

58209  
MDA B

# Radioecology: Nuclear Energy and the Environment

Volume I

Authors

F. Ward Whicker, Ph.D.

Professor

Department of Radiology and  
Radiation Biology  
Colorado State University  
Fort Collins, Colorado

Vincent Schultz, Ph.D.

Professor

Department of Zoology  
Washington State University  
Pullman, Washington



CRC Press, Inc.  
Boca Raton, Florida



10462

Library of Congress Cataloging in Publication Data

Whicker, F. Ward.

Radioecology: nuclear energy and the environment.

Includes bibliographies and index.

1. Radioecology. I. Schultz, Vincent, joint author. II. Title.

QH543.5.W48 574.5'222 80-26184

ISBN 0-8493-5353-X (v.1)

ISBN 0-8493-5354-8 (v.2)

This book represents information obtained from authentic and highly regarded sources. Reprinted material is quoted with permission, and sources are indicated. A wide variety of references are listed. Every reasonable effort has been made to give reliable data and information, but the author and the publisher cannot assume responsibility for the validity of all materials or for the consequences of their use.

All rights reserved. This book, or any parts thereof, may not be reproduced in any form without written consent from the publisher.

Direct all inquiries to CRC Press, Inc., 2000 Corporate Blvd., N.W., Boca Raton, Florida, 33431.

© 1982 by CRC Press, Inc.

International Standard Book Number 0-8493-5353-X (Volume I)  
International Standard Book Number 0-8493-5354-8 (Volume II)

Library of Congress Card Number 80-26184  
Printed in the United States

### III. BEHAVIOR OF SPECIFIC GROUPS OF RADIONUCLIDES

Individual radionuclides exhibit unique sets of characteristics, and therefore, somewhat unique behavior. However, certain groups of radionuclides, owing to their chemical properties, behave similarly in the environment. In this section, the authors discuss the ecological behavior of some of the more important radionuclides, and at the same time, point out some commonalities and differences in behavior that result from chemical attributes. They have chosen to treat this topic by element groups, including nonmetals, light metals, noble gases, heavy metals, rare earths, and the actinides. Example radionuclides that fall within each element group are discussed, with emphasis upon those which are particularly abundant in the environment and which have received substantial study. In Volume II, Appendix B lists several major reviews on the behavior of specific radionuclides in the environment.

#### A. Nonmetals (H, C, P, I)

Nonmetals are chemically distinguished from metals on the basis that nonmetals tend to gain valence electrons when they chemically combine with other elements, while metals tend to lose electrons. The nonmetals include some, but not all of the elements within groups IIIA through VIIA of the periodic table. Hydrogen can behave either as a metal or nonmetal, and is included in this group largely as a matter of convenience. Within the nonmetal group considerable diversity in chemical and environmental behavior is evident. Of the 15 or so elements which might be placed in this group, the authors shall consider four, each of which is biologically essential and has one or more significant radioisotopes.

The lightest and one of the most ubiquitously distributed radionuclides is tritium ( $^3\text{H}$ ). Tritium is produced continually in the atmosphere by cosmic rays and also by nuclear reactors and nuclear detonations.<sup>16</sup> Tritium decays with a half-life of about 12 years to  $^3\text{He}$  by the emission of beta particles of 18 keV or less. Tritium closely follows the reactions of ordinary hydrogen, but because of the large relative mass discrepancy between  $^3\text{H}$  and  $^1\text{H}$ , chemical reaction and diffusion rates show measurable differences. The vast majority of environmental tritium is in the form of tritiated water (HTO), but small amounts in the form of HT or tritiated carbon compounds also exist. Upon release to the environment, tritium generally follows the hydrologic cycle, but its initial behavior is determined by the source term. If released as a gas or vapor to the atmosphere, substantial dispersion can be expected and the rapidity of deposition is largely dependent upon climatic factors. Precipitation is the principal mechanism of deposition.<sup>17</sup> If released in liquid form, the HTO is diluted in surface waters and it is subject to physical dispersion, percolation, and evaporation.

Tritium exhibits ecological behavior unlike most other radionuclides. In aqueous bodies, it does not have the tendency to adsorb to sediments or biotic surfaces, which is so common for most other radionuclides. For this reason, HTO makes an excellent tracer for  $\text{H}_2\text{O}$ . Tritium can undergo exchange reactions with surfaces, but at normal environmental concentrations there are roughly  $10^{18}$  atoms of  $^1\text{H}$  for every few atoms of  $^3\text{H}$ .<sup>18</sup> Therefore, on hydrogen-saturated surfaces, there is little probability for exchange with  $^3\text{H}$  unless its specific activity is very high. Tritium enters plants through the roots, leaves, and stems, with high efficiency. It enters animals through ingestion, inhalation, and direct absorption through the skin. Unlike many radionuclides, however, tritium is seldom if ever concentrated in biological tissues to levels greater than those in the ambient air or water. There is, in fact, evidence that the steady-state concentration of tritium in biological tissues approaches but does not exceed the concentrations in ambient water or water vapor.<sup>19,20</sup> However, there is also some evidence that organically bound tritium may account for cases in which the T/H ratio in biota

can slightly exceed the ratio in ambient water.<sup>21</sup> Tritium is lost from land plants through transpiration. It is lost from animals through exhalation, secretions, excretions, and surface evaporation.

Carbon-14, like tritium, is produced by cosmic rays, reactors, and nuclear explosions and is ubiquitously distributed in the biosphere. Carbon-14 decays with a half-life of about 5600 years through emission of 158 keV (maximum) beta particles. Natural formation and anthropogenic releases of <sup>14</sup>C are mainly atmospheric and the predominant chemical form in the atmosphere is <sup>14</sup>CO<sub>2</sub>. Dilution of atmospheric <sup>14</sup>CO<sub>2</sub> with <sup>12</sup>CO<sub>2</sub> occurs rapidly, and, under natural, steady-state conditions, there are about 15 dpm/g carbon. This represents about one atom of <sup>14</sup>C per 10<sup>12</sup> atoms of <sup>12</sup>C. Carbon-14 enters food chains primarily through photosynthetic incorporation of CO<sub>2</sub>. Complex transformations convert <sup>14</sup>C to many other chemical forms in plants and animals. When oxidized, compounds release <sup>14</sup>C back to the atmosphere as carbon dioxide. Carbon dioxide is also converted to carbonate and bicarbonate ions which may exist as inorganic carbon deposits.

In terms of bioaccumulation, <sup>14</sup>C is found in biota in roughly the same ratio to <sup>12</sup>C as it occurs in air or water. Since the total amount of carbon in organisms is roughly constant, except for growth phases, the amount of environmental <sup>14</sup>C that can accumulate is somewhat fixed. However, if the global ratio of <sup>14</sup>C/<sup>12</sup>C were to increase through man's nuclear activities, the biological incorporation of <sup>14</sup>C would increase proportionately. On the other hand, burning of large quantities of fossil fuels, which are depleted of <sup>14</sup>C because of radioactive decay, tends to lower the <sup>14</sup>C/<sup>12</sup>C ratio in the biosphere, thus reducing the incorporation of <sup>14</sup>C by plants and animals. There could be a slight fractionation between <sup>14</sup>C and <sup>12</sup>C as these nuclides pass through the chemical transformations in food chains because of the small isotopic mass difference. It is well known that <sup>14</sup>C forms a slightly stronger chemical bonds than <sup>12</sup>C and <sup>12</sup>CO<sub>2</sub> diffuses slightly more rapidly than <sup>14</sup>CO<sub>2</sub>.<sup>22</sup> However, such isotopic effects do not appear sufficient to alter significantly the <sup>14</sup>C/<sup>12</sup>C ratio through food chains.

Phosphorus is an essential component of biological systems, in that it is a building block of various kinds of tissues and a key element in many biochemical transformations, and in particular, those involving energy transduction. Phosphorus-32, one of six radioisotopes of phosphorus, is of considerable radioecological interest because it is frequently introduced into the environment, either as an unwanted contaminant or as a tracer used to study behavior of the element.<sup>7</sup> Produced mainly by neutron activation of stable phosphorus, <sup>32</sup>P has a half-life of about 14 days and a relatively energetic beta emission (1.71 MeV, maximum). Because of its short half-life and energetic beta particle, <sup>32</sup>P is attractive as a tracer as it is easy to measure and does not persist very long in the environment.

