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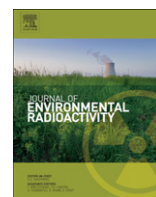
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Sources of anthropogenic radionuclides in the environment: a review

Qin-Hong Hu^{a,b,*}, Jian-Qing Weng^c, Jin-Sheng Wang^a^a College of Water Sciences, Beijing Normal University, 19 XinJieKouWai Street, Beijing 100875, PR China^b Department of Earth and Environmental Sciences, The University of Texas at Arlington, Arlington, TX 76019-0049, USA^c Radiation Monitoring Technical Center, National Environmental Protection Administration, 306 WenYi Road, Hangzhou 310012, PR China

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ABSTRACT

Studies of radionuclides in the environment have entered a new era with the renaissance of nuclear energy and associated fuel reprocessing, geological disposal of high-level nuclear wastes, and concerns about national security with respect to nuclear non-proliferation. This work presents an overview on sources of anthropogenic radionuclides in the environment, as well as a brief discussion of salient geochemical behavior of important radionuclides. We first discuss the following major anthropogenic sources and current developments that have lead, or could potentially contribute, to the radionuclide contamination of the environment: (1) nuclear weapons program; (2) nuclear weapons testing; (3) nuclear power plants; (4) uranium mining and milling; (5) commercial fuel reprocessing; (6) geological repository of high-level nuclear wastes that include radionuclides might be released in the future, and (7) nuclear accidents. Then, we briefly summarize the inventory of radionuclides ⁹⁹Tc and ¹²⁹I, as well as geochemical behavior for radionuclides ⁹⁹Tc, ¹²⁹I, and ²³⁷Np, because of their complex geochemical behavior, long half-lives, and presumably high mobility in the environment; biogeochemical cycling and environment risk assessment must take into account speciation of these redox-sensitive radionuclides.

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1. Introduction

During December 1938, nine months before the beginning of World War II, an Austrian physicist Lise Meitner, and two German chemists, Otto Hahn and Fritz Strassmann, discovered nuclear fission. This discovery culminated 40 years of research into radioactivity and ushered in a nuclear era wherein peaceful, or destructive, utilization of enormous nuclear energy has been entwined in the human civilization. Under the order of President Roosevelt in 1939, the United States of America (USA) first began to develop nuclear weapons during World War II, motivated by a fear of a potential race with Nazi Germany to develop such a weapon. After a slow start, in 1942 the program was officially operated under the U.S. Corps of Engineers and became known as the Manhattan Project, a mammoth effort to build the 1st atomic bomb. During the next five decades, before the Cold War ended in 1991, massive weapons production and testing programs, as well as the utilization of nuclear power for electricity generation, have produced a host of unwanted radionuclides in different waste forms and spread environmental contamination across the world.

Major sources of radioactive waste and contamination include the production of electrical power and weapons from nuclear fuels, nuclear weapons' tests, fuel reprocessing, and nuclear accidents. In the USA, the total volume of all radioactive waste is 5.5 million m³, with the radioactivity of about 1.2×10^9 TBq (tera becquerel; 1 TBq = 27.03 Ci) (Ahearne, 1997). In addition, there are 30–80 million m³ of contaminated soil and 1800–4700 million m³ of contaminated water. Among them, over 70 million m³ of soil and at least 1800 million m³ of water have been contaminated by releases from U.S. Department of Energy (DOE) facilities that were used for weapons production (Ewing, 2004). Despite the large volumes of contaminated soil and water from low- and intermediate-level wastes, these wastes only account for a few percent of the total radioactivity. The majority of radioactivity comes from high-level waste (HLW) and spent nuclear fuel (SNF).

Because of worldwide nuclear weapons' tests that began in New Mexico in 1945, over 2×10^8 TBq of radioactivity has been released into the atmosphere (Choppin, 2003). In addition, with the development of nuclear power industry from 1960 onwards, further emissions occurred to a minor extent from properly operating reactors. Nuclear accidents that destroyed reactors, particularly the Chernobyl accident in 1986, have contributed to most of the radioactive emissions in the atmosphere. Overall, controlled and uncontrolled releases of radioactivity from nuclear reactors' operations constitute less than 3% of the amount from atmospheric weapons testing (Choppin, 2003).

* Corresponding author. Department of Earth and Environmental Sciences, The University of Texas at Arlington, 500 Yates Street, Box 19049, Arlington, TX 76019-0049, USA. Tel.: +1 817 272 5398; fax: +1 817 272 2628.

E-mail address: maxhu@uta.edu (Q.-H. Hu).

This work will summarize major anthropogenic sources that contribute to the radionuclide contamination of the environment; the summary is focused on HLW because of its radioactivity over prolonged duration. A common suite of radionuclides, including ^3H , ^{14}C , ^{90}Sr , ^{99}Tc , ^{129}I , ^{137}Cs , ^{237}Np , ^{241}Am , as well as several uranium and plutonium isotopes, from the nuclear-related activities, are of particular environmental importance because of their abundance, mobility, or toxicity. We will therefore briefly discuss the salient geochemical behavior of these radionuclides, with particular focus on ^{99}Tc , ^{129}I , and ^{237}Np , because of their complex biogeochemical behavior, long half-lives, and presumably high mobility in the environment.

2. Major sources of anthropogenic radionuclides in the environment

2.1. Operations of nuclear weapons program

HLW liquid and sludge are generated from weapons production and reprocessing, by which fissile materials (uranium and plutonium) are separated from fission products in spent nuclear reactor fuels to produce weapons-grade fuel. In the USA, there is approximately 0.38 million m^3 of such HLW with a radioactivity of $3.4 \times 10^7 \text{ TBq}$ (Ahearne, 1997). Most of the HLW (0.34 million m^3) is stored in tanks at Hanford, Washington, and Savannah River, South Carolina; the rest is stored as a solid calcine in tanks at the Idaho National Laboratory and West Valley in New York where a reprocessing plant was once operated (Ahearne, 1997).

Established in 1943, the Hanford Site occupies 1517 km^2 in south-central Washington, which is approximately half the total area of the state of Rhode Island. Between 1944 (startup of the world's first full-scale plutonium-production reactor) and 1988, the Hanford Site has been a plutonium-production facility for the USA weapons program. Currently, the Hanford Site has been engaged in the world's largest environmental cleanup effort, with many challenges to be resolved in the face of overlapping technical, political, regulatory, and cultural interests (Gephart, 2003). Being another major nuclear operations' facility, the Savannah River Site is located along the Savannah River in South Carolina, with an area of 840 km^2 . The historical mission of the site was to produce tritium and plutonium for the weapons program in large nuclear production reactors. The processes generated high-level, low-level, transuranic, and mixed wastes that were managed in seeping and settling basins, unlined disposal pits, waste piles, burial grounds, and underground storage tanks. More than 400 individual contaminated sites and 659 contaminated buildings have been identified at the Savannah River Site (Eisenbud and Gesell, 1997).

As the reprocessing has removed much of the actinide content, over 99% of the present radioactivity from HLW liquid and sludge is from non-actinide radionuclides; yet contribution from long-lived actinides will become comparatively larger over time. In Hanford, about 2 million m^3 of highly radioactive and chemically toxic wastes were produced from two chemical processing plants for Pu. These wastes are currently stored in 177 underground tanks, with capacities ranging from 208 to 416 m^3 for each tank. Sixty-seven tanks are known or are suspected of having leaked, with an estimated 570 m^3 of waste containing $3.7 \times 10^4 \text{ TBq}$ of radioactivity released to the subsurface (Ewing, 2004). In addition to $7.2 \times 10^6 \text{ TBq}$ of radioactive tank waste, there are $6.8 \times 10^5 \text{ TBq}$ of nuclear materials stored on site, some radioactive solid waste, and contaminated groundwater and soil; a total radioactivity is $1.4 \times 10^7 \text{ TBq}$ in Hanford (Gephart, 2003). Nuclear operation at the Hanford Site has discharged a variety of radionuclides into the environment. Tables 1 and 2 present the release into the air and nearby Columbia River during 1944–1972; smaller releases have continued since 1972 (Washington State Department of Health,

Table 1

Radionuclides released into the air at Hanford during 1944–1972

Radionuclide	Radioactive half-life ^a	Radiation released into the air (TBq) ^b
^3H	12.32 a	7400
^{60}Co	5.271 a	0.037
^{85}Kr	10.76 a	703,000
^{89}Sr	50.52 d	25.9
^{90}Sr	28.78 a	2.37
^{95}Zr	64.02 d	44.4
^{103}Ru	39.27 d	44.4
^{106}Ru	1.02 a	14.4
^{129}I	15,700,000 a	1.70
^{131}I	8.04 d	27,380
^{132}Te	3.20 d	148
^{133}Xe	5.243 d	15,540
^{137}Cs	30.07 a	1.554
^{144}Ce	284.6 d	141
^{239}Pu	24,100 a	0.067

^a From Baum et al. (2002); a: year; d: day.

^b From Washington State Department of Health (2000).

2000). Most of the released radionuclides are relatively short-lived (Tables 1 and 2).

