

## Web Supplement 28.6.7

### 28.6.7 Radioactivity

Radioactivity is a form of energy emitted by radioactive elements (radioisotopes or radionuclides). Some of the radionuclides in the ocean are natural and others are the result of human activities. The most abundant of the naturally occurring radionuclides are listed in Table W28.5. Some are primordial, having been present at Earth's formation.

**Table W28.5** Main Naturally Occurring Radionuclides in the Oceans, Their Concentrations, and Inventories.

Radionuclide	Half Life (y)	Concentration (Bq L <sup>-1</sup> )	Approximate Inventory (TBq)
<b>Primordial:</b>			
<sup>40</sup> K	1.26 × 10 <sup>9</sup>	12	1.6 × 10 <sup>10</sup>
<sup>87</sup> Rb	4.88 × 10 <sup>10</sup>	0.11	1.5 × 10 <sup>8</sup>
<sup>234</sup> U	2.47 × 10 <sup>5</sup>	0.046	6.3 × 10 <sup>7</sup>
<sup>238</sup> U	4.47 × 10 <sup>9</sup>	0.040	5.5 × 10 <sup>7</sup>
<sup>226</sup> Ra	1.62 × 10 <sup>3</sup>	0.0035	4.8 × 10 <sup>6</sup>
<sup>210</sup> Pb	22.3	0.0030	4.1 × 10 <sup>6</sup>
<sup>235</sup> U	7.04 × 10 <sup>8</sup>	0.0022	3.0 × 10 <sup>6</sup>
<sup>210</sup> Po	138 days	0.0020	2.7 × 10 <sup>6</sup>
<b>Cosmogenic:</b>			
<sup>14</sup> C	5.73 × 10 <sup>3</sup>	0.006 <sup>a</sup>	8.0 × 10 <sup>6</sup>
<sup>3</sup> H	12.3	0.1 <sup>a</sup>	8.5 × 10 <sup>5</sup>

<sup>a</sup>Surface water values.

Source: After Pentreath, R. J. (1988). *Radionuclides in the aquatic environment. In Radionuclides in the Food Chain*, G. Silinii, ed. Springer-Verlag, pp. 99–119.

**Table W28.6** Properties of Some Ionizing Radiation.

Type	Relative Mass Ratio	Relative Energy	Relative Penetration	Relative Ionization
$\alpha$	7380	Moderate	1	$10^4$
$\beta$	1	Low to high	$10^2$	$10^2$
$\gamma$	0	High	$10^3$ – $10^4$	1

Others are currently being produced in the atmosphere by spallation reactions, which involve the bombardment of atoms with cosmic radiation, and on Earth by radioactive decay of chemicals present in the crust and hydrosphere. The total inventory of these naturally occurring radionuclides is about 16 million PBq, most of which is primordial  $^{40}\text{K}$  present in rocks.<sup>1</sup>

Radionuclides can present a health threat to humans and marine organisms because of the ionizing ability of the emitted radiation. As shown in Table W28.6,  $\gamma$  radiation is an effective penetrator and, thus, can induce harm just through exposure. The same is true for  $\beta$  particles. In comparison,  $\alpha$  particles have relatively low energy and, thus, are not capable of deep penetration. On the other hand, ingestion of  $\alpha$  particles is very harmful as they are very effective ionizing agents. The major pathway by which marine organisms and humans are exposed to radionuclides comes from consumption of fish and shellfish due to bioaccumulation of  $^{210}\text{Po}$  and  $^{210}\text{Pb}$ . Most of the  $^{210}\text{Po}$  and  $^{210}\text{Pb}$  in the ocean is natural, but as described next, human activities have increased their inputs to the coastal ocean.

The anthropogenic sources of radionuclides to the marine environment can be divided into two categories: (1) emissions of artificially generated isotopes and (2) mobilization of naturally occurring isotopes. These sources are summarized in Table W28.7. Emissions of the artificially generated isotopes result from activities of the nuclear industries. They include weapons testing, accidents, dumping, and discharges from power and fuel reprocessing plants, and well as disposal of medical wastes. Humans mobilize the naturally occurring isotopes during mineral mining, oil drilling, and fossil-fuel burning. The radionuclides associated with each of these activities are listed in Table W28.8 and their half-lives are given in Table W28.9. Note that the nuclear industry has generated a set of radionuclides quite different from those listed in Table W28.5. Human activities have elevated the concentrations of these isotopes in the marine environment. In the case, of the  $\beta$  and  $\gamma$  emitters, the human input has swamped the natural inventory and, hence, these isotopes are referred to as *artificial radionuclides*. The most abundant are  $^{137}\text{Cs}$ ,  $^{90}\text{Sr}$ ,  $^3\text{H}$ ,  $^{14}\text{C}$ , and  $^{241}\text{Pu}$ . Their half-lives range from days ( $^{131}\text{I}$  and  $^{60}\text{Co}$ ) up to millions of years ( $^{129}\text{I}$  and  $^{237}\text{Np}$ ).

<sup>1</sup> 1 Bq (becquerel) = 1 disintegration per second = 0.0167 disintegrations per minute.  $10^{15}\text{Bq} = 10^3\text{TBq} = 1\text{PBq}$ .

**Table W28.7** Sources of Marine Artificial Radionuclides.

<b>Nuclear Industry</b>	
Nuclear weapons	Fallout from testing Disposal of demilitarized units
Nuclear reactors	Dumping of liquid and solid wastes including spent fuels and reactor parts
Nuclear reprocessing and power plants	Discharge of liquid wastes
<b>Nonuclear Industry</b>	
Land-based discharges	Phosphorite mining, concrete and ceramics production Medical testing Burning of fossil fuels (coal and oil in power plants)
Offshore	Produced waters and drill muds from oil wells

**Table W28.8** Types of Radionuclides in Anthropogenic Emissions Listed by Source and Emission Type.

Source	Emission Type		
	Alpha	Beta	Gamma
Nuclear testing	$^{239}\text{Pu}$ , $^{240}\text{Pu}$ , $^{241}\text{Am}$ (all minor)	$^3\text{H}$ , $^{90}\text{Sr}$ , $^{137}\text{Cs}$ , $^{125}\text{Sb}$ , $^{241}\text{Pu}$	$^{137}\text{Cs}$ , $^{55}\text{Fe}$ and $^{14}\text{C}$
Disposal from nuclear industries	$^{241}\text{Am}$ , $^{238}\text{Pu}$	$^3\text{H}$ , $^{90}\text{Sr}$ , $^{137}\text{Cs}$ , $^{60}\text{Co}$	$^{134}\text{Cs}$ , $^{137}\text{Cs}$ , $^{55}\text{Fe}$ , $^{58}\text{Co}$ , $^{125}\text{I}$ and $^{14}\text{C}$
Discharges	$^{239}\text{Pu}$ , $^{240}\text{Pu}$ , $^{241}\text{Am}$	$^{137}\text{Cs}$ , $^{99}\text{Tc}$ , $^{129}\text{I}$	
Phosphate fertilizers <sup>a</sup>	$^{226}\text{Ra}$ , $^{232}\text{Th}$ , $^{238}\text{U}$ , $^{210}\text{Pb}$ , $^{210}\text{Po}$	$^{40}\text{K}$ , $^{210}\text{Pb}$	
Medical testing		$^{99}\text{Tc}$ , $^{131}\text{I}$	
Produced waters <sup>a</sup>	$^{210}\text{Pb}$ , $^{210}\text{Po}$ , $^{226}\text{Ra}$ ,	$^{228}\text{Ra}$	
Fossil fuel burning <sup>a</sup>	$^{232}\text{Th}$ , $^{238}\text{U}$	$^{40}\text{K}$	
Natural gas burning <sup>a</sup>	$^{226}\text{Ra}$ and $^{222}\text{Rn}$		

<sup>a</sup>Naturally occurring radionuclides mobilized by anthropogenic activities.

