RADIOACTIVE WASTES

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Introduction

The discovery and the history of radioactivity is closely connected to that of modern science. In 1896 Antoine Henri Becquerel observed and described the spontaneous emission of radiation by uranium and its compounds. Two years later, in 1898, the chemical research of Marie and Pierre Curie led to the discovery of polonium and radium.

In 1934 Frédéric Joliot and Irène Curie discovered artificial radioactivity. This discovery was soon followed by the discovery of fission and the enormous amounts of energy released by this process. However, few in the then limited community of scientists working with radioactivity believed that it would be possible within a fairly near future to establish enough resources to develop the fission process for commercial production of energy or even think about the development of mass-destruction weapons.

World War II made a dramatic change to this. The race that began in order to be the first to develop mass-destruction weapons based on nuclear energy is well known. Following this came the development of nuclear reactors for commercial production of electricity. From the rapidly growing nuclear industry, both military and commercial, radioactive waste was produced and became a problem. As with many other waste problems, discharges to the sea or ocean dumping were looked upon as the simplest and thereby the best and final solution.

The Sources

Anthropogenic radioactive contamination of the marine environment has several sources: disposal at sea, discharges to the sea, accidental releases and fallout from nuclear weapon tests and nuclear accidents. In addition, discharge of naturally occurring radioactive materials (NORM) from offshore oil and gas production is a considerable source for contamination.

The marine environment receives in addition various forms of radioactive components from medical, scientific and industrial use. These contributions are mostly short-lived radionuclides and enter the local marine environment through diffuse outlets like muncipal sewage systems and rivers.

Disposal at Sea

The first ocean dumping of radioactive waste was conducted by the USA in 1946 some 80 km off the coast of California. The International Atomic Energy Agency (IAEA) published in August 1999 an 'Inventory of radioactive waste disposal at sea' according to which the disposal areas and the radioactivity can be listed as shown in **Table 1**.

Figure 1 shows the worldwide distribution of disposal-points for radioactive waste.

The majority of the waste disposed consists of solid waste in various forms and origin, only 1.44% of the total activity is contributed by low-level liquid waste. The disposal areas in the north east Atlantic and the Arctic contain about 95% of the total radioactive waste disposed at sea.

Most disposal of radioactive waste was performed in accordance with national or international regulations. Since 1967 the disposals in the north-east Atlantic were for the most part conducted in accordance with a consultative mechanism of the Organization for Economic Co-operation and Development/Nuclear Energy Agency (OECD/NEA).

The majority of the north-east Atlantic disposals were of low-level solid waste at depths of 1500–5000 m, but the Arctic Sea disposals consist of various types of waste from reactors with spent fuel to containers with low-level solid waste dumped in fairly shallow waters ranging from about 300 m depth in the Kara Sea to less than 20 m depth in some fiords on the east coast of Novaya Zemlya.

Most of the disposals in the Arctic were carried out by the former Soviet Union and were not reported internationally. An inventory of the USSR disposals was presented by the Russian government in 1993. Already before this, the good collaboration between Russian and Norwegian authorities had led to a joint Norwegian–Russian expedition to the Kara Sea in 1992 followed by two more joint expeditions, in 1993 and 1994. The main purpose of these expeditions was to locate and inspect the most important dumped objects and to collect samples for assessing the present environmental impact and to assess the possibility for potential future leakage and environmental impacts.

Table 1 Worldwide disposal at sea of radioactive waste[a]

North-east Atlantic Ocean	42.32 PBq
North-west Atlantic Ocean	2.94 PBq
Arctic Ocean	38.37 PBq
North-east Pacific Ocean	0.55 PBq
West Pacific Ocean	0.89 PBq

[a]PBq (petaBq) = 10^{15} Bq (1 Bq = 1 disintegration s^{-1}). The old unit for radioactivity was Curie (Ci); 1 Ci = 3.7×10^{10} Bq.

