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SOURCES OF RADIOACTIVITY IN THE OCEAN ENVIRONMENT: FROM LOW LEVEL WASTE TO NUCLEAR POWERED SUBMARINES

KENNETH ALVIN SOLOMON

Rand Corporation, 1700 Main Street, Santa Monica, CA 90406-2138 (U.S.A.)

Summary

This paper summarizes both natural and man-made radioactivity in the marine environment. Radioactivity occurs naturally in both the sea water and in the ocean sediment. Radioactivity in the sea water is fairly uniform geographically and is dominated by the naturally occurring isotope ^{40}K (potassium-40). Unlike sea water, sediment radiation levels vary with sediment type and location. The primary source of natural radiation in the sediment results from deposition of insoluble thorium isotopes formed by the decay of water-soluble uranium.

Man-made sources of radioactivity arise from, in descending order of importance:

- Sinking of two U.S. and two Soviet nuclear submarines;
- Fallout from nuclear weapons testing;
- Dumping of primarily British and American low-level nuclear waste; and
- Dumping of reprocessing plant radiated effluents from the British Windscale facility and other European and Indian reprocessing facilities.

GLOSSARY

Activity: The radioactivity released from a given quantity of material measured in curies (or better in becquerels).

Activity-natural: Unstable nuclei which exist in nature such as ^{40}K.

Activity-induced: When a nucleus is made to react with a particle or electromagnetic radiation to form an unstable nucleus which did not previously exist in nature (e.g. ^{239}Pu).

Bioaccumulation: The build-up of radionuclides in organisms or in the food chain.

Curie: The quantity of a radioactive substance which undergoes 3.7×10^{10} radioactive transformations per second (becquerel, Bq). Curie is abbreviated as "Ci". A mCi is a thousandth of a curie (10^{-3} curie) and a pCi is a pico-curie or a millionth millionth of a curie (10^{-12} curie).

Decay Mechanisms: In their approach to a stable state, radioactive atoms may emit charged particles of various types: positive particles, (e.g., protons and positrons); uncharged par-

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ticles (neutrons, α); negative particles (electrons); or electromagnetic radiation (γ rays, X rays).

Alpha Decay: The α particle is identical with the nucleus of a helium atom. The emission of an α particle creates a new nucleus with an atomic number reduced by two (2) and an atomic weight decreased by three (3) or four (4) mass units.

Beta Decay: In β decay a negative electron is emitted from the nucleus of an atom. Since atomic number is defined by the amount of protons of the nucleus, ejection of a β^- particle increases the atomic number by unity (one neutron gives one proton + electron).

Positron Decay (β^+): The positron is a particle whose mass is the same as that of an electron and whose charge is equal in manitude but opposite in sign. Positron emission represents a mechanism whereby a nucleus can decrease its atomic number by unity.

Gamma-ray Emission: The γ ray is electromagnetic radiation originating in the nucleus, as distinguished from X rays, which usually are less energetic and which arises from energy adjustments as electrons move between orbital shells outside the nucleus. Gamma rays are usually emitted instantaneously when a neutron is captured in a nucleus (n, γ reaction). Gamma rays are frequently emitted following ejection of a particle from a radioactive nucleus. In a few cases (isomeric nuclear transitions), radioative nuclei emit γ rays alone.

Neutron Decay (n): A few cases are known in which unstable atoms emit neutrons. The fission products: ^{87}Kr and ^{137}Xe and the isotope ^{17}O are examples.

Fission product: A nuclide produced by the fission of a heavier element and daughters of fission products.

Half-life: The time required for a radioactive substance to decay to half its original activity level.

High-level waste: Includes reprocessing wastes, such as high-level liquid wastes and products from solidification of high-level liquid waste; also includes irradiated fuel elements, i.e., spent fuel.

Low-level waste: Any radioactive waste which is not high-level or transuranic.

Macrofauna: Nekton and large benthic fauna.

Microfauna: Plankton and microorganisms at all ocean depths and in the deep seabed.

Rem: (röntgen equivalent man) expresses the effective dose equivalent for all forms of ionizing radiation. It is the product of the absorbed dose

and factors related to biological significance. A mRem or millirem is one thousandth of a rem (1 rem = 10^{-2} Gy; Gy stands for Gray).

Spent fuel: nuclear reactor fuel that has been used and is ready for reprocessing or ultimate disposal.

Tonne: Metric ton, or 1000 kilograms.

Introduction

This paper is limited in scope; it reports on the amount of radiation — both natural and man made — that exists in the marine environment. It does not assess the health risk from this radioactivity. It draws from prior work of the author[1] and considers some more recent information.

