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Anthropogenic radionuclides in the environment

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1 Abstract

2 Studies of radionuclides in the environment have entered a new era with the renaissance
3 of nuclear energy and associated fuel reprocessing, geological disposal of high-level nuclear
4 wastes, and concerns about national security with respect to nuclear non-proliferation. This work
5 presents an overview of anthropogenic radionuclide contamination in the environment, as well as
6 the salient geochemical behavior of important radionuclides. We first discuss the following
7 major anthropogenic sources and current development that contribute to the radionuclide
8 contamination of the environment: (1) nuclear weapons program; (2) nuclear weapons testing;
9 (3) nuclear power plants; (4) commercial fuel reprocessing; (5) geological repository of high-
10 level nuclear wastes, and (6) nuclear accidents. Then, we summarize the geochemical behavior
11 for radionuclides ^{99}Tc , ^{129}I , and ^{237}Np , because of their complex geochemical behavior, long
12 half-lives, and presumably high mobility in the environment. Biogeochemical cycling and
13 environment risk assessment must take into account speciation of these redox-sensitive
14 radionuclides .

15

16 *Keywords:* contamination; environment; fuel reprocessing; nuclear power plant (NPP);
17 radionuclide; repository; waste

1 **1. Introduction**

2 During December 1938, nine months before the beginning of World War II, an Austrian
3 physicist Lise Meitner, and two German chemists, Otto Hahn and Fritz Strassmann, discovered
4 nuclear fission. This discovery culminated 40 years of research into radioactivity and ushered in
5 a nuclear era wherein peaceful, or destructive, utilization of enormous nuclear energy has been
6 entwined in the human civilization. The United States of America (USA) first began developing
7 nuclear weapons during World War II under the order of President Roosevelt in 1939, motivated
8 by a fear of a potential race with Nazi Germany to develop such a weapon. After a slow start, in
9 1942 the program was officially operated under the U.S. Corps of Engineers and became known
10 as the Manhattan Project, a mammoth effort to build the 1st atomic bomb. During the next five
11 decades, before the Cold War ended in 1991, massive weapons production and testing programs,
12 as well as the utilization of nuclear power for electricity generation, have produced a host of
13 unwanted radionuclides in different waste forms and spread environmental contamination across
14 the world.

15 Major sources of radioactive waste and contamination include the production of electrical
16 power and weapons from nuclear fuels, nuclear weapons tests, fuel reprocessing, and nuclear
17 accidents. In the USA, the total volume of all radioactive waste is 5.5 million m³ (MCM), with
18 the radioactivity of about 1.2×10^9 TBq (tera becquerel; 1 TBq=27.03 Ci) (Ahearne, 1997). In
19 addition, there are 30–80 MCM of contaminated soil and 1,800–4,700 MCM of contaminated
20 water. Among them, over 70 MCM of soil and at least 1,800 MCM of water have been
21 contaminated by releases from U.S. Department of Energy (DOE) facilities that were used for
22 weapons production (Ewing, 2004). Despite the large volumes of contaminated soil and water
23 from low- and intermediate-level wastes, these wastes only account for a few percent of the total

1 radioactivity. The majority of radioactivity comes from high-level waste (HLW) and spent
2 nuclear fuel (SNF).

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

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

19 **2. Major sources of anthropogenic radionuclides in the environment**

20 *2.1. Operations of nuclear weapons program*

21 HLW liquid and sludge are generated from weapons production and reprocessing, by
22 which fissile materials (uranium and plutonium) are separated from fission products in spent
23 nuclear reactor fuels to produce weapons-grade fuel. In the USA, there is approximately 0.38
24 MCM of such HLW with a radioactivity of 3.4×10^7 TBq. Most of the HLW (0.34 MCM) is

1 stored in tanks at Hanford, Washington, and Savannah River, South Carolina; the rest is stored as
2 a solid calcine in tanks at the Idaho National Laboratory and West Valley in New York where a
3 reprocessing plant was once operated (Ahearne, 1997).

4 Established in 1943, the Hanford Site occupies 1,517 km² in south-central Washington,
5 which is approximately half the total area of the state of Rhode Island. Between 1944 (startup of
6 the worlds' first full-scale plutonium production reactor) and 1988, the Hanford Site has been a
7 plutonium production facility for the USA weapons program. Currently, the Hanford Site is
8 engaged in the world's largest environmental cleanup, with many challenges to be resolved in the
9 face of overlapping technical, political, regulatory, and cultural interests (Gephart, 2003).

10 Another major nuclear operations facility, the Savannah River Site is located on 840 km² along
11 the Savannah River in South Carolina. The historical mission of the site was to produce tritium
12 and plutonium for the weapons program in large nuclear production reactors. The processes
13 generated high-level, low-level, transuranic, and mixed wastes that were managed in seeping and
14 settling basins, unlined disposal pits, waste piles, burial grounds, and underground storage tanks.
15 More than 400 individual contaminated sites and 659 contaminated buildings have been
16 identified at the Savannah River Site (Eisenbud and Gesell, 1997).

17 As the reprocessing has removed much of the actinide content, over 99% of the present
18 radioactivity from HLW liquid and sludge is from non-actinide radionuclides; contribution from
19 long-lived actinides will become comparatively larger over time. In Hanford, about 2 MCM of
20 highly radioactive and chemically toxic waste were produced from two chemical processing
21 plants for Pu. These wastes are currently stored in 177 underground tanks, with capacities
22 ranging from 208 to 416 m³ for each tank. Sixty-seven tanks are known or are suspected of
23 having leaked, with an estimated 570 m³ of waste containing 3.7×10^4 TBq of radioactivity
24 released to the subsurface (Ewing, 2004). In addition to 7.2×10^6 TBq of radioactive tank waste,

1 there are 6.8×10^5 TBq of nuclear materials stored on site, some radioactive solid waste, and
2 contaminated groundwater and soil; a total radioactivity is 1.4×10^7 TBq in Hanford (Gephart,
3 2003). Nuclear operation at the Hanford Site has discharged a variety of radionuclides into the
4 environment. Tables 1 and 2 present the release into the air and nearby Columbia River during
5 1944–1972; smaller releases have continued since 1972 (Washington State Department of
6 Health). Most of the released radionuclides are relatively short-lived, but 2.3×10^4 TBq of
7 extremely long-lived ^{237}Np has been discharged into the river (Table 2).

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

1 ^{137}Cs , was released to the Techa River (Christensen et al., 1997). More mobile radionuclides
2 such as ^{90}Sr have been found to be present more than 2,000 km downstream of the Techa River.

3 Since 1951, the liquid waste has been discharged into a small swampy lake, Lake
4 Karachai, covering less than 0.5 km^2 in area, inside the Mayak complex. Up to 1993, the lake
5 accumulated some 4.4×10^6 TBq of radioactivity (Figure 1), including 3.6×10^6 TBq of ^{137}Cs and
6 7.4×10^5 TBq of ^{90}Sr (Solodov et al., 1998). Lake Karachai is probably the "most polluted spot"
7 on Earth. For comparison, the Chernobyl disaster released about 1.2×10^7 TBq of total
8 radioactivity (UNSCEAR, 2000), and this radiation is not concentrated in one location. About
9 20% of the activity is in the water phase with a concentration of about 0.5 TBq/m^3 , whereas the
10 remainder is found in the lake sediments. The groundwater system near the lake has been
11 contaminated (Solodov et al., 1998; Novikov et al., 2006). Starting in the 1960s, Lake Karachai
12 began to dry out; its area dropped from 0.5 km^2 in 1951 to 0.15 km^2 by the end of 1993. In 1967,
13 following a drought in the region, the wind carried 22 TBq of radioactive dust away from the
14 dried area of the lake; an area of $1,800 \text{ km}^2$ was contaminated with more than 4 GBq (giga
15 becquerel) $^{90}\text{Sr}/\text{km}^2$ and $12 \text{ GBq } ^{137}\text{Cs}/\text{km}^2$ (Christensen et al., 1997). According to a report of
16 Natural Resources Defense Council, the radiation level at the shore of the lake was 600
17 röntgens/hr in 1990, high enough to give a lethal dose to a human within an hour (Cochran et al.,
18 1989). In addition to Techa River and Lake Karachai, about 1.2×10^4 TBq of radioactive waste
19 was released to five other reservoirs (Christensen et al., 1997).

20 In addition to staggering waste generation and release, operation in Mayak resulted in
21 many accidents. The most notable "Kyshtym" accident occurred on September 29, 1957, when
22 the failure of the cooling system for a tank storing tens of thousands of tons of dissolved nuclear
23 waste resulted in a violent explosion, which released 7.4×10^5 TBq of radioactivity; an area of
24 $20,000 \text{ km}^2$ (with 270,000 inhabitants) were contaminated with more than $4,000 \text{ Bq}/\text{km}^2$.

1 Approximately 70% of this activity consisted of the rather short-lived nuclides such as ^{95}Zr , ^{106}Ru
2 and ^{144}Ce , and 2.7% and 0.04% was ^{90}Sr and ^{137}Cs , respectively (Christensen et al., 1997).