A large quantity of radioactive waste and serious environmental contamination also resulted from the nuclear weapons program in the former Soviet Union. Since 1948 the production association Mayak, covering an area of 90 km^2 , has produced plutonium for nuclear weapons (Christensen et al., 1997; Balonov et al., 2006). As the first and still one of the largest and most diverse nuclear facilities in Russia, the Mayak complex includes plutonium and tritium production reactors; fuel reprocessing facilities; a plutonium processing, finishing, and component manufacturing plant; mixed-oxide (MOX) fuel fabrication plants; and nuclear waste treatment and storage facilities (Nuclear Threat Initiative, 2008). The Mayak plant is located within the Chelyabinsk area of approximately 200 km^2 in the southern Ural mountains of eastern Russia, with the town of Ozyorsk (population about 10,000) located 10 km northwest and the city of Chelyabinsk (population about 1 million) at 80 km north from the Mayak plant. The area has a number of natural and man-made reservoirs that have been used in various periods to dump nuclear waste (Christensen et al., 1997; Solodov et al., 1998). From 1949 to 1956, medium- and high-level radioactive wastes were discharged directly into the Techa River system, which flows via the Ob River into the Kara Sea. At a rate of 37 TBq/day , about 76 million m^3 liquid waste with a total radioactivity of $1.1 \times 10^5 \text{ TBq}$ (Fig. 1), including $1.2 \times 10^4 \text{ TBq}$ of ^{90}Sr and $1.3 \times 10^4 \text{ TBq}$ of ^{137}Cs , was released to the Techa River (Christensen et al., 1997; Balonov et al., 2006, 2007). As summarized in Balonov et al. (2007), about 95% of the radioactivity was released to the

Table 2

Radionuclides released into the Columbia River at Hanford during 1944–1972

Radionuclide	Radioactive half-life ^a	Radiation released into Columbia River (TBq) ^b
^{24}Na	14.96 h	481,000
^{32}P	14.28 d	8510
^{46}Sc	83.81 d	4440
^{51}Cr	27.702 d	266,400
^{56}Mn	2.578 h	2,960,000
^{65}Zn	243.8 d	18,130
^{72}Ga	14.1 h	136,900
^{76}As	26.3 h	92,500
^{90}Y	2.67 d	16,650
^{131}I	8.04 d	1778
^{239}Np	2.4 d	233,100

^a From Baum et al. (2002); d: day; h: hour.

^b From Washington State Department of Health (2000).

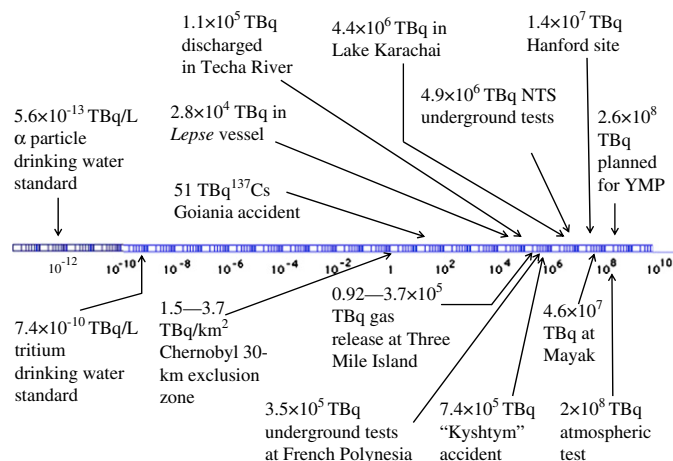


Fig. 1. Radioactivity in various contaminated sites with the USEPA drinking water standards for tritium and α particles. Note that the radioactivity data are at different times and only for relative comparison.

Techa River from March 1950 until November 1951; the release had the following radionuclide composition: ^{89}Sr —8.8%; ^{90}Sr —11.6%; ^{137}Cs —12.2%; $^{95}\text{Zr} + ^{95}\text{Nb}$ —13.6%; $^{103}\text{Ru} + ^{106}\text{Ru}$ —25.9%; radioisotopes of rare earth elements—26.8%. More mobile radionuclides such as ^{90}Sr have been found to be present more than 2000 km downstream of the Techa River. Recently, Balonov et al. (2007) presented an excellent study on the exposure assessment of the population living in the Techa area basin.

Since 1951, the liquid waste has been discharged into a small swampy lake, Lake Karachai, covering less than 0.5 km² in area, inside the Mayak complex. Up to 1993, the lake accumulated some 4.4×10^6 TBq of radioactivity (Fig. 1), including 3.6×10^6 TBq of ^{137}Cs and 7.4×10^5 TBq of ^{90}Sr (Solodov et al., 1998). Lake Karachai is probably the “most polluted spot” on Earth. For comparison, the Chernobyl disaster released about 1.2×10^7 TBq of total radioactivity (UNSCEAR, 2000), and this radiation is not concentrated in one location. According to a report of Natural Resources Defense Council, the radiation level at the shore of the lake was 600 röntgens/h in 1990, high enough to give a lethal dose to a human within an hour (Cochran et al., 1989). About 20% of the radioactivity is in the water phase with a concentration of about 0.5 TBq/m³, whereas the remainder is found in the lake sediments. The groundwater system near the lake has been contaminated (Solodov et al., 1998; Novikov et al., 2006). Starting in the 1960s, Lake Karachai began to dry out; its area dropped from 0.5 km² in 1951 to 0.15 km² by the end of 1993. In 1967, following a drought in the region, the wind carried 22 TBq of radioactive dust away from the dried area of the lake; an area of 1800 km² was contaminated with more than 4 GBq (giga becquerel) ^{90}Sr /km² and 12 GBq ^{137}Cs /km² (Christensen et al., 1997). In addition to Techa River and Lake Karachai, about 1.2×10^4 TBq of radioactive waste was released to five other reservoirs (Christensen et al., 1997).

In addition to staggering waste generation and release, operations in Mayak resulted in many accidents. The most notable “Kyshtym” accident occurred on September 29, 1957, when the failure of the cooling system for a tank storing tens of thousands of tons of dissolved nuclear waste resulted in a violent explosion, which released 7.4×10^5 TBq of radioactivity; an area of 20,000 km² (with 270,000 inhabitants) were contaminated with more than 4000 Bq/km². Approximately 70% of this activity consisted of the rather short-lived nuclides such as ^{95}Zr , ^{106}Ru and ^{144}Ce ; 2.7 and 0.04% were ^{90}Sr and ^{137}Cs , respectively (Christensen et al., 1997).

In addition to environmental problems from the production of nuclear weapons, the other very pressing environmental and

non-proliferation problem in Russia is the storage and disposal of the SNF from nuclear submarines and nuclear-powered surface ships. With the end of Cold War, 200 Russian nuclear submarines were decommissioned, and over 100 are awaiting dismantlement (Webster, 2003). The largest SNF storage facility at Andreeva bay, on the northernmost coast of the Kola Peninsula near the border with Norway, contains 21,000 spent fuel assemblies extracted during the 1970s and 12,000 m³ of solid and liquid radioactive wastes. Three makeshift storage tanks for spent nuclear fuel have been deteriorating because of poor maintenance and the harsh Arctic climate. Poor condition of the storage facility has also lead to contamination of a substantial amount of soil, water, and concrete at Andreeva bay (Ewing, 2004).

2.2. Nuclear weapons testing

The first nuclear weapons test was conducted on July 16, 1945, near Alamogordo, New Mexico. According to the U.S. Department of Energy's Nevada Site Office (DOE/NV, 2000), the explosive yield of this Trinity test was 21 kt (1 kt = 10^{12} calories). From this day to September 23, 1992, the United States maintained a program of vigorous nuclear testing, with the exception of a moratorium between November 1958 and September 1961. As released in DOE/NV (2000), a total of 1054 nuclear tests were conducted, with 106 tests taking place at sites in the Pacific Ocean, 904 at the Nevada Test Site (NTS), and 17 on miscellaneous sites in the continental USA (e.g., Alaska, Colorado, Mississippi, and New Mexico).

Occupying a 19,000 km² area in the northeast of the country, the Semipalatinsk test site in the Republic of Kazakhstan is located immediately south of Russia, and west of China. Following World War II, the steppes of Kazakhstan became the first center for nuclear weapons testing within the former Soviet Union. During 1949–1989, the Soviet Union conducted a total of about 460 nuclear weapons' tests in Semipalatinsk, including 133 atmospheric nuclear tests. Starting in 1961, more than 300 test explosions were conducted underground. Thirteen of the underground tests resulted in releases of radioactive gases to the atmosphere (IAEA, 2008).

In late 1940 to early 1960s, about 350 atmospheric atomic bomb tests were conducted at different test sites in the northern hemisphere (Eikenberg et al., 2004). These tests released into the atmosphere more than 9×10^5 TBq of residual ^{239}Pu , 9×10^5 TBq of fissionogenic ^{137}Cs , and 6×10^5 TBq of ^{90}Sr . Choppin (2003) reported that over 2×10^8 TBq of radioactivity has been released into the atmosphere from worldwide nuclear weapons' tests (Fig. 1).