Phosphorus is a comparatively scarce element in the biosphere,<sup>23</sup> yet because of its chemistry, it is usually quite mobile in ecosystems and is readily assimilated by plants and animals in its more soluble forms, such as phosphates. Because of its vital role in biological tissues and its comparative scarcity in the environment, organisms can concentrate phosphorus to levels that greatly exceed the concentrations in ambient media. For instance, Polikarpov<sup>13</sup> gives a mean concentration factor of 24,000 for <sup>32</sup>P accumulation in freshwater plants and a mean concentration factor of 8000 for freshwater animals. In marine organisms, concentration factors for <sup>32</sup>P range from 10 to 1000.<sup>13</sup> Mammals assimilate around 75% of ingested <sup>32</sup>P and some 90% of the total body burden locates in bone.<sup>6</sup> Phosphorus in the body is excreted slowly, particularly that which is deposited in bone.

Iodine is of great interest to radioecologists because it too is an essential nutrient element and several of its radioisotopes are produced in copious quantities by nuclear fission. Environmental releases have enabled ready detection of <sup>131</sup>I in air, plants, and

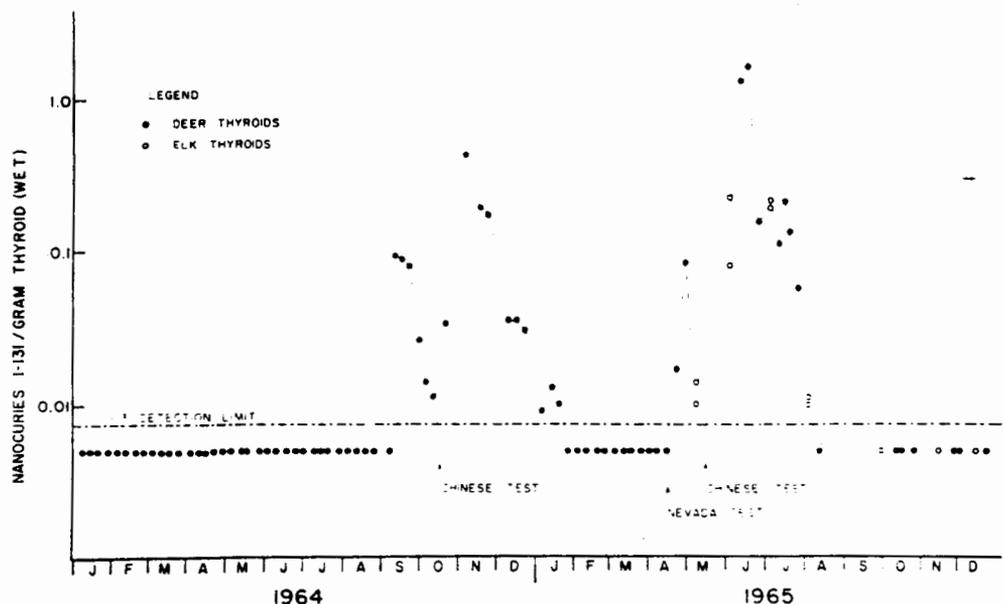


FIGURE 8. Iodine-131 concentrations of Colorado mule deer and elk thyroids in response to nuclear testing activities in 1964 and 1965. (From Whicker, F. W., Farris, G. C., and Dahl, A. H. *J. Wildl. Manage.*, 30(4), 781, 1966. With permission.)

animals on a global basis. Some 20 radioisotopes of iodine exist, but  $^{131}\text{I}$  has received the greatest attention. Iodine-131 has a half-life of 8 days and has beta as well as gamma emissions. Iodine-129 has received attention in recent years because it is also produced by fission and has a very long half-life ( $1.7 \times 10^7$  years) which enables it to accumulate in the environment over long periods of time. The biological incorporation of radioisotopes of iodine is largely governed by the abundance and physiology of stable iodine.

Iodine readily enters biological systems. When assimilated by higher animals, it selectively concentrates in the thyroid gland. This gland has such a remarkable ability to concentrate iodine that it is an excellent bioindicator of  $^{131}\text{I}$  fluctuations in the environment (Figure 8). Based upon  $^{131}\text{I}$  uptake and retention data from mule deer, the thyroid  $^{131}\text{I}$  concentration may exceed the forage  $^{131}\text{I}$  concentration by three or four orders of magnitude under conditions of chronic intake.<sup>25</sup> Because of the short half-life of  $^{131}\text{I}$ , the fact that it localizes in a small quantity of biological tissue, and that ingestion is usually a more important route of intake than inhalation, carnivore thyroids generally concentrate much less of the nuclide than herbivore thyroids. Iodine-131 exhibits concentration factors ranging from 10 to 1300 in freshwater organisms.<sup>13</sup> In marine organisms, iodine concentration factors range from 10 in fish tissues to over 1000 in sessile algae.<sup>26</sup> An excellent review of iodine in the environment was prepared by Hanson.<sup>27</sup>

The major commonality of the four radionuclides discussed under the nonmetal group is that they are all isotopes of essential biological nutrients. As such, their uptake and retention characteristics are largely controlled by the flux of these essential nutrients through biological processes. Radionuclides such as  $^{40}\text{K}$ ,  $^{45}\text{Ca}$ ,  $^{54}\text{Mn}$ ,  $^{59}\text{Fe}$ ,  $^{60}\text{Co}$ , and  $^{65}\text{Zn}$ , to be discussed later, are also isotopes of essential nutrients.

## B. Light Metals

### 1. Group IA (K, Rb, Cs)

This group includes elements known as the "alkali metals". The ecological and

physiological behavior of K, Rb, and Cs is remarkably similar. In general, the physiological behavior of Rb and Cs can be inferred reasonably well from that of K, an essential nutrient. The potassium radioisotope of greatest general interest is  $^{40}\text{K}$ . This primordial nuclide occurs naturally and has a half-life of  $1.3 \times 10^9$  years. It is homogeneously mixed with stable potassium and its isotopic abundance is about 0.0119% by mass. Owing to its ubiquitous distribution, abundance, and penetrating gamma rays,  $^{40}\text{K}$  accounts for a significant fraction of the natural background radiation exposure. The isotopic difference between  $^{40}\text{K}$  and stable K is so slight that behavior is essentially identical and the isotopic abundance is preserved in biotic and abiotic materials.

Rubidium has some 16 possible radioisotopes, most of which are short-lived and not particularly important. Rubidium-87, however, is a primordial, naturally occurring radionuclide with a half-life of about  $4.8 \times 10^{10}$  years. Also of some interest is 18.6 day  $^{86}\text{Rb}$ , which is a potential fission and neutron activation product. This nuclide exhibits concentration factors averaging 1500 in freshwater plants, 250 in freshwater animals, and 15 in marine animals.<sup>13</sup> These values are quite similar to comparative data for  $^{137}\text{Cs}$  as well as stable Cs and K.

Cesium-137 is one of the best-studied of all the radionuclides. It is a very abundant fission product, with a half-life of about 30 years. Its production in nuclear weapons detonations has resulted in wide distribution throughout the biosphere and its mobility and physiological properties have led to detectable concentrations in essentially all organisms. The fact that  $^{137}\text{Cs}$  emits energetic photons makes it simple to measure and, also, a potentially significant contributor to genetic dose in humans and other species. Because of its abundance, half-life and biological mobility,  $^{137}\text{Cs}$  is a critical component of nuclear fission wastes.

As with most other radionuclides, cesium can enter plants by aerial deposition or surface adsorption and root uptake, and animals by inhalation, ingestion, and surface absorption or adsorption. As already stated, Cs is a chemical analog of K and behaves similarly. However, it does not behave identically. In fact, in specific food chains, Cs concentrations tend to increase with trophic level and its ratio to K can increase likewise.<sup>14,28</sup> In numerous studies the observed ratio  $(\text{Cs}/\text{K})_{\text{consumer}} \div (\text{Cs}/\text{K})_{\text{food}}$  varies from one to four and averages about three. The apparent explanation for this phenomenon is that while Cs and K are both assimilated to approximately the same extent, Cs is retained in the body longer. In fact, the biological half-time of Cs exceeds that of K by a factor of two to five in humans,<sup>28</sup> and this trend probably also holds for most other animals as well. We are not certain to what extent the trophic level effect is unique to Cs. This phenomenon has not been generally observed with other radionuclides. On the other hand, the trophic level effect has been observed for the pesticide DDT<sup>29</sup> and continuing research may well uncover other substances which concentrate with trophic level.