3 In addition to environmental problems from the production of nuclear weapons, the other
4 very pressing environmental and nonproliferation problem in Russia is the storage and disposal
5 of the SNF from nuclear submarines and nuclear-powered surface ships. With the end of Cold
6 War, 200 Russian nuclear submarines were decommissioned, and over 100 are awaiting
7 dismantlement (Webster, 2003). The largest SNF storage facility at Andreeva bay, on the
8 northernmost coast of the Kola Peninsula near the border with Norway, contains 21,000 spent
9 fuel assemblies extracted during the 1970s and 12,000 m³ of solid and liquid radioactive wastes.
10 Three makeshift storage tanks for spent nuclear fuel are deteriorating due to poor maintenance
11 and the harsh Arctic climate. Poor condition of the storage facility has also lead to
12 contamination of a substantial amount of soil, water, and concrete at Andreeva bay (Ewing,
13 2004). Furthermore, called a “Radioactive Ghost Ship”, the *Lepse*, a 5,000-ton nuclear service
14 vessel moored on the Kola Peninsula near Murmansk, has accumulated 639 nuclear fuel
15 assemblies (about 320 are damaged) and 39 m³ of solid radioactive waste; the spent fuel contains
16 about 2.8×10^4 TBq radioactivity (Webster, 2003).

17 2.2. Nuclear weapons testing

18 The first nuclear weapons test was conducted on July 16, 1945, near Alamogordo, New
19 Mexico. According to the U.S. Department of Energy’s Nevada Site Office (DOE/NV) (2000),
20 the explosive yield of this Trinity test was 21 kT (1 kT=10¹² calories). From this day to
21 September 23, 1992, the United States maintained a program of vigorous nuclear testing, with
22 the exception of a moratorium between November 1958 and September 1961. As released in
23 DOE/NV (2000), a total of 1,054 nuclear tests were conducted, with 106 tests taking place at

1 sites in the Pacific Ocean, 904 at the Nevada Test Site (NTS), and 17 on miscellaneous sites in
2 the continental USA (e.g., Alaska, Colorado, Mississippi, and New Mexico).

3 Occupying a 19,000 km² area in the northeast of the country, the Semipalatinsk test site
4 in the Republic of Kazakhstan is located immediately south of Russia, and west of China.
5 Following World War II, the steppes of Kazakhstan became the first center for nuclear weapons
6 testing within the former Soviet Union. During 1949–89, the Soviet Union conducted a total of
7 about 460 nuclear weapons tests in Semipalatinsk, including 133 atmospheric nuclear tests.
8 Starting in 1961, more than 300 test explosions were conducted underground. Thirteen of the
9 underground tests resulted in releases of radioactive gases to the atmosphere (IAEA, 2007a).

10 In late 1940 to early 1960s, about 350 atmospheric atomic bomb tests were conducted at
11 different test sites in the northern hemisphere (Eikenberg et al., 2004). These tests released into
12 the atmosphere more than 9×10^5 TBq of residual ²³⁹Pu, 9×10^5 TBq of fissionogenic ¹³⁷Cs, and
13 6×10^5 TBq of ⁹⁰Sr. Choppin (2003) reported that over 2×10^8 TBq of radioactivity have been
14 released into the atmosphere from worldwide nuclear weapons tests.

15 At the NTS with an area about 3,500 km² (the area of Rhode Island is 3,144 km²), a large
16 inventory of radionuclides (tritium, fission products, activation products, and actinides) is present
17 in the subsurface environments as a result of 828 underground nuclear weapons tests (DOE/NV,
18 2000). A total of about 4.9×10^6 TBq of radioactivity decayed corrected to September 23, 1992,
19 the date of the last underground nuclear test, was produced (Smith et al., 2003). The inventory
20 includes only those radionuclides (a total of 43) whose concentrations dissolved in a cavity
21 (produced from an underground nuclear test) volume of water, decay corrected to 100 years in
22 the future, exceed 0.1 of the proposed maximum permissible concentrations of radionuclides in
23 drinking water set by the U.S. Environmental Protection Agency in 1991 (USEPA, 1991). This
24 selection criterion effectively excludes most radionuclides with half-lives less than 10 years.

1 Since 1973, various studies have been investigating the environmental effects of nuclear testing
2 at the NTS (e.g., Coles and Ramspott, 1982; Buddemeier and Hunt, 1988; Kersting et al., 1999;
3 Tompson et al., 2002).

4 Between 1966 and 1996, France had conducted nuclear weapons tests above and beneath
5 the atolls of Mururoa and Fangataufa in French Polynesia (Pfungsten et al., 2001). The total
6 French nuclear testing program comprised 210 tests, 50 in the atmosphere and 160 underground;
7 all French testing ceased on January 27, 1996 (IAEA, 1998). For the underground tests, 13 were
8 conducted at In Ecker in Algeria in the 1960s, and 147 (with a total nuclear yield of 3,192 kt) in
9 shafts at Mururoa and Fangataufa during 1975–1996. Estimated underground inventories, decay
10 corrected to May 1, 1996, of 36 radionuclides for the sites of Mururoa and Fangataufa were
11 published in IAEA (1998); a total radioactivity was 3.5×10^5 TBq.

12 Table 3 lists the radionuclide inventories from the underground nuclear tests conducted in
13 the NTS and French Polynesia; the inventory is decay corrected to January 1, 2008. In terms of
14 radioactivity, ^3H , ^{90}Sr , ^{137}Cs , ^{241}Am , and plutonium isotopes are currently the radionuclides of
15 great importance (Table 3). Over time (e.g., 1,000–10,000 years later), long-lived ^{14}C , ^{36}Cl , ^{99}Tc ,
16 ^{129}I , ^{237}Np , as well as several uranium and plutonium isotopes will become important.

17 2.3. Nuclear power plants

18 To generate electricity, nuclear reactors use fuel of solid ceramic pellets of enriched
19 uranium that are sealed in strong metal tubes; the tubes are bundled together to form a nuclear
20 fuel assembly. The uranium pellets are about the size of the tip of a person's little finger, yet one
21 pellet has the amount of energy equivalent to almost one ton of coal. After 3–4 years in a reactor,
22 the uranium pellets are no longer efficient for producing electricity and the assembly is removed
23 from the reactor; this SNF is highly radioactive. About 25–30 t of SNF are created per year per

1 GWe (gigawatt electricity). Thus, about 12,000 t of SNF are produced based on the world's
2 current nuclear energy capacity (Macfarlane and Miller, 2007).

3 As the number of nuclear power plants (NPP) increased during the 1960s, so did the
4 growing concern for the fate of the SNF. In 2002, there were about 150,000 tHM (metric tones of
5 heavy metal) of SNF in the world. By the end of 2004, there was about 190,000 tHM of SNF in
6 storage (IAEA, 2007b). Most of this fuel (90,000 t for 2002 data) is still at the 236 nuclear power
7 stations, which together have 439 reactors, where it was originally generated in 31 different
8 countries (Ewing, 2004; IAEA, 2007b). In 2000, the USA's SNF inventory was about
9 42,300 tHM, with a total radioactivity of 1.2×10^9 TBq (Figure 1), increasing by slightly less than
10 2,000 tHM every year. Since the mid-1940s, SNF and HLW have accumulated throughout the
11 USA. Currently, SNF and HLW are stored at some 125 sites in 39 states, with over 161 million
12 people residing within 121 km of temporarily stored nuclear waste. By the year 2020, the
13 inventory will be about 77,100 tHM with an activity of 1.3×10^9 TBq; the currently legislated
14 capacity for the potential high-level waste repository at Yucca Mountain is 70,000 t of SNF
15 equivalent.

16 As the world's economy grows, nuclear power is recognized as the only large-scale
17 emissions-free power source that is able to help meet the growing need for electricity. As the
18 third leading source of electricity, nuclear power provides 19% of electricity in the US (Table 4).
19 (Nuclear power provides 78% of electricity for France, 48% for Sweden, and 30% for the entire
20 European Union.) The DOE projects the U.S. will need 40% more electricity by 2030. In order to
21 keep pace with the nuclear energy needs, it will be necessary to build an average of three new
22 plants per year starting in 2015. The U.S. has not ordered a new nuclear power plant since the
23 1970s, partially as a result of constant litigation and overly complex regulations. The Nuclear
24 Power 2010 initiative, a partnership between industry and the U.S. government, was launched in

1 2002 to reduce regulatory and other barriers to the development of new nuclear power plants.
2 The Nuclear Regulatory Commission (NRC) is working to improve and streamline the regulatory
3 process to help accelerate the construction of nuclear plants. The comprehensive energy policy
4 also includes permanent disposal of SNF and HLW, and advanced technologies for nuclear fuel
5 reprocessing.

6 As recent as September 26, 2007, NRG Energy, Inc of New Jersey and the South Texas
7 Project Nuclear Operating Company filed an application to build and operate two new nuclear
8 power reactors at the South Texas Project nuclear power station site in Matagorda County
9 (Scientific America, 2007). This is the first full nuclear plant license application in the United
10 States in 29 years, since Pennsylvania's Three Mile Island reactor had a partial meltdown in 1979
11 that sparked a massive protest movement against nuclear plants; public acceptance of nuclear
12 power has changed with the increasing concern of global warming from greenhouse gas emission
13 of fossil fuel. If the application is approved, NRG expects to bring the units on line in 2014 and
14 2015.