At the NTS with an area about 3500 km² (the area of Rhode Island is 3144 km²), a large inventory of radionuclides (tritium, fission products, activation products, and actinides) is present in the subsurface environments as a result of 828 underground nuclear weapons' tests (DOE/NV, 2000). A total of about 4.9×10^6 TBq of radioactivity decayed corrected to September 23, 1992, the date of the last underground nuclear test, was produced (Smith et al., 2003). The inventory includes only those radionuclides (a total of 43) whose concentrations dissolved in a cavity (produced from an underground nuclear test) volume of water, decay corrected to 100 years in the future, exceed 0.1 of the proposed maximum permissible concentrations of radionuclides in drinking water set by the U.S. Environmental Protection Agency in 1991 (USEPA, 1991). This selection criterion effectively excludes most radionuclides with half-lives less than 10 years. Since 1973, various studies have been investigating the environmental effects of nuclear testing at the NTS (e.g., Coles and Ramspott, 1982; Buddemeier and Hunt, 1988; Kersting et al., 1999; Tompson et al., 2002).

Between 1966 and 1996, France had conducted nuclear weapons tests' above and beneath the atolls of Mururoa and Fangataufa in French Polynesia (Pfungsten et al., 2001). The total French nuclear testing program comprised of 210 tests, 50 in the

atmosphere and 160 underground; all French testing ceased on January 27, 1996 (IAEA, 1998). For the underground tests, 13 were conducted at In Ecker in Algeria in the 1960s, and 147 (with a total nuclear yield of 3192 kt) in shafts at Mururoa and Fangataufa during 1975–1996. Estimated underground inventories, decay corrected to May 1, 1996, of 36 radionuclides for the sites of Mururoa and Fangataufa were published in IAEA (1998) with a total radioactivity of 3.5×10^5 TBq (Fig. 1).

Table 3 lists the radionuclide inventories from the underground nuclear tests conducted in the NTS and French Polynesia; the inventory was decay corrected to January 1, 2008. In terms of

radioactivity, ^3H , ^{90}Sr , ^{137}Cs , ^{241}Am , and plutonium isotopes are currently the radionuclides of great importance (Table 3). Over time (e.g., 1000–10,000 years later), long-lived ^{14}C , ^{36}Cl , ^{99}Tc , ^{129}I , ^{237}Np , as well as several uranium and plutonium isotopes will become important.

2.3. Nuclear power plants

To generate electricity, nuclear reactors commonly use fuel of solid ceramic pellets of enriched uranium that are sealed in strong metal tubes; the tubes are bundled together to form a nuclear fuel assembly. The uranium pellets are about the size of the tip of a person's little finger, yet one pellet has the amount of energy equivalent to almost 1 t of coal. After 3–4 years in a reactor, the uranium pellets are no longer efficient for producing electricity and the assembly is removed from the reactor; this SNF is highly radioactive. About 25–30 t of SNF is created per year per GWe (gigawatt electricity). Thus, about 12,000 t of SNF has been produced based on the world's current nuclear energy capacity (Macfarlane and Miller, 2007).

As the number of nuclear power plants (NPP) increased during the 1960s, so did the growing concern for the fate of the civilian SNF. In 2002, there were about 150,000 tHM (metric tones of heavy metal) of SNF in the world. By the end of 2004, there was about 190,000 tHM of SNF in storage (IAEA, 2007). Most of this fuel (90,000 t for 2002 data) is still at the 236 nuclear power stations, which together have 439 reactors, where it was originally generated in 31 different countries (Ewing, 2004; IAEA, 2007). In 2000, the USA's SNF inventory was about 42,300 tHM, with a total radioactivity of 1.2×10^9 TBq (Fig. 1), increasing by slightly less than 2000 tHM every year. Since the mid-1940s, SNF and HLW have accumulated throughout the USA. Currently, SNF and HLW are stored at some 125 sites in 39 states, with over 161 million people residing within 121 km of temporarily stored nuclear waste. By the year 2020, the inventory will be about 77,100 tHM with an activity of 1.3×10^9 TBq; the currently legislated capacity for the potential high-level waste repository at Yucca Mountain is 70,000 tHM of SNF equivalent.

As the world's economy grows, nuclear power is recognized as the only large-scale emissions-free power source that is able to help meet the growing need for electricity. As the third leading source of electricity, nuclear power provides 19% of electricity in the US (Table 4). (Nuclear power provides 78% of electricity for France, 48% for Sweden, and 30% for the entire European Union.) The DOE projects the U.S. will need 40% more electricity by 2030. In order to keep pace with the nuclear energy needs, it will be necessary to build an average of three new plants per year starting in 2015. The U.S. has not ordered a new nuclear power plant since the 1970s, partially as a result of constant litigation and overly complex regulations. The Nuclear Power 2010 initiative, a partnership between industry and the U.S. government, was launched in 2002 to reduce regulatory and other barriers to the development of new nuclear power plants. The Nuclear Regulatory Commission (NRC) is working to improve and streamline the regulatory process to help accelerate the construction of nuclear plants. The comprehensive energy policy also includes permanent disposal of SNF and HLW, and advanced technologies for nuclear fuel reprocessing.

As recent as September 26, 2007, NRG Energy Inc of New Jersey and the South Texas Project Nuclear Operating Company filed an application to build and operate two new nuclear power reactors at the South Texas Project nuclear power station site in Matagorda County (Scientific American, 2007). This is the first nuclear plant license application in the United States in 29 years, since Pennsylvania's Three Mile Island reactor had a partial meltdown in 1979 that sparked a massive protest against nuclear plants; public acceptance of nuclear power has changed with the increasing

Table 3

Radionuclide inventory from underground nuclear tests at the Nevada Test Site and French Polynesia

Radio-nuclide	Half-life (yr) ^a	Radioactivity (TBq)	
		USA Nevada Test Site ^b	French Polynesia ^c
^3H	12.32	1.97E+06	1.45E+05
^{14}C	5715	1.05E+02	2.80E+01
^{26}Al	7.1E+05	4.01E–03	NA
^{36}Cl	3.01E+05	2.28E+01	1.70E+00
^{39}Ar	269	1.14E+02	NA
^{40}K	1.27E+09	3.00E+01	NA
^{41}Ca	1.03E+05	1.64E+02	1.30E+00
^{55}Fe	2.73	NA	3.93E+02
^{59}Ni	7.6E+04	4.20E+00	3.80E+00
^{60}Co	5.271	NA	5.60E+02
^{63}Ni	101	4.26E+02	4.15E+02
^{79}Se	2.90E+05	NA	1.10E–02
^{85}Kr	10.76	2.46E+03	4.72E+02
^{90}Sr	28.78	5.58E+04	8.15E+03
^{93}Zr	1.5E+06	2.83E+00	3.20E–01
$^{93\text{m}}\text{Nb}$	16.1	2.96E+02	NA
^{94}Nb	2.0E+04	1.48E+01	NA
^{99}Tc	2.13E+05	2.11E+01	2.50E+00
^{106}Ru	1.02	NA	2.63E+00
^{107}Pd	6.5E+06	1.27E–01	2.10E–01
$^{113\text{m}}\text{Cd}$	14.1	3.38E+01	1.86E+00
$^{121\text{m}}\text{Sn}$	55	2.19E+02	3.11E–01
^{125}Sb	2.758	NA	3.89E+01
^{126}Sn	2.3E+05	1.23E+00	1.80E–01
^{129}I	1.57E+07	6.51E–02	6.10E–03
^{134}Cs	2.065	NA	1.87E–02
^{135}Cs	2.3E+06	2.22E+00	2.70E–01
^{137}Cs	30.07	7.43E+04	1.13E+04
^{147}Pm	2.623	NA	5.04E+02
^{151}Sm	90	3.51E+03	4.57E+02
^{150}Eu	36	4.08E+02	NA
^{152}Eu	13.54	2.55E+03	1.82E+02
^{154}Eu	8.593	1.14E+03	1.95E+01
^{155}Eu	4.75	NA	8.56E+01
$^{166\text{m}}\text{Ho}$	1200	5.39E+00	NA
^{232}Th	1.40E+10	2.18E+00	NA
^{232}U	69.8	2.29E+01	NA
^{233}U	1.59E+05	1.73E+01	NA
^{234}U	2.46E+05	1.43E+01	NA
^{235}U	7.04E+08	3.18E–01	NA
^{236}U	2.34E+07	3.47E–01	1.40E–01
^{238}U	4.47E+09	1.65E+00	NA
^{237}Np	2.14E+06	1.80E+00	2.50E–01
^{238}Pu	87.7	1.30E+03	1.82E+02
^{239}Pu	2.41E+04	5.92E+03	1.10E+03
^{240}Pu	6560	1.55E+03	3.00E+02
^{241}Pu	14.4	1.05E+04	3.88E+03
^{242}Pu	3.75E+05	5.99E–01	9.20E–03
^{241}Am	432.7	1.34E+03	3.73E+02
^{243}Am	7370	2.62E–01	NA
^{244}Cm	18.1	1.55E+02	NA
Total		2.13E+06	1.74E+05

^a From Baum et al. (2002). Read $7.1\text{E}+5$ as 7.1×10^5 .

^b Radioactivities on September 23, 1992 reported in Smith et al. (2003) were decay corrected to January 1, 2008. NA: not available.

^c Radioactivities on May 1, 1996 reported in IAEA (1998) were decay corrected to January 1, 2008.

