams. Under some conditions, ground water may remain in the ground for very short periods of time; whereas under other conditions, it may remain in the ground for hundreds or thousands of years. In very deep geological formations, ground water may remain for an entire era of geologic time.

Flow of ground water and the transport of radionuclides in it are controlled by the characteristics of the geological formation in which the water resides. As a general rule, ground water flow in more permeable strata will be more rapid, and ion-exchange and fixation of radionuclides will be low.(19,31,39) Ground water occurs in materials ranging from gravel, sand, and clay to rocks, such as granite, basalt, sandstone, and limestone. Clays have the highest retention capacities and lowest permeabilities. This is particularly true for montmorillonitic clays.(35,57) Some rocks would be virtually impermeable if they did not contain cracks or other openings.(19,31,39) Some openings may be large enough to permit turbulent flow, and this flow will be as rapid as in surface streams. Such large channels occur in limestone (due to dissolution) and in basalt and other volcanic outflows (due to escape of dissolved volcanic gases).

Where dissolution is possible, it may be increased by non-radioactive pollutants injected into ground water.(71) This injection may occur during or after radionuclides have been placed into the ground. Other activities of man, such as pumping or construction of dams or irrigation canals, can influence direction and rate of ground water flow. Thus ground water flow rates vary greatly. At the Savannah River Plant in South Carolina, measured flow rates in Coastal Plain sedimentary soils range from 0.006 ft per day near the ground water divide between surface streams to 1 ft per day near the surface streams.(24) At the National Reactor Testing Station in Idaho, the rates in basalt range from 5 to 25 ft per day.(57)

Flow of ground water obeys Darcy's law, which was previously defined in describing soil moisture flow.(19,31,39) Use of this equation is simpler with ground water than with soil moisture, because there is no change of the hydraulic conductivity due to changing water content. Movement of ground water occurs when there is a difference of hydrostatic head between two points. Flow is in the direction of the lower hydrostatic head.

To predict movement of radionuclides in ground water and to monitor this movement requires a thorough knowledge of the flow path or flow pattern of the water.(19) The more heterogeneous the strata, the more difficult it is to define the flow. Where cracks or channels occur, it is extremely difficult. In relatively homogeneous strata, the flow pattern is more easily defined; however, proper location of monitoring wells is complicated by flow along curved rather than straight lines.(19,24) Thus, any radionuclides moving with ground water will follow a curved path to depths below the water table and then curve upwards to enter surface waters. Consequently, monitoring wells must extend to the proper depth. Near a point source of radionuclides entering ground water, monitoring wells must be closely spaced as compared to locations more distant from the source, because radionuclides will be dispersed both laterally and vertically as they migrate from the source.(24,57)

Although movement of radionuclides in ground water is complex,(75) digital computer modeling techniques do provide a means of predicting movement with a reasonable accuracy provided characteristics of flow and retention properties of the strata have been adequately defined.(21,53,57,59,75) Because of their great variation, these properties must be defined for each individual site. Thus, the regulatory agencies responsible for site selection require each site to define the rate at which radionuclides are transported through soil and ground water.(74) These rates must then be considered in calculating radiation doses to people from normal and abnormal operations that could cause radionuclides to enter the ground.

IV. PROBLEMS WHICH CAN OCCUR FROM RADIONUCLIDES IN SOIL OR GROUND WATER

From the previous discussion of movement of radionuclides in soil, it is obvious that problems that can occur will depend upon many factors. If no problems are to exist, then the radionuclides must move through the soil so slowly that they will not enter useable ground water or into surface waters such as streams or lakes in "unacceptable" concentrations (as specified by federal or state regulatory agencies). Also, radionuclides must be at a depth below the roots of vegetation and burrowing animals and located where there will be no need to excavate for any useful purpose before the radionuclides decay to innocuous levels. The location must be well-marked to prevent unintentional intrusion.

Intentional placement of radionuclides into soil has included ground disposal of liquid waste into seepage basins, trenches, and cribs, plus burial of solid waste. (These practices and the consequences of them are described in Section IV-E, Waste Management or Disposal.) There are more possible short-term problems with disposal of liquid waste to the ground than with the burial of solid waste. Addition of waste water to soil increases the rate of soil moisture flow to rates greater than those due to precipitation alone; because migration of radionuclides is proportional to water flow, they will move faster.(65)

Nonradioactive chemicals in liquid waste can improve the retention of some radionuclides while causing others to be more mobile. For instance, strontium in highly alkaline waste is less mobile than in acid waste; while ruthenium is extremely mobile in alkaline waste, but relatively immobile in acid waste.(10,52,54) If soil readily retains a particular radionuclide, high concentrations may develop in the soil, even if concentrations in the original waste liquid are low. This phenomena has been observed with plutonium in liquid waste placed into cribs at Hanford, Washington.(73) Concentration of ^{137}Cs by streambed sediments that have the same retention characteristics as soil are well documented. Such concentration has been observed in the Cattaragus Creek in New York State (6,8) and in streams at the Savannah River Plant, Aiken, South Carolina.(46) Once a ground disposal site for liquid waste is placed out of operation and covered with clean soil, possible problems are very similar to possible problems with solid waste burial.

