Radionuclides in the environment

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Abstract

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 green-house 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. This work focuses on the overview of radionuclide contamination from the nuclear operations, as well as the geochemical behavior of important radionuclides.

Introduction

Over 2 10^{20} Bq of radioactivity have been released into the atmosphere due to worldwide nuclear weapons tests since the first test in New Mexico in 1945. Controlled and uncontrolled releases of radioactivity from operation of nuclear reactors have been less than 3% of this amount (Choppin, 2003)

As a result of 828 underground nuclear tests conducted during 1951 to 1992 at Nevada Test Site (NTS) in USA, a total of about 4.9×10^{18} Bq of radioactivity was produced. In the proposed geological repository at Yucca Mountain, which is located within the NTS, about 2.6×10^{20} Bq of radioactivity from high-level nuclear waste and spent fuel will be stored for many generations to come.

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 in England and at La Hague in France. 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 4.5 and 6.0 TBq annually. 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 plutonium. Discharge of ^{99}Tc has been also high. In the three-year period from 1978 to 1980, Sellafield discharged almost 300 TBq of ^{99}Tc. And more importantly, current and future discharge of ^{99}Tc contributes a significant portion of the radioactivity, at about 90 TBq per year. Today it is the large releases of the radioactive ^{99}Tc from Sellafield that is largely responsible for the pollution of the Norwegian coast and the Barents Sea (The Bellona Foundation, 2003).

Materials and Methods

A suite of radionuclides, including ^{90}Sr, ^{99}Tc, ^{129}I, ^{137}Cs, ^{237}Np, ^{241}Am, as well as several uranium and plutonium isotopes, from the nuclear operations are of particular environmental importance because of their abundance, mobility, or toxicity. We compile the abundance and discuss their geochemical behavior of these important radionuclides at Yucca Mountain and the NTS of the United States.

Results

Table 1 presents the radionuclide inventory from the potential high-level geological repository at Yucca Mountain (BSC, 2001) and from the underground nuclear tests at the NTS, decayed corrected to September 23, 1992, the date of the last underground nuclear test (Smith *et al.*, 2003).

Discussion

^{90}Sr and ^{137}Cs are the major fission products, yet they do not pose long-term risk because of their 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.

Of particular importance to the environment and risk assessment are radionuclides ^{99}Tc, ^{129}I, and ^{237}Np, because of their long half-lives, presumably high mobility. These radionuclides possess dynamic sorption-precipitation behavior as they are redox-sensitive elements that will respond to the change of redox condition along groundwater flow path. Reducing zones in the subsurface will contribute to the delayed transport of important radionuclide, such as ^{99}Tc. Overall, ^{99}Tc may not necessarily exist as a mobile and conservative species $^{99}TcO_4^-$, as commonly assumed for the groundwater.

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Table 1. Radionuclide inventory at Yucca Mountain and the NTS

Radionuclide	Radioactive half life (yr)	Yucca Mountain		Nevada Test Site†	
		Isotope total (TBq)	Percentage of total inventory (%)	Isotope total (TBq)	Percentage of total inventory (%)
^{90}Sr	2.91E+01	9.58E+07	3.71E+01	8.06E+04	3.34E+01
^{99}Tc	2.13E+05	4.00E+04	1.55E-02	2.11E+01	8.75E-03
^{129}I	1.70E+07	8.71E+01	3.37E-05	6.51E-02	2.70E-05
^{137}Cs	3.02E+01	1.41E+08	5.47E+01	1.06E+05	4.38E+01
^{236}U	2.34E+7	7.46E+02	2.89E-04	3.47E-01	1.44E-04
^{237}Np	2.14E+06	9.82E+02	3.80E-04	1.80E+00	7.46E-04
^{239}Pu, ^{240}Pu	>6.5E+3	2.31E+06	8.92E-01	7.47E+03	3.10E+00
^{241}Am	4.32E+02	1.10E+07	4.24E+00	1.37E+03	5.69E-01

† Percentage does not consider the radioactivity from ^{3}H.