Table 4
Nuclear power and geological repository in major countries^a

Country	Operating plants	Nuclear electricity generation in 2006 (billions kWh)	Nuclear power reliance (%)	Nuclear power of world total (%)	Projected repository operation
Belgium	7	44.3	54	1.7	Between 2035 and 2080
Canada	18	92.4	16	3.5	After 2034
China	11	51.8	1.9	1.9	At earliest 2040
Czech Republic	6	24.5	31	0.9	After 2030
Finland	4	22	28	0.8	Site selected; emplacement in 2020
France	59	428.7	78	16	By 2025
Germany	17	158.7	32	6.0	No projected date
India	17	15.6	2.6	0.6	TBD
Japan	55	291.5	30	11	At earliest 2035
Korea (South)	20	141.2	39	5.3	TBD
Russia	31	144.3	16	5.4	After 2025
Slovakia	5	16.6	57	0.6	TBD
Spain	8	57.4	20	2.2	TBD
Sweden	10	65.1	48	2.4	Around 2015
Switzerland	5	26.4	37	1.0	After 2050
Ukraine	15	84.8	48	3.2	After 2020
United Kingdom	19	69.2	18	2.6	TBD
United States	104	787.2	19	30	2017

^a From Witherspoon and Bodvarsson (2001), OCRWM (2007) and World Nuclear Association (2007a); TBD: to be determined.

concern of global warming from greenhouse gas emission of fossil fuel. If the application is approved, NRG expects to bring the units on line in 2014 and 2015.

In the former Soviet Union and the present Russian Federation, nuclear waste has also been produced from commercial power generation. Within the Russian Federation, there are about 14,000 tHM (1.8×10^8 TBq) of HLW, and 30 operating NPP at 10 locations that produce spent fuel at 850 tHM/y. In addition, there are 38 former Soviet Union NPP from which Russia will take back the SNF for reprocessing; Russia's policy of closed fuel cycle is to reprocess most of the SNF. According to Ewing (2004), the total radioactive waste of more than 4×10^8 m³ at Minatom (Russia's Ministry of Atomic Energy) facilities has more than 7.4×10^7 TBq (Fig. 1). In addition, at least 5×10^6 m³ of waste ($>3.7 \times 10^6$ TBq of radioactivity) has been injected into geological formations.

A number of countries, specifically China, India, Japan, Pakistan, the Russian Federation, and the Republic of Korea, announced plans for significant expansion of nuclear energy. In addition, Argentina, France, South Africa, Ukraine and the USA have expressed an intention to expand their existing programs (IAEA, 2007). Asia has had the most significant expansion in nuclear power generation capacity. In East and South Asia, there are over 109 nuclear power reactors in operation, 18 under construction, and another 110 planning to be built. The great growth in nuclear energy generation is expected to be in China, Japan, South Korea, and India; their current electricity contribution is listed in Table 4. The Chinese economy has been growing at an annual rate of about 10%, with electricity demand growing twice that fast. Most of mainland China's electricity is produced from fossil fuels (about 80%, mainly coal) and hydropower (about 18%). Rapid growth in demand has given rise to power shortages, and the reliance on fossil fuels has led to much air pollution; the economic loss from pollution is about 3–7% of gross domestic product (World Nuclear Association, 2007b). While coal is the main energy source, most coal reserves are in the north or northwest of China, which presents an enormous logistics problem. Nuclear power has an important role, especially in the coastal areas that are remote from the coalfields and where the economy has been developing rapidly.

Moves to build nuclear power in mainland China commenced in 1970 and the industry has now moved to a steady development phase. The first two nuclear power plants in mainland China were at Daya Bay near Hong Kong and Qinshan, south of Shanghai, with construction starting in the mid-1980s. Currently, there are 11

operating nuclear power reactors in the provinces of Zhejiang, Guangdong, and Jiangsu (all are coastal areas with rapid economic development and electricity demand), with a total net capacity of 8.6 GWe. The nuclear power is currently providing 51.8 billion kWh (kilowatts-hours), which is 1.9% of the nation's total electricity. The Chinese government plans to increase nuclear generating capacity by fivefold to 40 GWe by 2020, which requires adding an average of 2 GWe/y. In May 2007, the National Development and Reform Commission further announced that its target for nuclear generation capacity in 2030 was 160 GWe. More than 16 provinces, regions and municipalities have announced intentions to build nuclear power plants in the 20th 5-year plan (2011–2015).

When China started to develop nuclear power, a closed fuel cycle strategy was also formulated and declared at an IAEA conference in 1987. The spent fuel activities involve at-reactor storage, away-from-reactor storage, and reprocessing. Based on expected installed capacity of 20 GWe by 2010 and 40 GWe by 2020, the annual SNF generation will amount to about 600 t in 2010 and 1000 t in 2020, the cumulative SNF increasing to about 3800 t and 12,300 t, respectively. Construction of a centralized spent fuel storage facility at Lanzhou Nuclear Fuel Complex in Gansu Province began in 1994. The initial stage of that project has a storage capacity of 550 t and could be doubled.

Over 200 radionuclides are produced during the operation of a typical reactor; most of the radionuclides are relatively short-lived and decay to low levels within a few decades (Crowley, 1997). A number of radionuclides are emitted from normal operation of NPP. For example, the annual discharge of gaseous ¹⁴C to the atmosphere from pressured water reactors in Germany was 280 ± 20 GBq/GWe in 1999, on average 30% is thought to have emitted in the form of CO₂, the rest in organic form (CH₄ in particular). In France, ¹⁴C discharges were estimated to be 140 GBq/y per unit of 900 MWe and 220 GBq/y per unit of 1300 MWe (Roussel-Debet et al., 2006). Based on combined worldwide operable nuclear reactors of 3.72×10^5 MWe (World Nuclear Association, 2007a), the annual discharge of ¹⁴C worldwide is about 60 TBq/y. As a comparison, all atmospheric nuclear tests emitted about 2.13×10^5 TBq of ¹⁴C, cosmogenic natural production in the upper atmosphere is at a rate of approximately 1.54×10^3 TBq/y.

To assess the potential environmental contamination from normal operation of nuclear power plants, monitoring of radionuclides in terrestrial and aquatic environments has been performed on various environmental samples, including soils, plants (pine

needle, tea, grass, moss, algae) and foodstuff (wheat, cabbage, rice) (e.g., Lu et al., 2006; Adliené et al., 2006; Jean-Baptiste et al., 2007). To address the growing nuclear energy in China, a Radiation Monitoring Technical Center has been established in the coastal Zhejiang Province by The *State Environment Protection Administration*. The Center is mainly responsible for radiological monitoring and radioactive waste management, which include national radiation monitoring network management, data collection and analyses, preparation of technical guidelines and standards, training, and quality assurance/quality control. The Center compiles and publishes an annual report about the radiation monitoring results near the NPP and other nuclear facilities in China.

2.4. Uranium mining and milling

As integral parts of nuclear fuel cycle, uranium mining (ore extraction) and milling (physical and chemical extraction of U from the ore) have generated the *largest* volume of radioactive waste. Worldwide, the total estimated volume of mill tailing is $938 \times 10^6 \text{ m}^3$ produced at about 4384 mines, and the radioactivity of these tailings depends on the grade of ore mined and varies from less than 1 Bq/g to more than 100 Bq/g (Abdelouas, 2006). With only 19 mines, Kazakhstan has produced by far the largest volume of tailings ($209 \times 10^6 \text{ m}^3$). With a total of 3900 mines, the tailing production in the USA is $120 \times 10^6 \text{ m}^3$, representing 13% of the global inventory. In another report of Ewing (2004), the radioactive waste inventory from uranium mine and mill tailings in 2010 was estimated to reach $438 \times 10^6 \text{ m}^3$, with a radioactivity of $1.1 \times 10^8 \text{ TBq}$; this compares to $9.3 \times 10^7 \text{ TBq}$ from weapons program SNF and $1.5 \times 10^9 \text{ TBq}$ from civilian SNF. Under the Uranium Mill Tailing Remedial Action (UMTRA) program established in 1979, the U.S. Department of Energy has completed the surface remediation of 24 sites in 10 states. By the end of 1999, the total accumulated remediation cost had reached US\$1.5 billion; active remediation will require another decade of effort (Abdelouas, 2006).

The radionuclides in uranium mill tailings include uranium isotopes (^{238}U , ^{235}U , and ^{234}U), ^{230}Th , ^{226}Ra , and ^{222}Rn ; both ^{238}U and ^{230}Th are long-lived α -emitters. ^{222}Rn , an inert radioactive gas with a short half-life (3.8 days), has been identified as an important carcinogen. Radon concentrations persist in mill tailings because ^{222}Rn is a decay product of the longer-lived ^{226}Ra (half-life 1600 years), with the gamma radiation constituting the principal radiation risks from uranium tailings. In addition to radioactivity, uranium mill tailings are often associated with elevated concentrations of highly toxic heavy metals. Because of the oxidation of high sulfide content (a few to tens of wt%), uranium tailings generate acidic waters and accelerates the releases of radioactive and hazardous elements (Abdelouas, 2006).

Typically, a light water reactor producing 900 MW of electrical power uses 20 t of metallic uranium per year, which requires the mining of 17,000 t of 1 wt% uranium ore (Abdelouas, 2006). In 2003, 36,000 t of metallic U was produced, which is about half of the annual utilities' requirements. After 2015, the mine production will increase because of diminishing uranium stockpiles. Even with the increasing build-up of nuclear power plants, it is estimated that the current uranium resource of more than 23 million tones is about twice the amount required for the worldwide needs up to 2100 (Macfarlane and Miller, 2007).

