

APPENDIX II

Radioactive Substances in the Environment

This appendix identifies potential sources, properties, and pathways of radioisotopes in the environment to provide the reader with a useful context for discussions of measurement techniques and their application to HRS scoring. In general, radioactive sources at Superfund sites contain either naturally occurring radionuclides or manmade radionuclides, or both, in varying concentrations and physical and chemical forms.

Radionuclides present in the natural environment can be divided into three groups according to origin:

- (1) **Naturally occurring radionuclides** are those terrestrial radionuclides (and their decay products) of primordial origin with half-lives comparable to the age of the earth (about 3 billion years);
- (2) **Cosmic radiation and cosmogenic radionuclides** consist of primary charged and neutral particles that bombard the earth's atmosphere and the secondary particles generated by the primary particles in the earth's atmosphere; and
- (3) **Ubiquitous manmade radionuclides** are those radionuclides generated by man's activities and widely distributed in the environment.

Group #1: Naturally Occurring Radionuclides

Naturally occurring terrestrial radionuclides include several dozen or more radionuclides of the uranium, thorium, and actinium series that decay in series to eventually form isotopes of stable lead. Also included among the naturally occurring radionuclides are a group of "non-series" radioisotopes, e.g., H-3, K-40, and Rb-87, that decay directly to a stable isotope. Uranium-238, U-235, and Th-232 head the uranium, actinium, and thorium series, respectively. Each of these series can be further divided into several subseries based on the differences in the radioactive and physical properties of their progeny, as discussed below. When the decay members of these series are not subjected to either chemical or physical separation processes in the environment, a state of secular equilibrium may be achieved whereby the all series members decay at the same rate as the parent nuclide heading the series. More often, however, series members separate from each other in the environment to some extent due to their differing physical and chemical properties. As a result, varying degrees of activity disequilibrium can occur among series members.

Uranium Series

The members of the uranium series are shown in Exhibit 1 along with their respective radioactive half-lives and principal decay modes. Uranium-238, which heads this series, constitutes 99.28% by weight of the four isotopes of uranium with mass numbers 230, 234, 235, and 238 found in nature. By comparison, the natural abundances of U-234 and U-235 are only 0.0058% and 0.71%, respectively.

The first uranium subseries consists of the radioisotopes U-238, Th-234, Pa-234m, and U-234. In general, all four isotopes are found together in equal activity concentrations (i.e., secular equilibrium) under a wide range of environmental settings. However, less than equal activity concentrations of U-238 and U-234 have been reported by several investigators, indicating that some separation of these isotopes may occur in the environment. For example, Rosholt et al. (Ro66) reported a $^{234}\text{U}/^{238}\text{U}$ activity ratio as low as 0.58 in a soil horizon weathered to clay, and Smith and Jackson (Sm69) reported activity ratios of 0.914 to 0.985 in 16 widely distributed sources. A uranium activity ratio of 1.1 in water was determined from samples taken from the Atlantic, Pacific, and Indian-Oceans (Ro64). Because of the large variability that can exist in uranium isotope activity ratios, it is very important to determine the degree of isotopic equilibrium between U-234 and U-238 in media samples on a site-specific basis.

Exhibit 1. Uranium Decay Series*

Radioisotope (atomic #)	Historical name	Half-life**	Major radiation energies (MeV) and intensities***		
			α	β	γ
U-238 (92) ↓	Uranium I	4.51 x 10 ⁹ y	4.15 (25%) 4.20 (75%)	---	---
Th-234 (90) ↓	Uranium X ₁	24.1 d	---	0.103 (21%) 0.193 (79%)	0.063c (4%) 0.093c (4%)
Pa-234m [†] (91) ↓	Uranium X ₂	1.17 m	---	2.29 (98%)	0.765 (0.3%) 1.001 (0.6%)
U-234 (92) ↓	Uranium II	2.47 x 10 ⁵ y	4.72 (28%) 4.77 (72%)	---	0.53 (0.2%)
Th-230 (90) ↓	Ionium	8.0 x 10 ⁴ y	4.82 (24%) 4.88 (76%)	---	0.068 (0.6%) 0.142 (0.07%)
Ra-226 (88) ↓	Radium	1602 y	4.60 (6%) 4.78 (95%)	---	0.186 (4%)
Rn-222 (86) ↓	Radon (gas)	3.82 d	5.49 (100%)	---	0.510 (0.07%)
Po-218 [†] (84) ↓	Radium A	3.05 m	6.00 (~100%)	0.33 (~0.02%)	---
Pb-214 (82) ↓	Radium B	26.8 m	---	0.65 (50%) 0.71 (40%) 0.98 (6%)	0.295 (19%) 0.352 (36%)
Bi-214 [†] (83) ↓	Radium C	19.7 m	5.45 (0.012%) 5.51 (0.008%)	1.0 (23%) 1.51 (40%) 3.26 (19%)	0.609 (47%) 1.120 (17%) 1.764 (17%)
Po-214 (84) ↓	Radium C'	164 μ s	7.69 (100%)	---	0.799 (0.014%)
Pb-210 (82) ↓	Radium D	21 y	---	0.016 (85%) 0.061 (15%)	0.047 (4%)
Bi-210 (83) ↓	Radium E	5.01 d	4.85 (0.00007%) 4.89 (0.00005%)	1.161 (~100%)	---
Po-210 (84) ↓	Radium F	138.4 d	5.305 (100%)	---	0.803 (0.0011%)
Pb-206 (82)	Radium G	Stable	---	---	---

* Source: Lederer and Shirley (1978) and Shleien and Terpilak (1984).