An important concept with regard to radionuclides such as  $^{137}\text{Cs}$ , which are chemically analogous to major nutrient elements, is that the tendency for the radionuclide to accumulate in the biota is reduced if there is an abundance of the analogous element in the environment. Conversely, a scarcity of the analogous element usually leads to increased accumulation of the radionuclide in the biota. This pattern was illustrated for Sr and Ca (Figure 7) and a similar effect has also been shown for  $^{137}\text{Cs}$  concentrations in fish from lakes in Finland which have a range of K concentrations in water.<sup>15</sup> This concept also applies to terrestrial ecosystems. For instance, increases in K intake in animals has been shown to reduce retention of Cs, which could effectively reduce the Cs body burden.<sup>30,31</sup> A comprehensive review of cesium and its relationships to potassium in ecosystems was prepared by Davis.<sup>32</sup>

As pointed out earlier in this chapter, the long-term availability of cesium and many other radionuclides depends heavily upon ecosystem characteristics, and in particular,

soil properties. Soils and sediments of high clay content can effectively immobilize Cs by chemical binding. In such systems, the soil acts like a sink for Cs and in time very little of the nuclide is available for biological incorporation. Other systems have sandy soils with a low cation exchange capacity and larger quantities of Cs can be recycled through the biota of such systems for long periods of time.

## 2. Group IIA (Ca, Sr, Ba, Ra)

This group, termed the "alkaline earth metals", contains elements which have two valence electrons and an oxidation state of plus 2. These elements are chemically reactive and seldom occur in the free state. They commonly form salts such as carbonates, sulfates, and chlorides. Calcium is by far the most abundant of the alkaline earths in the biosphere and is an essential biological nutrient. The ecological and physiological behavior of Sr, Ba, and Ra can be inferred approximately from the behavior of calcium. Our principal interest in calcium, from the standpoint of radioecology, is the fact that its flow through ecological systems markedly affects important radionuclides such as  $^{90}\text{Sr}$ ,  $^{140}\text{Ba}$ , and  $^{226}\text{Ra}$ . The most important radioisotope of calcium,  $^{45}\text{Ca}$ , is seldom produced by man or nature in sufficient quantities to be considered a biological hazard; rather, it is of greater interest as a tracer to study the behavior of Ca. A specific review of some of the radioecological aspects of calcium is available.<sup>33</sup>

The strontium radioisotopes  $^{89}\text{Sr}$  and  $^{90}\text{Sr}$  received great notoriety and study during the era of atmospheric nuclear testing in the 1950s and early 1960s. Strontium-90, like  $^{137}\text{Cs}$ , is produced in high yields from nuclear fission and is persistent in the biosphere because of its 28-year half-life. Strontium has a tendency to form compounds that are comparatively soluble. Because of this and its chemical similarity to calcium, Sr isotopes are comparatively mobile in ecosystems and deposit in calcium-bearing tissues such as bone and shells, where they can remain for years. The beta particles of  $^{90}\text{Sr}$ , and its radioactive daughter  $^{90}\text{Y}$ , irradiate portions of the bone marrow, the site of blood cell formation, and thus are considered potential internal hazards.

Owing to its metabolic control by calcium, Sr uptake by organisms has often been expressed as an "observed ratio",<sup>34</sup> which is defined as the Sr/Ca ratio in an organism divided by the Sr/Ca ratio in the diet (or soil in the case of plant uptake). Observed ratios for soil to plants are usually close to unity, suggesting little discrimination between Sr and Ca by plants.<sup>35</sup> On the other hand, observed ratios describing food to animal tissue transfers are usually less than unity, indicating discrimination against Sr in favor of Ca.<sup>35</sup> Many studies have demonstrated that factors such as soil composition, diet, and age can substantially modify observed ratios. Therefore, the observed ratio is not a predictive panacea. Observed ratios describing Sr/Ca transport from water to fish muscle and bone also suggest discrimination against Sr.<sup>36</sup> Nevertheless, concentration factors for Sr in aquatic organisms can be very high.<sup>13</sup> In summary, strontium isotopes in the environment readily enter food chains, but tissue concentrations do not appear to increase with trophic level. High available calcium budgets in ecosystems tend to reduce the bioaccumulation of strontium.

The general behavior of isotopes of barium is not unlike that of strontium. Of several barium radioisotopes,  $^{140}\text{Ba}$  is the most important.<sup>37</sup> A high-yield fission product,  $^{140}\text{Ba}$  has a half-life of 12.8 days and emits both beta and gamma radiation. Because of the comparatively short life of  $^{140}\text{Ba}$ , it will enter food chains primarily by the more direct pathways, such as foliar deposition and inhalation. Also because of its short half-life,  $^{140}\text{Ba}$  generally does not build up in the calcium-bearing tissues to the same extent as, say,  $^{90}\text{Sr}$ . If deposited in bone,  $^{140}\text{Ba}$  will likely be removed mainly by radioactive decay rather than by excretion. In summary,  $^{140}\text{Ba}$  is biologically mobile, but important for only a limited time following its entry into the environment.

Isotopes of radium constitute the most important naturally occurring radionuclides

of the elements in Group IIA. Two are of overwhelming significance, namely,  $^{226}\text{Ra}$  and  $^{228}\text{Ra}$ . These radionuclides are generated in the decay chains of  $^{238}\text{U}$  and  $^{232}\text{Th}$ , respectively. Radium-226 is an alpha, gamma emitter with a 1620-year half-life while  $^{228}\text{Ra}$  is a beta, gamma emitter with a 6.7 year half-life. Both are reasonably mobile in the environment, and, due to their chemical similarity to calcium, are considered bone seekers. The release of radium from bone is very slow, thus chronic intake can lead to comparatively high concentrations. Short-lived radioactive progeny of  $^{226}\text{Ra}$  and  $^{228}\text{Ra}$  can grow toward secular equilibrium with these nuclides during their residence in bony tissues, contributing substantially to radiation dose.<sup>6</sup>

Geological deposits serve as the major reservoirs of radium, but measurable quantities occur in soil, ground water, plants, and animals. The geographic distribution of Ra, like its primordial precursors U and Th, is ubiquitous yet heterogenous, with well-known "hot-spots" in many specific areas of the earth's surface. Although radium is obtained from soil by plants through root uptake, the degree of uptake appears somewhat less than for Sr or Ca.<sup>38,39</sup> The concentration of Ra in most plants is normally one or two orders of magnitude less than the soil upon which they grow,<sup>39</sup> but soil characteristics and species can modify uptake dramatically. An interesting example is the Brazil nut, which concentrates Ra to levels comparable to those found in the soil.<sup>40</sup> Ingested Ra is absorbed from the gastrointestinal tract to varying degrees; however, the International Commission on Radiological Protection (ICRP)<sup>6</sup> cites an assimilation fraction of 0.3. Polikarpov<sup>13</sup> indicates a general lack of information on the accumulation of Ra in aquatic organisms, but cites limited data which indicate concentration factors of 15 to 650 for marine animals and values of 100 to 2750 for plankton. A good review of the concentrations of  $^{226}\text{Ra}$  in natural waters and foods is provided by Eisenbud.<sup>41</sup>

### C. Noble Gases (A, Kr, Xe, Rn)

Elements of group VIIIA are called the "noble" or "inert gases". Under natural conditions, these elements exist in the gaseous state and because of their electronic configurations, do not enter into chemical combination with other elements. The inert gases therefore exist as monoatomic molecules. Each of the noble gases occurs naturally in the earth's atmosphere, with argon being the most abundant. Stable isotopes of He, Ne, A, Kr, and Xe occur in nature, whereas all isotopes of Rn are unstable and radioactive. Even though the longest-lived radon isotope, 3.8-day  $^{222}\text{Rn}$ , is quite unstable, it is readily measurable in the atmosphere because it is continuously generated by the decay of  $^{226}\text{Ra}$  in rock and soil.