2.5. Commercial fuel reprocessing

At present, the worldwide capacity to reprocess SNF is between one-third and one-half of the annual production rate of SNF ($\sim 10,000 \text{ tHM/y}$). In operation since 1976, La Hague plant located on the Cotentin Peninsula in Normandy, France is the world's

largest commercial fuel reprocessing facility. With a capacity of 1650 tHM/y, the plant processed a total of 1100 t in 2005, and recovered plutonium is sent to Marcoule nuclear site (which served French defense programs and also reprocessed gas-cooled reactor fuel) where MOX fuel is fabricated. La Hague treats spent nuclear fuel from France, Japan, Germany, Belgium, Switzerland and the Netherlands. The non-renewable waste is eventually sent back to the user nation, as established under international law.

With a 900 t/y capacity, THORP (Thermal Oxide Reprocessing Plant), designed for reprocessing uranium oxide fuel, in Sellafield, England has reprocessed a total of 8000 t nuclear fuel, including those from the United Kingdom, Japan, Germany, and Switzerland (Bellona, 2003). Construction of THORP started in the 1970s and was completed in 1994; the plant went into operation in August 1997. In addition, the Magnox Reprocessing Plant (B205) in Sellafield began commercial operation in January 1964 and has an annual capacity of 1500 t to reprocess other nuclear fuels (metallic uranium fuel, and now fuels from Britain's Magnox reactors) (Bellona, 2003). Webb et al. (2006) summarized incidents and accidents, at Sellafield over the period 1950–2000, which had off-site radiological consequences.

In addition to these main commercial reprocessing facilities in France and England, there are several major reprocessing facilities in the world. The Mayak facility in Russia can reprocess 400 t nuclear fuels each year. Kalpakkam atomic reprocessing plant in India has an annual reprocessing capacity of 275 t. Since 1993, about 2 billion U.S. dollars have been invested in constructing the most recent major reprocessing plant in Rokkasho, Japan. With a capacity of 800 t/y, the Rokkasho plant is currently undergoing test operations, separating a small amount of used nuclear fuel. It is the successor to a smaller reprocessing plant located in Tokai, Ibaraki. A pilot (50 t/y) reprocessing plant using the Plutonium–Uranium Extraction (PUREX) process was opened in 2006 in Lanzhou, China. This plant could be fully operational in 2008 and is capable of expansion to 100 t/y. A large commercial reprocessing plant based on indigenous advanced technology is planned to follow and begin operation in China about 2020 (World Nuclear Association, 2007b).

Radionuclides have been discharged, with government authorization, over the past forty years into the sea and atmosphere from the nuclear fuel reprocessing plants at Sellafield and at La Hague. Past discharges from Sellafield are well documented in Gray et al. (1995), Jackson et al. (1998), and Bellona (2003). Based on a number of assumptions about effluent management decisions, regulatory requirements and the nature of future business at Sellafield, Jones et al. (2004) calculated expected future discharges for the period 2001–2028. During the years between 1974 and 1978, for example, the Sellafield plant's annual discharge of ^{137}Cs was 4000 TBq. The plutonium discharge in the same period was between 45 and 60 TBq annually (Bellona, 2003). Over those five years, more than twice as much plutonium was discharged from Sellafield as was released during the 1986 Chernobyl accident, where discharges amounted to about 100 TBq of long-lived plutonium (UNSCEAR, 2000). Moreover, La Hague has significantly increased liquid radioactive carbon release over the past two decades from less than 1 TBq/y prior to 1983 up to about 10 TBq/y in 1999, and the release was still above 7 TBq/y up to 2004 (Fiévet et al., 2006).

Discharge of ^{99}Tc from Sellafield has also been high. In the three-year period from 1978 to 1980, Sellafield discharged almost 300 TBq of ^{99}Tc (Bellona, 2003). From an average of 4–6 TBq/y throughout the entire 1980s, the discharges of ^{99}Tc increased to 190 TBq in 1995. In the five-year period 1994–1998, a staggering 530 TBq of ^{99}Tc was discharged from Sellafield. Such increased discharges of technetium come as a result of reprocessing of Magnox fuel. At a rate of about 90 TBq/y, current and future discharge of ^{99}Tc contributes a significant portion of the total released radioactivity (Jones et al., 2004).

Today, it is the large release of the radioactive ^{99}Tc from Sellafield that is largely responsible for the pollution of the Norwegian coast and the Barents Sea. To a less extent, La Hague has also been discharging ^{99}Tc , which increased slowly from ~ 0.4 TBq/y in 1970 to a peak of ~ 25 TBq/y in 1985. By 1991 the discharge had fallen to ~ 0.9 TBq/y and have since remained relatively stable at < 1 TBq/y (Keogh et al., 2007).

From 1966 to 1999, a total of 2300 kg (15 TBq) of liquid ^{129}I was discharged into the marine environment from La Hague and another 64.2 kg (0.42 TBq) gaseous ^{129}I released to the atmosphere (Fréchou and Calmet, 2003). Up until 1998, 2600 kg (17 TBq) of ^{129}I was discharged into the sea by La Hague and Sellafield, an amount that is 50 times the total release from nuclear weapons tests' and three orders of magnitude higher than the Chernobyl accident release (Hou et al., 2001). After 1998, ^{129}I has been discharged at about 0.2 TBq/y from Sellafield and negligibly from La Hague (Keogh et al., 2007). The total release of ^{129}I was estimated to be about 255 kg (1.7 TBq) and 4000 kg (26 TBq) from gaseous and liquid discharges, respectively (Aldahan et al., 2006). From 1944 through 1972, the plutonium-production operation at the Hanford Site released about 260 kg (1.7 TBq) of ^{129}I into the air. In comparison, the operation of production reactors from 1953 to about 1990 at the Savannah River Site released about 32 kg (0.21 TBq) of ^{129}I into the air.

In addition to environmental contamination, a principal concern with fuel reprocessing has always been the possibility of the diversion of fissile material, mainly ^{235}U and ^{239}Pu , for weapons production. However, other fissile nuclides, such as ^{237}Np and Am, may be separated during reprocessing. In 1997, the global inventory of ^{237}Np and $^{241, 242, 243}\text{Am}$, with the principal isotope of interest being ^{241}Am from the decay of ^{241}Pu , was estimated to be 80 t (enough for several thousand nuclear weapons), and this inventory grows at a rate of approximately 10 t/y (Ewing, 2004).

As the bare critical mass of ^{239}Pu is less than 10 kg, the potential diversion of civil-source Pu, originally destined for fabrication of mixed-oxide (U + Pu) fuel, to the production of a nuclear weapon is an extremely important proliferation and environmental problem (Ewing, 2004). France and Britain have the largest inventories, with 72 t (of which 33.6 t is foreign owned) and 60 t (6.1 t foreign owned), respectively. Both the USA and Japan have 5 t of civil-source Pu, but Japan has another 24.1 t in other countries (mostly in the La Hague reprocessing facility of France). Of equal importance are the stockpiles of highly enriched ^{235}U (HEU), with 645 t in the USA and 1050 t in Russia. The USA agreed to purchase 500 t of weapons uranium from Russia to blend down to low enriched ^{235}U (LEU) for use in commercial reactors. As of 2003, 193 t of bomb-grade HEU has been converted to 5700 t LEU of 4% ^{235}U for use in light water reactors (LWRs). This amount of HEU could have been used to produce over 7700 nuclear weapons (Ewing, 2004). Moreover, during the past decade, plutonium from dismantled nuclear weapons has emerged as a new waste type with proliferation implications (Ewing, 2004). The disarmament process, under Strategic Arms Reduction Treaties between the USA and Russia, of thousands of nuclear weapons will produce 30–40 t of weapons-grade plutonium in each country, as well as hundreds of tons of HEU.

The Nuclear Fuel Service in the U.S. operated a nuclear fuel reprocessing center from 1966 to 1972 in West Valley, New York and processed 640 t of SNF. During the operation, 2500 m³ of highly radioactive liquid waste was generated and stored in an underground waste tank, which was vitrified during 1996–2002. In 1977, fear of nuclear weapons proliferation (especially after India demonstrated nuclear weapons capabilities using reprocessing technology) led President Carter to issue a Presidential directive to indefinitely suspend the commercial reprocessing and recycling of plutonium in the U.S.

Initiated in 2006 by the USA, the Global Nuclear Energy Partnership (GNEP) is a comprehensive strategy to expedite the development of nuclear power around the world while improving the use of resources and providing greater disincentives to the proliferation of nuclear weapons. The GNEP involves both political and technological initiatives. A global nuclear energy partnership involves nations with advanced civilian nuclear energy programs, such as France, Japan, China, and Russia, aiming to improve the proliferation-resistance of the nuclear fuel cycle while guaranteeing access to fuel supplies. Technological initiatives include reprocessing spent uranium fuel for use in advanced reactors to help extract more energy and to have the potential of reducing storage requirements for nuclear waste by up to 90 percent. The GNEP calls for construction of the world's largest nuclear fuel reprocessing facility by 2020, capable of reprocessing 2000–3000 t of SNF a year; large enough to reprocess all the waste generated annually by the U.S.'s 104 commercial nuclear power plants (Johnson, 2007).