** Half-life given in seconds (s), minutes (m), days (d), or years (y).

*** Intensities refer to percentage of disintegrations of the nuclide itself, not to the parent of the series.

† Approximately 0.13% of all Pa-234m β particle emissions form an intermediate radioisotope, Pa-234 (6.75 hrs: β -emitter), before decaying to U-234. For Po-218, 0.02% decays through At-218 (~2 sec: α -emitter) before forming Bi-214. For Bi-214, 0.02% decays through Tl-210 (1.3 m: β -emitter) to Pb-210.

The second uranium subseries, headed by U-234, includes Th-230 and Ra-226. In general, the activity concentrations of Th-230 and Ra-226 measured in most soils and rocks are comparable to those of U-238 and U-234, suggesting that Th and Ra do not tend to migrate from either of their uranium precursors under stable conditions. This may not be true in the case of ground water, surface water, or sediments. For example, Rosholt et al. (Ro66) reported that the disequilibrium between Th-230 and U-238 or U-234 may range by a factor of two in sea water and enhanced in sediments. Other evidence suggests that Ra-226 is readily mobile in natural waters, either due to recoil or breakdown of entrapping solids. A common place for accumulation of radium isotopes is in the calcium carbonate "sinter" deposited at the orifices of, and with the out-wash from, hot springs. Such locations typically show little activity from the uranium precursors. In other environmental settings, Ra-226 demonstrates a strong affinity for anions, particularly sulfate. Thus, in uranium deposits that have been subjected to strong sulfuric acid water produced by the oxidation of ferrous sulfide, low concentrations of Ra-226 are present.

The third subseries, headed by Ra-226, consists of Rn-222, a noble gas, and its short half-life progeny, Po-218, Pb-214, Bi-214, and Po-214. Due to its inert gas structure and relatively long radioactive half-life, Rn-222 is highly mobile in the environment. The short-lived radon progeny are readily ionized and are attracted to dust particles in the air or to clay minerals in soil. In general, Rn-222 and its short half-life progeny quickly establish equilibrium activity concentrations in most samples.

The final subseries consists of the longer-lived radon decay products, Pb-210, Bi-210, and Po-210, and terminates with the formation of stable Pb-206. Due primarily to the migration of Rn-222, Pb-210 concentrations in environmental media are highly variable. Variable concentrations of Po-210 are also common due to its chemical properties.

Actinium Series

Uranium-235 heads the actinium series shown in Exhibit 2. Similar to the uranium series, the actinium series also includes radionuclides with half-lives long enough to permit disequilibrium conditions. Rosholt (Ro59) considers all progeny of U-235 to be a single group headed by Pa-231, which he has shown to be out of equilibrium with U-235. The short half-life of Ra-223 (11.4 days) usually precludes any significant disequilibrium between itself and its parent Pa-231. For the case of radium deposits from ground water, a separate subgroup headed by Ra-223 and ending with stable Pb-207 is often considered. Disequilibrium due to migration of the noble gas Rn-219 is local due to its 4 second half-life.

Thorium Series

The thorium series (Exhibit 3), headed by Th-232, comprises a number of somewhat short-lived progeny. Given no migration of these progeny, the series reaches secular equilibrium in 60 years in minerals, rocks, and soils of low permeability. In highly permeable soils, waters, natural gas, petroleum, and the atmosphere, the chemical and physical properties of the progeny can cause disequilibrium.

The thorium series may be divided into three subseries. The first subseries consists of Th-232 only, the least mobile of the series radionuclides. This radioisotope exists naturally as a very stable oxide and is strongly adsorbed on silicates (Cl76). The second subseries consists of Ra-228, Ac-228, Th-228, and Ra-224. The equilibrium of this subseries is governed by radioactive recoil, adsorption, and changes in carrier compounds with which the radionuclides become associated. Thoron, Rn-220, and its progeny down to stable Pb-208 make up the third possible subseries. As with the actinium series, disequilibrium caused by migration of the noble gas Rn-220 is unlikely due to the short half-life of Rn-220 (55 second).

Non-Series Radionuclides

Exhibit 4 lists 7 of the 17 naturally occurring radionuclides that decay to stable isotopes. Of the 17, 15 have combinations of half-lives, isotopic abundances, and elemental abundances which result in their having insignificant specific activities. Only K-40, Rb-87 and H-3 occur in significant concentrations in nature. K-40 and Rb-87 are alkali metals and Rb-87 is found in nature as a replacement for potassium in minerals.