From the standpoint of radioecological interest, radioisotopes of A, Kr, Xe, and Rn have dominated our attention. The argon radioisotopes of principal interest are  $^{37}\text{A}$  and  $^{41}\text{A}$ , both of which are produced by neutron activation of stable argon. Krypton-85 is the most important radioisotope of Kr because of its comparatively high fission yield and 10-year half-life. Xenon-133 has received some attention because it is also a common fission product and its 5.3-day half-life is sufficient for the nuclide to travel considerable distances in the atmosphere. As mentioned earlier in this volume,  $^{222}\text{Rn}$  and  $^{220}\text{Rn}$  are the most significant radon isotopes because they are produced in the decay chains of primordial  $^{238}\text{U}$  and  $^{232}\text{Th}$ , respectively.

In terms of biospheric transport, the noble gases exhibit similar behavior. They all behave as true gases and disperse through the atmosphere according to the laws of molecular and turbulent diffusion. These gases, unlike some of the diatomic or triatomic gases, are chemically inert and transport, therefore, is not altered by chemical transformations. The inert gases are not transported through food chains and they do not generally concentrate in biological tissues. An exception to the latter statement is

$^{222}\text{Rn}$ , which can reside in fatty tissues if generated within an organism by the decay of  $^{226}\text{Ra}$ . Also, atmospheric noble gases can permeate nonpolar substances, such as fatty compounds, but only to a limited extent. In almost all circumstances, the principal radiation exposure mode to plants and animals from the noble gases is submersion, or external irradiation from atoms decaying in the immediate atmosphere surrounding the organism. The radiation exposure from the noble gases is, therefore, controlled by atmospheric concentration, the types and energies of the radiations emitted, and the physical half-life of the radionuclide.

Radon-220 and 222 present a more complex radiation exposure problem than the A, Kr, and Xe isotopes discussed, in that  $^{220}\text{Rn}$ ,  $^{222}\text{Rn}$  generate a series of radioactive decay products. These decay products are not gaseous and tend to attach to small dust particles which can settle on or attach to biological surfaces. Of particular concern is the inhalation of radon progeny. Residence of these progeny in the lung leads to irradiation of lung tissues, which at sufficient levels has induced cancer in uranium miners.

#### D. Heavy Metals (Cr, Mn, Fe, Co, Zn, Zr, Tc, Ru, Pb, Po)

The heavy metals as a group exhibit complex and varied chemistry, therefore, few broad statements can be made for this group as a whole. Some of these elements play essential roles in biological processes, while others have no known biological function. Some of the important radioisotopes of these elements are naturally occurring, while others are strictly activation or fission products which result primarily from nuclear activities. In the following paragraphs, some of the more important radionuclides of the heavy metals are briefly discussed.

The only radioisotope of chromium of radioecological significance is 28-day  $^{51}\text{Cr}$ , a neutron activation product of stable  $^{50}\text{Cr}$ . In the natural environment, chromium occurs largely in the form of chromite deposits in rocks. It generally forms insoluble oxides and plays no known essential role in the physiology of organisms.<sup>42</sup> Very low concentrations of  $\text{CrO}_4^{--}$  ions occur in the hydrosphere. Chromium, especially in its more soluble chromate and dichromate forms, is considered a toxic metal. Chromium-51 has been used as a radiotracer in scientific investigations and is produced by reactors and nuclear detonations in quantities dependent on the amount of stable chromium accessible to thermal neutrons. In aquatic systems,  $^{51}\text{Cr}$  exhibits concentration factors of 200 to 4000 in freshwater organisms<sup>42</sup> and values of 2 to 80 in marine biota.<sup>13</sup> The uptake of  $^{51}\text{Cr}$  from soil by terrestrial plants is generally very low, and assimilation of the nuclide following ingestion by animals is also low, probably less than 0.5%.<sup>6</sup> Because of its short half-life, low biological mobility, and mostly weak radiations,  $^{51}\text{Cr}$  is not generally considered an important biological hazard.

The most consequential radioisotope of manganese is 300-day  $^{54}\text{Mn}$ , which is produced by nuclear weapons and reactors through the reactions  $^{54}\text{Cr}(p,n)^{54}\text{Mn}$  and  $^{54}\text{Fe}(n,p)^{54}\text{Mn}$ .<sup>41</sup> Stable manganese, which occurs in igneous rock at a concentration of about 0.01%,<sup>43</sup> finds its way into the biosphere through geologic weathering and other geochemical processes. The chemistry of Mn is fairly complex, but large quantities in the environment tend to form insoluble  $\text{MnO}_2$  in oxidizing conditions. Acidic or reducing conditions can convert oxide forms to  $\text{Mn}^{++}$  which can undergo exchange reactions with surfaces, especially clays, and which can be taken up by plants. Manganese can replace Ca in carbonates and probably in other compounds. Manganese is an essential element which is required for many biochemical processes in both plants and animals. Because of this, and certain of its chemical forms which are soluble, it enters and passes through food chains. The entry of  $^{54}\text{Mn}$  into organisms is affected by availability of stable Mn, as well as other substances, and chemical form. It is assimilated by mammals to the extent of about 10%,<sup>6</sup> and the principal internal target

organ is the liver.<sup>6,43,44</sup> Aquatic organisms also readily accumulate <sup>54</sup>Mn from the environment.<sup>13</sup>

Neutron activation of iron forms two radioisotopes commonly found in nuclear fallout debris and some reactor effluents, namely <sup>55</sup>Fe and <sup>59</sup>Fe. Iron-55 (2.6-year half-life) is somewhat longer-lived than <sup>59</sup>Fe and usually produced in greater quantities. Because of its mode of decay and weak radiations, <sup>55</sup>Fe is comparatively difficult to measure and not particularly hazardous. Its behavior in the environment is largely controlled by the geochemistry of stable iron, which is both abundant and biologically essential. The major biospheric reservoir of iron is the lithosphere, which contains the element at about 2 atom percent.<sup>23</sup> In general, the bioaccumulation of <sup>55</sup>Fe and <sup>59</sup>Fe is inversely related to the concentrations of biologically available iron in the environment. In the oceans, iron tends to form ferric hydroxide, an insoluble precipitate, resulting in very low concentrations of the element (~10 ppb) in sea water.<sup>45</sup> As a result, marine organisms accumulate substantial quantities of <sup>55</sup>Fe from worldwide fallout and certain marine food chains provide a significant vector of the nuclide to man.<sup>46</sup> Radioisotopes of iron are also accumulated readily by freshwater and terrestrial organisms. Persson,<sup>47</sup> who measured <sup>55</sup>Fe in the lichen-reindeer-human food chain, found readily measurable levels of the nuclide in each biotic component; however, the concentrations appeared to decline with trophic level. As one would expect, radioisotopes of iron tend to concentrate in red blood cells and the spleen is one of the critical organs in man.<sup>6</sup>

Several radioisotopes of cobalt are produced through neutron activation by reactors and nuclear detonations. Of these, the most commonly observed are 267-day <sup>57</sup>Co, 71-day <sup>58</sup>Co, and 5.2-year <sup>60</sup>Co. Cobalt-60 is probably of greatest overall interest because of its comparatively long half-life, penetrating gamma rays, and its uses in medicine, industry, and research. Cobalt-60 is produced mainly by neutron activation of stable <sup>59</sup>Co and <sup>60</sup>Ni. Cobalt is generally considered a trace element in the environment, but it occurs as a microconstituent of plants and animals and plays a role in certain biochemical reactions.<sup>45</sup> Cobalt radionuclides are readily accumulated from the environment by aquatic as well as terrestrial organisms. In marine ecosystems, cobalt concentration factors generally run on the order of 10 to 1000, while values of 100 to 100,000 have been observed in freshwater organisms.<sup>13,48</sup> Data on radiocobalt behavior are more limited for terrestrial than for aquatic ecosystems, but <sup>60</sup>Co concentration ratios of the order of 0.01 to 2.0 have been observed for plants/soil<sup>49</sup> and values of 0.1 to 100 were found for rat (*Rattus rattus*) tissues/soil at Eniwetak Atoll.<sup>50</sup> In mammals, cobalt is readily assimilated from the gut, and it is eliminated from the body comparatively rapidly. Major target organs for assimilated Co usually include the liver and kidney.