These Arctic disposals differ significantly from the rest of the reported sea disposals in other parts of the world oceans as most of the dumped objects are found in shallow waters and some must be characterized as high-level radioactive waste, i.e. nuclear reactors with fuel. Possible releases from these sources may be expected to enter the surface circulation of the Kara Sea and from there be transported to important fisheries areas in the Barents and Norwegian Seas. **Figures 2** and **3** give examples of some of the radioactive waste dumped in the Arctic. Pictures were taken with a video camera mounted on a ROV (remote operated vehicle). The ROV was also equipped with a NaI-detector for gamma-radiation measurements and a device for sediment sampling close to the actual objects.

The Global Convention on the Prevention of Marine Pollution by Dumping of Wastes and Other Matter was adopted by an Intergovernmental Conference in London in 1972. The convention named the London Convention 1972, formerly the London Dumping Convention (LDC), addressed from the very beginning the problem of radioactive waste. But it was not until 20 February 1994 that a total prohibition on radioactive waste disposal at sea came into force.

Discharges to the Sea

Of the total world production of electricity about 16% is produced in nuclear power plants. In some countries nuclear energy counts for the majority of the electricity produced, France 75% and Lithuania 77%, and in the USA with the largest production of nuclear energy of more than 96 000 MWh this accounts for about 18% of the total energy production.

Routine operations of nuclear reactors and other installations in the nuclear fuel cycle release small amounts of radioactive material to the air and as liquid effluents. However, the estimated total releases of ^{90}Sr, ^{131}I and ^{137}Cs over the entire periods of operation are negligible compared to the amounts released to the environment due to nuclear weapon tests.

Some of the first reactors that were constructed used a single-pass cooling system. The eight reactors constructed for plutonium production at Hanford, USA, between 1943 and 1956, pumped water from Columbia River through the reactor cores then delayed it in cooling ponds before returning it to the
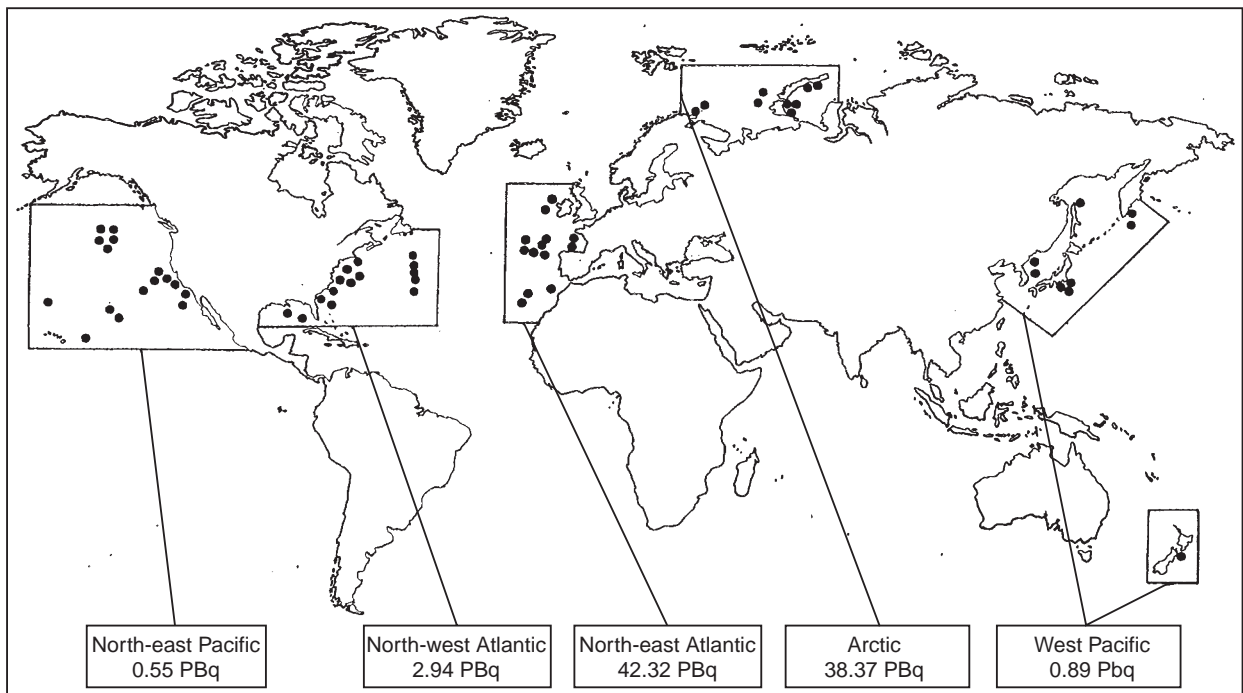