2.6. Geological repository of high-level nuclear wastes

Altogether, currently there are 439 nuclear power plants around the world, operating in 31 countries. All countries using nuclear power must deal with high-level radioactive waste, regardless of the number of reactors they have in service. Nuclear energy production eventually creates waste in the form of spent nuclear fuel, which is depleted or used fuel from nuclear power plants and research facilities. Spent nuclear fuel is highly radioactive, and remains so for thousands of years. Isolating this high-level waste from people and the environment has been an important and challenging issue for all countries that use nuclear power. Social, economic, political, technical, and geographic considerations shape a country's radioactive waste management decisions.

High-level waste makes up the smallest volume of radioactive waste, around 3% of the world's total, but it contains approximately 95% of all the radioactivity in low- and high-level waste combined. In the United States, high-level radioactive waste primarily comes from defense-related reprocessing of spent nuclear fuel at Hanford and Savannah River sites. All countries with high-level radioactive waste and spent nuclear fuel plan to eventually dispose of these materials deep underground, in a geologic disposal facility called a repository (NRC, 1957; Mobbs et al., 1988; Witherspoon and Bodvarsson, 2001).

The HLW is destined for vitrification in a borosilicate glass and then emplaced in a geological repository for permanent disposal, along with the SNF. Over 500 t of HLW has been vitrified in France and Germany. At the Mayak facility, about 1.1×10^7 TBq HLW from reprocessing has been immobilized into a Na–Al phosphate glass. In the USA during 1996–2001, the HLW was vitrified into 275 stainless steel canisters (each canister weighs about 2-t, and is 0.61 m in diameter and almost 3.05 m high) in the West Valley Plant of New York. The Defense Waste Processing Facility at Savannah River started operation in 1996 after 18 years of planning and \$2 billion in construction costs (Crowley, 1997), and have produced about 1600 canisters by February 2004 (Ewing, 2004). Each Savannah River canister holds about 1.68 t of glass, of which about 42.6 kg will be HLW; radioactivity of the waste in an individual canister will be as high as 8.7×10^3 TBq. It is estimated that 6000–8000 such canisters will be required to hold existing and projected waste at Savannah River. Another vitrification plant is under construction at Hanford, wherein all the HLW will be vitrified into about 20,000 canisters for final disposal in the proposed Yucca Mountain repository, which is located on the western edge of the NTS in a remote desert on federally protected land.

The U.S. DOE began studying Yucca Mountain, Nevada, in 1978 to determine whether it would be suitable for the nation's first

long-term geologic repository for spent nuclear fuel and high-level radioactive waste. On July 23, 2002, President Bush signed a Resolution, allowing the DOE to take the next step in establishing a safe repository in which to store the nation's nuclear waste. The DOE is currently in the process of preparing an application to obtain the NRC license to proceed with the construction of the repository. In July of 2006, the DOE announced plans to submit a license application to NRC by June 30, 2008, and to initiate repository operations in 2017.

Based on the currently legislated repository capacity of 70,000 MTHM, there will be a total of 7860 packages of commercial spent nuclear fuel, 3910 packages of defense spent nuclear fuel, and 3910 packages of defense high-level nuclear waste for disposal in the proposed Yucca Mountain geological repository (BSC, 2001). With the reported radionuclide (a total of 26) inventory in each package, about 2.6×10^8 TBq of radioactivity from high-level nuclear waste and spent fuel will be stored for many generations to come (Table 5; Fig. 1). Similar to the radionuclides from underground nuclear tests, ^{90}Sr , ^{137}Cs , ^{241}Am , and plutonium isotopes will be of great importance over the short-term, while long-lived ^{14}C , ^{36}Cl , ^{99}Tc , ^{129}I , ^{237}Np , ^{243}Am , and several uranium and plutonium isotopes will become important over the long-term.

After forty years of commercial nuclear power generation, no country has disposed of HLW (Table 4). Along with the USA, Belgium, Canada, China, Finland, France, Germany, Japan, Russia, Spain, Sweden, Switzerland and the United Kingdom have invested significant resources in their radioactive waste management programs because of their historic or anticipated reliance on nuclear energy. Deep geologic disposal has been accepted as the best method for isolating highly radioactive, long-lived waste (Witherspoon and Bodvarsson, 2001). Many countries have been developing their repository at different paces to cope with the HLW. For example, high-level wastes in China will be vitrified, encapsulated, and put into a geological repository (probably granite) some 500 m deep. Site selection is focused on six candidate locations and

will be completed by 2020. An underground research laboratory will then operate for 20 years and actual disposal is anticipated to occur from 2040 (Wang et al., 2001).

2.7. Nuclear accidents

Radioactive contamination of the environment has occurred as a result of nuclear accidents, particularly by the U.S. and former Soviet Union (e.g., the accidents in Mayak facility in Section 2.1) considering the magnitude of their nuclear weapons' programs and nuclear power generation. The most notable is the catastrophic accident that destroyed Unit 4 of the Chernobyl nuclear complex in the Ukraine in April of 1986. It was estimated that 1.2×10^7 TBq of radioactivity was released in the Chernobyl accident (UNSCEAR, 2000), and the noble gases contributed about 50% of the total release; the principal radionuclides and their inventories are presented in Table 6. Eikenberg et al. (2004) compared the total atmospheric release of long-lived dose-relevant fission radionuclides and actinides from the atomic bomb tests and the Chernobyl reactor explosion. Compared to the sum of all previously performed atmospheric bomb tests, the values for ^{90}Sr , ^{137}Cs , and $^{239+240}\text{Pu}$ from the Chernobyl accident were in the order of 10% and much higher for ^{238}Pu and ^{241}Am . Fallout of hot particles in the vicinity of the reactor caused a considerable contamination of the soil surface, with ^{137}Cs up to 10^6 Bq/m², and 116,000 people had to be evacuated within a zone of 30 km distance from the reactor (Balonov, 2007; Izrael, 2007). The contribution of ^{137}Cs from the Chernobyl plume was significant even 2000 km away; however, the total surface contamination was at least two orders of magnitude below the level within the 30-km exclusion zone. In all, there were 28 deaths from the acute radiation syndrome as a result of the Chernobyl accident (Eisenbud and Gesell, 1997).

Being the world's first major accident at a civilian power plant, the Three Mile Island Unit II accident in 1979 in Pennsylvania has played a significant role in effectively stopping the growth of

Table 5
Radionuclide inventory in potential Yucca Mountain repository

Radio-nuclide	Half-life (yr)	Radioactivity (TBq)			Total radioactive mass (kg)	Total radioactivity (TBq)	Percentage of total inventory (%)
		Commercial spent nuclear fuel	Defense spent nuclear fuel	Defense high-level waste glass			
^{14}C	5715	1.78E+03	3.03E+01	4.16E+00	1.10E+01	1.81E+03	6.98E-04
^{90}Sr	28.78	9.00E+07	1.11E+06	5.76E+06	1.89E+04	9.69E+07	37.3
^{99}Tc	2.13E+05	3.79E+04	2.82E+02	1.79E+03	6.37E+04	4.00E+04	1.54E-02
^{129}I	1.57E+07	9.25E+01	6.41E-01	1.23E+00	1.44E+04	9.43E+01	3.63E-05
^{137}Cs	30.07	1.35E+08	1.41E+06	5.67E+06	4.42E+04	1.42E+08	54.6
^{210}Pb	22.6	0.00E+00	1.53E-04	1.24E-03	5.00E-07	1.39E-03	5.37E-10
^{226}Ra	1599	0.00E+00	2.68E-04	2.39E-03	7.26E-05	2.66E-03	1.02E-09
^{228}Ra	5.76	0.00E+00	2.75E-01	1.26E-01	3.98E-05	4.00E-01	1.54E-07
^{227}Ac	21.77	6.50E-02	1.18E+00	4.89E+00	2.29E-03	6.13E+00	2.36E-06
^{231}Pa	3.25E+04	1.37E-01	2.24E+00	5.48E+00	4.46E+00	7.86E+00	3.02E-06
^{229}Th	7880	0.00E+00	7.62E-01	1.17E-01	1.20E-01	8.79E-01	3.38E-07
^{230}Th	7.54E+04	1.10E+00	3.16E-02	2.33E-02	1.52E+00	1.16E+00	4.45E-07
^{232}Th	1.40E+10	0.00E+00	2.37E-01	1.16E-01	8.68E+04	3.54E-01	1.36E-07
^{232}U	69.8	6.48E+01	4.69E+02	2.63E+00	6.57E-01	5.37E+02	2.07E-04
^{233}U	1.59E+05	1.96E-01	2.99E+02	1.55E+01	8.81E+02	3.14E+02	1.21E-04
^{234}U	2.46E+05	3.30E+03	5.14E+01	4.24E+01	1.48E+04	1.30E+03	1.31E-03
^{235}U	7.04E+08	3.95E+01	2.60E+00	5.31E-01	5.33E+05	4.26E+01	1.64E-05
^{236}U	2.34E+07	7.37E+02	7.98E+00	3.72E-01	3.12E+05	7.46E+02	2.87E-04
^{238}U	4.46E+09	7.74E+02	2.47E+00	1.27E+01	6.35E+07	7.89E+02	3.04E-04
^{237}Np	2.14E+06	9.70E+02	4.87E+00	7.36E+00	3.77E+04	9.82E+02	3.78E-04
^{238}Pu	87.7	7.52E+06	1.57E+04	2.31E+05	1.23E+04	7.76E+06	2.99E+00
^{239}Pu	2.41E+04	7.90E+05	2.06E+04	7.53E+04	3.86E+05	8.86E+05	3.41E-01
^{240}Pu	6560	1.38E+06	1.61E+04	1.25E+04	1.68E+05	1.41E+06	5.42E-01
^{242}Pu	3.73E+05	6.22E+03	6.35E+00	4.45E+00	4.26E+04	6.24E+03	2.40E-03
^{241}Am	432.7	1.09E+07	5.80E+04	3.26E+04	8.64E+04	1.10E+07	4.21E+00
^{243}Am	7370	7.49E+04	4.30E+01	1.15E+01	1.01E+04	7.49E+04	2.88E-02
Total		2.46E+08	2.63E+06	1.18E+07	6.53E+07	2.60E+08	100