As previously discussed, leaching of radionuclides from buried solid waste should be very slow because soil moisture flow from precipitation alone is very slow. Observed migration rates at the various burial sites are discussed in Section IV-E, <u>Waste Management or Disposal</u>. Where migration beyond the burial site has occurred as at Oak Ridge, Tennessee,(20) Maxey Flats, Kentucky,(30) and West Valley, New York,(30) it can be attributed to poor site selection or improper layout of trenches. Even with these conditions, the quantities moving are small. Knowledge is presently available to improve site selection.(75)

Leakage from high-level waste tanks is also discussed in Section IV-E, <u>Waste Management or Disposal.</u> Although it is highly undesirable to have such waste free in the soil, no exposures to man from migration of radionuclides through soil have been reported. In the arid regions of the Western United States where local precipitation does not recharge the water table, and if leakage and soil drainage stop before waste reaches the water table, further movement is not likely due to the lack of a driving force.(22) In the humid eastern United States, the soils are naturally acid; therefore if the waste is alkaline, the clays in the soil can become peptized, causing the waste-containing soils to become quite impermeable and significantly reducing waste migration.(23,51) In humid regions, the rate of movement above the water table can be further reduced by the use of some impermeable material to prevent infiltration of precipitation.(35) If the site for waste tanks has been properly selected, the movement of high-level waste through soil should be sufficiently slow to permit recovery by excavation, if necessary. If radionuclides in soil are in the root zone of vegetation growing on the soil, the ions will be absorbed by the vegetation and transported above the ground surface within the vegetation.(3,25,47,55,62) The radionuclides can then enter the food chain of animals, including humans, and will be subjected to biological recycling.(3,18,42,61) Vegetation not consumed will eventually die. Some dead parts of plants will be dispersed by wind or surface runoff of precipitation. As dead parts of plants decay, the radionuclides will be released to the surface where they can be transported by wind(18,70) or water.(17,18,48) Erosion by either wind or water can also transport radionuclides bound to soil particles. Soil covering radionuclides can be removed by erosion so that the exposed radionuclides are subject to dispersion by wind and water. Thus radionuclides in vegetation or exposed surface soil will begin moving beyond the area in which they were originally placed.

Concern has been expressed that changes in environmental conditions could increase movement of radionuclides in soil.(56) Such changes could result from man's activities including construction of dams, increased irrigation, or higher withdrawal of ground water by pumping. Climatic changes in future centuries could also alter the hydrology of a region. Changes from aerobic to anaerobic conditions could change the soil chemistry in a manner similar to that which occurs annually in stratified lakes. These changes are known to influence adsorption and release by bottom sediments.(49)

The most important problem concerning radionuclides in soil is likely to be cost of surveillance and management of the area to ensure that the radionuclides do not migrate to the soil surface or to flowing surface water(70) because their transport would then become far more rapid. Management of burial sites will require constant vigilance to prevent growth of deep-rooted vegetation, while at the same time maintaining a soil cover of shallow-rooted vegetation or non-vegetated soil covers to prevent erosion.(38) The area must be monitored to determine whether migration of the radionuclides is within acceptable limits, and if not, protective action must be taken.(70) Protective action may require excavation. If this is deemed necessary, the slow movement and dispersion in soil provide time for the necessary corrective action to be taken; although in some cases, the cost may be tremendous.

Once radionuclides move from unsaturated soil into ground water, considerable control of them has been lost. Ground water flow is apt to be more rapid than soil moisture flow. If the flow is through solution cavities or fractures in rock, retention by ion exchange and fixation may be negligible. Thus, the possibility of recovery is greatly diminished if not impossible. Also, prevention of ground water flow through strata containing radionuclides would be much more difficult than stopping soil moisture flow by interrupting infiltration of precipitation.

As the technology for volume reduction and containment has developed and the desire for reducing damages of any kind to the environment has grown, the intentional release of radioactivity to soil has decreased. Disposal of liquid waste to the ground was once a common practice, but is now restricted to amounts that are biologically acceptable for release to flowing surface streams in uncontrolled areas.(72) Burial of low-level solid waste contaminated with radionuclides continues, but is more carefully controlled than in the past.(70) Solid waste that contains greater than 10 nCi of transuranic elements per gram is now placed into retrievable storage.(70) Accidental spills or leaks may never be completely eliminated, but recovery of radionuclides from soil is more readily accomplished than recovery from spills into flowing surface water.