Most radionuclides of zinc are short-lived and seldom abundant in the environment. The major exception is 245-day <sup>65</sup>Zn, which is produced, sometimes in copious quantities, by neutron activation of stable Zn in nuclear reactors and detonations. Zinc-65 has also been used widely as an experimental tracer for stable zinc in biological and ecological studies. Stable zinc, which heavily influences the behavior of <sup>65</sup>Zn in the environment, is a comparatively rare constituent of the biosphere, yet it is considered essential for growth and function of organisms.<sup>51</sup> A substantial fraction of the Zn in both aquatic and terrestrial ecosystems ties up with soil and sediment where it is at least temporarily unavailable to all but those organisms which ingest or reside in soils and sediments. However, a measurable fraction of environmental Zn is accumulated by organisms and transported through aquatic and terrestrial food chains.<sup>51</sup> Zinc-65 is taken up through leaves and roots by plants, but uptake is strongly modified by chemical form of the nuclide and soil properties. When ingested by animals, <sup>65</sup>Zn

shows intermediate assimilation, but fairly long retention,<sup>6</sup> hence substantial tissue accumulation is possible under chronic intake. Liver and prostate represent critical organs in man. Weapons testing in the Pacific led to substantial <sup>65</sup>Zn contamination of marine biota, with concentration factors of up to 10<sup>3</sup> reported for algae and 10<sup>5</sup> for certain mollusks.<sup>51</sup> Polikarpov<sup>13</sup> cites <sup>65</sup>Zn concentration factors generally in the range of 10<sup>3</sup> to 10<sup>4</sup> for both marine and freshwater biota, but large variations in the values occur between specific organisms and circumstances. Because of its half-life and biological mobility, <sup>65</sup>Zn can be transported through food chains to man.<sup>52</sup>

Zirconium, though rare and lacking apparent biological function, is of interest because of 65-day <sup>95</sup>Zr, a high-yield fission product with energetic beta and gamma radiations. Most stable zirconium is associated with oxide compounds of the mineral zircon.<sup>53</sup> The chemistry of Zr in solutions is complicated by its tendency to form colloids and to undergo hydrolysis and polymerization. In general, the chemical forms of Zr in the environment are not very soluble and as a result, mobility of the nuclide in biological systems is low. Although <sup>95</sup>Zr has been measured in biological samples in areas which received fission product debris, the material is largely associated with external surfaces. Root uptake of <sup>95</sup>Zr from soil is very low and the assimilation fraction of the nuclide following ingestion is generally <10<sup>-4</sup>, according to ICRP.<sup>6</sup> Because of its low assimilation, the critical organ for continuous ingestion of <sup>95</sup>Zr is considered to be the gastrointestinal tract. The small quantities of Zr which are assimilated, are generally retained for long periods of time. Despite low solubility and poor ability to enter organisms, reasonably high concentration factors, probably resulting from surface adsorption, have been observed for <sup>95</sup>Zr in aquatic biota. Concentration factors of the order of 10<sup>2</sup> to 10<sup>4</sup> have been documented for freshwater plants and animals, while values in the range of 1 to 10<sup>3</sup> in marine biota have been recorded.<sup>13</sup> Because of its high fission yield and penetrating gamma rays, <sup>95</sup>Zr and its 35-day daughter, <sup>95</sup>Nb, usually contribute importantly to external gamma fields following deposition of fresh fission debris.

Technetium is interesting because the element has no stable isotopes and <sup>99</sup>Tc is an abundant, long-lived fission product which appears to be very mobile in ecosystems. This element has received little attention until the last few years and few papers have appeared on the subject. Of substantial interest is the indication that the element is poorly retained by soil and that plant uptake is quite high.<sup>54</sup> Plant/soil concentration ratios as high as 10<sup>3</sup> have been reported for <sup>99</sup>Tc supplied to soil in the pertechnetate form.<sup>55</sup> Assimilation of Tc following ingestion appears high (~50%), but retention of the nuclide by animal tissues appears low, with biological half-times of 1 to 30 days, depending upon tissue type.<sup>6</sup> Since 2 × 10<sup>5</sup>-year <sup>99</sup>Tc is a pure beta emitter, its importance as an external hazard is minimal. However, its abundance, long half-life, and ecological mobility may pose special problems, especially in regard to nuclear waste disposal.

Of the 10 or so radioisotopes of ruthenium, <sup>103</sup>Ru and <sup>106</sup>Ru are of greatest significance in that both are high-yield, intermediate-lived fission products which exhibit some mobility and biological concentration. Stable ruthenium is a rare element in the environment and it does not appear to have significant biological function. Therefore, radioisotopes of Ru are not considered as having essential nutrient analogs, even though they are absorbed to some extent by living tissues. Ruthenium can exhibit several oxidation states and form a wide variety of compounds and complexes, which complicates the matter of its uptake and retention by plants or animals.<sup>56</sup> In general, Ru appears to exhibit substantially greater mobility through soils and groundwater than other fission products; however, its mobility is strongly influenced by chemical form. Instances of substantial subsurface migration of <sup>106</sup>Ru have been documented

at nuclear waste disposal sites. Ruthenium can be accumulated from the soil by plants, as shown by Auerbach and Olson who measured plant/soil concentration ratios up to 15 for bean leaves.<sup>56</sup> Ruthenium-106 has been found in substantial concentrations in muskrat (*Ondatra zibethica*) kidney tissue at Oak Ridge waste disposal areas and in measurable concentrations from fallout in deer liver.<sup>44,56</sup> Tabulated data indicate that <sup>106</sup>Ru has a moderate assimilation fraction in mammals and fairly rapid biological elimination. In terms of radiological assessment of <sup>103</sup>Ru and <sup>106</sup>Ru, account must be taken of the respective short-lived radioactive daughters, <sup>103m</sup>Rh and <sup>106</sup>Rh. These rhodium daughters rapidly reach secular equilibrium with <sup>103,106</sup>Ru and generally accompany the parent nuclides through ecological and biological systems.

Of the heavy metals discussed in this section, all but lead and polonium result primarily from the nuclear activities of man. The element lead consists of four stable isotopes of mass 204, 206, 207, and 208, and it has some 13 radioisotopes. The lead radioisotopes of mass 210, 211, 212, and 214 are all naturally occurring beta emitters generated by decay of radon isotopes. Of these four, <sup>210</sup>Pb has received the greatest attention because of its ubiquitous nature and 20-year half-life. One of the significant features of lead is that it is the heaviest element having stable isotopes and it marks the cutoff for alpha emitters, all of which are heavier than lead. Lead is present in all biospheric media, but concentrations vary widely according to sample type and location. It is not considered biologically essential, but it behaves in physiologically similar fashion to the alkaline earths and is incorporated in biological material.<sup>40,41</sup> The industrial activities of man have resulted in substantial global contamination by this potentially toxic element.<sup>57</sup> Radioactive <sup>210</sup>Pb is generated by decay of <sup>222</sup>Rn in the atmosphere and lithosphere. A major environmental pathway of this nuclide is aerial deposition, rates of 10<sup>11</sup> atoms/m<sup>2</sup>-year having been documented.<sup>58</sup> Measurable levels therefore occur on foliage,<sup>59</sup> particularly long-lived species such as lichens and mosses.<sup>60</sup> Herbivorous animals continually ingest <sup>210</sup>Pb and because it is moderately soluble and is tenaciously retained in the body, measurable body burdens have been documented in reindeer,<sup>60</sup> humans,<sup>58</sup> and other species. <sup>210</sup>Pb is also inhaled by animals and it has been documented that people who smoke have considerably higher body burdens than nonsmokers.<sup>61</sup> Assimilated lead primarily lodges in bone, but the kidney and liver are also important target organs. Though comparatively little data on the subject are available, the senior author has documented concentration factors of 10<sup>2</sup> to 10<sup>4</sup> for stable lead in freshwater biota.