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

1 A number of countries announced plans for significant expansion of nuclear energy,
2 specifically China, India, Japan, Pakistan, the Russian Federation, and the Republic of Korea. In
3 addition, Argentina, France, South Africa, Ukraine and the USA have expressed an intention to
4 expand their existing programs (IAEA, 2007b). Asia has had the most significant expansion in
5 nuclear power generation capacity. In East and South Asia, there are over 109 nuclear power
6 reactors in operation, 18 under construction, and another 110 planning to be built. The great
7 growth in nuclear energy generation is expected to be in China, Japan, South Korea, and India;
8 their current electricity contribution is listed in Table 4. The Chinese economy has been growing
9 at an annual rate of about 10%, with electricity demand growing twice that fast. Most of
10 mainland China's electricity is produced from fossil fuels (about 80%, mainly coal) and
11 hydropower (about 18%). Rapid growth in demand has given rise to power shortages, and the
12 reliance on fossil fuels has led to much air pollution; the economic loss due to pollution is about
13 3–7% of gross domestic product (World Nuclear Association, 2007b). While coal is the main
14 energy source, most reserves are in the north or northwest of China, which presents an enormous
15 logistics problem. Nuclear power has an important role, especially in the coastal areas that are
16 remote from the coalfields and where the economy has been developing rapidly.

17 Moves to build nuclear power in mainland China commenced in 1970 and the industry
18 has now moved to a steady development phase. The first two nuclear power plants in mainland
19 China were at Daya Bay near Hong Kong and Qinshan, south of Shanghai, with construction
20 starting in the mid-1980s. Currently, there are 11 operating nuclear power reactors in the
21 provinces of Zhejiang, Guangdong, and Jiangsu (all are coastal areas with rapid economic
22 development and electricity demand), with a total net capacity of 8.6 GWe. The nuclear power is
23 currently providing 51.8 billion kWh (kilowatts-hours), which is 1.9% of the nation's total
24 electricity. The Chinese government plans to increase nuclear generating capacity by five-fold to

1 40 GWe by 2020, which requires adding an average of 2 GWe per year. In May 2007, the
2 National Development and Reform Commission further announced that its target for nuclear
3 generation capacity in 2030 was 160 GWe. More than 16 provinces, regions and municipalities
4 have announced intentions to build nuclear power plants in the 20th 5-year plan (2011-15).

5 When China started to develop nuclear power, a closed fuel cycle strategy was also
6 formulated and declared at an IAEA conference in 1987. The spent fuel activities involve at-
7 reactor storage, away-from-reactor storage, and reprocessing. Based on expected installed
8 capacity of 20 GWe by 2010 and 40 GWe by 2020, the annual SNF generation will amount to
9 about 600 t in 2010 and 1,000 t in 2020, the cumulative SNF increasing to about 3,800 t and
10 12,300 t, respectively. Construction of a centralized spent fuel storage facility at Lanzhou
11 Nuclear Fuel Complex in Gansu Province began in 1994. The initial stage of that project has a
12 storage capacity of 550 t and could be doubled.

13 Over 200 radionuclides are produced during the operation of a typical reactor; most of the
14 radionuclides are relatively short-lived and decay to low levels within a few decades (Crowley,
15 1997). A number of radionuclides are emitted from normal operation of NPP. For example, the
16 annual discharge of gaseous ^{14}C to the atmosphere from pressured water reactors in Germany
17 was 280 ± 20 GBq per GWe in 1999, on average 30% is thought to have emitted in the form of
18 CO_2 , the rest in organic form (CH_4 in particular). In France, ^{14}C discharges were estimated to be
19 140 GBq/y per unit of 900 MWe and 220 GBq/y per unit of 1,300 MWe (Roussel-Debet et al.,
20 2006). Based on combined worldwide operable nuclear reactors of 3.72×10^5 MWe (World
21 Nuclear Association, 2007a), the annual discharge of ^{14}C worldwide is about 60 TBq/y. As a
22 comparison, all atmospheric nuclear tests emitted about 2.13×10^5 TBq of ^{14}C , cosmogenic
23 natural production in the upper atmosphere is at a rate of approximately 1.54×10^3 TBq/y.

1 To assess the potential environmental contamination from normal operation of nuclear
2 power plants, monitoring of radionuclides in terrestrial and aquatic environments has been
3 performed on various environmental samples, including soils, plants (pine needle, tea, grass,
4 moss, algae) and foodstuff (wheat, cabbage, rice) (e.g., Lu et al., 2006; Adlienè et al., 2006;
5 Jean-Baptiste et al., 2007). To address the growing nuclear energy in China, a Radiation
6 Monitoring Technical Center has been established in the coastal Zhejiang Province by The State
7 Environment Protection Administration. The Center is mainly responsible for radiological
8 monitoring and radioactive waste management, which include national radiation monitoring
9 network management, data collection and analyses, preparation of technical guidelines and
10 standards, training, and quality assurance/quality control. The Center compiles and publishes an
11 annual report about the radiation monitoring results near the NPP and other nuclear facilities in
12 China.

13 2.4. *Commercial fuel reprocessing*

14 At present, the worldwide capacity to reprocess SNF is between one-third and one-half of
15 the annual production rate of SNF (~10,000 tHM/y). In operation since 1976, La Hague plant
16 located on the Cotentin Peninsula in Normandy, France is the world's largest commercial fuel
17 reprocessing facility. With a capacity of 1,650 tHM/y, the plant processed a total of 1,100 tons in
18 2005, and recovered plutonium is sent to Marcoule nuclear site (which served French defense
19 programs and also reprocessed gas-cooled reactor fuel) where MOX fuel is fabricated. La Hague
20 treats spent nuclear fuel from France, Japan, Germany, Belgium, Switzerland and the
21 Netherlands. The non-renewable waste is eventually sent back to the user nation, as established
22 under international law.

23 With a 900 t/y capacity, THORP (Thermal Oxide Reprocessing Plant), designed for
24 reprocessing uranium oxide fuel, in Sellafield, England has reprocessed a total of 8,000 t,

1 including those from the United Kingdom, Japan, Germany, and Switzerland (Bellona, 2003).
2 Construction of THORP started in the 1970s and was completed in 1994; the plant went into
3 operation in August 1997. In addition, the Magnox Reprocessing Plant (B205) in Sellafield
4 began commercial operation in January 1964 and has an annual capacity of 1,500 t to reprocess
5 other nuclear fuels (metallic uranium fuel, and now fuels from Britain's Magnox reactors)
6 (Bellona, 2003). In addition to these main commercial reprocessing facilities in France and
7 England, there are several major reprocessing facilities in the world. The Mayak facility in
8 Russia can reprocess 400 t nuclear fuels each year. Kalpakkam atomic reprocessing plant in
9 India has an annual reprocessing capacity of 275 t. Since 1993, about 2 billion U.S. dollars have
10 been invested in constructing the most recent major reprocessing plant in Rokkasho, Japan. With
11 a capacity of 800 t/y, the Rokkasho plant is currently undergoing test operations, separating a
12 small amount of used nuclear fuel. It is the successor to a smaller reprocessing plant located in
13 Tokai, Ibaraki. A pilot (50 t/y) reprocessing plant using the Plutonium-Uranium Extraction
14 (PUREX) process was opened in Lanzhou, China. This plant could be fully operational
15 in 2008 and is capable of expansion to 100 t/y. A large commercial reprocessing plant based on
16 indigenous advanced technology is planned to follow and begin operation in China about 2020
17 (World Nuclear Association, 2007b).

18 Radionuclides have been discharged, with government authorization, over the past forty
19 years into the sea and atmosphere from the nuclear fuel reprocessing plants at Sellafield and at
20 La Hague. Tables 5 and 6 present the authorized annual limits for aerial and liquid discharges,
21 respectively, from the Sellafield plant, wherein the THORP operation is currently the major
22 source. During the years between 1974 and 1978, for example, the Sellafield plant's annual
23 discharge of ^{137}Cs was 4,000 TBq. The plutonium discharge in the same period was between 45
24 and 60 TBq annually (Bellona, 2003). Over those five years, more than twice as much plutonium

1 was discharged from Sellafield as was released during the 1986 Chernobyl accident, where
2 discharges amounted to about 100 TBq of long-lived plutonium (UNSCEAR, 2000). Moreover,
3 La Hague has significantly increased liquid radioactive carbon release over the past two decades
4 from less than 1 TBq/y prior to 1983 up to about 10 TBq/y in 1999, and were still above 7 TBq/y
5 up to 2004 (Fiévet et al., 2006).

6 Discharge of ^{99}Tc from Sellafield has also been high. In the three-year period from 1978
7 to 1980, Sellafield discharged almost 300 TBq of ^{99}Tc (Bellona, 2003). More importantly, at a
8 rate of about 90 TBq per year, current and future discharge of ^{99}Tc contributes a significant
9 portion of the total released radioactivity. Today, it is the large releases of the radioactive ^{99}Tc
10 from Sellafield that is largely responsible for the pollution of the Norwegian coast and the
11 Barents Sea. To a less extent, La Hague has also been discharging ^{99}Tc , which increased slowly
12 from ~ 0.4 TBq/y in 1970 to a peak of ~ 25 TBq/y in 1985. By 1991 the discharge had fallen to
13 ~ 0.9 TBq/y and have since remained relatively stable at < 1 TBq/y (Keogh et al., 2007).