Exhibit 2. Actinium Decay Series*

Radioisotope (atomic #)	Historical name	Half-life**	Major radiation energies (MeV) and intensities***		
			α	β	γ
U-235 (92) ↓	Actinouranium	7.1 x 10 ⁸ y	4.37 (18%) 4.40 (57%) 4.58c (8%)	---	0.143 (11%) 0.185 (54%) 0.204 (5%)
Th-231 (90) ↓	Uranium Y	25.5 h	---	0.140 (45%) 0.220 (15%) 0.305 (40%)	0.026 (2%) 0.084c (10%)
Pa-231 (91) ↓	Protactinium	3.25 x 10 ⁴ y	4.95 (22%) 5.01 (24%) 5.02 (23%)	---	0.027 (6%) 0.29c (6%)
Ac-227[†] (89) ↓	Actinium	21.6 y	4.86c (0.18%) 4.95 (1.2%)	0.043 (~99%)	0.70 (0.08%)
Th-227 (90) ↓	Radioactinium	18.2 d	5.76 (21%) 5.98 (24%) 6.04 (23%)	---	0.050 (8%) 0.237c (15%) 0.31c (8%)
Ra-223 (88) ↓	Actinium X	11.43 d	5.61 (26%) 5.71 (54%) 5.75 (9%)	---	0.149c (10%) 0.270 (13%) 0.33c (6%)
Rn-219 (86) ↓	Actinon (gas)	4.0 s	6.42 (8%) 6.55 (11%) 6.82 (81%)	---	0.272 (9%) 0.401 (5%)
Po-215[†] (84) ↓	Actinium A	1.78 ms	7.38 (~100%)	0.74 (~0.0002%)	---
Pb-211 (82) ↓	Actinium B	36.1 m	---	0.29 (1.4%) 0.56 (9.4%) 1.39 (87.5%)	0.405 (3.4%) 0.427 (1.8%) 0.832 (3.4%)
Bi-211[†] (83) ↓	Actinium C	2.15 m	6.28 (16%) 6.62 (84%)	0.60 (0.28%)	0.351 (14%)
Tl-207 (81) ↓	Actinium C''	4.79 m	---	1.44 (99.8%)	0.897 (0.16%)
Pb-207 (82)	Actinium D	Stable	---	---	---

* Source: Lederer and Shirley (1978) and Shleien and Terpilak (1984).

** Half-life given in seconds (s), minutes (m), days (d), or years (y).

*** Intensities refer to percentage of disintegrations of the nuclide itself, not to the parent of the series.

† Approximately 1.4% of all Ac-227 emissions form an intermediate radioisotope, Fr-223 (22 m; β -emitter), before decaying to Ra-223. For Po-215, 0.00023% decays through At-215 (~ 0.1 msec; α -emitter), before forming Bi-211. For Bi-211, 0.28% decays through Po-211 (0.52 sec; β -emitter) to Pb-207.

Exhibit 3. Thorium Decay Series*

Radioisotope (atomic #)	Historical name	Half-life**	Major radiation energies (MeV) and intensities***		
			α	β	γ
Th-232 (90) ↓	Thorium	1.41 x 10 ¹⁰ y	3.95 (24%) 4.20 (75%)	---	---
Ra-228 (88) ↓	Mesothorium I	6.7 y	---	0.005 (100%)	---
Ac-228 (89) ↓	Mesothorium II	6.13 h	---	1.18 (35%) 1.75 (12%) 2.09 (12%)	0.34c (15%) 0.908 (25%) 0.96c (20%)
Th-228 (90) ↓	Radiothorium	1.910 y	5.34 (28%) 5.43 (71%)	---	0.084 (1.6%) 0.214 (0.3%)
Ra-224 (88) ↓	Thorium X	3.64 d	5.45 (6%) 5.68 (94%)	---	0.241 (3.7%)
Rn-220 (86) ↓	Thoron (gas)	55 s	6.29 (100%)	---	0.55 (0.07%)
Po-216 (84) ↓	Thorium A	0.15 s	6.78 (100%)	---	---
Pb-212 (82) ↓	Thorium B	10.64 h	---	0.346 (81%) 0.586 (14%)	0.239 (47%) 0.300 (3.2%)
Bi-212 [†] (83) ↓ ↓ (64%) (36%) ↓ ↓	Thorium C	60.6 m	6.05 (25%) 6.09 (10%)	1.55 (5%) 2.26 (55%) 0.98 (6%)	0.040 (2%) 0.727 (7%) 1.620 (1.8%)
Po-212 ↓ (84) ↓ ↓ ↓	Thorium C'	304 ns	8.78 (100%)	---	---
↓ Tl-208 ↓ (81) ↓ ↓	Thorium C''	3.01 m	---	1.28 (25%) 1.52 (21%) 1.80 (50%)	0.511 (23%) 0.583 (86%) 0.860 (12%) 2.614 (100%)
Pb-208 (82)	Thorium D	Stable	---	---	---

* Source: Lederer and Shirley (1978) and Shleien and Terpilak (1984).

** Half-life given in seconds (s), minutes (m), hours (h), days (d), or years (y).

*** Intensities refer to percentage of disintegrations of the nuclide itself, not to the parent of the series.

† Percentages in brackets are branching fractions.

Exhibit 4. Non-Series Naturally Occurring Radioisotopes*

Radioisotope (atomic #)	Name (elemental abundance)	Half-life**	Major radiation energies (MeV) and intensities***		
			α	β	γ
K-40 (19)	Potassium (0.0118%)	1.3×10^9 y	---	1.314 (89%)	1.46 (11%)
Rb-87 (37)	Rubidium (27.85%)	4.7×10^{10} y	---	0.274 (100%)	---
La-138 (57)	Lanthanum (0.089%)	1.1×10^{11} y	---	0.21 (100%)	0.81 (30%) 1.43 (70%)
Sm-147 (62)	Samarium (15.07%)	1.1×10^{11} y	2.2 (100%)	---	---
Lu-176 (71)	Lutetium (2.6%)	2.2×10^{10} y	---	0.43 (100%)	0.088 (15%) 0.202 (85%) 0.306 (95%)
Re-187 (75)	Rhenium (62.9%)	4.3×10^{10} y	---	0.043 (100%)	---

* Source: Lederer and Shirley (1978).