**Table W28.9** Half-Lives (y) of Artificial Radionuclides.

$^3\text{H}$	12.33	$^{125}\text{Sb}$	2.76	$^{239}\text{Pu}$	24,100	$^{131}\text{I}$	8 d
$^{14}\text{C}$	5730	$^{129}\text{I}$	15,700,000	$^{240}\text{Pu}$	6500	$^{125}\text{I}$	59 d
$^{55}\text{Fe}$	2.73	$^{137}\text{Cs}$	30.7	$^{241}\text{Pu}$	14.4	$^{58}\text{Co}$	72 d
$^{90}\text{Sr}$	28.78	$^{237}\text{Np}$	2,200,000	$^{242}\text{Pu}$	375,000	$^{60}\text{Co}$	5.3
$^{99}\text{Tc}$	211,100	$^{238}\text{Pu}$	87.7	$^{241}\text{Am}$	433		

The inventories of anthropogenic emissions are provided in Table W28.10 by source. In 2000, the total oceanic inventories of the most abundant artificial radionuclides was 60,000 PBq of  $^3\text{H}$ , 300 PBq of  $^{137}\text{Cs}$ , 189 PBq of  $^{90}\text{Sr}$ , 54 PBq of  $^{239+240+241}\text{Pu}$ , and 5.1 PBq of  $^{241}\text{Am}$ . The global dispersion and deposition of debris from atmospheric nuclear weapons is by far the largest source of artificial radioactivity to the terrestrial and marine environment. Nevertheless, this caused only a slight increase in the total global inventory of radioactivity, with the major contributor being tritium ( $^3\text{H}$ ). Evidence for significant radionuclide exposures to humans and marine life seems to be limited to a few areas where inputs have been geographically concentrated, such as around the discharge outfalls serving the nuclear fuel reprocessing plants.

The fate of radionuclides in the marine environment is similar to that of the stable isotopes, being dependent on chemical speciation, including redox state, solubility, and tendency to form complex ions. For example,  $^{238,239,240}\text{Pu}$  and  $^{241}\text{Am}$  are particle reactive, whereas  $^{137}\text{Cs}$ ,  $^{99}\text{Tc}$ , and  $^{129}\text{I}$  are not. The particle-reactive radionuclides are strongly concentrated in marine sediments as shown in Table W28.11. Some, such as  $^{210}\text{Po}$ , also have high bioaccumulation factors in fish and shellfish. The artificial radionuclides that exhibit conservative behavior, such as  $^3\text{H}$ ,  $^{90}\text{Sr}$ , and  $^{137}\text{Cs}$ , have been used by oceanographers as tracers of ocean currents because most of their injection occurred over a very short period as a result of above-ground nuclear-bomb testing.

### **Fallout**

Above-ground nuclear weapons testing was conducted from 1945 through 1980 (Figure W28.5). A total of 543 atmospheric detonations took place mostly in the equatorial Pacific on remote oceanic islands and in the polar north. 159 and 240 Mt (respectively) of radioactive debris was injected into the upper troposphere and lower stratosphere, composing 90% of the total injected from weapons testing. The period of most intense testing took place between 1957 and 1963 (259 tests). Since 1980, all testing has been confined to underground sites. Some underwater testing was conducted by the former USSR.

Most of the radionuclide fallout was in the form of sub-micron-sized particles that solubilized shortly after deposition on the sea surface. The majority thus delivered were  $^{137}\text{Cs}$ ,  $^{90}\text{Sr}$ , and  $^{241}\text{Pu}$  (Table W28.10a). Some of the particles settled gravitationally and others were washed out by incorporation into rain droplets. The transuranium

**Table W28.10(a)** Inventories of the Long-Lived Artificial Radionuclides: (a) Nuclear Weapons Testing and Breakup of the SNAP-9A Satellite.

Nuclide	Half-life (years)	Production (TBq)	Deposited Inventory (TBq)		
			Northern Hemisphere	Southern Hemisphere	North Atlantic Ocean
$^3\text{H}$	$1.2 \times 10^1$	$2.4 \times 10^8$	$6.0 \times 10^7$	$1.5 \times 10^7$	$1.3 \times 10^7$
$^{90}\text{Sr}$	$2.9 \times 10^1$	$6.0 \times 10^5$	$3.0 \times 10^5$	$9.5 \times 10^4$	$6.4 \times 10^4$
$^{137}\text{Cs}$	$3.0 \times 10^1$	$9.6 \times 10^5$	$4.7 \times 10^5$	$1.5 \times 10^5$	$9.4 \times 10^4$
$^{238}\text{Pu}^a$	$8.8 \times 10^1$	$3.3 \times 10^2$	$3.2 \times 10^2$	$4.8 \times 10^2$	$6.4 \times 10^1$
$^{239+240}\text{Pu}$	$2.4 \times 10^4^b$	$1.3 \times 10^4$	$1.0 \times 10^4$	$2.6 \times 10^3$	$2.0 \times 10^3$
$^{241}\text{Pu}$	$1.4 \times 10^1$	$1.7 \times 10^{5c}$	$5.0 \times 10^4$	$1.2 \times 10^4$	$1.0 \times 10^4$
$^{241}\text{Am}$	$4.3 \times 10^{2d}$	$5.5 \times 10^3$	$2.8 \times 10^3$	$7.0 \times 10^2$	$5.6 \times 10^2$

<sup>a</sup>Includes SNAP-9A satellite<sup>b</sup> $^{239}\text{Pu}$  included<sup>c</sup>Assumed input in 1963<sup>d</sup>From ultimate decay of  $^{241}\text{Pu}$ 

See Table W28.10d for data sources.

**Table W28.10(b)** Inventories of the Long-Lived Artificial Radionuclides: Discharges from the Nuclear Fuel Reprocessing Plants at Sellafield and La Hague.

Radionuclide	Sellafield (to 1986)		La Hague (to 1985)	
	Total	Cumulative Inventory	Total	Cumulative Inventory
$^3\text{H}$	25,230	16,735	10,190	8470
$^{106}\text{Ru}$	27,545	285	4,905	800
$^{137}\text{Cs}$	41,080	33,480	940	760
$^{90}\text{Sr}$	6,120	4,690	755	675
$^{239/240}\text{Pu}$	680	680	—	—
$^{238}\text{Pu} + ^{239/240}\text{Pu}$	—	—	3	3
$^{241}\text{Am}$	535	815 <sup>e</sup>	—	—
$^{125}\text{Sb}$	170 <sup>f</sup>	65 <sup>f</sup>	1,000	795
$^{99}\text{Tc}$	305 <sup>f</sup>	305 <sup>f</sup>	—	—

<sup>e</sup>Includes grow-in from  $^{241}\text{Pu}$ <sup>f</sup>Since 1978

See Table W28.10d for data sources.

**Table W28.10(c)** Inventories of the Long-Lived Artificial Radionuclides: Emissions from the Chernobyl Nuclear Reactor Accident in 1986.

Nuclide	Quantity (TBq)	Nuclide	Quantity (TBq)
<sup>90</sup> Sr	8,000	<sup>238</sup> Pu	30
<sup>95</sup> Zr	140,800	<sup>239</sup> Pu	25.5
<sup>106</sup> Ru	58,000	<sup>240</sup> Pu	36
<sup>134</sup> Cs	19,000	<sup>241</sup> Pu	5100
<sup>137</sup> Cs	37,700	<sup>242</sup> Cm	780
<sup>144</sup> Ce	89,600		

See Table W28.10d for data sources.

**Table W28.10(d)** Inventories of the Long-Lived Artificial Radionuclides: Waste Dumping at Sea from 1946 to 1993 prior to Adoption of London Convention Amendments of 1993.

Disposal Site	Inventory (PBq)		Wastes (X denotes usage contributing to inventory)			
	Northwest	Northeast	Liquid Wastes	Solid Wastes, <sup>g</sup> Mostly Packaged	Nuclear Reactors without Fuel	Nuclear Reactors with Fuel
Atlantic	2.94	42.32		X	X	
Pacific	0.89	0.55	X	X	X	
Arctic		38.37	X	X	X	X (Kara Sea)

<sup>g</sup>Packages were required to remain intact for a period of time after they reached the seabed.

Source: From Pentreath, R. J. (1988). Sources of artificial radionuclides in the marine environment, pp. 12–34. In Radionuclides: A Tool for Oceanography, J. C. Guarj, P. Guegueniat, and R. J. Pentreath, eds. Elsevier Applied Science; Linsley, G., K.-L. Sjoblom, and T. Cabianca (2004). Overview of point sources of anthropogenic radionuclides in the ocean, pp. 109–138. In Marine Radioactivity, H. D. Livingston, ed. Elsevier Ltd.

radionuclides tended to sorb to particles while others, such as <sup>137</sup>Cs and <sup>90</sup>Sr remained in solution.

The fraction deposited locally depended on whether the explosion was conducted on the ground or in the atmosphere as an airburst. Of the total injected, 12% was deposited close to the test sites and 10% in latitudinal bands. The remaining 78% was entrained in Hadley cells that enabled cross-latitudinal transport, but deposition was generally within the same hemisphere as the explosion. The combined influence of the detonation site locations and atmospheric circulation caused most of the fallout to be deposited between 40 to 50° in the northern and southern hemispheres. The majority of the inventory fell onto the Pacific Ocean.