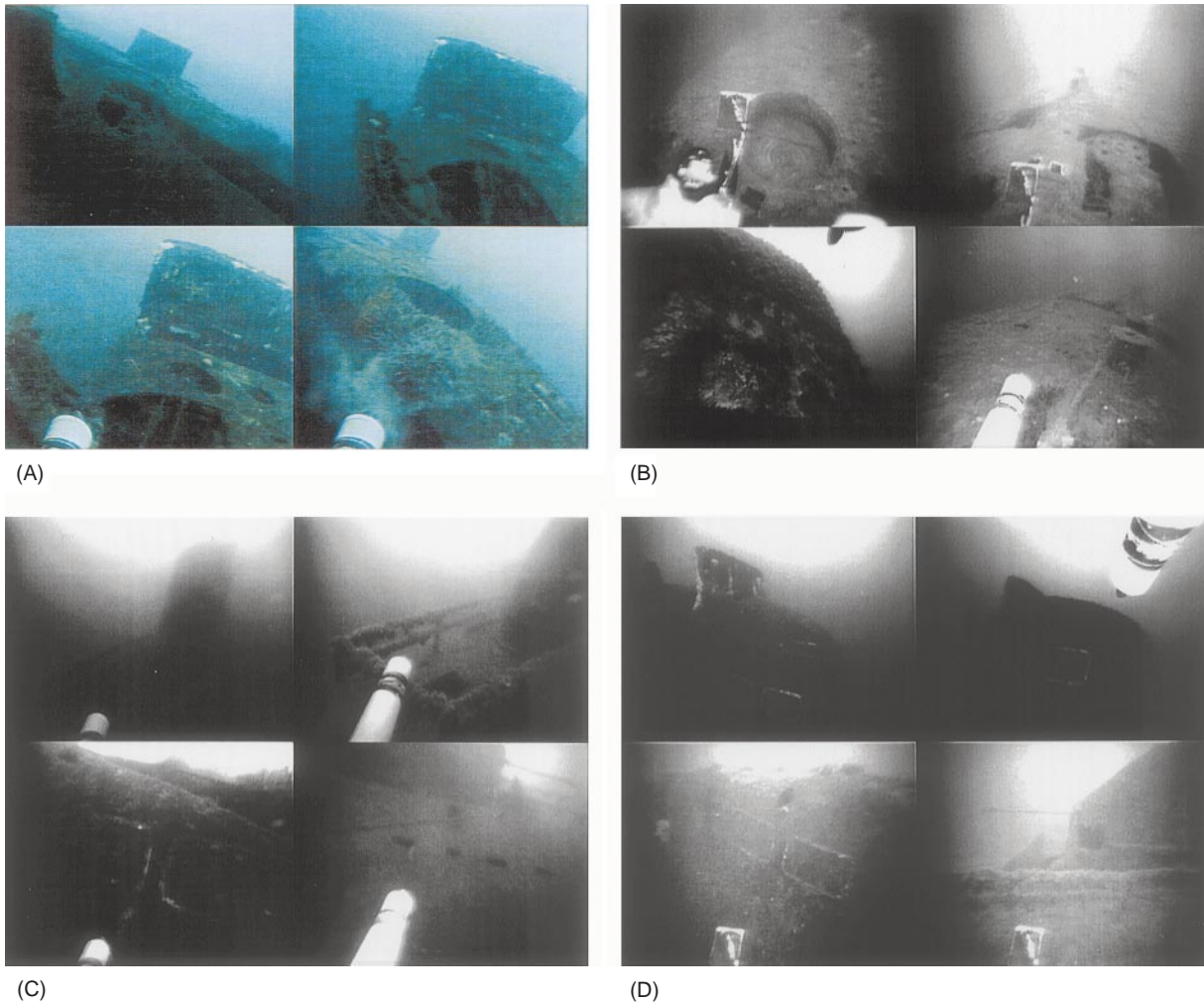**Figure 1** The worldwide location for disposal of radioactive waste at sea.