14 From 1966 to 1999, a total of 2,300 kg (15 TBq) of liquid ^{129}I was discharged in the
15 marine environment at La Hague and another 64.2 kg (0.42 TBq) gaseous ^{129}I released to the
16 atmosphere, an amount that is 50 times the total release from nuclear weapons tests (Fréchet and
17 Calmet 2003). Up until 1998, 2,600 kg (17 TBq) of ^{129}I was discharged into the sea by La Hague
18 and Sellafield, an amount that is 50 times the total release from nuclear weapons tests and three
19 orders of magnitude higher than the Chernobyl accident release (Hou et al., 2001). After 1998,
20 ^{129}I has been discharged at about 0.2 TBq/y from Sellafield and negligibly from La Hague
21 (Keogh et al., 2007). The total release of ^{129}I was estimated to be about 255 kg (1.7 TBq) and
22 4,000 kg (26 TBq) from gaseous and liquid discharges, respectively (Aldahan et al., 2006). From
23 1944 through 1972, the plutonium-production operation at the Hanford Site released about 260

1 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.

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

10 As the bare critical mass of ^{239}Pu is less than 10 kg, the potential diversion of civil-source
11 Pu, originally destined for fabrication of mixed-oxide (U+Pu) fuel, to the production of a
12 nuclear weapon is an extremely important proliferation and environmental problem (Ewing,
13 2004). France and Britain have the largest inventories, with 72 t (of which 33.6 t is foreign
14 owned) and 60 t (6.1 t foreign owned), respectively. Both the USA and Japan have 5 t of civil-
15 source Pu, but Japan has another 24.1 t in other countries (mostly in the La Hague reprocessing
16 facility of France). Of equal importance are the stockpiles of highly enriched ^{235}U (HEU), with
17 645 t in the USA and 1,050 t in Russia. The USA agreed to purchase 500 t of weapons uranium
18 from Russia to blend down to low enriched ^{235}U (LEU) for use in commercial reactors. As of
19 2003, 193 t of bomb-grade HEU have been converted to 5,700 t LEU of 4% ^{235}U for use in light
20 water reactors (LWRs). This amount of HEU could have been used to produce over 7,700
21 nuclear weapons (Ewing, 2004). Moreover, during the past decade, plutonium from dismantled
22 nuclear weapons has emerged as a new waste type with proliferation implications (Ewing, 2004).
23 The disarmament process, under Strategic Arms Reduction Treaties between the USA and

1 Russia, of thousands of nuclear weapons will produce 30–40 t of weapons-grade plutonium in
2 each country, as well as hundreds of tons of HEU.

3 The Nuclear Fuel Service in the U.S. operated a nuclear fuel reprocessing center from
4 1966 to 1972 in West Valley, New York and processed 640 t of SNF. During the operation,
5 2,500 m³ of highly radioactive liquid waste were generated and stored in an underground waste
6 tank, which was vitrified during 1996–2002. In 1977, fear of nuclear weapons proliferation
7 (especially after India demonstrated nuclear weapons capabilities using reprocessing technology)
8 led President Carter to issue a Presidential directive to indefinitely suspend the commercial
9 reprocessing and recycling of plutonium in the U.S.

10 Initiated in 2006 by the USA, the Global Nuclear Energy Partnership (GNEP) is a
11 comprehensive strategy to expedite the development of nuclear power around the world while
12 improving the use of resources and providing greater disincentives to the proliferation of nuclear
13 weapons. The GNEP involves both political and technological initiatives. A global nuclear
14 energy partnership involves nations with advanced civilian nuclear energy programs, such as
15 France, Japan, China, and Russia, aiming to improve the proliferation-resistance of the nuclear
16 fuel cycle while guaranteeing access to fuel supplies. Technological initiatives include
17 reprocessing spent uranium fuel for use in advanced reactors to help extract more energy and to
18 have the potential of reducing storage requirements for nuclear waste by up to 90 percent. The
19 GNEP calls for construction of the world's largest nuclear fuel reprocessing facility by 2020,
20 capable of reprocessing 2,000 to 3,000 t of SNF a year; large enough to reprocess all the waste
21 generated annually by the U.S.'s 104 commercial nuclear power plants (Johnson, 2007).

22 *2.5. Geological repository of high-level nuclear wastes*

23 Altogether, currently there are 439 nuclear power plants around the world, operating in
24 31 countries. All countries using nuclear power must deal with high-level radioactive waste,

1 regardless of the number of reactors they have in service. Nuclear energy production eventually
2 creates waste in the form of spent nuclear fuel. Spent nuclear fuel is depleted or used fuel from
3 nuclear power plants and research facilities. Spent nuclear fuel is highly radioactive, and remains
4 so for thousands of years. Isolating this high-level waste from people and the environment has
5 been an important and challenging issue for all countries that use nuclear power. Social,
6 economic, political, technical, and geographic considerations shape a country's radioactive waste
7 management decisions.

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

15 The HLW is destined for vitrification in a borosilicate glass and then emplaced in a
16 geological repository for permanent disposal, along with the SNF. Over 500 t of HLW have been
17 vitrified in France and Germany. At the Mayak facility, about 1.1×10^7 TBq HLW from
18 reprocessing has been immobilized into a Na-Al phosphate glass. In the USA during 1996 to
19 2001, the HLW was vitrified into 275 stainless steel canisters (each canister weighs about two-
20 tons, and is 0.61 m in diameter and almost 3.05 m high) in the West Valley Plant of New York.
21 The Defense Waste Processing Facility at Savannah River started operation in 1996 after 18
22 years of planning and \$2 billion in construction costs (Crowley, 1997), and had produced about
23 1,600 canisters by February 2004 (Ewing, 2004). Each Savannah River canister holds about 1.68
24 t of glass, of which about 42.6 kg will be HLW; radioactivity of the waste in an individual

1 canister will be as high as 8.7×10^3 TBq. It is estimated that 6,000–8,000 such canisters will be
2 required to hold existing and projected waste at Savannah River. Another vitrification plant is
3 under construction at Hanford, wherein all the HLW will be vitrified into about 20,000 canisters
4 for final disposal in the proposed Yucca Mountain repository, which is located on the western
5 edge of the NTS in a remote desert on federally protected land.

6 The U.S. DOE began studying Yucca Mountain, Nevada, in 1978 to determine whether it
7 would be suitable for the nation's first long-term geologic repository for spent nuclear fuel and
8 high-level radioactive waste. On July 23, 2002, President Bush signed a Resolution, allowing the
9 DOE to take the next step in establishing a safe repository in which to store the nation's nuclear
10 waste. The DOE is currently in the process of preparing an application to obtain the NRC license
11 to proceed with construction of the repository. In July of 2006, the DOE announced plans to
12 submit a license application to NRC by June 30, 2008, and to initiate repository operations in
13 2017.

14 Based on the currently legislated repository capacity of 70,000 MTHM, there will be a
15 total of 7,860 packages of commercial spent nuclear fuel, 3,910 packages of defense spent
16 nuclear fuel, and 3,910 packages of defense high-level nuclear waste for disposal in the proposed
17 Yucca Mountain geological repository (BSC, 2001). With the reported radionuclide (a total of
18 26) inventory in each package, about 2.6×10^8 TBq of radioactivity from high-level nuclear waste
19 and spent fuel will be stored for many generations to come (Table 7; Figure 1). Similar to the
20 radionuclides from underground nuclear tests, ^{90}Sr , ^{137}Cs , ^{241}Am , and plutonium isotopes will be
21 of great importance over the short-term, while long-lived ^{14}C , ^{36}Cl , ^{99}Tc , ^{129}I , ^{237}Np , ^{243}Am , and
22 several uranium and plutonium isotopes will become important over the long-term.

23 After forty years of commercial nuclear power, no country has disposed of HLW (Table
24 4). Along with the USA, Belgium, Canada, China, Finland, France, Germany, Japan, Russia,

1 Spain, Sweden, Switzerland and the United Kingdom have invested significant resources in their
2 radioactive waste management programs because of their historic or anticipated reliance on
3 nuclear energy. Deep geologic disposal has been accepted as the best method for isolating highly
4 radioactive, long-lived waste (Witherspoon and Bodvarsson, 2001). Many countries have been
5 developing their repository at different paces to cope with the HLW. For example, high-level
6 wastes in China will be vitrified, encapsulated and put into a geological repository (probably
7 granite) some 500 m deep. Site selection is focused on six candidate locations and will be
8 completed by 2020. An underground research laboratory will then operate for 20 years and
9 actual disposal is anticipated from 2040 (Wang et al., 2001).

10 2.6. Nuclear accidents

11 Contamination of the environment by radioactivity has occurred as a result of nuclear
12 accidents, particularly by the U.S and former Soviet Union (e.g., the accidents in Mayak facility
13 in Section 2.1) considering the magnitude of their nuclear weapons programs and nuclear power
14 generation. The most notable is the catastrophic accident that destroyed Unit 4 of the Chernobyl
15 nuclear complex in the Ukraine in April of 1986. It is estimated that 1.2×10^7 TBq of
16 radioactivity was released in the Chernobyl accident (UNSCEAR, 2000), and the noble gases
17 contributed about 50% of the total release; the principal radionuclides and their inventories are
18 presented in Table 8. Eikenberg et al. (2004) compared the total atmospheric release of long-
19 lived dose-relevant fission radionuclides and actinides from the atomic bomb test and the
20 Chernobyl reactor explosion. Compared to the sum of all previously performed atmospheric
21 bomb tests, the values for ^{90}Sr , ^{137}Cs , and $^{239+240}\text{Pu}$ from the Chernobyl accident are in the order
22 of 10% and much higher for ^{238}Pu and ^{241}Am . Fallout of hot particles in the vicinity of the
23 reactor caused a considerable contamination of the soil surface, with ^{137}Cs up to 10^6 Bq/m², and
24 116,000 people had to be evacuated within a zone of 30 km distance from the reactor (Balonov,

1 2007). The contribution of ^{137}Cs from the Chernobyl plume was significant even 2,000 km away;
2 however, the total surface contamination was at least two orders of magnitude below the level
3 within the 30-km exclusion zone. In all, there were 28 deaths from the acute radiation syndrome
4 as a result of the Chernobyl accident (Eisenbud and Gesell, 1997).