Table 6
Estimate of principal radionuclides released in the Chernobyl accident

Radionuclide		Radioactive half-life ^a	Activities released (TBq) ^b
Noble gases	¹³³ Xe	5.243 d	6.50E+06
Volatile	¹³² Te	3.20 d	1.15E+06
	¹³¹ I	8.04 d	1.76E+06
	¹³⁴ Cs	2.065 a	5.40E+04
	¹³⁷ Cs	30.07 a	8.50E+04
Intermediate	⁸⁹ Sr	50.52 d	1.15E+05
	⁹⁰ Sr	28.78 a	1.00E+04
	¹⁰³ Ru	39.27 d	1.68E+05
	¹⁰⁶ Ru	1.02 a	7.30E+04
	¹⁴⁰ Ba	12.75 d	2.40E+05
	⁹⁵ Zr	64.02 d	1.96E+05
Refractory (including fuel particles)	⁹⁹ Mo	2.7476 d	1.68E+05
	¹⁴¹ Ce	32.50 d	1.96E+05
	¹⁴⁴ Ce	284.6 d	1.16E+05
	²³⁹ Np	2.355 d	9.45E+05
	²³⁸ Pu	87.7 a	35
	²³⁹ Pu	24,100 a	30
	²⁴⁰ Pu	6560 a	42
	²⁴¹ Pu	14.4 a	6.00E+03
	²⁴² Cm	162.8 d	900

^a From Baum et al. (2002).

^b From UNSCEAR (2000).

nuclear power in the U.S. With $0.92\text{--}3.7 \times 10^5$ TBq radioactive gas release, this accident did not cause notable radiation effects on individuals living in the vicinity of the reactor (25,000 people lived within 8 km of the site at the time of the accident), yet the socio-political ramifications of the accident were evident. The Three Mile Island accident was a significant turning point in the global development of nuclear power. From 1963 to 1979, the number of reactors under construction globally increased every year except in 1971 and 1978. However, following the Three Mile Island accident, the number of reactors under construction declined every year from 1980 to 1998.

As of October 2000, there were about 400 reactors in nuclear-powered vessels around the world (IAEA, 2001). Of these, the Russian Federation had 75 active military vessels with about 150 reactors, and the USA had about 117 nuclear vessels. There are six confirmed nuclear submarine accidents since 1963 at various sites in the Atlantic Ocean (Table 7). Though there were some radioactive sources in these submarines, the released radioactivity was relatively small (Table 7). With the exception of the Russian submarine Kursk, which lies at a depth of 108 m in the Barents Sea, all other submarines are too deep for the submarine or reactor(s) to be recovered.

The world's first nuclear icebreaker Lenin was in operation from 1959 until 1989. During that time, there have been two serious accidents onboard (Bellona, 2008). The *Lepse*, a 5000-t nuclear service vessel moored on the Kola Peninsula near Murmansk, has accumulated 639 nuclear fuel assemblies (about 320 are damaged)

Table 7
Radioactivity involved in the nuclear submarine accident^a

Country	Date	Vessel involved	Radioactive material involved	Total activity (TBq)	Estimated activity released
USA	4/10/1963	SNN-593 "Thresher"	Nuclear reactor	1150	0.04 GBq
	5/22/1968	SNN-583 "Scorpion"	Nuclear reactor; two nuclear warheads	1300	0.04 GBq
Soviet Union	4/8/1970	K-8	2 Reactors	9250	–
			Warheads(s)	0.03	
	10/6/1986	K-219	2 Reactors	9250	–
	4/7/1989	K-278 "Komsomolets"	Reactor core	3590	<370 GBq
Russian Federation	8/12/2000	K-141 "Kursk"	2 Reactors	$1\text{--}2 \times 10^6$	–

^a IAEA (2001).

and 39 m^3 of solid radioactive waste; the spent fuel contains about 2.8×10^4 TBq radioactivity (Webster, 2003).

Nuclear energy sources are also used in some spacecraft, satellites, and deep sea acoustic signal transmitters for generation of heat or electricity, with two common types of nuclear energy sources: radioisotope thermoelectric generators (RTGs) and nuclear reactors. A typical RTG contains approximately 1000 TBq ²³⁸Pu or 10,000 TBq ⁹⁰Sr. On April 21, 1964, the U.S. navigational satellite "Transit 5BN-3" with a radioisotope generator containing 630 TBq (about 1 kg) of ²³⁸Pu failed to achieve orbit and burned up over the West Indian Ocean; the nuclear fuel was vaporized and dispersed worldwide (IAEA, 2001). By mid-1970, 95% of the ²³⁸Pu was found to deposit on the Earth's surface, which almost tripled the global deposit of this Pu isotope by 1970 (Hardy et al., 1973).

Lighthouses in remote Russian waters are often powered by RTGs, which may contain up to several thousand TBq of ⁹⁰Sr; there are some 500 RTGs in use. There were two recorded incidents in 1987 and 1997 and where RTGs were lost at sea and contained about 2.5×10^4 and 1.3×10^3 TBq of sealed ⁹⁰Sr sources, respectively (IAEA, 2001). Spread of orphan nuclear sources and their potential use in radiological dispersion devices (RDD) by terrorist groups have been a continuing concern (e.g., Sohier and Hardeman, 2006). Because of the radiotoxicity, widespread use and sufficiently long half-life, 7 radionuclides are of particular concern in the RDD: ⁶⁰Co, ⁹⁰Sr, ¹³⁷Cs, ¹⁹²Ir, ²³⁸Pu, ²⁴¹Am, and ²⁵²Cf. Commercial radioactive sources for potential RDD include RTG (⁹⁰Sr), teletherapy and irradiators (⁶⁰Co and ¹³⁷Cs), industrial radiography (⁶⁰Co and ¹⁹²Ir), well logging and moisture detectors (¹³⁷Cs, ²⁴¹Am, and ²⁵²Cf). Social, economic, and environmental effects of deliberate use of RDD were reflected in two serious accidents in which abandoned teletherapy units caused extensive contamination, acute radiation effects, and, in one case, fatalities. In December 1983, a teletherapy unit that contained 16.7 TBq of ⁶⁰Co was dismantled to be sold as scrap in Juarez, Mexico; rupture of a radioactive source caused serious contamination (Eisenbud and Gesell, 1997). A more serious accident occurred in September 1987 in the state of Goiania, Brazil. A teletherapy unit that contained 51 TBq of ¹³⁷Cs in the form of CsCl₂ powder was left in an abandoned medical clinic (Fig. 1). From various attempts of scavenging of this unknown object, CsCl₂ was found to be distributed over an area of about 1 km²; four people ultimately died of acute radiation dose, one person required amputation of an arm, and 129 persons had measurable body contamination (of whom 21 were hospitalized from serious skin burns or blood changes) (Eisenbud and Gesell, 1997).

3. Salient geochemical behavior of important radionuclides

Of the suite of important radionuclides (³H, ¹⁴C, ⁹⁰Sr, ⁹⁹Tc, ¹²⁹I, ¹³⁷Cs, ²³⁷Np, ²⁴¹Am, as well as several uranium and plutonium isotopes), ³H is considered an ideal water tracer, although small amounts of tritium retardation have been reported in transport studies, and ¹⁴C may be retarded by its isotopic exchange with

carbonate minerals in the aquifer (Hu et al., 2008). ^{90}Sr and ^{137}Cs are the major fission products, yet they do not pose long-term risk because of their relatively short half-lives and strong sorption in the subsurface. In general, the mobility of actinides in aqueous systems is low, dependent on (1) their thermodynamic properties, which determine solubility and speciation as a function of pH and redox potential, (2) the availability of inorganic ligands to form soluble complexes, and (3) the composition and abundance of minerals and mineral colloids present in the system (Dozol and Hagemann, 1993; Silva and Nitsche, 1995).