Figure 2 (A)–(D) Pictures of a disposed submarine at a depth of *c.* 30 m in the Stepovogo Fiord, east coast of Novaya Zemyla. The submarine contains a sodium-cooled reactor with spent fuel. Some of the hatches of the submarine are open which allows for 'free' circulation of water inside the vessel.

river. The river water and its contents of particles and components were thereby exposed to a great neutron flux and various radioactive isotopes were created. In addition corrosion of neutron-activated metal within the reactor structure contributed to the radioactive contamination of the cooling water. Only a limited number of these radionuclides reached the river mouth and only ^{32}P, ^{51}Cr, ^{54}Mn and ^{65}Zn were detected regularly in water, sediments and marine organisms in the near-shore coastal waters of the US Pacific Northwest.

Reactors operating today all have closed primary cooling systems that do not allow for this type of contamination. Therefore, under normal conditions production of electricity from nuclear reactors does not create significant amounts of operational discharges of radionuclides. However, the 434 energy-producing nuclear plants of the world in 1998

created radioactive waste in the form of utilized fuel. Utilized fuel is either stored or reprocessed.

Only 4–5% of the utilized nuclear fuel worldwide is reprocessed. Commercial, nonmilitary, reprocessing of nuclear fuel takes place in France, Japan, India and the United Kingdom. Other reprocessing plants defined as defense-related are in operation and producing waste but without discharges. For example in the USA, at the Savannah River Plant and the Hanford complex, about 83 000 m^3 and 190 000 m^3, respectively, of high-level liquid waste was in storage in 1985.

Reprocessing plants and the nuclear industry in the former Soviet Union have discharged to the Ob and Yenisey river systems ending up in the Arctic ocean. In 1950–51 about 77×10^6 m^3 liquid waste of 100 PBq was discharged to the River Techa. The Techa River is connected to the River Ob as is the
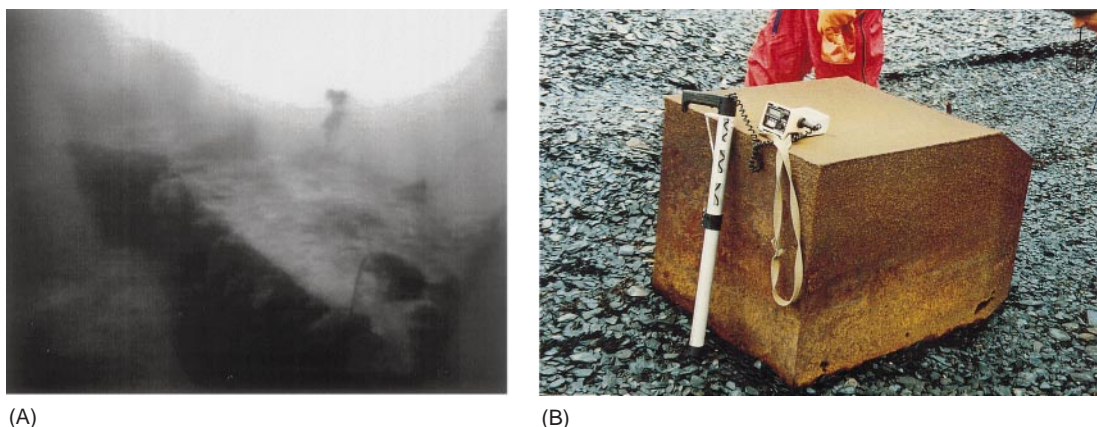
(A)   (B)

Figure 3 (A) Containers of low level solid waste at the bottom of the Abrosimov Fiord at a depth of *c*.15 m and (B) a similar container found washed ashore.

Tomsk River where the Tomsk-7, a major production site for nuclear weapons plutonium, is situated. Other nuclear plants, such as the Krasnoyarsk industrial complex, have discharged to the Yenisey river. Large amounts of radioactive waste are also stored at the sites.