5 Being the world's first major accident at a civilian power plant, the Three Mile Island
6 Unit II accident in 1979 in Pennsylvania has played a significant role in effectively stopping the
7 growth of nuclear power in the U.S. With $0.92\text{--}3.7\times 10^5$ TBq radioactive gas release, this
8 accident did not cause notable radiation effects on individuals living in the vicinity of the reactor
9 (25,000 people lived within 8 km of the site at the time of the accident), yet the sociopolitical
10 ramifications of the accident are evident. The Three Mile Island accident was a significant
11 turning point in the global development of nuclear power. From 1963 to 1979, the number of
12 reactors under construction globally increased every year except in 1971 and 1978. However,
13 following the event, the number of reactors under construction declined every year from 1980 to
14 1998.

15 As of October 2000, there were about 400 reactors in nuclear powered vessels around the
16 world (IAEA, 2001). Of these, the Russian Federation had 75 active military vessels with about
17 150 reactors, and the USA had about 117 nuclear vessels. There are six confirmed nuclear
18 submarine accidents since 1963 at various sites in the Atlantic Ocean (Table 9). With the
19 exception of the Russian submarine Kursk, which lies at a depth of 108 m in the Barents Sea, all
20 other submarines are too deep for the submarine or reactor(s) to be recovered.

21 Nuclear energy sources are also used in some spacecraft, satellites, and deep sea acoustic signal
22 transmitters for generation of heat or electricity, with two common types of nuclear energy
23 sources: radioisotope thermoelectric generators (RTGs) and nuclear reactors. A typical RTG
24 contains approximately 1,000 TBq ^{238}Pu or 10,000 TBq ^{90}Sr . On April 21, 1964, the U.S.

1 navigational satellite “Transit 5BN-3” with a radioisotope generator containing 630 TBq (about 1
2 kg) of ^{238}Pu failed to achieve orbit and burned up over the West Indian Ocean; the nuclear fuel
3 was vaporized and dispersed worldwide (IAEA, 2001). By mid-1970, 95% of the ^{238}Pu was
4 found deposited on the Earth’s surface, which almost tripled the global deposit of this Pu isotope
5 by 1970 (Hardy et al., 1973).

6 Lighthouses in remote Russian waters are often powered by RTGs, which may contain up
7 to several thousand TBq of ^{90}Sr ; there are some 500 RTGs in use. There were two recorded
8 incidents where RTGs were lost at sea and released about 2.5×10^4 and 1.3×10^3 TBq of ^{90}Sr ,
9 respectively (IAEA, 2001). Spread of orphan nuclear sources and their potential use in
10 radiological dispersion devices (RDD) by terrorist groups have been a continuing concern (e.g.,
11 Sohler and Hardeman, 2006). Due to the radiotoxicity, widespread use and sufficiently long half-
12 life, 7 radionuclides are of particular concern in the RDD: ^{60}Co , ^{90}Sr , ^{137}Cs , ^{192}Ir , ^{238}Pu , ^{241}Am ,
13 and ^{252}Cf . Commercial radioactive sources for potential RDD include RTG (^{90}Sr), teletherapy
14 and irradiators (^{60}Co and ^{137}Cs), industrial radiography (^{60}Co and ^{192}Ir), well logging and
15 moisture detectors (^{137}Cs , ^{241}Am , and ^{252}Cf). Social, economic, and environmental effects of
16 deliberate use of RDD are reflected in two serious accidents in which abandoned teletherapy
17 units caused extensive contamination, acute radiation effects, and, in one case, fatalities. In
18 December 1983, a teletherapy unit that contained 16.7 TBq of ^{60}Co was dismantled to be sold as
19 scrap in Juarez, Mexico; rupture of a radioactive source caused serious contamination (Eisenbud
20 and Gesell, 1997). A more serious accident occurred in September 1987 in the state of Goiania,
21 Brazil. A teletherapy unit that contained 51 TBq of ^{137}Cs in the form of CsCl_2 powder was left in
22 an abandoned medical clinic (Figure 1). From various attempts of scavenging, of this unknown
23 object, the CsCl_2 was found to be distributed over an area of about 1 km^2 ; four people ultimately
24 died of acute radiation injury, one person required amputation of an arm, and 129 persons had

1 measurable body contamination (of whom 21 were hospitalized from serious skin burns or blood
2 changes) (Eisenbud and Gesell, 1997).

3 **3. Salient geochemical behavior of important radionuclides**

4 Of the suite of important radionuclides (^3H , ^{14}C , ^{90}Sr , ^{99}Tc , ^{129}I , ^{137}Cs , ^{237}Np , ^{241}Am , as
5 well as several uranium and plutonium isotopes), ^3H is considered an ideal water tracer, although
6 small amounts of tritium retardation have been reported in transport studies, and ^{14}C may be
7 retarded by its isotopic exchange with carbonate minerals in the aquifer (Hu et al., 2007b). ^{90}Sr
8 and ^{137}Cs are the major fission products, yet they do not pose long-term risk because of their
9 short half-lives and strong sorption in the subsurface. In general, the mobility of actinides in
10 aqueous systems is low, dependent on (1) their thermodynamic properties, which determine
11 solubility and speciation as a function of pH and redox potential, (2) the availability of inorganic
12 ligands to form soluble complexes, and (3) the composition and abundance of minerals and
13 mineral colloids present in the system (Dozol and Hagemann, 1993; Silva and Nitsche, 1995).

14 Of particular importance to the environment and risk assessment are radionuclides ^{99}Tc ,
15 ^{129}I , and ^{237}Np , because of their long half-lives and presumably high mobility (de Marsily et al.,
16 1977; Bondietti and Francis, 1979; Hu and Smith, 2004). These nuclides are present in high
17 abundance in underground nuclear test, fuel reprocessing, and in spent reactor fuel (Table 10).
18 Due to their long half-lives (2.13×10^5 and 1.57×10^7 years for ^{99}Tc and ^{129}I , respectively) and
19 presumed mobile behavior in groundwater, both ^{99}Tc and ^{129}I are important dose contributors to
20 the calculated health risk for many U.S. DOE nuclear facilities, including the NTS (Smith et al.,
21 2003), Hanford Site (Kaplan and Serne, 1998; Um et al., 2004), Savannah River Site (Beals and
22 Hayes, 1995), and Idaho National Laboratory (Beasley et al., 1998). ^{237}Np also has a long half-
23 live (2.14×10^7 years), and weak sorption with high mobility potential in the subsurface (Arnold
24 et al., 2006).

1 With a high-abundance (6%) fission yield, ^{99}Tc has been estimated to have 25–30 ton
2 produced worldwide up to the mid-1980s with approximately 1% of that amount (150–200 TBq)
3 released to the environment (Dowdall et al., 2005). Activities conducted at European nuclear
4 reprocessing facilities (mainly Sellafield and La Hague) have led to the increased radioactivity in
5 the Arctic marine environment. Increased discharge of ^{99}Tc from Sellafield beginning in 1994
6 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
7 (Dowdall et al., 2005).

8 Depending upon the redox conditions, Tc exists in two stable oxidation states. It forms a
9 reduced species [predominantly Tc(IV)] at redox potential (Eh) values below about 220 mV with
10 respect to standard hydrogen electrode in neutral pH conditions. At higher Eh, it occurs as
11 Tc(VII)O_4^- . Due to its weak interaction with mineral surfaces, TcO_4^- is considered one of the
12 most mobile radionuclides in the environment. In contrast, transport of Tc(IV) species ($\text{TcO}_2 \cdot$
13 $n\text{H}_2\text{O}$) are expected to be strongly retarded due to sorption and/or precipitation; the solubility of
14 $\text{TcO}_2 \cdot n\text{H}_2\text{O(s)}$ in carbonate-containing groundwater was reported to be about 10^{-8} M (Eriksen et
15 al., 1992). Similarly, chemical equilibrium modeling of groundwater at Yucca Mountain
16 indicated a maximum Tc solubility of $4\times 10^{-9}\text{ M}$ under reducing conditions (Arnold et al., 2006).
17 Experiments conducted on sediment-groundwater samples in Germany showed dramatically
18 different sorption coefficients for Tc among aerobic and anaerobic conditions (Lieser and
19 Bauscher, 1987). By varying the redox potential, they observed a change in the K_d value of about
20 three orders of magnitude over a small range of Eh at $170 \pm 60\text{ mV}$ and a pH of 7 ± 0.5 .