**Table W28.11** Metal Enrichment Factors for Sediment, Marine Fish, and Shellfish.

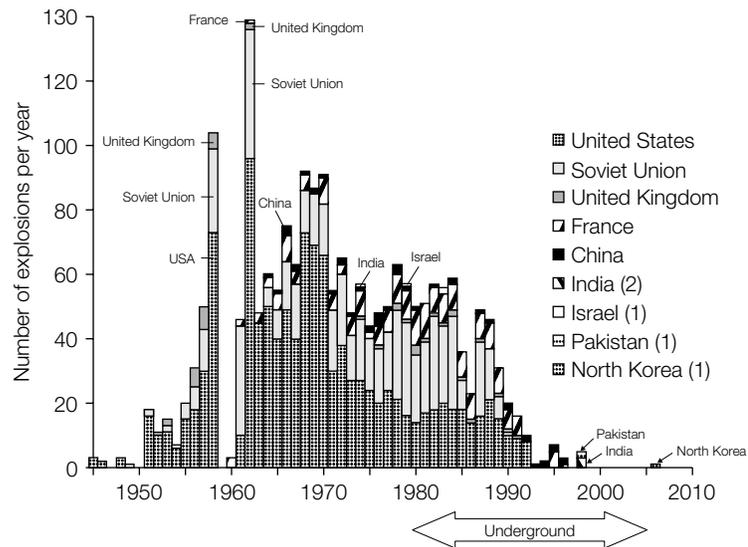
Element	Sediment Concentrating Factor (Bq kg <sup>-1</sup> per Bq L <sup>-1</sup> )	Bioaccumulation Factor (Bq kg <sup>-1</sup> per Bq L <sup>-1</sup> )	
		Marine Fish	Shellfish
Ag	1,000	500	10,000
Am	2,000,000	50	20,000
Ca	500	2	5
Ce	2,000,000	50	5,000
Cl	0.03	0.05	0.05
Cm	2,000,000	50	30,000
Co	200,000	1,000	5,000
Cr	50,000	200	800
Cs	3,000	100	30
Fe	50,000	3,000	30,000
Hg	10,000	20,000	20,000
I	20	10	10
Ir	100,000	20	100
Mn	200,000	400	5,000
Na	1	0.1	0.3
Nb	500,000	10	1,000
Np	5,000	10	400
Pb	200,000	200	1,000
Pm	2,000,000	500	5,000
Po	20,000,000	2,000	50,000
Pu	100,000	40	3,000
Ra	5,000	500	1,000
Ru	300	2	2,000
S	0.5	2	4
Sb	1,000	400	400
Se	1,000	6,000	6,000
Sn	1,000	50,000	50,000
Sr	1,000	2	2

*(Continued)*

**Table W28.11** (Continued)

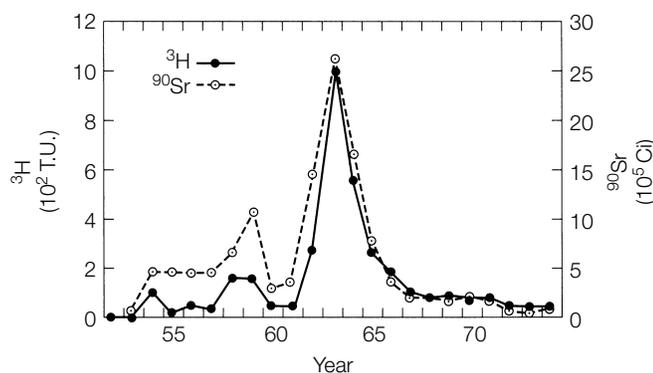
Element	Sediment Concentrating Factor (Bq kg <sup>-1</sup> per Bq L <sup>-1</sup> )	Bioaccumulation Factor (Bq kg <sup>-1</sup> per Bq L <sup>-1</sup> )	
		Marine Fish	Shellfish
Tc	100	30	1,000
Th	2,000,000	600	1,000
Tl	20,000	5,000	5,000
U	1,000	1	30
Zn	20,000	1,000	50,000
Zr	1,000,000	20	5,000

Source: From IAEA (2000). *Guidance on radiological assessment procedures to determine if materials for disposal at sea are within the scope of the London Convention*. IAEA, <http://imo.amsa.gov.au/public/2000/lc22.html>.

**FIGURE W28.5**

Number of nuclear weapons explosions per year. Source: After Elert, G. (1998–2007) *Half-life*, The Physics Hypertextbook, <http://hypertextbook.com/physics/modern/half-life/>.

While in the atmosphere, the bomb tritium underwent equilibrium exchange reactions with water vapor, leading to the formation of tritiated water ( ${}^3\text{H}^1\text{HO}$ ). This was carried to Earth's surface as rain. The rain depositional histories of  ${}^{90}\text{Sr}$  and  ${}^3\text{H}$  are shown in Figure W28.6. Penetration of the tritiated water into the deep ocean was fastest at the



**FIGURE W28.6**

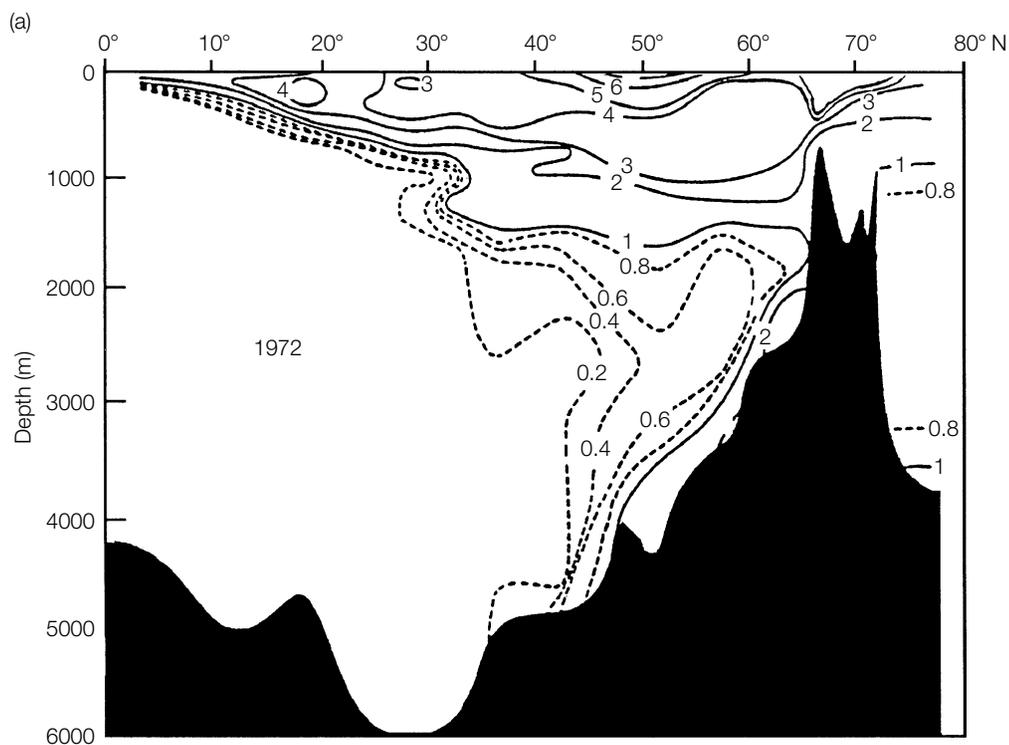
Plot of the  $^3\text{H}$  content of rain at the west coast of Ireland, from 1952 to 1974 (solid circles). Tritium levels are reported in tritium units (T.U.), which are equal to  $10^{18} \times (^3\text{H}/^1\text{H})$ . Also given is the total annual northern hemisphere  $^{90}\text{Sr}$  deposition (open circles) in  $10^5$  Ci ( $10^5$  Ci = 3.7 PBq). Source: From Dreisigacker, E., and W. Roether (1978). Tritium and  $^{90}\text{Sr}$  in North Atlantic surface water. *Earth and Planetary Science Letters*, 38, 301–312, p. 307.

site of NADW formation (Figure W28.7). The bomb  $^3\text{H}$  has provided a clear tracer of the pathway of NADW through the deep sea and has been used to estimate the rate of movement of this water mass.

The steep vertical concentration gradients seen in the depth profiles of  $^{90}\text{Sr}$ ,  $^{137}\text{Cs}$ , and  $^{239+240}\text{Pu}$  also demonstrate that these radionuclides have penetrated into the ocean (Figure W28.8). These gradients have diminished over time due to declining surface water concentrations. Similarly, latitudinal concentration gradients have blurred over time. Since above-ground testing ceased in 1980, sea surface deposition of these radionuclides has been fueled by the remaining atmospheric stock and resuspension of debris from land, especially during dust events. Some of the changes over time are also due to physical transport via advection and turbulent mixing. The diminishing source of bomb-derived radionuclides has been recorded by coral that incorporate  $^{90}\text{Sr}$  into their calcareous hard parts. Figure W28.9 clearly records the peak input of artificial radionuclides to seawater as having occurred in the 1960s.