Radioactive waste stored close to rivers has the potential of contaminating the oceans should an accident happen to the various storage facilities.

The commercial reprocessing plants in France at Cap de la Hague and in the UK at Sellafield have for many years, and still do, contributed to the radioactive contamination of the marine environment. They both discharge low-level liquid radioactive effluents to the sea. Most important, however, these discharges and their behavior in the marine environment have been and are still thoroughly studied and the results are published in the open literature. The importance of these discharges is extensive as radionuclides from Sellafield and la Hague are traced throughout the whole North Atlantic. Most important is Sellafield; **Table 2** summarizes the reported discharge of some important radionuclides.

In addition a range of other radionuclides have been discharged from Sellafield, but prior to 1978 the determination of radionuclides was, for many

Table 2 Total discharges of some radionuclides from Sellafield 1952–92

^{3}H	39 PBq
^{90}Sr	6.3 PBq
^{134}Cs	5.8 PBq
^{137}Cs	41.2 PBq
^{238}Pu	0.12 PBq
^{239}Pu	0.6 PBq
^{241}Pu	21.5 PBq
^{241}Am	0.5 PBq

components, not specific. Technetium (^{99}Tc) for instance was included in the 'total beta' determinations with an estimated annual discharge from 1952 to 1970 below 5 TBq and from 1970 to 1977 below 50 TBq. Specific determination of ^{99}Tc in the effluents became part of the routine in 1978 when about 180 TBq was discharged followed by about 50 TBq in 1979 and 1980 and then an almost negligible amount until 1994.

The reason for mentioning ^{99}Tc is that this radionuclide, in an oceanographic context, represents an almost ideal tracer in the oceans. Technetium is most likely to be present as pertechnetate, TcO_4^-, totally dissolved in seawater; it acts conservatively and moves as a part of the water masses. In addition the main discharges of technetium originate from point sources with good documentation of time for and amount of the release. The discharges from Sellafield are a good example of this. From 1994 the UK authorities have allowed for a yearly ^{99}Tc discharge of up to 200 TBq.

Based on surveys before and after the discharges in 1994, 30 TBq (March–April) and 32 TBq (September–October), the transit time for technetium from the Irish Sea to the North Sea was calculated to be considerably faster than previous estimations of transit times for released radionuclides. This faster transport is demonstrated by measurements indicating that the first discharge plume of ^{99}Tc had reached the south coast of Norway before November 1996 in about 2.5 years compared to the previously estimated transit time of 3–4 years.

Other reprocessing plants may have discharges to the sea, but without a particular impact in the world oceans. The reprocessing plant at Trombay, India, may, for example, be a source for marine contamination.

Accidental Releases

Accidents resulting in direct radioactive releases to the sea are not well known as most of them are connected to wreckage of submarines. Eight nuclear submarines with nuclear weapons have been reported lost at sea, two US and six former USSR. The last known USSR wreck was the submarine *Komsomolets* which sank in the Norwegian Sea southwest of Bear Island, on 7 April 1989. The activity content in the wreck is estimated by Russian authorities to be 1.55–2.8 PBq ^{90}Sr and 2.03–3 PBq ^{137}Cs and the two nuclear warheads on board may contain about 16 TBq ^{239,240}Pu equivalent to 6–7 kg plutonium. Other estimates indicate that each warhead may contain 10 kg of highly enriched uranium or 4–5 kg plutonium.

On August 12th 2000, the Russian nuclear submarine *Kursk* sank at a depth of 108 meters in the Barents Sea north of the Kola peninsula. Vigorous explosions in the submarine's torpedo-chambers caused the wreckage where 118 crew-members were entrapped and lost their lives. *Kursk*, and Oscar II attack submarine, was commissioned in 1995 and was powered by two pressurized water reactors. *Kursk* had no nuclear weapons on board. Measurements close to the wreck in the weeks after the wreckage showed no radioactive contamination indicating that the primary cooling-systems were not damaged in the accident. A rough inventory calculation estimates that the reactors at present contain about 56 000 TBq. Russian authorities are planning for a salvage operation where the submarine or part of the submarine will be lifted from the water and transported to land. Both a possible salvage operation or to leave the wreck where it is will create a demand for monitoring as the location of the wreck is within important fishing grounds.