V. SUMMARY

The material in this chapter differs from that in all other chapters in that there are no major controversies, for the basic fundamentals are widely accepted. However, the inhomogeneities that exist in soil and other geological formations have at times led to a divergent opinion in the scientific community on the reliability of predicting movement of radionuclides in soil and ground water.

Soils are formed by physical and chemical weathering of rock plus incorporation of organic matter. The dominant factors determining the characteristics of soils are climate and parent rock. Thus, soils change greatly from north-to-south and from east-to-west within the contiguous United States. Superimposed upon this are many local factors influencing the nature of soils; large variations frequently occur within a few feet.

Soil moisture flow provides the main driving force for movement of radionuclides in soil, while ion-exchange and various chemical and mineralogical reactions which convert radionuclides from very soluble to less soluble forms retard movement. The end result of these forces is determined by the precipitation and evaporation of water and by the mineralogical and chemical characteristics of soil. As with the characteristics of soil, rates of precipitation and evaporation change greatly from north-to-south and from east-to-west. In the eastern half of the country, precipitation exceeds evaporation, so that ground water is recharged by local precipitation, and radionuclides can potentially be leached through soil to ground water. In most of the western half of the country, evaporation exceeds precipitation, so that no local precipitation flows to ground water. Under the latter conditions, it is always possible that infiltrating precipitation could dissolve radionuclides in soil and deposit them on the soil surface as evaporation occurs. Retention of radionuclides by soil generally increases from south-to-north due to an increased organic matter content, and increases from east-to-west due to mineralogical differences.

Adsorption of radionuclides by soil by ion-exchange obeys the same basic rules as adsorption of ions by ion-exchange resins. Also, several reactions occur in soils that can convert certain radionuclides from soluble and readily ion-exchangeable forms to relatively insoluble and not readily ion-exchangeable forms. Examples include the precipitation of strontium as phosphates, carbonates, or hydroxides, and the entrapment of cesium by mica-like minerals. These reactions are commonly called "fixation" processes and greatly reduce the movement of radionuclides and their uptake by vegetation.

Movement of most radionuclides in soil is of little or no concern because of their short half-lives. Many others are very immobile. Those relatively long-lived nuclides with some degree of mobility include ^{3}H, ^{54}Mn, $^{57,58,60}Co$, ^{65}Zn, $^{55,59}Fe$, ^{51}Cr, $^{103,106}Ru$, $^{89,90}Sr$, $^{137,140}Ba$, and ^{137}Cs. Movement of transuranic radionuclides in soil needs much more research because some of them have extremely long half-lives. They are very insoluble in soil, and their mobility is so low that some of these radionuclides have not received much attention until recently.

The same chemical factors which retard movement of radionuclides in soil are applicable to movement of radionuclides in ground water. However, flow of ground water and conditions under which it exists are quite different from those of soil moisture.

Whereas soil moisture is the water present in the unsaturated zone above the water table, ground water is the water present in the saturated zone below the water table. Soil moisture flow occurs when water infiltrates the soil surface and displaces downward the water held in soil by capillary forces. When infiltration is due only to precipitation, the flow is very slow and seldom exceeds a few feet per year. Ground water flow may also be extremely slow; but under some conditions, flow may be as rapid as that in surface streams.

The characteristics of earth materials that contain ground water are most important in determining movement of radionuclides. As a general rule, flow in more permeable strata will be rapid, and ion-exchange and fixation of radionuclides will be low. Clays have the highest retention capacities for radionuclides and the lowest permeabilities of common soil types. Fractured rocks have low retention capacities and frequently very high flow rates, especially if the channels in the rock are large.

Flow of both soil moisture and ground water obey Darcy's law. However, applying this law to calculate soil moisture flow is more complicated because the hydraulic conductivity varies with water content and changes as flow occurs. Transport and dispersion equations have been developed and can be used in conjunction with Darcy's law to describe the movement of radionuclides in both soils and ground water. The accuracy of these calculations is relatively good provided the characteristics of flow and retention properties are defined accurately for the environment of concern. These characteristics must be determined for each site. Where soil and earth materials containing ground water are extremely heterogeneous, the characteristics may be extremely difficult to define accurately.

If no problems are to exist with radionuclides in soil or ground water, the radionuclides must move so slowly that they will not move into useable ground water or surface waters in concentrations prohibited by federal or state regulatory agencies. Also, the radionuclides must be below the depth of plant roots and burrowing animals and must be located where excavations for useful purposes will not be required until after the radionuclides decay to innocuous levels.

There are more short-term problems with disposal of liquid waste to the ground than with burial of solid waste. The addition of waste water increases both soil moisture and ground water flow. The addition of nonradioactive chemicals in liquid waste can improve retention of some radionuclides while causing others to be more mobile. If a radionuclide is readily retained by soil, highly localized concentrations may develop in soil although the original concentrations in the liquid are low. The leaching of radionuclides from buried solids should be very slow.

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