The vertical depth profile of  $^{239+240}\text{Pu}$  shown in Figure W28.8 exhibits a subsurface maximum. This feature is caused by a reversible incorporation of Pu into sinking particles that is thought to be biologically mediated because the subsurface maximum is most pronounced in areas with high surface productivity. A likely mechanism is incorporation into fecal pellets followed by release during remineralization in the subsurface waters. In the northwest Pacific, the subsurface Pu maximum has been decreasing over time and deepening, reflecting a decreasing source and continued removal from the surface waters. A gradient across the sediment-water interface has also developed that is consistent with a postdepositional remineralization of Pu.

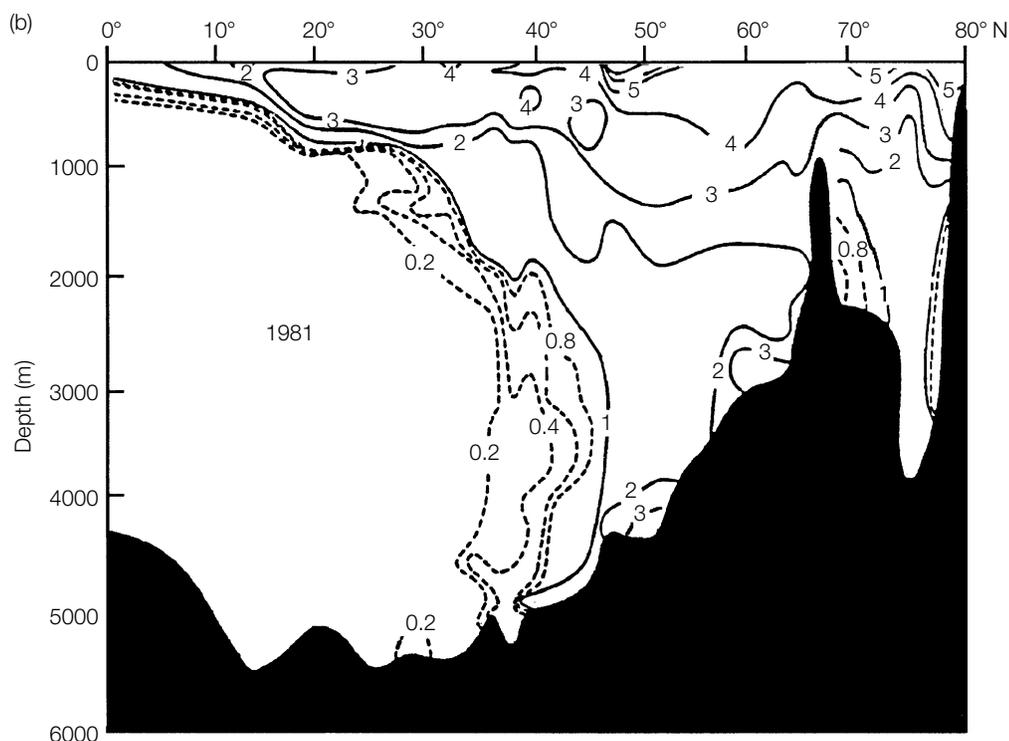
Although radiocarbon ( $^{14}\text{C}$ ) was a minor component of the bomb radionuclide inventory, its fate in the ocean has been intensely studied. Like tritium, bomb radiocarbon



**FIGURE W28.7** (Continued)

underwent isotopic equilibrium exchange reactions in the atmosphere, in this case, with  $\text{CO}_2$ . The radiocarbon-labeled  $\text{CO}_2$  was transferred into the ocean primarily via gas exchange. Bomb testing more than doubled the natural atmospheric inventory of radiocarbon. As shown in Figure W28.10, the oceanic reservoir has been significantly increased. The radiocarbon content of seawater is measured as a ratio relative to  $^{12}\text{C}$ .<sup>2</sup> Part of the increase in the relative abundance of radiocarbon due to bomb testing has been offset by release of “dead” carbon from fossil-fuel burning. This “dead” carbon is millions of years old and has no radiocarbon (half-life is 5730 y). As with  $^{90}\text{Sr}$ , the time trend of injection of radiocarbon into seawater can be ascertained from the radiocarbon content of calcite deposited by coral. Figure W28.11 shows that the coral record for  $^{14}\text{C}$  is similar to that of  $^{90}\text{Sr}$ . Radiocarbon measurements have also been used to estimate water mass ages and mixing rates.

<sup>2</sup> Radiocarbon data are reported as  $\Delta^{14}\text{C}$ , which is the difference (in parts per thousand, or “per mil”) between a sample’s  $^{14}\text{C}/^{12}\text{C}$  and the ratio that was present in atmospheric  $\text{CO}_2$  prior to the onset of anthropogenic inputs.

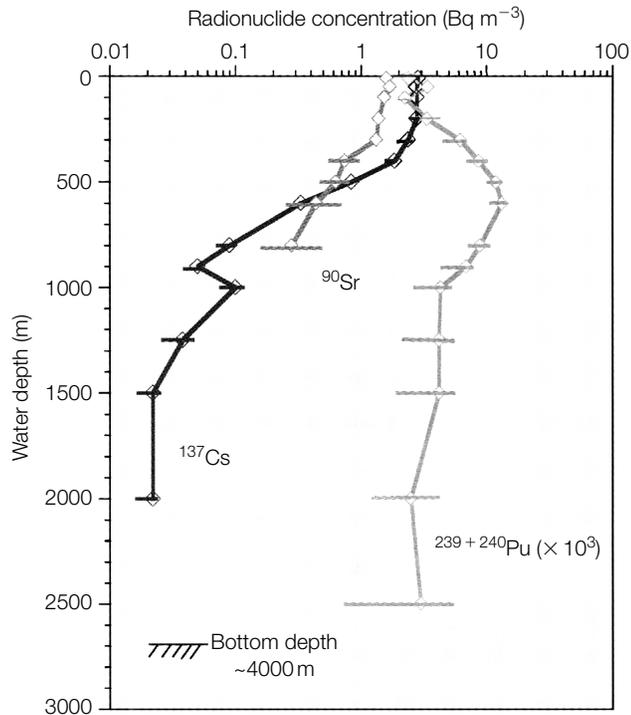


**FIGURE W28.7**

Distribution of tritium units (T.U.) in the western Atlantic Ocean (a) 1972 and (b) 1981. (See Figure W28.6 for unit definition.) This diagram indicates the extent to which bomb-produced material has penetrated the deep ocean since aboveground testing began in the 1950s. *Source:* From Östlund, H. G., and C. G. H. Rooth (1990). North Atlantic tritium and radiocarbon transients 1972–1983. *Journal of Geophysical Research*, 95, 20147–20165, p. 20154.

### **Accidental Losses**

Some radionuclides have been released into the ocean and atmosphere as a result of accidental losses from: (1) nuclear-powered military surface or underwater vessels; (2) nuclear weapons and military vessels carrying these weapons; (3) nuclear-powered civilian ships; (4) nuclear energy sources used in spacecraft, satellites, and in the deep sea as acoustic signal transmitters; (5) power generators, such as used for lighthouses; (6) cargos of nuclear material in transit; and (7) sealed radiation sources. Military vessels now collectively carry around 400 nuclear reactors. Six nuclear submarines have been lost to accidents since 1963 at various sites in the Atlantic Ocean. The first was the U.S.S. *Thresher*, which sank off the New England coast in 1963. Only one, the USSR's *Kursk*, has sunk in water depths shallow enough to permit recovery. So far no leakage of radioactivity to adjacent sediments has been detected.