** Half-life given in years (y).

*** Intensities refer to percentage of disintegrations of the nuclide itself.

Distribution of Naturally Occurring Radionuclides:

In Rocks

The source of the primordial radionuclides is the earth's crust and underlying plastic mantle. Because of sedimentary processes sorting the products of weathering, several major types of sedimentary rock (shales, sandstones, and carbonate rocks) develop that differ significantly in radionuclide concentrations:

- Shales are composed of fine grains of clay (normally 35%), silt, or mud obtained from the breakdown of other rock. A significant fraction of shale contains potassium as a major constituent. All shale can adsorb the series radionuclides. The radionuclides also may be present in the cement that binds the shale together. Mean values for common shales are 2.7 percent potassium, 12 ppm thorium, and 3.7 ppm uranium (Cl66).
- Sandstones are composed of medium-sized grains, usually of quartz (SiO_2), that contain little in the way of radioactive impurities. Sandstone consisting of quartz grains bound with quartz cement is one of the least radioactive rocks. Such sandstone may contain less than 1 percent potassium, less than 2 ppm thorium, and less than 1 ppm uranium. Arkoses - sandstones that contain greater than 25 percent potassium-bearing feldspar - may contain upwards of two to three percent potassium. Clark et al. (Cl 66) report averages of 6.4 ppm thorium and 3.0 ppm uranium for modern beach sands. Thus, sandstone made from beach sand may be high in the series nuclides. In general, sandstones are low in both series and non-series radionuclides.
- Carbonate rocks (limestone and dolomites), derived by chemical precipitation from water or by accumulation of shells, bones, and teeth of organisms, are low in radionuclide content. Still the intergranular spaces contain a variety of elements characteristic of the sea water where most radionuclides may be deposited. Carbonate rocks are low in potassium due to the high solubility of potassium salts, and are low in thorium because it is highly depleted in sea water. Uranium becomes fixed by the reducing conditions prevailing in the decaying organic matter at the sea bottom and thus becomes incorporated in the carbonate rocks.

Exhibit 5 provides summary data on the average concentrations of K-40, Rb-87, Th-232, and U-238 in various types of rocks and sediments.

In Soil

Radionuclides in soil are derived from source rock. In most cases, soil activity concentrations are often less than source rock concentrations due to water leaching, dilution as a result of the soil's increased porosity, and the addition of organic matter and water. In addition, biochemical processes taking place during soil development also tend to reduce the radionuclide concentrations in comparison to the source rock. However, in some cases, soil radioactivity may be augmented by sorption or precipitation of radionuclides from incoming water, by redistribution of wind-blown soils, or by activities such as adding fertilizer or importing top soil to a location. Exhibit 5 provides summary data on average concentrations of K-40, Rb-87, Th-232, and U-238 in soil.

In the Hydrosphere

The concentrations of naturally occurring radionuclides in water are several orders of magnitude less than those in rocks and soils. Potassium-40 is one of the more abundant radionuclides in most water systems. For uranium and thorium series isotopes, there is a shift away from equilibrium between parent radionuclides and progeny. Concentrations of uranium and Rn-222 daughters are frequently observed to be elevated compared to Ra-226 levels.

Exhibit 5. Concentrations of Naturally Occurring Radioisotopes in Rock and Soil^a

Type of Rock or Soil	Potassium-40		Rubidium-87		Thorium-232		Uranium-238	
	Percent	pCi/g	ppm	pCi/g ^b	ppm	pCi/g ^b	ppm	pCi/g ^c
Igneous rocks								
Basalt (crustal average)	0.8	8	40	0.8	3-4	0.3-0.4	0.5-1	0.2-0.3
Mafic ^d	0.3-1.1	2-11	10-50	0.03-1	1.6-2.7	0.2-0.3	0.5-0.9	0.2-0.3
Sialic ^d	4.5	30-41	170-200	4-5	16-20	1.6-2.2	3.9-4.7	1.4-1.6
Granite (crustal average)	> 4	> 30	170-200	4-5	17	1.9	3	1.1
Sedimentary rocks								
Shale sandstones	2.7	22	120*	3	12	1.4	3.7	1.1
Clean quartz	< 1	< 8	< 40*	< 1	< 2	< 0.2	< 1	< 0.3
Dirty quartz	2.7	117	90?	2.7	3-67	0.3-0.7	2-37	< 1.1
Arkose	2-3	16-24	80-120	2	2.7	< 0.2	1-2.7	0.3-0.7?
Beach sands (unconsolidated)	< 1	< 8	< 40	< 1	6	0.7	3	1.1
Carbonate rocks	0.3	2	10	0.2	2	0.2	2	0.7
Continental upper crust Average ^e	2.8	23	112	3	10.7	1.2	2.8	1.0
Soils ^a	1.5	11	65	1	9	1.0	1.8	1.8
Soils ^b	---	3-19	---	3.5	---	0.2-1.4	---	0.2-1.4