Several radioisotopes of polonium are generated by the natural decay series of <sup>232</sup>Th, <sup>238</sup>U, and <sup>235</sup>U, but most are very short-lived and therefore exhibit behavior that is controlled largely by that of the parent nuclides. The important exception is 138-day <sup>210</sup>Po, which is generated as a radon daughter in the <sup>238</sup>U decay series. Because of its half-life, abundance, physiological behavior, and alpha emission, <sup>210</sup>Po is considered "very hazardous" in comparison to most other radionuclides.<sup>62,63</sup> Since there are no stable isotopes of polonium we do not have this usual yardstick for understanding its geochemistry or biology. As with <sup>210</sup>Pb, <sup>210</sup>Po enters the biosphere largely through aerial deposition on foliage and ingestion or inhalation by terrestrial animals. In long-lived biological tissues, such as lichens, <sup>210</sup>Po activity usually approaches secular equilibrium with <sup>210</sup>Pb. While absorption of <sup>210</sup>Po from the gut is comparable to lead, the distribution is more heavily oriented to the soft tissues, notably kidney, spleen, and liver.<sup>6</sup> Because of this, <sup>210</sup>Po has greater opportunity than lead to enter higher trophic levels of food chains. Since <sup>210</sup>Po accumulates on foliage, it is present in tobacco and smokers contain up to 3 times the lung burden of this nuclide as nonsmokers.<sup>58</sup> Polonium-210 is also accumulated by aquatic organisms and concentration factors of the order of 10<sup>3</sup> have been reported for marine biota.<sup>13</sup>

### E. Rare Earths (Y, La, Ce, Pr, Pm)

The rare earth elements include the lanthanide series (atomic numbers 57 through 71) and because of their similar chemical properties, scandium and yttrium. The principal radioisotopes of interest in this group include  $^{90,91}\text{Y}$ ,  $^{140}\text{La}$ ,  $^{141,144}\text{Ce}$ ,  $^{144}\text{Pr}$ , and  $^{147}\text{Pm}$ . These radionuclides are produced as principal fission products and as daughters of other fission products. They share numerous important chemical and biological attributes. The stable isotopes of the rare earths occur in trace quantities in nature, and apparently no essential biological role for them has been demonstrated. In most aqueous solutions, the rare earths usually form +3 and +2 ions.<sup>64</sup> In normal aquatic environments, however, the rare earths mainly form hydroxides and occur as insoluble particulate forms. One of the chemical characteristics of the lanthanide series is that electrons can enter the 4f inner shell in addition to the valence shell. A comprehensive review of the chemistry of the rare earths is available.<sup>65</sup>

The rare earth radionuclides in the environment have largely been formed by nuclear weapons detonations. The initial chemical form of such material was principally the oxides, which are comparatively insoluble. Because of the insolubility and lack of biological function, the rare earth nuclides generally exhibit low biotic uptake and minimal food chain transport.<sup>64</sup> These nuclides, however, attach readily to biological surfaces and may be ingested in significant quantities by filter-feeding aquatic organisms, but concentrations usually diminish markedly at higher trophic levels. Rare earths are generally retained strongly by soils and uptake by terrestrial plants is very low, usually lower than uptake of strontium, cesium, and ruthenium. Measurable levels may, nevertheless, occur on terrestrial vegetation following aerial deposition of fallout debris. As a group, the rare earths are expected to be assimilated from the gut in animals to an extent of less than 0.01%.<sup>6</sup> However, the small fraction assimilated seeks bone, liver, and kidney as principal target organs, and excretion is very slow, with biological half-times of the order of  $10^2$  to  $10^4$  days. Because of low assimilation, the critical organ for continuous ingestion is usually the gastrointestinal tract. For inhalation of insoluble particles, the lung is sometimes listed as the critical organ.

Yttrium-90 and 91 are of interest because  $^{91}\text{Y}$  is a fairly abundant radionuclide at a few months post-fission and  $^{90}\text{Y}$  is a daughter of  $^{90}\text{Sr}$ . Both have energetic beta emissions. Yttrium-90 contributes little to the total radioactivity until several years post-fission because its production is largely from 28-year  $^{90}\text{Sr}$ , with which it grows into secular equilibrium. Because of this, the biological importance of  $^{90}\text{Y}$  is largely dependent upon the ecological persistence and mobility of  $^{90}\text{Sr}$ . In bone, the critical organ for  $^{90}\text{Sr}$ , most of the radiation dose is actually delivered by the higher energy beta particles of  $^{90}\text{Y}$ . By itself,  $^{90}\text{Y}$  is neither persistent nor mobile in biological systems. Being longer-lived, 58-day  $^{91}\text{Y}$  can accumulate on biological surfaces. Concentration factors ranging from 10 to over  $10^4$  have been measured for this nuclide in aquatic organisms.<sup>13</sup>

Lanthanum-140 is the only radionuclide of La that deserves discussion, and it is of importance primarily because it is generated by the decay of 13-day  $^{140}\text{Ba}$ . The 40-hr half-life of  $^{140}\text{La}$  precludes significant environmental persistence as a principal fission product. Since  $^{140}\text{Ba}$  is also comparatively short-lived,  $^{140}\text{La}$  is of relative significance only for a month or two post-fission. Similar to the situation for  $^{90}\text{Sr}$ - $^{90}\text{Y}$ ,  $^{140}\text{La}$  behavior and importance is largely dependent on its longer-lived parent; however, most of the radiation dose from  $^{140}\text{Ba}$  actually arises from the energetic radiations of  $^{140}\text{La}$ .

Cerium has two abundant fission-produced radioisotopes, 33-day  $^{141}\text{Ce}$  and 285-day  $^{144}\text{Ce}$ . As one would expect,  $^{141}\text{Ce}$  is of relative importance from a few weeks to a few months post-fission, while  $^{144}\text{Ce}$  peaks in relative importance between  $10^2$  and  $10^3$  days post-fission. During the latter period,  $^{144}\text{Ce}$  and its 17-minute daughter,  $^{144}\text{Pr}$ , contribute more of the total fission product beta activity than any other nuclide.<sup>56</sup> The  $^{144}\text{Ce}$ -

$^{144}\text{Pr}$  pair emits a mixture of beta particles and gamma rays, but the 3 MeV beta of  $^{144}\text{Pr}$  is the dominant contributor to dose from internally deposited  $^{144}\text{Ce}$ . Despite the generally low solubility and mobility of  $^{144}\text{Ce}$ , the nuclide has been found in fish tissues following U.S. nuclear testing in the Pacific,<sup>64</sup> and in mule deer livers from worldwide fallout.<sup>44</sup> In the latter case, 71% of the estimated dose to deer liver resulting from several fallout radionuclides, was contributed by  $^{144}\text{Ce}$ - $^{144}\text{Pr}$ . In view of the very low assimilation expected for  $^{144}\text{Ce}$ , the only reasonable explanation for significant liver burdens would be chronic ingestion for a long period, coupled with tenacious retention by the organ. In aquatic ecosystems,  $^{144}\text{Ce}$  concentration factors range from 1 to  $10^4$ , with somewhat higher values for plants than animals and generally higher values for freshwater than marine biota.<sup>13</sup>

Promethium has 10 radioisotopes, but no stable isotopes. The isotope  $^{147}\text{Pm}$  is a comparatively high-yield fission product which contributes a significant fraction of the total beta activity 2 to 10 years post-fission. Its peak relative abundance occurs 5 years after fission. Since  $^{147}\text{Pm}$  is essentially a pure beta emitter, it does not contribute significantly to external exposure from fallout fields and is mainly of interest as an internal emitter. Its detection and measurement normally requires chemical separation, therefore it has not received as much study as some of the other rare earths. Like most of the other rare earths, the fraction of ingested  $^{147}\text{Pm}$  assimilated is  $<10^{-4}$ , and 80% of the body burden is likely to be found in bone, with about 6% in the liver and 2% in the kidney.<sup>6</sup> Mean concentration factors of roughly 4000 and 700 have been reported for freshwater plants and animals, respectively.<sup>13</sup>

#### F. Actinides (Th, U, Pu)

The actinide elements include number 89 (actinium) and higher numbered elements. Included are the naturally occurring elements Ac, Th, Pa, and U, as well as the transuranium elements, which are largely produced by artificial neutron bombardment of natural uranium. All actinide elements can exist in the (III) oxidation state, but this state is most stable only for Ac, Am, and higher-numbered elements.<sup>66</sup> The (IV) oxidation state is most stable for Th, Np, and Pu, while the (VI) state is most stable for U. Elements 91 through 95 can exist in several oxidation states, complicating their chemistry. The actinides are chemically similar to the rare earths in a number of ways which can make their separation difficult. Like the rare earths, electrons can be added to a subshell of actinides, in this case the 5f subshell.