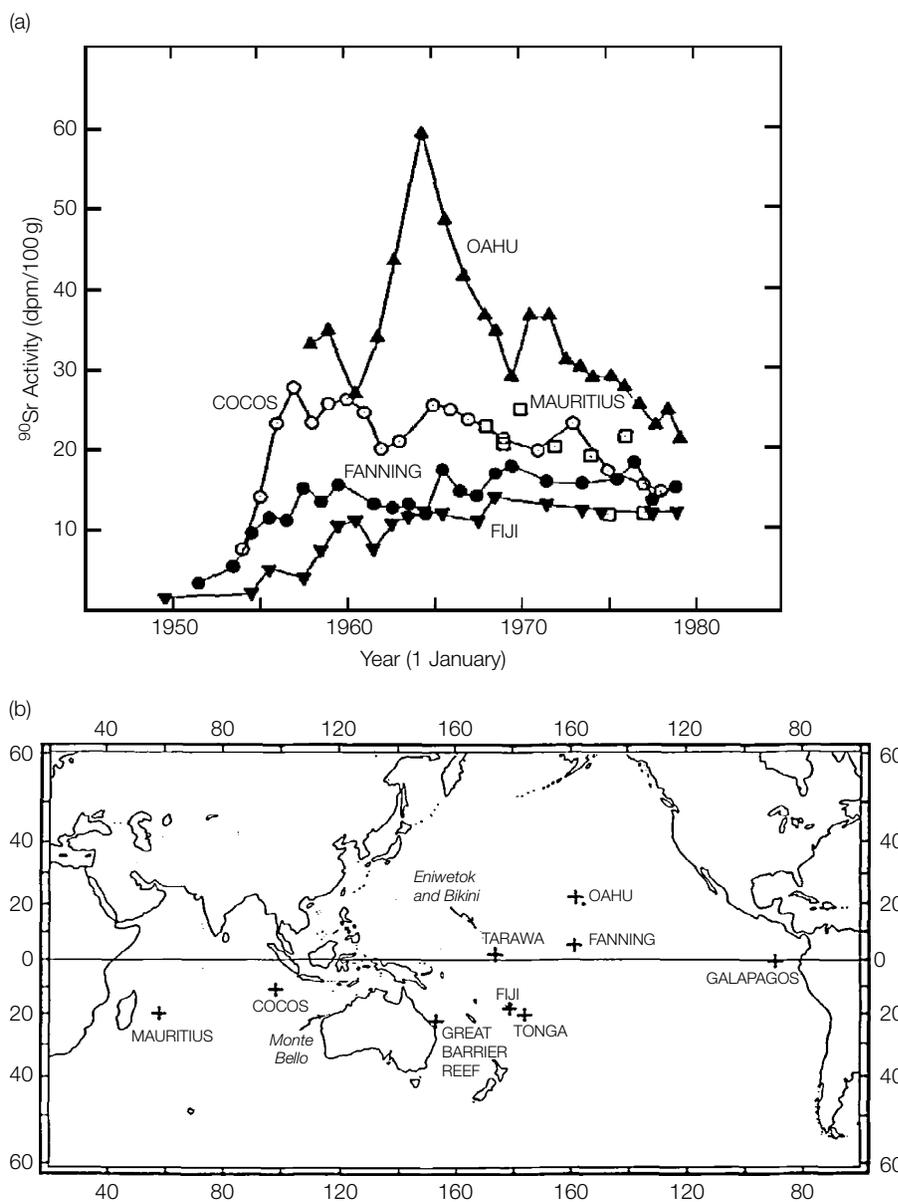


**FIGURE W28.8**

Depth distribution of  $^{90}\text{Sr}$ ,  $^{137}\text{Cs}$ , and  $^{239+240}\text{Pu}$  in the Equatorial Pacific. *Source:* From Hamilton, T. F., J.-C. Milliès-LaCroix, and G. H. Hong (1996).  $^{90}\text{Sr}$  ( $^{137}\text{Cs}$ ) and Pu isotopes in the Pacific Ocean: Sources and trends. In *Radionuclides in the Ocean: Inputs and Inventories*, P. Guegueniat, P. Germain, and H. Metivier, eds. Les Ulis cedex A, France: Les Éditions de Physique.

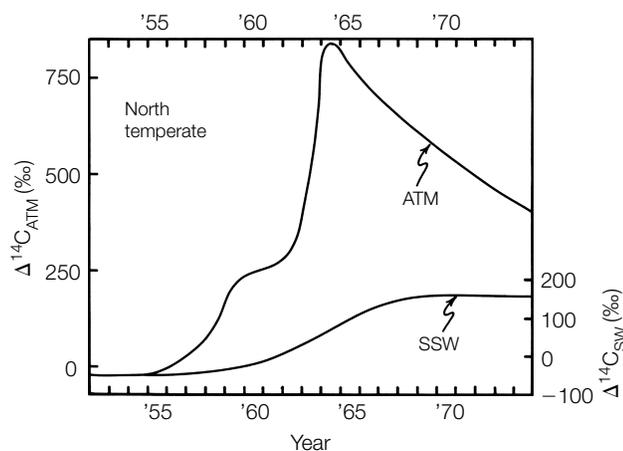
Seven recorded accidents have involved the loss of nuclear weapons. In two, the weapons were destroyed and radioactivity was released, some of which has entered the ocean. One civilian nuclear-powered vessel has had an accidental loss. This was a USSR ice breaker whose reactor vessels were salvaged and intentionally disposed of by dumping in the Kara Sea. Five accidents involving nuclear-powered satellites and spacecraft have resulted in radionuclide releases. The loss of  $^{238}\text{Pu}$  from the breakup of the SNAP-9A satellite in 1964 was triple that injected by nuclear testing (Table W28.10a)! Other accidents include the loss at sea of two USSR radionuclide thermoelectric generators that contain  $^{90}\text{Sr}$  and three incidents involving loss of cargo that included spent fuel. An unknown number of sealed radiation sources used in association with oil and gas prospecting and extraction have been left on the seabed.

Other land-based emissions have resulted from accidents at nuclear-power plants: (1) the explosion of a nuclear waste dump in the Ural Mountains (USSR) in 1957; (2) the partial core meltdown at the Three-Mile Island plant (Harrisburg, PA) in 1979;



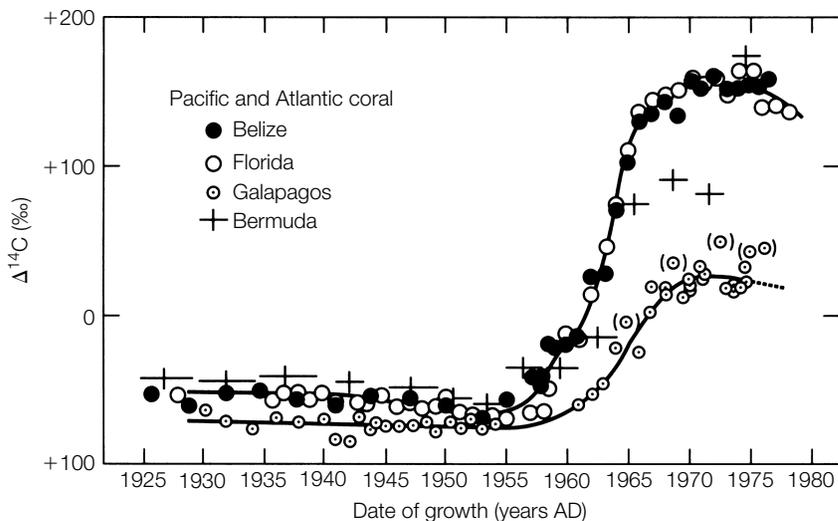
**FIGURE W28.9**

(a) Time histories of  $^{90}\text{Sr}$  activity (dpm/100 g) as recorded in the coral at Oahu (triangle), Cocos (open circle), Fanning (closed circle), and Fiji (inverted triangle). Indian Ocean data are plotted in open symbols, Pacific data in closed symbols. Tick marks on the time axis represent the first day of the year and (b) Sample site location map. *Source:* From J. R. Toggweiler, and S. Trumbore (1985). Bomb-test  $^{90}\text{Sr}$  in Pacific and Indian Ocean surface water as recorded by banded corals. *Earth and Planetary Science Letters* 74, 306–314.



**FIGURE W28.10**

$\Delta^{14}\text{C}$  for atmospheric  $\text{CO}_2$  (ATM) and surface ocean TDIC (SSW) from 1952 to 1974 for the north temperate zone. Source: From Broecker, W. S., and T.-H. Peng (1982). *Tracers in the Sea*. Lamont Doherty Geophysical Observatory, Columbia University, New York (p. 389). See footnote 7 for definition of  $\Delta^{14}\text{C}$ .



**FIGURE W28.11**

$\Delta^{14}\text{C}$  values obtained from corals. The Galapagos coral comes from an area of intense upwelling in the western equatorial Atlantic that has evidently supplied a significant amount of nonradiogenic DIC to the coral. Source: From Druffel, E. M., and H. E. Suess (1983). On the radiocarbon record in banded corals. *Journal of Geophysical Research* 88, 1271–1280, p. 1273. See footnote 7 for definition of  $\Delta^{14}\text{C}$ .