a. References cited in text unless otherwise noted; single values are average; values estimated in the absence of reference are followed by a question mark.
 b. To obtain series equilibrium alpha, beta, or approximate gamma activity (excluding bremsstrahlung and X rays), multiply by 6, 4, or 3, respectively.
 c. To obtain series equilibrium alpha, beta, or approximate gamma activity (excluding bremsstrahlung or X rays), multiply by 8, 6, or 3, respectively.
 d. From CI66 for potassium and rubidium, the range of values for rocks within the class is given; for thorium and uranium, the median and mean values are given, respectively.
 e. Estimated by application of crustal abundance ratio with respect to potassium.
 f. From Ta85.
 g. In-situ gamma spectral measurements at 200 locations by Lowder et al. (1964).
 h. Potassium, thorium, and uranium from Annex, 1, UN82; rubidium from NCRP (1976).

Elevated Rn-222 concentrations, ranging from several hundreds to several thousands of pCi/L, are often found in ground water samples, whereas Ra-226 concentrations in the same sample are typically a factor of 1000 lower. Radium and thorium isotopes tend to concentrate in bottom sediments.

Radionuclide concentrations of fresh water bodies and urban water supplies vary widely depending on local geology, hydrology, geochemistry, and radionuclide soil concentrations. Sea water, on the other hand, exhibits a rather narrow range of activity concentrations (Ko62, Ch86).

In the Atmosphere

The level of radioactivity in air and soil water is due primarily to Rn-222, Rn-220, Rn-219, and their decay products. Approximately 35 percent of the Rn-222 produced from Ra-226 in soil emanates into soil pore spaces, resulting in a Rn-222 concentration of about 500 pCi/L of pore fluid per ppm of U-238 in equilibrium with Ra-226 (NCRP87b). At a soil concentration of 1-2 ppm of U-238, Rn-222 levels in soil pores range 10^2 to 10^3 pCi/L, several orders of magnitude greater than typical atmospheric levels. Atmospheric radon concentrations depend on the amount of radon exhaled by the soil and on atmospheric factors that control its upward dispersion. Rn-222 measurements outdoor show that the mean concentrations can range from 100 to 1100 pCi/m³ (NCRP87b). Exhibit 6 summarizes typical concentrations of naturally occurring radionuclides in the atmosphere.

In the Biosphere

Potassium-40 is the most abundant radionuclide in the biosphere. Concentrations of other naturally occurring radionuclides in plants and animals are highly variable and are almost never in equilibrium (NCRP76). For example, Ra-226 is preferentially taken up by plants relative to U-238 or U-234. In general, activity concentrations in plants range from 1 to 50 pCi/g for ⁴⁰K, from 0.01 to 10 pCi/g for Po-210, and are about 0.1 pCi/g for Rb-87 (NCRP76), as shown in Exhibit 7.

Group #2: Cosmic Radiation and Cosmogenic Radionuclides

Cosmic radiation consists of primary charged and neutral particles that bombard the earth's atmosphere and the secondary particles (e.g., H-3 and C-14) generated by the primary particles in the earth's atmosphere. Primary cosmic radiation, produced by supernovas and solar flares, is composed of approximately 87 percent photons, 11 percent alpha particles, 1 percent heavier nuclei, and 1 percent electrons with energies up to at least 10^{20} eV (average energy is 10^8 to 10^{11} eV). Secondary cosmic particles are produced by a variety of spallation and neutron activation reactions, mostly with the nuclei of argon, nitrogen, and oxygen.

Cosmic radiation increases with altitude as the mass of the atmosphere decreases. Cosmic flux density is least near the geomagnetic equator and increases with latitude. At sea level, the flux density is about 10% lower at the equator than at high latitudes. Energetic solar flares generate large numbers of photons that can penetrate the earth's magnetic field and add to the cosmic ray flux density incident on the atmosphere. These bursts seldom produce significant effects at ground level. There is evidence for an 11-year cycle in mean solar activity that produces a modulation of the cosmic radiation reaching the earth's atmosphere. At ground altitudes, the effect is about 10 percent.

Exhibit 8 shows the typical environmental radiation field at 1 meter above sea level due to cosmic and terrestrial radionuclides.

A total of 20 radionuclides are produced by cosmic rays in the earth's atmosphere. From the point of view of radiation measurements and doses, only carbon-14 (C-14) and, to a lesser extent, tritium (H-3) are worth considering.

Exhibit 6. Radionuclides In The Atmosphere*

Radionuclide	Surface air content	
	Typical range (pCi/m ³)	Mean value (pCi/m ³)
Uranium series:		
Rn-222		
Pb-214	20 - 500	120
Bi-214	0 - 500	100
Pb-210	0 - 500	100
Po-210	0.003 - 0.03	0.01
	--	0.003
Thorium series:		
Rn-220		
Pb-212	--	100
	0.5 - 10	2
Others:		
Kr-85	--	17
Be-7	0.02 - 0.20	0.06

* Source: NCRP (1976): Table 2-8.