Of particular importance to the environment and risk assessment are radionuclides ^{99}Tc , ^{129}I , and ^{237}Np , because of their long half-lives and presumably high mobility (de Marsily et al., 1977; Bondietti and Francis, 1979; Hu and Smith, 2004). These nuclides are present in high abundance in underground nuclear test, fuel reprocessing, and in spent reactor fuel (Table 8). Because of their long half-lives (2.13×10^5 and 1.57×10^7 years for ^{99}Tc and ^{129}I , respectively) and presumed mobile behavior in groundwater, both ^{99}Tc and ^{129}I are important dose contributors to the calculated health risk for many U.S. DOE nuclear facilities, including the NTS (Smith et al., 2003), Hanford Site (Kaplan and Serne, 1998; Um et al., 2004), Savannah River Site (Beals and Hayes, 1995), and Idaho National Laboratory (Beasley et al., 1998). ^{237}Np also has a long half-life (2.14×10^7 years), and weak sorption with high mobility potential in the subsurface (Arnold et al., 2006).

With a high-abundance (6%) fission yield, ^{99}Tc has been estimated to have 25–30 t produced worldwide up to the mid-1980s with approximately 1% of that amount (150–200 TBq) released to the environment (Dowdall et al., 2005). Activities conducted at European nuclear reprocessing facilities (mainly Sellafield and La Hague) have led to the increased radioactivity in the Arctic marine environment. Increased discharge of ^{99}Tc from Sellafield beginning in 1994 has yielded an average ^{99}Tc level of $1.3 \pm 0.3 \text{ Bq/m}^3$ in seawater and $320 \pm 70 \text{ Bq/m}^3$ in seaweed (Dowdall et al., 2005).

Dependent upon the redox conditions, Tc exists in two stable oxidation states. It forms a reduced species [predominantly Tc(IV)] at redox potential (Eh) values below about 220 mV with respect to standard hydrogen electrode in neutral pH conditions. At higher Eh, it occurs as Tc(VII)O_4^- . Due to its weak interaction with mineral surfaces, TcO_4^- is considered one of the most mobile radionuclides in the environment. In contrast, transport of Tc(IV) species ($\text{TcO}_2 \cdot n\text{H}_2\text{O}$) are expected to be strongly retarded because of sorption and/or precipitation; the solubility of $\text{TcO}_2 \cdot n\text{H}_2\text{O(s)}$ in carbonate-containing groundwater was reported to be only about 10^{-8} M (Eriksen et al., 1992). Similarly, chemical equilibrium modeling of groundwater at Yucca Mountain indicated a maximum Tc solubility of $4 \times 10^{-9} \text{ M}$ under reducing conditions (Arnold et al.,

2006). Experiments conducted on sediment–groundwater samples in Germany showed dramatically different sorption coefficients for Tc among aerobic and anaerobic conditions (Lieser and Bauscher, 1987). By varying the redox potential, they observed a change in the K_d value of about three orders of magnitude over a small range of Eh at $170 \pm 60 \text{ mV}$ and a pH of 7 ± 0.5 .

As with ^{99}Tc , ^{129}I has a unique and complex chemistry in the environment. The fate and transport of ^{129}I is dictated by its chemical speciation. Aqueous iodine usually occurs as the highly mobile iodide anion (I^-). Under more oxidizing conditions, iodine may be present as the iodate anion (IO_3^-), which is more reactive than iodide and could be sorbed onto positively charged sites existing locally in clays and organic matter (Couture and Seitz, 1983; Sheppard and Thibault, 1992; Hu et al., 2005). In contrast to ^{99}Tc , iodine has a minimal retardation under reducing conditions when I^- is the predominant form, and multiple iodine species (I^- , IO_3^- , and organic iodine species) are known to coexist in various aqueous systems (cf., Hu et al., 2005).

Eisenbud and Gesell (1997) reported that 93 TBq of ^{129}I was produced by the nuclear power industry up to the year 2000. Over the duration of 1956–1988, the PUREX plant at Hanford processed more fuel than all the other Hanford separation plants combined. With the cumulative release of 0.873 TBq ^{129}I , the PUREX plant was responsible for the majority of ^{129}I emissions at the Hanford Site; more than 90% of the total ^{129}I was present as vapor phase material (Fritz and Patton, 2006). From the monitoring of ^{129}I activity in air and milk samples, atmospheric emissions were found to be the major sources of ^{129}I in environmental samples.

Study of short-lived iodine isotopes provides useful information about iodine species released from nuclear facilities and its subsequent environmental behavior. According to UNSCEAR (2000), the ^{131}I (half-life 8.04 days) inventory in the accidental reactor core at Chernobyl was estimated to be $3.2 \times 10^6 \text{ TBq}$ and the total ^{131}I release during the course of the accident was about $1.7 \times 10^6 \text{ TBq}$ (Table 6). From four air samples collected at 14.5 m above the ground surface in Japan 10–23 days after the Chernobyl nuclear reactor accident date, Noguchi and Murata (1988) reported the proportions of airborne ^{131}I species during the Chernobyl accident: $19 \pm 9\%$ particulate iodine (iodine attached to particulate matter of mean diameter of $0.8 \mu\text{m}$), $5 \pm 2\%$ I_2 , $6 \pm 3\%$ HIO and other non- I_2 inorganic iodines, and $70 \pm 11\%$ organic iodines (e.g., CH_3I). The depositional velocity of radioactive iodine from air to vegetation and water bodies, and subsequent migration, varied with the iodine speciation. Other studies on species distribution of radioactive iodine in and around nuclear power plants and of stable iodine in the environment have also indicated the important proportion of organic iodines. Aldahan et al. (2006) reported that the main forms of iodine released into the environment from European reprocessing facilities are alkyl iodides, CH_3I and iodides (e.g., HI) that transform into water-soluble molecules and/or IO_3^- , which are adsorbed on and/or nucleate into aerosols and return to Earth's surface as wet and dry fallouts.

Careful attention must be given to the iodine speciation when interpreting the biogeochemical behavior of iodine in the environment. Hu et al. (2008) studied the content and speciation of stable iodine in representative surface soils, and sorption and transport behavior of different iodine species (iodide, iodate, and 4-iodoaniline as a representative refractory organic iodine) in sediments collected at numerous nuclear facilities in the United States. In natural soils, iodine is mostly (nearly 90% of total iodine) present as organic species, while inorganic iodine becomes important (up to 50%) only in sediments with low organic matter. Results from laboratory column studies showed much greater retardation of 4-iodoaniline than iodide or iodate.

The most striking feature of neptunium chemistry in aqueous systems is the large stability range for Np(V) (e.g., Lieser and

Table 8
Inventory of ^{99}Tc and ^{129}I from various sources^a

Source	^{99}Tc		^{129}I	
	Mass (kg)	Radioactivity (TBq)	Mass (kg)	Radioactivity (TBq)
Natural hydrosphere			80	0.52
Natural atmosphere			0.0005	0.000003
Atmospheric testing			80	0.52
Chernobyl			6	0.04
Savannah River Site			32	0.21
Hanford Site	19,700	12,400	260	1.7
NTS underground nuclear testing	33.6	21.1	10	0.065
Proposed Yucca Mountain repository	63,700	40,000	14,400	94
Spent fuel reprocessing (Europe)	1340	840	4000	26

^a From Aldahan et al. (2006), Keogh et al. (2007), Moran et al. (1999), Smith et al. (2003), and Washington State Department of Health (2000).

Muhlenweg, 1988). The pentavalent NpO_2^+ species is dominant at pH values <8 whereas Np(V) carbonate complexes tend to dominate at higher pH values (Kaszuba and Runde, 1999). Since Np(V) solid phases are relatively soluble and Np(V) aqueous species do not easily sorb onto common minerals, Np(V) is relatively mobile in the environment. Under reducing conditions, Np(IV) is present as the low solubility Np(OH)_4 (aq) species at pH values >5 (Kaszuba and Runde, 1999). Np(IV) shows a strong tendency for sorption to mineral surfaces (Lieser and Muhlenweg, 1988; Nakata et al., 2002), which limits its mobility in aqueous systems. There are relatively small numbers of environmental ^{237}Np measurements, which will be changed with the interest in its long-term behavior and development of sensitive analytical methods (Keith-Roach et al., 2001; Kenna, 2002).

4. Conclusions

In this study we provide an overview of nuclear waste and contamination of anthropogenic radionuclides in the environment, as well as the salient geochemical behavior of important radionuclides. Radioactive waste legacy produced and environmental contamination by anthropogenic radionuclides from weapons' programs during the Cold War arms race are staggering and have been posing long-term political, socioeconomic, and technical challenges.

Studies of radionuclides in the environment have entered a new era, in the face of the renaissance of nuclear energy and concerns about national security with respect to nuclear non-proliferation. With the perspective of reducing the reliance on fossil fuels and the emissions of greenhouse gases, nuclear power will likely undergo rebirth in North America and Western Europe, while it has been enjoying rapidly increasing importance in the energy structure in Asia.

Knowledge of the speciation and reactions of multi-valent radionuclides, such as ^{99}Tc , ^{129}I and ^{237}Np , is very important for understanding their transport behavior in the environment and for managing and remediating radionuclide contamination at legacy nuclear operations' facilities. These radionuclides possess dynamic sorption–precipitation behavior as they are redox-sensitive elements that will respond to the change of redox condition along transport pathways. Moreover, iodine released to the environment has multiple species (inorganic and organic) with different hydrophilic, atmophilic, and biophilic characteristics, which have to be taken into in the study of environmental radioactivity. Understanding biogeochemical behavior of radionuclides is critical in meeting the challenges of the new nuclear era.

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