(3) leakage from the Savannah River Nuclear Reactor Site (Aiken, SC) over the past four decades; and (4) the fire that occurred in the Chernobyl reactor (Kiev, USSR) in 1986. The most serious of these was the accident at Chernobyl, which injected highly radioactive debris into the atmosphere (Table W28.10c). This caused widespread contamination over much of Europe and led to significant inputs to the Baltic, Black, Irish, and Mediterranean Seas. External exposures to humans were from airborne deposition and rainout, mainly of  $^{137}\text{Cs}$ . Internal exposures resulted from fish and shellfish consumption.

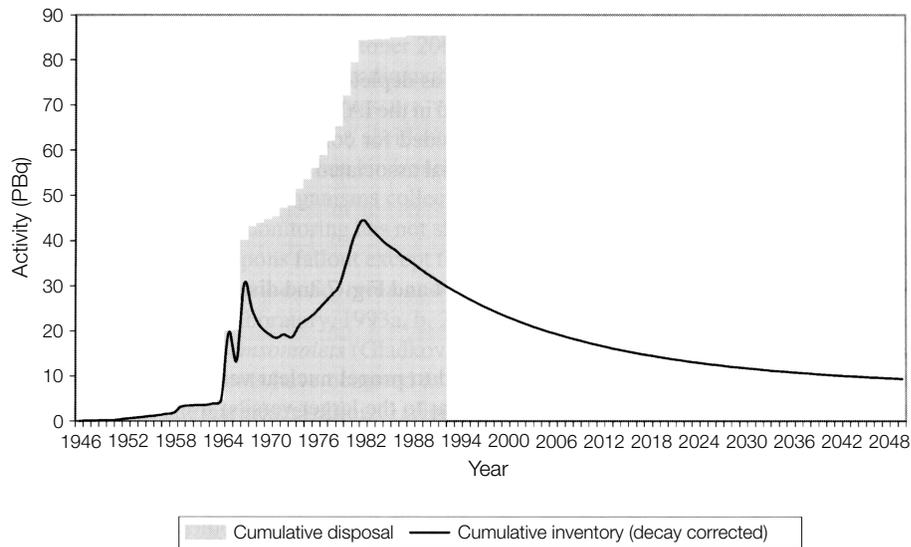
### ***Dumping***

Starting in the 1940s, the oceans have intentionally been used as a disposal option for artificial radionuclides. Two techniques have been employed: (1) dumping and (2) discharge via outfalls into rivers or directly into coastal waters. Dumping was necessitated by the creation of wastes from the production of nuclear weapons. The first offshore dumping took place in 1946 at a site 80 km off the coast of California. Through 1967, a number of shallow sites were used for the disposal of relatively small amounts of wastes. After 1967, disposal moved to deeper waters and was coordinated by the Nuclear Energy Agency of the Organization for Economic Cooperation and Development (NEA/OECD) (Table W28.10d).

The waste was supposed to be packaged in a manner that would ensure its intact delivery to the seafloor. The packaging was also designed to remain intact for a period sufficient to ensure that the bulk of the isotopes had decayed. Over the next 35 y, approximately 85 PBq of radioactivity was disposed of at 80 dumpsites. The wastes consisted mostly of low-level materials generated by: (1) nuclear plant operations, (2) fuel fabrication and reprocessing, (3) radionuclide use in medicine, research, and industry, and (4) decontamination of related parts and equipment. Some of the waste was liquid and some solid, including nuclear reactor vessels.

Most of the nuclear waste came from the UK (41%) and the former USSR (45%). Only 4.2% of the dumped waste came from the United States. Of the USSR's contribution, most was in the form of reactors containing spent nuclear fuel that were dumped in the Arctic Ocean at two very shallow sites (12 to 135 m and 380 m). About half of all the dumped wastes were low-level solids discharged into the northeastern Atlantic, primarily by the UK. This was disposed of at about 10 different sites, all located no further west than 20°W. These disposals took place from 1949 to 1982. As shown in Figure W28.12, the radioactivity from the dumped wastes reached a peak in 1982 (45 PBq). Following passage of a dumping moratorium in 1983, radioactive decay has reduced the size of the total inventory from 85 to 21 PBq (circa 2004). By 2050, it should be down to 10 PBq. The peaks in 1965 and 1967 are due to dumping by the former USSR of reactors containing nuclear fuel into the Arctic Ocean.

One-third of the radioactivity of the dumped wastes was contributed by tritium. Other isotopes of importance were  $^{90}\text{Sr}$ ,  $^{134}\text{Cs}$ ,  $^{137}\text{Cs}$ ,  $^{55}\text{Fe}$ ,  $^{58}\text{Co}$ ,  $^{125}\text{I}$ , and  $^{14}\text{C}$ . These are beta and gamma emitters. Less than 2% of the activity came from alpha emitters, primarily Pu and Am. Monitoring of the dumpsites has demonstrated that: (1) although radioactivity is locally elevated in water and sediments, leakages thus far



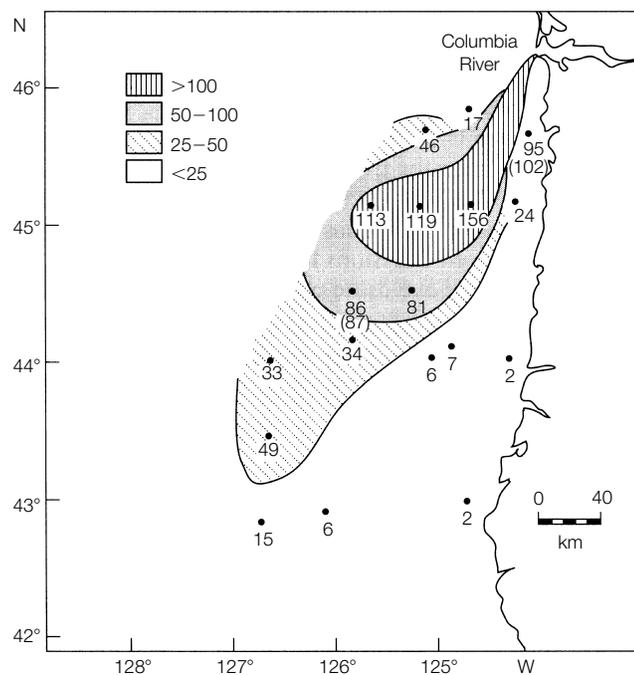
**FIGURE W28.12**

Cumulative inventory of radioactive waste disposed in the world oceans. *Source:* From Linsley, G., K.-L. Sjöblom, and T. Cabianca (2004). Overview of point sources of anthropogenic radionuclides in the oceans, pp. 109–138 in *Marine Radioactivity*, H. D. Livingston, ed. Elsevier Ltd.

seem insignificant, and (2) biotic threats to marine life and humans are not present even in the Arctic Ocean. Nevertheless, dumping in the northeastern Atlantic Ocean was halted via a global moratorium enacted in 1983 as part of the United Nation's London Convention. An amendment to the Convention was approved in 1993 to prohibit dumping of all types of radioactive waste as of 1994. This amendment also included a prohibition on dumping of all industrial wastes as of 1996. The only remaining route by which waste can legally enter the marine environment is by effluent discharges to rivers and from coastal locations. The last recorded case of ocean disposal of radionuclides occurred in 1993 when the USSR released low-level liquid wastes into the Sea of Japan.

### ***Discharges from Nuclear Industry***

The first and most significant authorized release of radionuclides to the sea was from nuclear fuel cycle installations, particularly spent fuel reprocessing plants. The first such discharge was introduced into the northeastern Pacific Ocean by the United States. Starting in the 1940s, radionuclides were directly discharged into the Columbia River from the Hanford Reservation, which is located 575 km upstream from the coastline. During its peak in the 1950s and 1960s, nine plutonium production reactors were operating at this site. Their primary coolant fluid was river water, which following use was discharged directly back into the river. The daily release of radioactivity into the Columbia River was on the order of thousands of curies. This practice was halted in 1971 and the reactors were eventually shut down.



**FIGURE W28.13**

The Columbia River plume as defined by the specific activity of  $^{51}\text{Cr}$  (dpm/100 L) of surface seawater. Numbers in parentheses are the results from duplicates. *Source:* From Osterberg, C., N. Cutshall, and J. Cronin (1965). Chromium-51 as a radioactive tracer of Columbia River water at sea. *Science* 150, 1585–1587, p. 1586.

As shown in Figure W28.13, the entry of these radionuclides into the coastal ocean was traceable as a plume of water enriched in  $^{51}\text{Cr}$ . Soluble chromium exists in seawater primarily as the chromate ion. The radioactive plume was traceable for 350 km downstream of the river mouth. Other radionuclides, such as  $^{65}\text{Zn}$  and  $^{60}\text{Co}$ , were rapidly transported to the sediments via adsorption onto sinking particles. Their distributions were used to trace the movement of sediments across the continental shelf.