Exhibit 7. Total Natural Radioactivity In Plants*

Radiation	Concentration (pCi/g gross weight)	Source
Gross alpha	0.14 - 3.1	mainly as Po-210; other U + Th series nuclides
Gross beta	7.8 - 123	mainly as K-40; Pb-210; Bi-210; other U + Th series nuclides
K-40	1 - 50	--
Rb-87	~0.1	--
Po-210	0.01 - 10	--

* Source: NCRP (1976): Table 2-9b.

Exhibit 8. Typical Environmental Radiation Field (One Meter Height)*

Radiation	Energy (MeV)	Source	Absorbed dose rate in free air (microrad/hr)
alpha	1 - 9	radon (atm)	2.7
beta	0.1 - 2 0.1 - 2 2 - 200	radon (atm) K, U, Th, Sr (soil) cosmic rays	0.2 2.5 0.7
gamma	<2.4 <1.5 <2.4 <2.6 <0.8	radon (atm) K (soil) U (soil) Th (soil) Cs + other fallout (soil)	0.2 2.0 1.0 2.4 0.3
neutron proton muons	0.1 - 100 10 - 2,000 100 - 30,000	cosmic rays	0.1
			Total: 14.5

* Source: NCRP (1976): Table 2-10.

Tritium (H3)

Tritium, a radioactive isotope of hydrogen, is a beta emitter (average energy 5.69 keV) with a radioactive half-life of 12.3 years. It occurs naturally in the surface waters of the earth as a product of the atmospheric interaction of high-energy cosmic rays with nitrogen and oxygen gases (UN72, NCRP79). Its annual production rate is approximately 2 megacuries (MCi), resulting in a steady-state inventory of about 30 MCi in the biosphere. Since 1954, large amounts of manmade tritium have been released into the environment primarily from nuclear weapons testings, discharges from nuclear power plants (Exhibit 10), and some nuclear weapons production plants. Tritium is used as a radioactive luminizing material in consumer products, such as watches, clocks, and emergency signs, and as a component of nuclear weapons.

Prior to the injection into the biosphere from nuclear tests, levels of H-3 in waters of the mid-latitude regions of the earth were in the range of 6 to 24 pCi/L. The amount of tritium added to the global inventory as a result of nuclear weapons testing is discussed under the next section on manmade radionuclides. About 90% of natural H-3 resides in the hydrosphere, 10% in the stratosphere, and only 0.1% in the troposphere. The low inventory of H-3 in the troposphere is due to the fact that tritium in the form of HTO is rapidly washed out by rain, with an estimated residence time of between 20 to 40 days.

Carbon-14 (C-14)

Carbon-14 is one of the three isotopes of carbon: C-12 (99.8%), C-13 (1.1%), and C-14 (0.1%). It is a pure beta-emitting radionuclide (average energy 50 keV) with a radioactive half-life of 5,730 years. Natural C-14 is produced in the upper atmosphere by interaction of cosmic-ray neutrons with nitrogen. Its production rate is not accurately known, but may correspond to about 0.03 MCi per year with a steady-state inventory of approximately 280 MCi (UN72). Similar to tritium, C-14 has been produced in significant quantities by nuclear weapons testing and discharges from nuclear power plants (see the section on manmade radionuclides).

As an isotope of carbon, C-14 is involved with all biological and geochemical processes on earth. It is present in the atmosphere as carbon dioxide, in the terrestrial biosphere as incorporated carbon, and in surface waters as dissolved bicarbonates. The concentration of C-14 in the environment varies widely. At present, the United Nations assumes a specific activity of 6.1 pCi/g in the terrestrial biosphere (UN 72).

Group #3: Ubiquitous Manmade Radionuclides

Manmade radioisotopes that are widely distributed in the environment are due primarily to releases from nuclear weapons testing and nuclear power facilities. Exhibits 9 and 10 list some of the important radionuclides produced by these processes.

Radionuclides released during nuclear weapons testing: Since the first test of a nuclear weapon at Alamogordo, New Mexico, in 1945, approximately 450 additional nuclear weapons have been detonated in the atmosphere. These detonations resulted in the production and global dispersal of several millions of curies of radioactive fission and activation products, transuranic elements, and unfissioned uranium and plutonium isotopes.

These detonations also significantly increased natural concentrations of H-3 and C-14. Between 1,900 to 8,000 MCi of H-3 were added to the northern hemisphere by nuclear weapons testing through 1963 (Er65, Mi71). As a result, average concentrations of H-3 in surface waters in the U.S. rose from 3 to 16 pCi/L to about 4,000 pCi/L in 1963 (Be73). Today, tritium concentrations due to fallout H-3 have decreased below the level due to natural H-3 (NCRP79). By the end of 1962, nuclear testing had increased the atmospheric concentration of C-14 to about twice its pre-1950 concentration of 6 pCi/g. Because of exchange with the ocean and to a lesser extent the biosphere, C-14 concentrations in the atmosphere due to weapons testing dropped to about 3 pCi/g by the end of 1970 (NCRP87b). The increase in C-14 concentrations in the ocean has been greatest in the surface waters since C-14 has a residence time of three to eight years in the mixing layers before it is transferred below the thermocline. Because it takes a few thousand years before C-14 reaches the ocean floor, there is no increase in C-14 concentrations for deep ocean sediments.