This paper was inspired by the October 13, 1986 sinking of a nuclear-powered Soviet submarine about 1500 kilometers off the Southeast Coast of the United States. Concern has been raised in a number of people's minds as to how damaging this sinking is to the environment. The author hopes to address this question, in part, by comparing the amount of radioactivity on board this submarine with what already exists in the marine environment.

Radiation in the marine environment is both nature-induced and human-induced. That which is naturally induced persists in both the sea water and the sediment. That which is human-induced also persists both in the sea water and the sediment and originates from four primary source categories: sinking of nuclear-powered submarines; atmospheric and ocean testing of nuclear weapons; dumping of low-level radioactive waste; and releasing of effluents from spent-fuel reprocessing facilities.

Natural radiation

Radioactivity in sea water is fairly uniform geographically and is dominated by the naturally occurring isotope ^{40}K (potassium-40). The activity level of sea water is 330 pCi/l. Measured as a radioactive dose, it is about 0.1 mRem/h.[2] Unlike sea water, sediment radiation levels vary with sediment type and location. In part, this variation results from the deposition of insoluble thorium isotopes formed from decaying of water soluble uranium. Thus, deep ocean sediments with a thick overlying water column tend to have larger concentra-

[1]See: M.B. Triplett and K.A. Solomon, *Monitoring Technologies for Ocean Disposal of Radioactive Waste*, R-2773-NOAA, The RAND Corporation, January 1982; K.A. Solomon and K. Wolf, "A Summary of Monitoring Requirements for Sub-Seabed Disposal of Radioactive Waste", *Journal of Hazardous Materials*, (10), 1985, pp. 205-226.

[2]A pCi/l is a pico-curie per liter. A pico-curie is one trillionth (or one millionth, millionth, or 10^{-12} curies). A mRem is one thousandth of a Rem (i.e. 10^{-3} Rem). B comparison, a chest X-Ray is 50 mRem and a dental X-Ray is about 5 or 10 mRem.

tion of alpha-emitting thorium isotopes than coastal waters have. Dose[3] rates for biological species in deep ocean red clays range from 9.9 to 38 mRem/h for alpha emitters. Because of the short range of alpha particles, this dose/rate may most significantly affect microorganisms, microfauna and sediment feeders.

Man-made radiation

Humans have been responsible for dumping radioactivity in the ocean environment both intentionally and unintentionally.

Intentional insertion into the marine environment

Intentional dumping occurred from three primary sources: low-level radioactive waste (LLW) disposal; spent-fuel reprocessing plants effluents; and nuclear weapons testing debris. The United States and Western Europe disposed of the majority of the low level waste in the marine environment. Virtually all U.S. dumping occurred between 1946 and 1961. A limited amount of U.S. dumping also took place between 1961 and 1970. European countries, under the auspices of the Nuclear Energy Agency (NEA), continue dumping LLW at a single site in the northeast Atlantic Ocean. Through 1970, the United States had disposed of more than 86,000 containers (55-gallon drums), or roughly 61,000 Ci of LLW; approximately 33,000 Ci of induced radioactivity in the *Sea Wolf* reactor vessel brings the U.S. total to 94,000 Ci. The United Kingdom and other NEA members through 1976 have disposed of 114,000 tonnes of material containing 340,000 Ci. Thus, in 30 years, waste disposal programs of the United States and Europe have dumped less than 500,000 Ci of mostly packaged waste at sites ranging in depth from 900 to 3800 meters.

By contrast, a single nuclear fuel reprocessing plant in the United Kingdom releases more than 225,000 Ci annually to coastal waters. Reprocessing plants separate plutonium and reusable uranium from spent nuclear fuel, and in the process, generate high-level waste and a large volume of low-level waste. The Windscale plant in the United Kingdom releases its low-level wastes directly to coastal waters. The French reprocessing plant at La Hague also releases LLW to coastal waters (the English Channel), but data are not available on the amount. Italy and India also have reprocessing plants adjacent to coastal waters, but they are considerably smaller than Windscale.