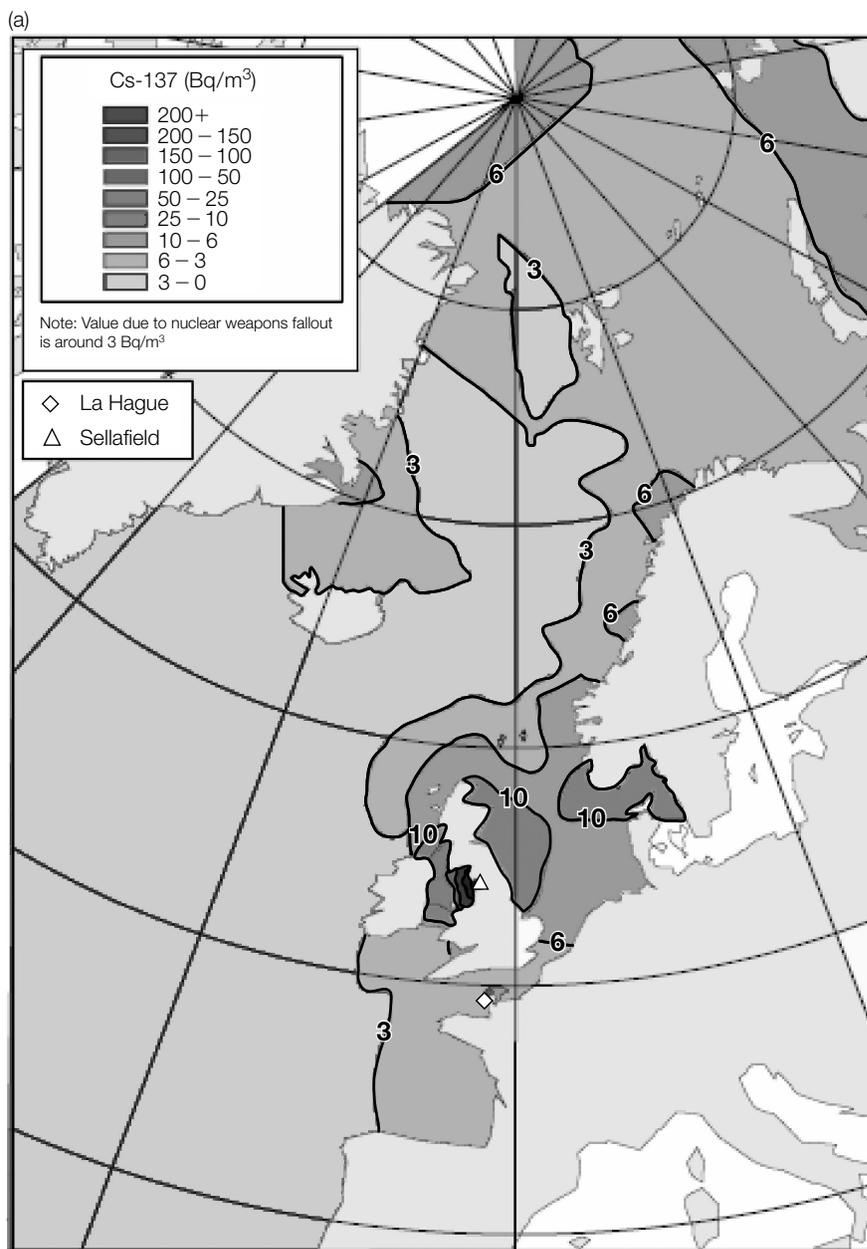
The first direct release of liquid radioactive effluents into the ocean started in 1952 with discharges from nuclear-production reactors in the UK located at Sellafield, La Hague, and Dounreay (Table W28.10b). Sellafield is on the midwestern coast of Great Britain and contains the world's largest nuclear-reprocessing plants, as well as the oldest operating commercial nuclear-power plant. Nuclear wastes are stored in ponds whose overflow water is discharged through two outfalls that extend about 2.5 km from the shoreline. The discharge contains low levels of  $^{137}\text{Cs}$ ,  $^{106}\text{Ru}$ ,  $^{90}\text{Sr}$ ,  $^{239,240}\text{Pu}$ ,  $^{241}\text{Am}$ , and  $^{99}\text{Tc}$ , which collectively account for 90% of the “normal” emissions from Sellafield. The peak annual discharge, 11 PBq, occurred in 1975. From 1952 to 2000, this plant put 22 PBq of  $^{241}\text{Pu}$  into the Irish Sea. The reprocessing plant at Cap de La Hague has

discharged 19 PBq (excluding  $^3\text{H}$ ) since 1965, primarily as  $^{90}\text{Sr}$ ,  $^{125}\text{Sb}$ ,  $^{137}\text{Cs}$ , and  $^{106}\text{Ru}$ . If  $^3\text{H}$  is included, annual releases are now 10 to 15 PBq. In comparison, three of the USSR's plants (Myak, Tomsk-7, and Krasnoyarsk-26) have collectively released 2.4 PBq of  $^{90}\text{Sr}$  via river discharges to the Arctic Ocean between 1961 and 1994, with half going to the Kara Sea.

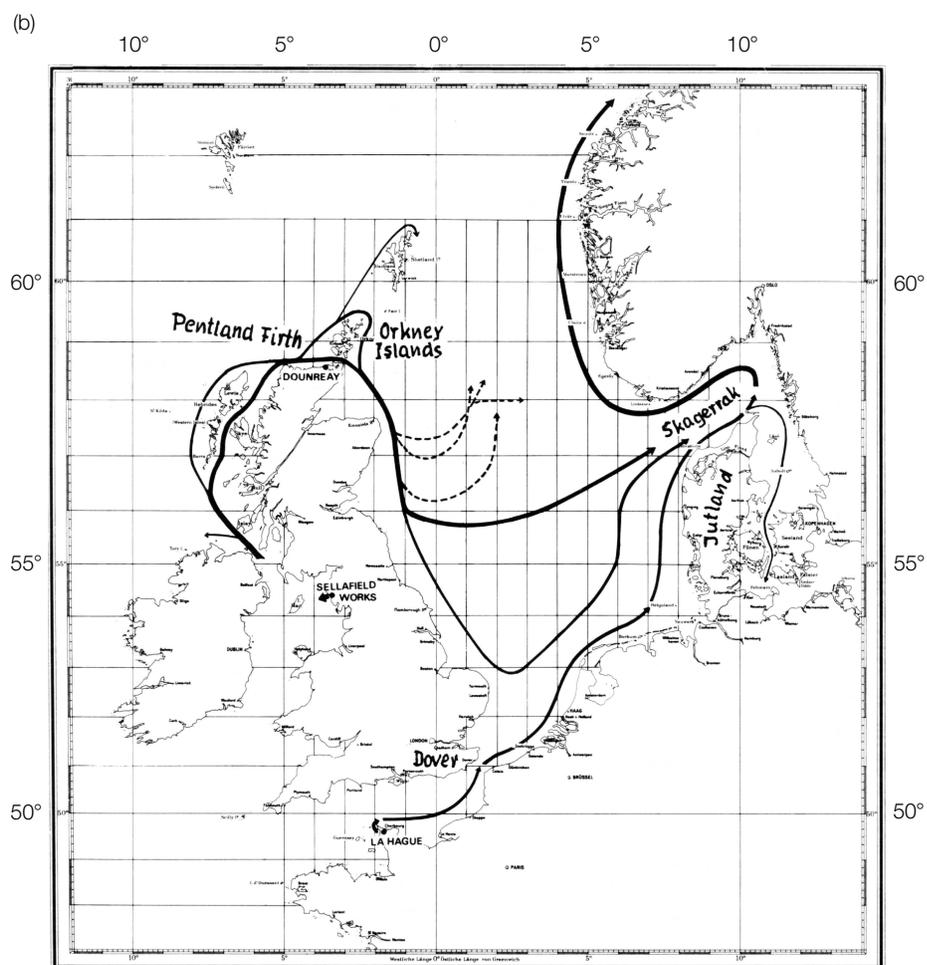
The soluble radionuclides, such as  $^{137}\text{Cs}$ , have proven useful as tracers of currents in the Irish and North Seas. The concentration gradient (Figure W28.14) indicates that  $^{137}\text{Cs}$  from the Sellafield reprocessing facilities is entrained in the Norwegian Coastal Current and carried into the Arctic Ocean via the North Sea. The input from La Hague is carried by currents through the English Channel to the North Sea and subsequently the Arctic.