Strontium-90 and Cs-137 are two of the most important fission products that were widely distributed in near-surface soils because of the weapons testing. Measurable concentrations of Sr-90 and Cs-137 in soil exist today. These concentrations are distributed almost exclusively in the upper 15 cm of soil and decrease roughly exponentially with depth.

Radionuclides released from nuclear power stations: Releases of radionuclides produced by nuclear fission in boiling water reactors (BWRs) and in pressurized water reactors (PWRs) occur because of periodic fuel failure, defects, or corrosion that results in transfer of some fission and activation products into the reactor coolant. In PWRs, the primary coolant is in a sealed loop that is continually purged for control of chemical composition and purification. Gaseous wastes released in the process are held in tanks for between 30 to 120 days to allow short-lived nuclides to decay prior to release. Other gaseous effluent streams originate from the condenser exhaust on the steam circuit, secondary coolant blowdown, reactor building ventilation (including containment purges), and turbine plus ancillary building ventilation (UN82). In BWRs, the main condenser air-ejector system continuously removes non-condensable gases from the steam flow. This is the main source of noble gases released with the gaseous waste stream. Secondary pathways include the purging system for the turbine gland seals, the condenser mechanical vacuum pump, and any process fluid leaks to ventilated buildings.

Radionuclides released to the atmosphere include noble gases (argon, krypton, and xenon), C-14, tritium, iodines, and particulates. Radionuclides discharged in liquid effluents include tritium, fission products, and activated corrosion products. Exhibit 10 lists the nuclide composition of typical liquid and gaseous effluents for PWRs and BWRs in the U.S. Compositions often vary depending on waste treatment methods employed, the age and condition of the plant, etc. Release rates are not listed for the nuclides since these data vary greatly from plant to plant. Environmental monitoring programs typically show that the nuclides in the effluents are not readily detectable in the environment except near the point of release.

Exhibit 9. Ubiquitous Manmade Radioisotopes*

Radioisotope (atomic #)	Name (Origin)†	Half-life**	Major radiation energies (MeV) and intensities***		
			α	β	γ
H-3 (1)	Tritium (NE, NF)	12.3 y	---	0.0186 (100%)	---
C-14 (6)	Carbon (NE, FF)	5730 y	---	0.156 (100%)	---
Mn-54 (25)	Manganese (NE)	303 d	---	---	0.835 (100%)
Fe-55 (26)	Iron (NE)	2.6 y	---	---	0.23 (0.004%)
Co-60 (27)	Cobalt (NE, NF)	5.26 y	---	1.48 (0.12%) 0.314 (99%)	1.17 (100%) 1.33 (100%)
Zn-65 (30)	Zinc (NE, NF)	245 d	---	$\beta+$: 0.327 (1.4%)	0.511 (3.4%) 1.12 (49%)
Kr-85 (36)	Krypton (NE, NF)	10.76 y	---	0.173 (0.4%) 0.687 (99.6%)	0.514 (0.4%)
Sr-90 (38)- Y-90 (39)	Strontium - Yttrium (NE, NF)	28 y (Sr) 64 h (Y)	--	0.546 (100% Sr) 2.27 (100% Y)	---
Zr-95 (40)	Zirconium (NE)	65.5 d	---	0.366 (55%) 0.398 (44%)	0.724 (49%) 0.756 (49%)
Nb-95 (41)	Niobium (NE)	35 d	---	0.160 (99.9%)	0.765 (100%)
Ru-106 (44) - Rh-106 (45)	Ruthenium - Rhenium (NE, NF)	368 d (Ru) 30 s (Rh)	---	0.039 (100% Ru) 3.54 (79% Rh)	0.512 (21%) 0.622 (11%) 1.05 (1.5%)
Sb-125 (51) - Te-125m (52)	Antimony - Tellurium (NE)	2.77 y (Sb) 58 d (Te)	---	0.61 (14% Sb)	0.153 (82% Te) 0.176 (6% Sb) 0.270 (25% Te) 0.427 (10% Sb) 0.599 (24% Sb) 0.634 (11% Sb) 0.66 (3% Sb) 0.92-1.14 (36% Te) 1.22 (67% Te) 2.09 (4% Te)
I-129 (53)	Iodine (NF)	1.7 x 10 ⁷ y	---	0.150 (100%)	0.040 (9%)
Cs-134 (55)	Cesium (NE, NF)	2.05 y	---	0.662 (100%)	0.57 (23%) 0.61 (98%) 0.796 (99%)