21 As with ^{99}Tc , ^{129}I has a unique and complex chemistry in the environment. The fate and
22 transport of ^{129}I is dictated by its chemical speciation. Aqueous iodine usually occurs as the
23 highly mobile iodide anion (I^-). Under more oxidizing conditions, iodine may be present as the
24 iodate anion (IO_3^-), which is more reactive than iodide and could be sorbed onto positively-

1 charged sites existing locally in clays and organic matter (Couture and Seitz, 1983; Sheppard and
2 Thibault, 1992; Hu et al., 2005). In contrast to ^{99}Tc , iodine has a minimal retardation under
3 reducing conditions when I^- is the predominant form, and multiple iodine species (I^- , IO_3^- , and
4 organic iodine species) are known to coexist in various aqueous systems (cf., Hu et al., 2005).

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

12 Study of short-lived iodine isotopes provides useful information about iodine species
13 released from nuclear facilities and its subsequent environmental behavior. According to
14 UNSCEAR (2000), the ^{131}I (half-life 8.04 days) inventory in the accidental reactor core at
15 Chernobyl was estimated to be 3.2×10^6 TBq and the total ^{131}I release during the course of the
16 accident was about 1.7×10^6 TBq (Table 8). From four air samples collected at 14.5 m above the
17 ground surface in Japan 10–23 days after the Chernobyl nuclear reactor accident date, Noguchi
18 and Murata (1988) reported the proportions of airborne ^{131}I species during the Chernobyl
19 accident: $19 \pm 9\%$ particulate iodine (iodine attached to particulate matter of mean diameter of 0.8
20 μm), $5 \pm 2\%$ I_2 , $6 \pm 3\%$ HIO and other non- I_2 inorganic iodines, and $70 \pm 11\%$ organic iodines (e.g.,
21 CH_3I). The depositional velocity of radioactive iodine from air to vegetation and water bodies,
22 and subsequent migration, varies with the iodine speciation. Other studies on species distribution
23 of radioactive iodine in and around nuclear power plants and of stable iodine in the environment
24 have also indicated the important proportion of organic iodines. Aldahan et al. (2006) reported

1 that the main forms of iodine released into the environment from European reprocessing
2 facilities are alkyl iodides, CH_3I and iodides (e.g., HI) that transform into water-soluble
3 molecules and/or IO_3^- , which are adsorbed on and/or nucleate into aerosols and return to Earth's
4 surface as wet and dry fallouts.

5 Careful attention must be given to the iodine speciation when interpreting the
6 biogeochemical behavior of iodine in the environment. Hu et al. (2007a) studied the content and
7 speciation of stable iodine in representative surface soils, and sorption and transport behavior of
8 different iodine species (iodide, iodate, and 4-iodoaniline as a representative refractory organic
9 iodine) in sediments collected at numerous nuclear facilities in the United States. In natural soils,
10 iodine is mostly (nearly 90% of total iodine) present as organic species, while inorganic iodine
11 becomes important (up to 50%) only in sediments with low organic matter. Results from
12 laboratory column studies showed much greater retardation of 4-iodoaniline than iodide or
13 iodate.

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

1 **4. Conclusions**

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

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

12 Knowledge of the speciation and reactions of multi-valent radionuclides, such as ^{99}Tc ,
13 ^{129}I and ^{237}Np , is very important for understanding their transport behavior in the environment
14 and for managing and remediating radionuclide contamination at legacy nuclear operations
15 facilities. These radionuclides possess dynamic sorption-precipitation behavior as they are redox-
16 sensitive elements that will respond to the change of redox condition along transport pathways.
17 Moreover, iodine released to the environment has multiple species (inorganic and organic) with
18 different hydrophilic, atmophilic, and biophilic characteristics, which have to be taken into in the
19 study of environmental radioactivity. Understanding biogeochemical behavior of radionuclides is
20 critical in meeting the challenges of the new nuclear era.

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 2 (Figure 1).

3
 4 **References**

- 5 Adlienè, D., Rääf, C., Magnusson, Å., Behring, J., Zakaria, M., Adlys, G., Skog, G., Stenström,
 6 K., Mattsson, S., 2006. Assessment of the environmental contamination with long-lived
 7 radionuclides around an operating RBMK reactor station. *J. Environ. Radioact.* 90, 68–77.
- 8 Ahearne, J.F., 1997. Radioactive waste: The size of the problem. *Physics Today* 50(6), 24–29.
- 9 Aldahan, A., Kekli, A., Possnert, G., 2006. Distribution and sources of ¹²⁹I in rivers of the Baltic
 10 region. *J. Environ. Radioact.* 88, 49–73.
- 11 Arnold, B.W., Meijer, A., Kalinina, E., Robinson, B.A., Kelkar, S., Jove-Colon, C., Kuzio, S.P.,
 12 James, S., Zhu, M., 2006. Impacts of reducing conditions in the saturated zone at Yucca
 13 Mountain. Proceedings of the 11th International High-Level Radioactive Waste
 14 Management Conference (IHLRWM), pp. 345–352. Las Vegas, NV.
- 15 Balonov, M.I., 2007. The Chernobyl Forum: Major findings and recommendations. *J. Environ.*
 16 *Radioact.* 96, 6–12.
- 17 Baum, E.M., Knox, H.D., Miller, T.R., 2002. *Nuclides and Isotopes*. 16th Edition. Knolls Atomic
 18 Power Laboratory, Inc.
- 19 Beals, D.M., Hayes, D.W., 1995. Technetium-99, iodine-129 and tritium in the waters of the
 20 Savannah River Site. *Sci. Total Environ.* 173/174, 101–115.
- 21 Beasley, T.M., Dixon, P.R., Mann L.J., 1998. Tc-99, U-236, and Np-237 in the Snake River
 22 Plain aquifer at the Idaho National Engineering and Environmental Laboratory, Idaho Falls,
 23 Idaho. *Environ. Sci. Technol.* 32, 3875–3881.
- 24 Bellona (The Bellona Foundation), 2003. Sellafield.
 25 http://bellona.org/filearchive/fil_sellaengweb.pdf (accesses on 10/30/2007)

- 1 BSC (Bechtel SAIC Company), 2001. Total System Performance Assessment - Analyses for
2 Disposal of Commercial and DOE Waste Inventories at Yucca Mountain - Input to Final
3 Environmental Impact Statement and Site Suitability Evaluation. Las Vegas, Nevada.
4 <http://www.ocrwm.doe.gov/documents/sl986m3b/index.htm> (accessed on 10/30/2007)
- 5 Bondietti, E.A., Francis, C.W., 1979. Geologic migration potentials of Tc-99 and Np-237.
6 *Science* 203, 1337–1340.
- 7 Buddemeier, R.W., Hunt, J.R., 1988. Transport of colloidal contaminants in groundwater:
8 Radionuclide migration at the Nevada Test Site. *Appl. Geochem.* 3, 535–548.
- 9 Christensen, G.C., Romanov, G.N., Strand, P., Salbu, B., Malyshev, S.V., Bergan, T.D.,
10 Oughton, D., Drozhko, E.G., Glagolenko, Y.V., Amundsen, I., Rudjord, A.L., Bjerk, T.O.,
11 Lind, B., 1997. Radioactive contamination in the environment of the nuclear enterprise
12 'Mayak' PA. Results from the joint Russian-Norwegian field work in 1994. *Sci. Total*
13 *Environ.* 202, 237-248.
- 14 Choppin, G.R., 2003. Actinide speciation in the environment. *Radiochim. Acta* 91(11), 645–649.
- 15 Coles, D., Ramspott, L., 1982. Migration of ruthentium-106 in a Nevada Test Site aquifer:
16 Discrepancy between field and laboratory results. *Science* 215, 1235–1237.
- 17 Cochran, T.B., Arkin, W.M., Norris, R.S., Sands, J.I., 1989. Nuclear Weapons Databook Series
18 Volume IV: Soviet Nuclear Weapons. Natural Resource Defense Council, Washington, DC.
- 19 Couture, R.A., Seitz, M.G., 1983. Sorption of anions of iodine by iron oxides and kaolinite.
20 *Nucl. Chem. Waste Manag.* 4, 301–306.
- 21 Crowley, K.D., 1997. Nuclear waste disposal: The technical challenges. *Physics Today* 50(6),
22 32–39.
- 23 de Marsily, G., Ledoux, E., Barbreau, A., Margat., J., 1977. Nuclear waste disposal: can the
24 geologist guarantee isolation? *Science* 197(4303), 519–527.