The particle reactive radionuclides, such as  $^{239,240}\text{Pu}$ ,  $^{241}\text{Am}$ , and  $^{106}\text{Ru}$ , have been rapidly transported to the sediments by adsorption onto sinking particles. As a result, more than 90% of the discharged Pu has been deposited in sediments located within 30 km of the outfall site at Sellafield (Figure W28.15). Some of these radionuclides have been remobilized and returned to land by (1) bioaccumulation in shellfish that are ingested by humans and (2) resuspension of sediment by storms and bottom currents. The latter has transported radioactive muds up into estuaries and salt marshes. At low tide, the fine-grained particles are exposed to the air, during which time the winds can resuspend them, leading to inhalation. Subtidal sediments contain the highest proportion of the estimated inventory of plutonium in the Irish Sea. The specific activity of the sediments is expected to increase over time because radioactive decay is producing daughter radionuclides that are stronger emitters than the parent isotopes. For example, the decay of  $^{241}\text{Pu}$  produces relatively low-energy  $\beta$  particles and  $^{241}\text{Am}$  with decay of the latter releasing higher-energy  $\alpha$  particles. Therefore, the sediments are a repository of old radionuclides, as well as a source of new ones. Some radionuclides, such as  $^{137}\text{Cs}$  and  $^{99}\text{Tc}$ , are concentrated in seaweeds as a result of bioaccumulation. Levels decrease with increasing distance from Sellafield and have fallen in response to reductions in radionuclide discharge.

Due to improved safety practices, emissions from Sellafield and Cap de la Hague have declined. Radioactive decay and surface currents have caused the specific activity of the waters to decrease. Nevertheless, in its 2003 report, the UN Scientific Committee on the Effects of Atomic Radiation found that in the northeast Atlantic, most of the discharged artificial radionuclides still comes from reprocessing plants (83%). Increasing radionuclide concentrations in the Arctic Ocean are attributed to advective export of  $^{129}\text{I}$  and  $^{99}\text{Tc}$  from the North Sea discharges and remobilization of  $^{137}\text{Cs}$  and  $^{239+240}\text{Pu}$  from the sediments around the Sellafield plant. Releases from nuclear power plants account for 13% of the radionuclides now being discharged into the ocean. Worldwide, liquid radionuclide discharges from nuclear-power plants (other than tritium) have declined since the 1970s, although tritium releases have increased by a factor of 4 to 5. This report was unable to evaluate discharges from land-based nuclear-power plant installations located in Central America, the Indian subcontinent, West Asia, and the USSR due to lack of documentation. These discharges are likely significant.

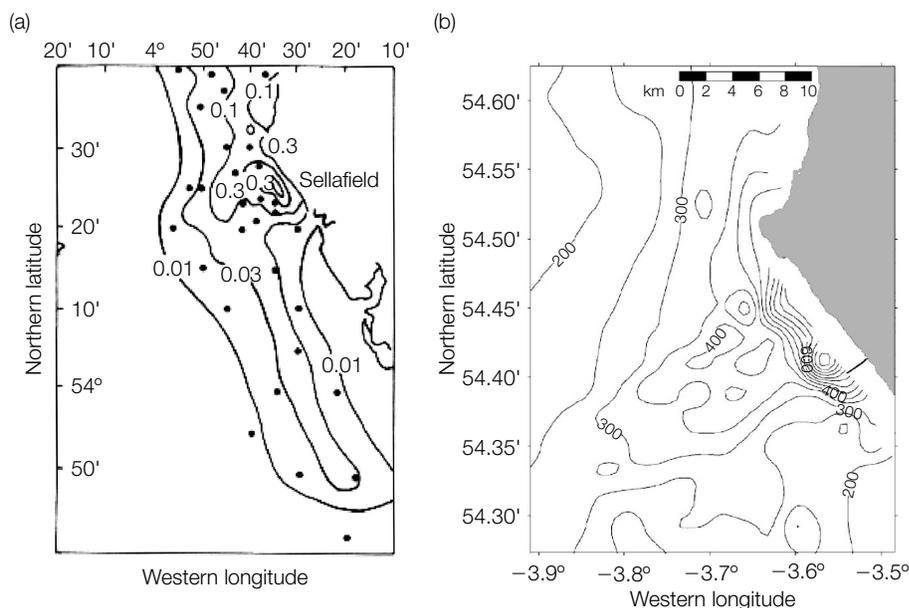


**FIGURE W28.14** (Continued)



**FIGURE W28.14**

(a) Activity of  $^{137}\text{Cs}$  in seawater ( $\text{Bq}/\text{m}^3$ ) in the northeast Atlantic based on measurements made from 1992 to 1996 and (b) transport routes of  $^{137}\text{Cs}$  deduced from measurements made from 1971 to 1984. Dotted lines indicate shifting transport routes. *Source:* After (a) OSPAR Commission (2000). *Quality Status Report 2000*. OSPAR Commission, London; (b) Kautsky, H. (1988). Determination of distribution processes, transport routes and transport times in the North Sea and the northern North Atlantic using artificial radionuclides as tracers, pp. 271–280 in *Radionuclides: A Tool for Oceanography*, J. C. Guary, P. Guegueniat, and R. J. Pentreath, eds. Elsevier Applied Science. (See companion website for color version.)



**FIGURE W28.15**

Radionuclide inventory (MBq/m<sup>2</sup>) in the top 150 cm of sediment collected from the northeast Irish Sea in 1983. (a)  $^{137}\text{Cs}$  and (b)  $^{239}\text{Pu} + ^{240}\text{Pu}$ . Dots mark sampling sites. *Source:* From Woodhead, D. S. (1988). Mixing processes in near-shore marine sediments as inferred from the distributions of radionuclides discharged into the northeast Irish Sea from Sellafield, pp. 331–340 in *Radionuclides: A Tool for Oceanography*, J. C. Guary, P. Guegueniat, and R. J. Pentreath, eds. Elsevier Applied Science.

### ***Discharges from Nonnuclear Industry***

Discharges of radioactive substances from the nonnuclear sector come from (1) mining and ore processing, (2) offshore oil production, (3) burning of coal, oil, or natural gas in thermal power stations, (4) the production of phosphate fertilizers, and (5) other miscellaneous industries such as concrete and ceramics production. These activities are mobilizing natural radionuclides. Many of these discharges are routed into rivers, which transport the radionuclides into the ocean. For example, phosphate fertilizer production is an important source of  $^{40}\text{K}$ ,  $^{226}\text{Ra}$ ,  $^{232}\text{Th}$ ,  $^{238}\text{U}$ ,  $^{210}\text{Pb}$ , and  $^{210}\text{Po}$ . Offshore oil production is a significant source of  $^{226}\text{Ra}$ ,  $^{228}\text{Ra}$ ,  $^{210}\text{Pb}$ , and  $^{210}\text{Po}$ . In this case, the radionuclides are mobilized from the seabed during drilling and become incorporated into the drill muds and production waters.

Although technically part of the nuclear industry, another important land-based source that ends up in river water is medical waste that contains radionuclides. Radioisotopes are used by the medical professions to produce images for diagnostic purposes ( $^{99}\text{Tc}$ ) and in therapeutic treatments ( $^{131}\text{I}$ ).

### ***Overall Assessment of Threats to Human Health of Artificial Marine Radionuclides***

For humans, radiation exposure from unenhanced sources of natural radioactivity is in most cases higher than those from anthropogenically derived sources. The most significant exposure derives from consumption, at high rates, of molluscan shellfish due to natural  $^{210}\text{Po}$  bioaccumulation. The second largest exposure is from anthropogenically enhanced radionuclides mobilized by phosphorite mining, primarily  $^{210}\text{Po}$ . The third largest, in terms of total exposure to people, is associated with exposure to the Sellafield discharges. Although produced waters are sources of  $^{226}\text{Ra}$ ,  $^{228}\text{Ra}$ , and  $^{210}\text{Pb}$ , these are considered to be a minor exposure to humans.

The radionuclides that posed the greatest health threat were  $^{137}\text{Cs}$ ,  $^{99}\text{Tc}$ ,  $^{239,240}\text{Pu}$ , and  $^{241}\text{Am}$ . Fortunately, radionuclides from nuclear fallout peaked in 1963. Exposures from this legacy radiation are now mostly from: (1)  $^{14}\text{C}$  and  $^{90}\text{Sr}$  that have been passed up the food chain and (2) from ingestion and external exposure to  $^{137}\text{Cs}$ . The only places where this exposure is still likely to be unacceptably high are those in close proximity to the test sites and only for humans whose diets are entirely local. As noted earlier, extensive studies of radionuclides disposed of at offshore dumpsites has indicated no significant impacts are posed to human health, even in the Arctic Ocean.