Exhibit 9 - Continued¹

Radioisotope (atomic #)	Name (Origin) [†]	Half-life ^{**}	Major radiation energies (MeV) and intensities ^{***}		
			α	β	γ
Cs-137 (55) - Ba-137m (56)	Cesium - Barium (NE, NF)	30 y (Cs) 2.55 m (Ba)	---	0.514 (95% Cs) 1.176 (5% Cs)	0.428 (30% Ba) 0.463 (11% Ba) 0.601 (18% Ba) 0.836 (12% Ba) 0.662 (89% Ba)
Ce-144 (58) - Pr-144 (59)	Cerium - Praseodymium (NE)	284 d (Ce) 17.3 m (Pr)	---	0.31 (76% Ce) 2.99 (98% Pr)	0.080 (2% Ce) 0.134 (11% Ce) 0.695 (1.5% Pr) 1.487 (0.3% Pr) 2.186 (0.7%)
Pu-238 (94)	Plutonium (SNAP, NE)	87 y	5.50 (72%) 5.46 (28%)	---	0.145 (2%)
Pu-239 (94)	Plutonium (NE, NF)	2.439 x 10 ⁴ y	5.155 (73%) 5.143 (15%) 5.105 (12%)	---	0.039 (0.007%) 0.052 (0.020%) 0.129 (0.005%) 0.375 (0.0012%)
Pu-240 (94)	Plutonium (NE, NF)	6580 y	5.1683 (76%) 5.1238 (24%)	---	---
Pu-241 (94) - Am-241 (95)	Plutonium - Americium (NE, NF)	13 d (Pu) 458 y (Am)	4.90 (0.002% Pu) 4.85 (0.003% Pu) 5.3884 (1.6% Am) 5.443 (12.8% Am) 5.486 (85% Am)	---	0.0264 (2.5% Am) 0.0595 (36% Am)

¹ Source: Lederer and Shirley (1978) and NCRP (1976).

^{**} Half-life given in minutes (m), hours (h), days (d), or years (y).

^{***} Intensities refer to percentage of disintegrations of the nuclide itself.

[†] "NE" = Nuclear explosions; "NF" = Nuclear facilities; "SNAP" = SNAP-9a (System for Nuclear Auxiliary Power) which was a satellite which dispersed 1 kg of Pu-238 in the earth's atmosphere when it burned up upon re-entry; and "FF" = Fossil fuel power plants and other industries.

Exhibit 10. Radioisotopes in Nuclear Reactor Effluent*

Effluent Type	Reactor Type	
	PWR	BWR
Gaseous	<p>H-3, K-40, Ar-41, Co-57, Co-58, Co-60, Ni-63, Br-82, Kr-85, Kr-85m, Kr-87, Kr-88, Sr-90, Nb-95, Zr-95, Zr-97, Tc-99m, I-132, I-133, Xe-133m, Cs-134, I-135, Xe-135, Xe-135m, Cs-137, Xe-138, Ce-139, Ce-143, Ce-144, Bi-214, Rn-222, Ra-226, Th-228, Th-232</p>	<p>H-3, N-13, Ne-24, Ar-41, Cr-51, Mn-54, Mn-56, Co-58, Fe-59, Co-60, Zn-65, Br-82, Kr-83m, Kr-85, Kr-85m, Kr-87, Kr-88, Kr-89, Sr-89, Sr-90, Sr-90, Sr-91, Y-91, Nb-95, Mo-99, Ru-103, Rh-106, Ag-110m, I-131, Xe-131m, I-132, Xe-133, Xe-133m, I-132, I-133, Xe-133, Xe-133m, Cs-134, I-135, Xe-135, Xe-135m, Cs-136, Cs-137, Xe-138, Ba-139, Xe-139, Ba-140, La-140, Ce-141, Ce-144, Hg-203</p>
Liquid	<p>H-3, Be-7, Na-24, K-40, Ar-41, Cr-51, Mn-54, Fe-55, Mn-56, Co-57, Co-58, Fe-59, Co-60, Ni-63, Cu-64, Zn-65, Zn-69m, Se-75, As-76, Ge-77, Br-82, Kr-88, Rb-88, Y-88, Rb-89, Sr-89, Mo-90, Sr-90, Y-91, Y-91m, Sr-92, Y-92, Nb-94, Nb-95, Zr-95, Nb-97, Zr-97, Mo-99, Tc-99m, Ru-103, Ru-105, Ru-106, Ag-108m, Cd-109, Ag-110m, Sn-113, Cd-115, In-115m, Sn-117m, Sb-112, Sb-124, Sb-125, Sb-127, I-131, Xe-131m, I-132, Te-132, Ba-133, Cd-133m, I-133, Xe-133, Xe-133m, Cs-134, I-134, I-135, Xe-135, Xe-135m, Cs-136, Cs-137, Cs-138, Ba-139, Ce-139, Ba-140, La-140, Ce-141, Ce-144, Pr-144, W-187, Hg-203, Bi-214, Pb-214, Ra-226, Th-228, Np-239</p>	<p>F-18, Ne-24, P-32, Cl-38, Cr-51, Mn-54, Fe-55, Mn-54, Mn-56, Co-57, Co-58, Fe-59, Co-60, Cu-64, Zn-65m, Zn-69m, As-76, Br-84, Kr-85, Kr-85m, Kr-88, Sr-89, Sr-90, Y-91m, Sr-90, Y-91m, Sr-92, Y-92, Nb-95, Zr-95, Nb-97, Mo-99, Tc-99m, Ru-103, Tc-104, Rh-103, Tc-104, Rh-105, Ru-105, Rh-106, Ru-106, Ag-110m, Ag-111, Sb-124, Sb-125, Te-129, Te-129m, I-131, Xe-131m, Te-132, I-133, Xe-133, Xe-133m, Cs-134, I-135, Xe-135, Xe-135m, Cs-136, Cs-137, Cs-138, Ba-139, Ba-140, La-140, Ce-141, La-142, Ce-144, Hg-203, Np-239</p>

* Radioisotope composition of gaseous and liquid effluent from Pressurized Water Reactors (PWRs) and Boiling Water Reactors (BWRs) in the United States for 1979 (NCRP 1987a).

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