- 1 Dowdall, M., Gerland, S., Karcher, M., Gwynn, J.P., Rudjord, A.L., Kolstad, A.K., 2005.
2 Optimisation of sampling for the temporal monitoring of technetium-99 in the Arctic marine
3 environment. *J. Environ. Radioac.* 84, 111–130.
- 4 Dozol, M., Hagemann, R., 1993. Radionuclide migration in groundwaters: Review of the
5 behaviour of actinides. *Pure and Appl. Chem.* 65, 1081-1102.
- 6 Eikenberg, J., Beer, H., Bajo, S., 2004. Anthropogenic radionuclides emissions into the
7 environment. R. Gieré and P. Stille (Editors) *Energy, Waste and the Environment: A
8 Geochemical Perspective*. Geological Society Special Publication 236, pp. 143-151. The
9 Geological Society, London.
- 10 Eisenbud, M., Gesell, T., 1997. *Environmental Radioactivity from Natural, Industrial, and
11 Military Sources*. 4th Edition, Academic Press, San Diego, CA.
- 12 Eriksen, T.E., Ndalamba, P., Bruno, J., Caceci, M., 1992. The solubility of $TcO_2 \cdot nH_2O$ in
13 neutral to alkaline solutions under constant pCO_2 . *Radiochim. Acta* 58/59, 67–70.
- 14 Ewing, R.C., 2004. Environmental impact of the nuclear fuel cycle. R. Gieré and P. Stille
15 (Editors) *Energy, Waste and the Environment: A Geochemical Perspective*. Geological
16 Society Special Publication 236, pp. 7–23. The Geological Society, London.
- 17 Fiévet, B., Voiseux, C., Rozet, M., Masson, M., Bailly du Bois, P., 2006. Transfer of radiocarbon
18 liquid releases from the AREVA La Hague spent fuel reprocessing plant in the English
19 Channel. *J. Environ. Radioact.* 90(3): 173–196.
- 20 Fréchou, C., Calmet, D., 2003. ^{129}I in the environment of the La Hague nuclear fuel reprocessing
21 plant-from sea to land. *J. Environ. Radioact.* 70, 43–59.
- 22 Fritz, B.G., Patton, G.W., 2006. Monitoring iodine-129 in air and milk samples collected near
23 the Hanford Site: an investigation of historical iodine monitoring data. *J. Environ. Radioact.*
24 86, 64–77.
- 25 Gephart, R.E., 2003. *Hanford: A Conversation about Nuclear Waste and Cleanup*. Battelle Press,
26 Columbus, Ohio.

- 1 Hardy, E.P., Krey, P.W., Volchok, H.L., 1973. Global inventory and distribution of fallout
2 plutonium. *Nature* 241, 444–446.
- 3 Hou, X., Dahlgaard, H., Nielsen, S.P., 2001. Chemical speciation analysis of ^{129}I in seawater and
4 a preliminary investigation to use it as a tracer for geochemical cycle study of stable iodine.
5 *Marine Chem.* 74, 145–155.
- 6 Hu, Q.H., Smith, D.K., 2004. Field-scale migration of ^{99}Tc and ^{129}I at the Nevada Test Site. J.M.
7 Hanchar, S. Stroes-Gascoyne, L. Browning (eds.) *Scientific Basis for Nuclear Waste*
8 *Management XXVIII*, Materials Research Society Symposium Proceedings, Volume 824,
9 pp. 399–404. Materials Research Society, Pittsburgh, PA, USA.
- 10 Hu, Q.H., Moran, J.E., Blackwood, V., 2007a. Geochemical cycling of iodine species in soils.
11 *Comprehensive Handbook of Iodine: Nutritional, Endocrine and Pathological*. V.R. Preedy,
12 G.N. Burrow and R.R. Watson, Editors, Elsevier, Inc, San Diego, USA (under review).
- 13 Hu, Q.H., Zhao, P., Moran, J.E., Seaman, J.C., 2005. Sorption and transport of iodine species in
14 sediments from the Savannah River and Hanford Sites. *J. Contam. Hydrol.* 78(3), 185–205.
- 15 Hu, Q.H., Rose, T.P., Zavarin, M., Smith, D.K., Moran, J.E., Zhao, P.H., 2007b. Assessing field-
16 scale migration of radionuclides at the Nevada Test Site: “Mobile” species. *J. Contam.*
17 *Hydrol.*, (under review).
- 18 IAEA (International Atomic Energy Agency), 1998. The radiological situation at the atolls of
19 Mururoa and Fangataufa: Inventory of radionuclides underground at the atolls. Technical
20 Report, Vol. 3, Vienna.
- 21 IAEA (International Atomic Energy Agency), 2001. Inventory of accidents and losses at sea
22 involving radioactive materials. IAEA-TECDOC-1242.
- 23 IAEA (International Atomic Energy Agency), 2007a. The Semipalatinsk test site, Kazakhstan.
24 <http://www-ns.iaea.org/appraisals/semipalatinsk.htm> (accessed 10/30/2007)
- 25 IAEA (International Atomic Energy Agency), 2007b. Annual Report 2006.
26 http://www.iaea.org/Publications/Reports/Anrep2006/anrep2006_full.pdf (accessed
27 10/30/2007)

- 1 Jean-Baptiste, P., Baumier, D., Fourre, E., Dapoigny, A., Clavel, B., 2007. The distribution of
2 tritium in the terrestrial and aquatic environments of the Creys-Malville nuclear power plant
3 (2002-2005). *J. Environ. Radioac.* 94, 107–118.
- 4 Johnson, J., 2007. Reprocessing key to nuclear plan. *Chemical and Engineering News*, pp. 48–
5 54, June 18, 2007.
- 6 Kaplan, D.L., Serne, R.J., 1998. Pertechnetate exclusion from sediments. *Radiochim. Acta* 81,
7 117–124.
- 8 Kaszuba, J.P., Runde, W.H., 1999. The aqueous geochemistry of neptunium: Dynamic control of
9 soluble concentrations with applications to nuclear waste disposal. *Environ. Sci. Technol.*
10 33, 4427–4433.
- 11 Keith-Roach, M.J., Day, J.P., Fifield, L.K., Livens, F.R., 2001. Measurement of Np-237 in
12 environmental mater samples by accelerator mass spectrometry. *Analyst* 126, 58–61.
- 13 Kenna, T.C., 2002. Determination of plutonium isotopes and neptunium-237 in environmental
14 samples by inductively coupled plasma mass spectrometry with total sample dissolution.
15 *J. Anal. At. Spectrom.* 17, 1471–1479.
- 16 Keogh, S. M., Aldahan, A., Possnert, G., Finegan, P., Leon Vintro, L., Mitchell, P.I., 2007.
17 Trends in the spatial and temporal distribution of ^{129}I and ^{99}Tc in coastal waters surrounding
18 Ireland using *Fucus vesiculosus* as a bio-indicator. *J. Environ. Radioact.* 95, 23–38.
- 19 Kersting, A.B., Efurud, D.W., Finnegan, D.L., Rokop, D.J., Smith, D.K., Thompson, J.L., 1999.
20 Migration of plutonium in groundwater at the Nevada Test Site. *Nature* 397, 56–59.
- 21 Lieser, K.H., Bauscher, C.H., 1987. Technetium in the hydrosphere and in the geosphere. I.
22 Chemistry of technetium and iron in natural waters and influence of the redox potential on
23 the sorption of technetium. *Radiochim. Acta* 42, 205–213.
- 24 Lieser, K.H., Hill, R., Muhlenweg, U., Singh, R.N., Tu, S.D., Steinkopff, T., 1991. Actinides in
25 the environment. *J. Radioanal. Nucl. Chem.* 147, 117–131.

- 1 Lu, J.-G., Huang, Y., Li, F., Wang, L., Li, S., Hsia, Y., 2006. The investigation of ^{137}Cs and ^{90}Sr
2 background radiation levels in soil and plant around Tianwan NPP, China. *J. Environ.*
3 *Radioact.* 90(2): 89–99.
- 4 Macfarlane, A.M., Miller, M., 2007. Nuclear energy and uranium resources. *Element*, 3(3): 185–
5 192.
- 6 Moran, J.E., Oktay, S., Santschi, P.H., Schink, D.R., 1999. Atmospheric dispersal of ^{129}I iodine
7 from nuclear fuel reprocessing facilities. *Environ. Sci. Technol.* 33, 2536–2542.
- 8 Nakata, K., Nagasaki, S., Tanaka, S., Sakamoto, Y., Tanaka, T., Ogawa, H. 2002. Sorption and
9 reduction of neptunium (V) on the surface of iron oxides. *Radiochim. Acta* 90, 665–669.
- 10 Noguchi, H., Murata, M., 1988. Physicochemical speciation of airborne I-131 in Japan from
11 Chernobyl. *J. Environ. Radioact.* 7, 65–74.
- 12 Novikov, A.P., Kalmykov, S.N., Utsunomiya, S., Ewing, R.C., Horreard, F., Merkulov, A.,
13 Clark, S.B., Tkachev, V.V., Myasoedov, B.F., 2006. Colloid transport of plutonium in the
14 far-field of the Mayak Production Association, Russia. *Science* 314(5799), 638-641.
- 15 Nuclear Threat Initiative, 2007.
16 <http://www.nti.org/db/nisprofs/russia/fissmat/pumayak/pomayak.htm> (accessed on
17 10/30/2007).
- 18 OCRWM (Office of Civilian Radioactive Waste Management), 2007. Radioactive waste: an
19 international concern. <http://www.ocrwm.doe.gov/factsheets/doeymp0405.shtml> (accessed
20 on 10/30/2007).
- 21 Pfingsten, W., Hadermann, J., Perrochet, P., 2001. Radionuclide release and transport from
22 nuclear underground tests performed at Mururoa and Fangataufa - Predictions under
23 uncertainty. *J. Contam. Hydrol.* 47, 349–363.
- 24 Roussel-Debet, S., Gontier, G., Siclet, F., Fournier, M., 2006. Distribution of carbon 14 in the
25 terrestrial environment close to French nuclear power plants. *J. Environ. Radioact.* 87, 246–
26 259.