Nuclear reactor accidents

While reprocessing plants are the principal contributors to marine radioactivity from the nuclear fuel cycle, nuclear power plants also routinely discharge

[3]Dose refers to the product of absorbed radiation dose and appropriate factors to account for differences in biological response due to the quality of radiation and its distribution in the body. The unit of measure is the rem (or in Gray, 1 rem = 10^{-2} Gy).

from 1 to 10 pCi/l in their cooling water. For a 1000 MWe (megawatt electric) reactor, this corresponds to an annual release of roughly 1 to 10 curies. At the other extreme, Three Mile Island released from 2.4×10^6 (2.4 million) to 13×10^6 (thirteen million) curies of ^{135}Xe (Xenon 135) to the air during the accident. By May 6, 1986 the April 25, 1986 accident at Chernobyl released 45×10^6 curies of ^{133}Xe (which is about the equivalent to the Chernobyl core inventory of ^{133}Xe.) Both the Three Mile Island and Chernobyl accidents released other isotopes. Specifically, the accident at Chernobyl released about 7.3×10^6 curies of ^{131}I (Iodine-131); 4.3×10^6 curies of ^{40}Ba (Barium-40); 2.2×10^6 curies of ^{89}Sr (Strontium-89); 0.22×10^6 curies of ^{90}Sr; and approximately 20×10^6 curies of about a dozen other isotopes.

Nuclear weapons testing

Another source of intentional release is from nuclear weapons testing. Fallout from nuclear weapons tests contributes to marine radioactivity principally from deposition of airborne contaminants and from underwater bursts. ^{90}Sr (Strontium-90) and ^{137}Cs (Cesium-137) are two of the more prevalent fission products. The National Academy of Sciences estimates[4] that 21×10^6 curies of ^{90}Sr and 34×10^6 curies of ^{137}Cs were released from airborne and surface nuclear tests. Roughly 15×10^6 curies of ^{90}Sr were deposited on the earth's surface between 1945 and 1966.

Submarine sinkings

Another man-made source of marine radioactivity resulted from the sinking of two U.S. nuclear-powered submarines, the *Thresher* in 1963, and the *Scorpion* in 1968. Two Soviet nuclear submarines sank as well, the more recent one on October 13, 1986. While actual curie content of their reactor core depends on the fuel mix, inventory, and burn-up (which I do not know), based on a small conventional reactor, these submarines could have from 10^8 to 10^9 (one hundred million to one billion) curies. However, as of 1979 measurements of water, sediment, and debris at both U.S. sites have not shown any evidence of radioactivity released from either submarine.[5] The reactor vessel itself or the fuel rods — at the time the measurements were taken — may have still been intact, or sufficiently intact, to prevent contamination of the surrounding environment. Whether they still remain intact today is not known; however, we do know that metallic fuel elements will not easily release their radionuclides, except during fuel melting.

We may in fact assume that the reactors on board the Soviet submarine have

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[4]A.B. Joseph et al. "Sources of Radioactivity and Their Characteristics" in *Radioactivity in the Marine Environment*, National Academy of Sciences, 1971, p. 19, Table 7.

[5]*Environmental Monitoring and Disposal of Radioactive Wastes from U.S. Naval Nuclear Powered Ships and Their Support Facilities*, NAVSEA Report, NT-79-1, Jan. 1979, pp. 8-9.

TABLE 1

Radioactivity in the marine environment (Illustrative examples)

Source	Description	Activity level[a]
Natural radioactivity		
Sea water	^{40}K accounts for most of the activity. ^{87}Rb and ^{3}H contribute significant but lesser amounts. Activity level is fairly constant in all parts of ocean.	330 pCi/l[b] (about 0.1 mRem/h)
Sediments	Activity levels vary significantly, with ^{40}K a significant contributor throughout. Thorium isotopes and ^{226}Ra are major constituents in deep sea.	Coastal sediments 2-32 pCi/g[c] Deep ocean red clay 30-100 pCi/g (about 10-38 mRem/h) Globigerina ooze 6-20 pCi/g
Radioactive waste disposal		
United States	1946-1970: Deposited over 86,000 containers (34,000 in the Atlantic dumpsites and 52,000 in the Pacific dumpsites[d])	61,000 Ci (46,000 Ci, Atlantic sites; 15,000 Ci, Pacific sites)
	Pressure vessel of the *Sea Wolf* reactor.	33,000 Ci Atlantic site[e]
United Kingdom and the Nuclear Energy Agency (NEA)	1951-1978: 14,000 metric tons	435,830 Ci[f]
Nuclear power programs		
Reprocessing plants[g]	Windscale in the United Kingdom is restricted to a total beta activity release of 300,000 Ci/y to coastal waters; total alpha of 6000 Ci/y. Principal components of total activity are ^{137}Cs, ^{106}Ru, ^{90}Sr, ^{241}Pu, and ^{3}H.	225,000 Ci/y[h]
Nuclear weapons tests	Through 1968, more than 350 weapons were tested, either above ground or in the ocean. These tests include: 2 underwater, 11 over the open ocean, 113 over or on coral islands, 79 on arctic islands.[i]	Releases to the atmosphere and earth's surface include: 21×10^6 Ci ^{90}Sr 34×10^6 Ci ^{137}Cs

Miscellaneous sources		
Nuclear submarine losses	*USS Thresher* sank in 2590 meters of water in 1963. *USS Scorpion* sank in 3050 meters of water in 1968.	Submarine nuclear fuel inventories are classified, but similar sized land-based power reactors contain from 10^8 Ci to 10^9 Ci.[j]

[a]All activity data in this table are reported in curies (1 Ci = 3.7×10^{10} nuclear transformations per second) to facilitate comparisons. This is an imperfect measure, as noted in the text, since the half-lives of isotopes vary and the resulting radiation (alpha, beta, gamma) also differs in the hazard it poses to biological species.