A very important feature of the actinides is that they are all radioactive, yet half-lives of the actinide nuclides vary widely. It is noteworthy that isotopes of all actinides can arise from the decay or neutron capture of primordial uranium. Certain isotopes of Ac, Th, Pa, and U arise in the decay chains of long-lived, primordial  $^{235}\text{U}$ ,  $^{238}\text{U}$ , and  $^{232}\text{Th}$ . Although some of the less massive transuranium nuclides, such as  $^{239}\text{Np}$  and  $^{239}\text{Pu}$ , are formed naturally in small quantities through neutron capture of  $^{238}\text{U}$ , the overwhelming production of transuranics is the result of man's activities. Elements heavier than uranium arise principally through neutron capture and through beta decay, which effectively adds a proton to the nucleus. The high neutron fluxes associated with nuclear reactors and nuclear detonations can lead to successive neutron captures by heavy nuclei, leading to yet heavier, neutron-rich nuclides. Greater stability of such neutron-rich nuclides is frequently achieved through beta decay and the formation of a new element of higher atomic number. Prior to 1940, the transuranium elements were unknown. Early investigations were conducted with microgram quantities of these elements, and nowadays very large quantities are produced by the nuclear industry.

Many of the actinide nuclides emit alpha particles, which increases their potential hazard as internal emitters. In addition to presenting potential radiation hazards, sev-

eral of the longer-lived actinides such as  $^{232}\text{Th}$ ,  $^{238}\text{U}$ , and  $^{237}\text{Np}$  can occur in sufficient mass quantities to be chemically toxic. Beta particle and photon emissions are also common among the actinides and a few nuclides, such as  $^{242}\text{Cm}$  and  $^{244}\text{Cm}$ , undergo spontaneous fission, which leads to emission of neutrons.

In general, the actinide nuclides form comparatively insoluble compounds in the environment and are therefore not considered biologically mobile. Furthermore, their interaction with biological systems is largely passive, and the heavier actinides, at least, have no known essential biological function. The actinides are transported in ecosystems mainly by physical and sometimes chemical processes. They tend to attach, sometimes strongly, to surfaces; and they tend to accumulate in soils and sediments which ultimately serve as storage reservoirs. Subsequent movement is largely associated with geological processes such as erosion and sometimes leaching. Organisms can effect actinide movement to a minor extent, usually through their own movements which cause rearrangement of abiotic materials.

Organisms are exposed to the actinides largely through surface contact, inhalation, and ingestion. Ordinarily, very little of these substances passes biological membranes to lodge internally, unless inhalation is the route of exposure. Uptake of the actinides by terrestrial plants from soil is generally considered to be low, especially if one considers plutonium, the element for which most data exist. Plant/soil concentration ratios for true uptake of Pu generally run of the order of  $10^{-4}$  or less, especially for the oxide and hydroxide forms which usually occur in the environment.<sup>67</sup> There is scattered evidence, however, that a somewhat higher plant/soil concentration ratios exist for U and Am.<sup>67,68</sup> There is considerable variation in plant uptake of actinides, according to the specific element, soil characteristics, and plant species. Uranium is apparently taken up to a greater extent than thorium and some plant species accumulate uranium to the extent that they may serve as bioindicators of uranium ore deposits.<sup>40</sup> In field situations, actinides associated with vegetation are frequently attached to the surfaces to a much greater extent than they are biologically incorporated.

In the case of terrestrial animals, the actinides are taken in mainly through ingestion and inhalation, the latter pathway generally being regarded as the more important of the two. This is generally true for recent environmental deposits which can be readily resuspended in the airstream. As deposits age and weather into the soil, however, the ingestion pathway may become relatively more significant. For submicron particles, the fraction of inhaled actinides which enters the blood exceeds the fraction of ingested material which enters the blood. Furthermore, submicron, insoluble radioactive particles may reside in the deep lung for considerable periods of time. Larger particles are usually swallowed following inhalation and thus enter the gastrointestinal tract where they encounter the same physiological environment as ingested material. Assimilation of all actinide elements from the gastrointestinal tract is assumed to be less than 0.01%.<sup>6</sup> In addition, the actinides deposit within the body in similar fashion and based on animal experiments, some 80% or more of the total body burden is expected to be found in bone, with 1 to 10% in both the kidney and liver.<sup>6</sup> Retention of all the actinides except uranium is normally long, with whole body biological half-times of  $10^4$  days or greater assumed for predictive purposes. Uranium is excreted somewhat more rapidly from the body, as indicated by a biological half-time of the order of  $10^2$  days. Under conditions of continuous ingestion of most actinides, bone or the gastrointestinal tract are usually considered critical organs for dosimetry purposes. For inhalation of small, insoluble particles, the lung is usually the critical organ.

In aquatic systems, the actinides are readily adsorbed on the surfaces of plants and small animals such as zooplankton, and thus they enter the food chain. However, concentration factors generally decline substantially with trophic level and in the internal as compared to external tissues by virtue of membrane discrimination. Concentra-

tion factors for Th, U, Pu, Am, and Cm have been reported, but most data are for  $^{239}\text{Pu}$ . Noshkin<sup>69</sup> summarized published  $^{239}\text{Pu}$  concentration factors for marine environments, and most values for invertebrates and algae were in the range of  $10^2$  to  $10^4$ . Concentration factors for vertebrate tissues ranged from 1 to  $10^3$ . Concentration factors for other actinides can generally be expected to fall into these ranges.

Because of their high natural abundance in certain localities and ubiquitous presence in the biosphere, Th and U deserve some additional discussion. So does plutonium, because it has been studied to a far greater extent than the other transuranics. In terms of total quantities of transuranic radioactivity generated by the nuclear industry, several radionuclides, including  $^{239}\text{Np}$ ,  $^{241}\text{Pu}$ ,  $^{241}\text{Am}$ , and  $^{242}\text{Cm}$  are considerably more abundant than the well-studied fissile nuclide  $^{239}\text{Pu}$ .<sup>70</sup> However, the environmental behavior of these nuclides is not well studied, but some general inferences to them can be made, with caution, from data on  $^{239}\text{Pu}$ .

Thorium occurs ubiquitously throughout the lithosphere and is found at an average concentration of about 10 ppm in crustal material.<sup>66</sup> Its geographic distribution, however, shows many "hot spots", the more notable of which occur in Brazil and India.<sup>41</sup> Such hot spots are usually characterized by monazite-bearing sands, or certain types of volcanic intrusives. Thorium has a potential role in the nuclear fuel cycle because neutron capture by primordial  $^{232}\text{Th}$  produces  $^{233}\text{Th}$ , which decays through  $^{233}\text{Pa}$  to form fissile  $^{233}\text{U}$ . In addition, thorium has certain industrial uses, such as in the manufacture of incandescent gas mantles and welding rods. Of the 13 isotopes of Th, three are very long-lived with half-lives  $\gg 1000$  years. The isotopes of predominant importance are  $^{232}\text{Th}$  and  $^{228}\text{Th}$  of the natural thorium series, and  $^{234}\text{Th}$  and  $^{230}\text{Th}$ , which are decay products in the natural uranium series (see Table 2 Chapter 4).

Mobility of Th in biological systems is extremely low. Plant uptake of the element is essentially negligible,<sup>40,68</sup> as is its absorption by animals, and there is apparently no evidence for measurable quantities of  $^{232}\text{Th}$  in foods or in marine biota.<sup>71,72</sup> However, there is evidence for small quantities of  $^{228}\text{Th}$  in plant and animal tissues.<sup>72,73</sup> This nuclide probably is generated within biological tissues by the decay of  $^{228}\text{Ra}$ , which is taken up by plants and animals to a far greater extent than thorium. This may be of some significance because Th is retained tenaciously by bone and the carcinogenic effects of  $^{228}\text{Th}$  at higher levels appear to be substantially greater than  $^{226}\text{Ra}$ , and perhaps even greater than  $^{239}\text{Pu}$ .<sup>74</sup>

Uranium is undoubtedly the most significant element of the nuclear era. It is the raw material from which is generated, either through fission or neutron activation, the majority of radionuclides discussed in this book. Natural uranium is present in the earth's crust at an average concentration of roughly 4 ppm and high grade ores in the Belgian Congo and Canada contain up to 1 to 4%  $\text{U}_3\text{O}_8$ .<sup>66</sup> Medium grade ores, ranging from 0.05 to 1%  $\text{U}_3\text{O}_8$  occur more frequently in the U.S. and elsewhere. Natural uranium is 99.27%  $^{238}\text{U}$ , which is fertile and 0.72%  $^{235}\text{U}$ , which is fissile and therefore a primary fuel for nuclear reactors. Because of the long ( $4.5 \times 10^9$  year) half-life of  $^{238}\text{U}$ , it is of low specific activity. Because of this, and its generally low biological mobility in ecological systems, it has received less attention from a radioecological point of view than it probably deserves.