- 1 Scientific American, 2007. Nuclear Power Reborn. 09/26/2007.
2 <http://www.sciam.com/article.cfm?chanID=sa025&ref=feedburner&articleId=43D0C499->
3 [E7F2-99DF-30F52701AD131FD0](http://www.sciam.com/article.cfm?chanID=sa025&ref=feedburner&articleId=43D0C499-E7F2-99DF-30F52701AD131FD0).
- 4 Sheppard, M.I., Thibault, D.H., 1992. Chemical behavior of iodine in organic and mineral soils.
5 *Appl. Geochem.* 7, 265–272.
- 6 Silva, R.J., Nitsche, H., 1995. Actinide environmental chemistry. *Radiochim. Acta* 70/71, 377–
7 396.
- 8 Smith, D.K., Finnegan, D.L., Bowen, S.M., 2003. An inventory of long-lived radionuclides
9 residual from underground nuclear testing at the Nevada test site, 1951–1992. *J. Environ.*
10 *Radioact.* 67, 35–51.
- 11 Sohler, A., Hardeman, F., 2006. Radiological Dispersion Devices: Are we prepared? *J. Environ.*
12 *Radioac.* 85,171–181.
- 13 Solodov, I.N., Zotov, A.V., Khoteev, A.D., Mukhamet-Galeev, A.P., Tagirov, B.R. and Apps, J.
14 A., 1998. Geochemistry of natural and contaminated subsurface waters in fissured bed rocks
15 of the Lake Karachai area, Southern Urals, Russia. *Appl. Geochem.* 13, 921–939.
- 16 Tompson, A.F.B., Bruton, C.J., Pawloski, G.A., Smith, D.K., Bourcier, W.L., Shumaker, D.E.,
17 Kersting, A.B., Carle, S.F., Maxwell R.M., 2002. On the evaluation of groundwater
18 contamination from underground nuclear tests. *Environ. Geol.* 42, 235–247.
- 19 Um, W., Serne, R.J., Krupka, K.M., 2004. Linearity and reversibility of iodide adsorption on
20 sediments from Hanford, Washington under water saturated conditions. *Water Res.* 38,
21 2009–2016.
- 22 UNSCEAR (United Nations Scientific Committee on the Effects of Atomic Radiation), 2000.
23 Sources and effects of ionizing radiation; Annex J. Exposure and effects of Chernobyl
24 accident. Report to General Assembly, United Nations, New York.
- 25 USDOE/NV (U.S. Department of Energy, Nevada Operations Office), 2000. United States
26 Nuclear Tests, July 1945 through September 1992. Las Vegas, U.S. Department of Energy,
27 Nevada Operations Office.

- 1 USEPA (U.S. Environmental Protection Agency), 1991. 40 CFR Parts 141 and 142; National
2 Primary Drinking Water Regulations; Radionuclides; Proposed Rule. Federal Register. Vol.
3 56, No. 138, pp. 33050-33127. Thursday, July 18, 1991.
- 4 Wang, J., Xue, W.M., Zheng, H.L., Su, R., 2001. Preliminary site characterization at Beishan,
5 Northwest China—A potential site for China’s high-level radioactive waste repository.
6 Witherspoon, P. A. and G. S. Bodvarsson, Eds. (2001). Geological Challenges in
7 Radioactive Waste Isolation. Lawrence Berkeley National Laboratory, University of
8 California, Berkeley, CA, LBNL–49767.
- 9 Washington State Department of Health, 2007. The release of radioactive materials from
10 Hanford: 1944–1972.
11 <http://www.doh.wa.gov/hanford/publications/history/release.html#Estimates> (accessed
12 10/31/2007).
- 13 Webster, P., 2003. Haunted by Red October. *Science* 301, 1460–1463.
- 14 Witherspoon, P.A., Bodvarsson, G.S. (Eds.), 2001. Geological Challenges in Radioactive Waste
15 Isolation. Lawrence Berkeley National Laboratory, University of California, Berkeley, CA,
16 LBNL–49767.
- 17 World Nuclear Association, 2007a. World Nuclear Power Reactors 2006-07 and Uranium
18 Requirements. <http://www.world-nuclear.org/info/reactors.html> (accessed on
19 10/30/2007).
- 20 World Nuclear Association, 2007b. Nuclear Power in China. [http://www.world-](http://www.world-nuclear.org/info/inf63.html)
21 [nuclear.org/info/inf63.html](http://www.world-nuclear.org/info/inf63.html) (accessed on 10/30/2007).

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Table 1. Radionuclides released into the air at Hanford during 1944–1972

Radionuclide	Radioactive half-life ¹	Radiation released into the air (TBq) ²
³ H	12.32 a	7400
⁶⁰ Co	5.271 a	0.037
⁸⁵ Kr	10.76 a	703000
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	27380
¹³² Te	3.20 d	148
¹³³ Xe	5.243 d	15540
¹³⁷ Cs	30.07 a	1.554
¹⁴⁴ Ce	284.6 d	141
²³⁹ Pu	24,100 a	0.067

¹ From Baum et al. (2002); a: year; d: day.

² from Washington State Department of Health (2007).

Table 2. Radionuclides released into the Columbia River at Hanford during 1944–1972

Radionuclide	Radioactive half-life ¹	Radiation released into Columbia River (TBq) ²
²⁴ Na	14.96 h	481000
³² P	14.28 d	8510
⁴⁶ Sc	83.81 d	4440
⁵¹ Cr	27.702 d	266400
⁵⁶ Mn	2.578 h	2960000
⁶⁵ Zn	243.8 d	18130
⁷² Ga	14.1 h	136900
⁷⁶ As	26.3 h	92500.0
⁹⁰ Y	2.67 d	16650
¹³¹ I	8.04 d	1776
²³⁹ Np	2140000 a	233100

¹ From Baum et al. (2002); a: year; d: day; h: hour.

² from Washington State Department of Health (2007).

Table 3. Radionuclide inventory from underground nuclear tests at the Nevada Test Site and French Polynesia

Radio-nuclide	Half-life (yr) ¹	Radioactivity (TBq)	
		USA Nevada Test Site ²	French Polynesia ³
³ H	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
^{93m} 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
^{113m} Cd	14.1	3.38E+01	1.86E+00
^{121m} 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
^{166m} 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

¹ From Baum et al. (2002). Read 7.1E+5 as 7.1×10^5 .

² Radioactivities on September 23, 1992 reported in Smith et al. (2003) were decay corrected to January 1, 2008. NA: not available.

³ 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 countries¹

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 2035–2080
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

¹From Witherspoon and Bodvarsson (2001), OCRWM (2007) and World Nuclear Association (2007a); TBD: to be determined.

Table 5. Discharge permits from Sellafield to the air before 2001¹

Radionuclide	Present aerial discharge limit (GBq/y)
³ H	1500000
¹⁴ C	7300
³⁵ S	210
⁴¹ Ar	3700000
⁶⁰ Co	0.92
⁸⁵ Kr	590000000
⁹⁰ Sr	9.4
¹⁰⁶ Ru	56
¹²⁵ Sb	5
¹²⁹ I	70
¹³¹ I	55
¹³⁷ Cs	18
Pu-alpha	1.2
²⁴¹ Pu	17
²⁴¹ Am+ ²⁴² Cm	0.74
Total alpha	2.5
Total beta	340

¹ from Bellona (2003).

Table 6. Discharge to the sea from Sellafield¹

Radionuclide	Discharge in 1999 (TBq)	Discharge authorization (TBq)
¹⁴ C	5.8	20.8
⁹⁰ Sr	31	48
⁹⁹ Tc	69	90
¹⁰⁶ Ru	2.7	63
¹²⁹ I	0.48	2
²⁴¹ Pu	2.9	27
²⁴¹ Am	0.03	0.3
Pu-alpha	0.11	0.7
Total alpha	0.13	1
Total beta	110	400
Uranium (kg)	540	2040

¹ from Bellona (2003).

Table 7. 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	5,715	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	1,599	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	7,880	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	3.40E+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	6,560	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	7,370	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 8. Estimates of principal radionuclides released in the Chernobyl accident

Radionuclide		Radioactive half-life ¹	Activities released (TBq) ²
Noble gases	¹³³ Xe	5.243 d	6.50E+06
	¹³² Te	3.20 d	1.15E+06
Volatile	¹³¹ I	8.04 d	1.76E+06
	¹³⁴ Cs	2.065 a	5.40E+04
	¹³⁷ Cs	30.07 a	8.50E+04
	⁸⁹ Sr	50.52 d	1.15E+05
Intermediate	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	6,560 a	42
	²⁴¹ Pu	14.4 a	6.00E+03
	²⁴² Cm	162.8 d	900

¹ From Baum et al., (2002).

² From UNSCEAR (2000).

Table 9. Radioactivity involved in the nuclear submarine accident¹

Country	Date	Vessel involved	Radioactive material involved	Total activity (TBq)	Estimated activity released
USA	4/10/1963	SNN-593 "Thresher"	Nuclear reactor	1,150	0.04 GBq
	5/22/1968	SNN-583 "Scorpion"	Nuclear reactor; two nuclear warheads	1,300	0.04 GBq
Soviet Union	4/8/1970	K-8	2 reactors warheads(s)	9,250 0.03	-
	10/6/1986	K-219	2 reactors	9,250	-
	4/7/1989	K-278 "Komsomolets"	Reactor core	3,590	<370 GBq
Russian Federation	8/12/2000	K-141 "Kursk"	2 reactors	1–2×10 ⁶	-

Table 10. Inventory of ^{99}Tc and ^{129}I from various sources¹

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)	1,340	840	4,000	26

¹From Aldahan et al. (2006), Keogh et al. (2007), Moran et al. (1999), Smith et al. (2003), and Washington State Department of Health (2007).

Figure Captions

Figure 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.

Figure 1