[b]The average dose rate to fish from this activity level is about 0.1 mRem/h. D.W. Woodhead and R.J. Pentreath, "A Provisional Assessment of Radiation Regime in Deep Ocean Environments," *Second International Ocean Dumping Symposium*, Woods Hole, Mass., April 15–18, 1980. Note: 1 pCi = 1×10^{-12} Ci.

[c]The activity levels in coastal sediments yield dose rates of 270 to 3300 μRem/h (alpha activity), 1.6 to 21 μRem/h (beta activity), and 1.5 to 16 μRem/h (gamma activity). Activity levels in deep ocean red clays yield dose rates of 9900 to 38,000 μRem/h (alpha activity), 18 to 65 μRem/h (beta activity), and 23 to 86 μRem/h (gamma activity). Activity levels in globigerina ooze yield dose rates of 2200 μRem/h (alpha), 3.7 μRem/h (beta), and 5.2 μRem/h (gamma). Woodhead and Pentreath, op. cit.

[d]Activity levels have been rounded off from the data reported in D.A. Deese, *Nuclear Power and Radioactive Waste*, D.C. Heath and Co., Lexington, Mass., 1978, p. 50 (Table 2-1).

[e]Estimated in A.B. Joseph et al., "Sources of Radioactivity and Their Characteristics," in *Radioactivity in the Marine Environment*, National Academy of Sciences, 1971, p. 37 (Table 22).

[f]The United Kingdom was responsible for waste dumping from 1951 to 1966. The NEA took control of dumping operations in 1967. The United Kingdom, France, Belgium, and Switzerland have recently used this site. See Deese, op. cit., and "Demonstrations Against Low-Level Sea Dumping," *Nuclear News*, August 1980, pp. 72–73.

[g]Data for reprocessing plants is illustrative and not meant to be complete. France, Italy, and India also have reprocessing plants adjacent to coastal waters, but Windscale is the largest and has readily accessible reports on its releases.

[h]This figure is based on average releases to coastal waters for 1977 and 1978. Included in the figure are approximately 32,000 Ci/y of ^{3}H and ^{241}Pu, which are not specifically regulated. British Nuclear Fuels Limited, "Annual Report on Radioactive Discharges and Monitoring of the Environment 1978," Health and Safety Directorate, Risley, Warrington, Cheshire, U.K., July 1979, pp. 10–11 (Tables 1–3).

[i]Joseph, op. cit., p. 9.

[j]Core activity depends on fuel mix, fuel inventory, and burnup which we do not know, but we have based our estimate on a 1000 MWe Pressurized Water Reactor after 550 full power days. For a reactor of this size, a core activity of 4×10^9 Ci results. Source: WASH-1400, "The Reactor Safety Study."

a similar radionuclide inventory and may in fact remain intact for some period of time.

The multiple missile warheads on board the Soviet submarine which recently sank probably present far less risk to the environment than does the reactor core itself. This is true because the weapons contain primarily isotopes of uranium and/or plutonium as the only active isotopes and the activities of these two isotopes is less than the combined activity of all isotopes in the burned reactor core.

Recaption

Table 1 summarizes the contribution from both man-made and natural sources. The data in Table 1 do not reflect all of the man-made sources.[6]

I use a common unit of radioactivity for comparison purposes, the curie. However, this is a less than perfect measure, especially when comparing isotopes that differ in their half-lives. For example, 10^6 curies of ^{3}H (tritium with a half life of 12 years) decays quickly, while 10^6 curies of ^{239}Pu (plutonium-239 with a half life of 24,000 years) decays far more slowly. Moreover, the type of radiation released (alpha, beta, gamma) and its energy spectrum combine to make comparisons by total activity less meaningful.

In Table 1, I also present a rough estimate of the millirem dose.

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[6]For instance, release data for only the Windscale reprocessing plant are shown. France, Italy, and India also have smaller reprocessing plants adjacent to or near coastal waters, but I do not have data for them.