Although of generally "low" biological mobility, uranium is taken up by plants to a considerably higher degree than thorium. In particular, certain perennial plants concentrate U in excess of 100 ppm, with plant/soil concentration ratios ranging from  $10^{-4}$  to values in excess of  $10^{-1}$ , and considerable effort has been spent investigating the use of certain species in prospecting for uranium.<sup>75-77</sup> The availability of U in soil apparently varies over a wide range according to soil chemistry and this undoubtedly contributes to high variability in plant uptake. There is some evidence that uranium stimulates plant growth at low concentrations, that it becomes toxic at slightly higher

levels, and that certain species appear to be associated with soils containing uranium, selenium, and sulfur.<sup>75</sup> Through a comprehensive series of studies in both aquatic and terrestrial ecosystems, Kovalsky and co-workers found that uranium occurs in detectable concentrations in a variety of biota, but concentrations generally diminish with trophic level.<sup>76</sup> Uranium is present in the human diet to the extent that some 1 to 2  $\mu\text{g}/\text{day}$  may be ingested per person.<sup>19</sup> In some localities, where spring waters contain elevated levels of U, 10 to 300  $\mu\text{g}/\text{day}$  can be ingested by people.<sup>39</sup> Concentration-factors ranging from 10 to 400 have been reported for U in marine biota.<sup>13</sup> Natural uranium is chemically toxic because of its low specific activity, and it is frequently difficult to distinguish radiation and chemical effects from the element.

Some 14 isotopes of plutonium have been documented, but most interest has centered around fissile  $^{239}\text{Pu}$ , with secondary attention focused upon  $^{238}\text{Pu}$  and  $^{241}\text{Pu}$ . Because  $^{239}\text{Pu}$  is a long-lived, bone-seeking alpha emitter of comparatively high radiotoxicity, and because of its potential importance as a nuclear fuel for reactors as well as a component of nuclear weapons, it has generated much respect, and from the public, much fear. It has been labeled as "one of the most toxic substances known to man". This has led to widespread adoption of the phrase, "the most toxic substance known to man", and frequent hysteria in connection with the politics of nuclear power and national defense. In actuality, while  $^{239}\text{Pu}$  is comparatively hazardous under certain circumstances, it is no more toxic than naturally occurring  $^{228}\text{Th}$ ,<sup>74</sup> and probably far less toxic than arsenic, or certain biological toxins.<sup>79</sup> Despite much general misunderstanding of plutonium, "perhaps no single element has ever been so intensively studied,"<sup>80</sup> and consequently, a great deal is known about its physics, chemistry,<sup>81</sup> and biology.<sup>82</sup> The ecology of plutonium has also been investigated rather intensively, particularly within the last decade,<sup>87</sup> and a number of recent reviews on the subject are available.<sup>83-87</sup>

A fundamental reason for the attractiveness of  $^{239}\text{Pu}$  as a nuclear fuel is that it can be chemically separated from its precursor,  $^{238}\text{U}$ , in rather pure form. In contrast, the other fissile isotopes,  $^{233}\text{U}$  and  $^{235}\text{U}$ , require much more effort to separate from  $^{238}\text{U}$  and to obtain in sufficient purity to serve as a reactor fuel or weapon component. Although plutonium can be chemically isolated from other elements, its chemistry is extremely complex. This complexity is due in large measure to the ability of Pu to exist in four oxidation states (III, IV, V, and VI), frequently simultaneously, in appreciable concentrations in aqueous systems.<sup>81</sup> It also occurs in six allotropic forms as a metal. In general, plutonium in the environment can be expected to undergo hydrolysis and oxidation, with  $\text{PuO}_2$  a more common form. However, Pu can form complexes with a variety of organic and inorganic compounds.<sup>88-89</sup> When complexed with chelating agents, such as with DTPA for example, plutonium solubility in soil, uptake by plants, and general biological mobility is significantly increased.<sup>88,90</sup> Under most environmental conditions, however, plutonium occurs in forms which are comparatively insoluble and which are poorly transferred across biological membranes.

As a result of nuclear weapons testing, some 0.3 to 0.5 million curies of plutonium have been introduced into the biosphere.<sup>92</sup> This has resulted in very low levels of Pu, ubiquitously distributed in the environment. Additionally, accidents, inadvertent releases, and experiments have caused higher level Pu contamination in local areas. Examples include U.S. Air Force bomber accidents in Spain and Greenland, atmospheric reentry of a  $^{238}\text{Pu}$ -powered satellite, releases at Rocky Flats, Oak Ridge, Hanford, and Mound Laboratory, and experiments at the Nevada Test Site.<sup>91</sup> Cleanup operations were instituted at several sites having the higher levels of Pu contamination. Residual contamination in undisturbed sites has provided opportunity to study the ecological behavior of plutonium in several kinds of environments.

In terrestrial ecosystems, well over 90%, and usually over 99% of the plutonium is found in the soil.<sup>92</sup> The remainder is distributed among the litter and biotic components of the ecosystem. An exception to this trend may be noted immediately following a contamination event, when a large fraction may be associated with vegetation. Distribution of Pu in aquatic systems is normally comparable, with sediments rapidly becoming the dominant reservoir of the material. Movement of Pu from soil and sediments to plants and animals is greatly inhibited by its insolubility and strong discrimination at biological membranes. As a rule of thumb,  $10^{-4}$  is a reasonable discrimination factor for Pu to be applied at each step in the soil-plant-animal mineral chain. Concentrations of Pu in plants are normally of the order of  $10^{-6}$  to  $10^{-3}$  the concentrations in the soil in which they grow, when aerial deposition does not contribute to the plant activity.<sup>90</sup> Resuspension of soil particles and subsequent deposition on foliage surfaces can result in much higher plant/soil concentration ratios, up to  $10^{-1}$  in some cases. Normally, less than  $10^{-4}$  of the plutonium ingested by consumers is absorbed from the intestinal tract.<sup>82</sup> If inhaled, however, a somewhat larger fraction of the Pu can enter the body. For instance, some 5% of a pulmonary deposit can be absorbed into the blood stream and 15% can enter the lymphatic system.<sup>93</sup>

Plutonium deposits in the body are retained tenaciously.<sup>93</sup> For instance, pulmonary and lymph node deposits are cleared with half-times of the order of 500 and 1000 days, respectively. Skeletal deposit retention half-times increase with body weight, and range from roughly 1 year in small mammals to over 100 years in humans. The distribution of plutonium in biological tissues tends to be nonuniform, and "hot spots" are commonly observed. This complicates the dosimetry and has led to considerable controversy regarding the biological consequences of internal deposits.<sup>94</sup>

It is important to be aware of the fact that a tremendous amount of research has been conducted on the biological effects of internally deposited plutonium, and more is known about the biomedical aspects of this element than most other hazardous materials. An excellent review on this is available.<sup>82</sup> From an ecological viewpoint, the isotopes of plutonium do not appear to be as hazardous as some of the more biologically mobile radionuclides such as <sup>137</sup>Cs or <sup>90</sup>Sr. Based upon substantial research efforts, no specific physical injury to plants, animals, or man has been shown to be caused by plutonium, even though such exposures have occurred.

### G. Summary: General Properties of Selected Radionuclides

A general idea of the ecological behavior of specific radionuclides can be obtained through tabulation of various attributes. Such attributes include general chemistry, half-life, distribution in ecosystems and organisms, modes of exposure to organisms, and absorption and retention by organisms. Table 2 represents an attempt at such a tabulation. Because of the wide variations in the quantification of such attributes, qualitative or semiquantitative descriptions are used, with the intent of giving generalized expectations for selected radionuclides. The literature should be consulted for more specific data which are to be applied to specific cases.

Much lumping and oversimplification are necessary for any generalized discussion of radionuclide behavior. For example, ecological transport is usually difficult to describe in terms that apply to all steps in a food chain, which apply to aquatic as well as terrestrial ecosystems, or which cover all chemical forms of the radionuclide in question. With such caveats in mind, we can proceed. In Table 2, important exposure modes or processes are described for animals and plants, both terrestrial and aquatic, with the kinds of organisms being implied by the name of the process. The categories describing degree of food chain transport and successive trophic level concentration are particularly subject to variations between specific trophic levels and between