The wreckage of *Komsomolets* in 1989 and the attempts to raise money for an internationally financed Russian led salvage operation became very public. The Russian explanation for the intensive attempts of financing the salvage was said to be the potential for radioactive pollution. The wreck of the submarine is, however, located at a depth of 1658 m and possible leaching of radionuclides from the wreck will, due to the hydrography of the area, hardly have any vertical migration and radioactive components will spread along the isopycnic surfaces gradually dispersing the released radioactivity in the deep water masses of the Nordic Seas. An explanation for the extensive work laid down for a salvage operation and for what became the final solution, coverage of the torpedo-part of the hull, may be that this submarine was said to be able to fire its torpedo missiles with nuclear warheads from a depth of 1000 m.

In 1990, the Institute of Marine Research, Bergen, Norway, started regular sampling of sediments and water close to the wreck of *Komsomolets*. Values of ^{137}Cs were in the range 1–10 Bq per kg dry weight sediment and 1–30 Bq per m^3 water. No trends were found in the contamination as the variation between samples taken at the same date were equal to the variation observed from year to year. Detectable amounts of ^{134}Cs in the sediment samples indicate that there is some leaching of radioactivity from the reactor.

Accidents with submarines and their possible impact on the marine environment are seldom noticed in the open literature and there is therefore little common knowledge available. An accident, however, that is well known is the crash of a US B-52 aircraft, carrying four nuclear bombs, on the ice off Thule air base on the northwest coast of Greenland in January 1968. Approximately 0.4 kg plutonium ended up on the sea floor at a depth of 100–300 m. The marine environment became contaminated by about 1 TBq ^{239,240}Pu which led to enhanced levels of plutonium in benthic animals, such as bivalves, sea-stars and shrimps after the accident. This contamination has decreased rapidly to the present level of one order of magnitude below the initial levels.

Fallout from Nuclear Weapon Tests and Nuclear Accidents

Nuclear weapon tests in the atmosphere from 1945 to 1980 have caused the greatest man-made release of radioactive material to the environment. The most intensive nuclear weapon tests took place before 1963 when a test-ban treaty signed by the UK, USA and USSR came into force. France and China did not sign the treaty and continued some atmospheric tests, but after 1980 no atmospheric tests have taken place.

It is estimated that 60% of the total fallout has initially entered the oceans, i.e. 370 PBq ^{90}Sr, 600 PBq ^{137}Cs and 12 PBq ^{239,240}Pu. Runoff from land will slightly increase this number. As the majority of the weapon tests took place in the northern hemisphere the deposition there was about three times as high as in the southern hemisphere.

Results from the GEOSECS expeditions, 1972–74, show a considerable discrepancy between the measured inventories in the ocean of 900 PBq ^{137}Cs, 600 PBq ^{90}Sr and 16 PBq ^{239,240}Pu and the estimated input from fallout. The measured values are far higher than would be expected from the assumed fallout data. Thus the exact input of anthropogenic radionuclides may be partly unknown or the geographical coverage of the measurements in

the oceans were for some areas not dense enough for accurate calculations.

Another known accident contributing to marine contamination was the burn-up of a US satellite (SNAP 9A) above the Mozambique channel in 1964 which released 0.63 PBq ^{238}Pu and 0.48 TBq ^{239}Pu; 73% was eventually deposited in the southern hemisphere.

The Chernobyl accident in 1986 in the former USSR is the latest major event creating fallout to the oceans. Two-thirds of the c.100 PBq ^{137}Cs released was deposited outside the Soviet Union. The total input to the world oceans of ^{137}Cs from Chernobyl is estimated to be from 15–20 Pbq, i.e. 4.5 PBq in the Baltic Sea; 3–5 PBq in the Mediterranean Sea, 1.2 PBq in the North Sea and about 5 PBq in the northeast Atlantic.

Natural Occurring Radioactive Material

Oil and gas production mobilize naturally occurring radioactive material (NORM) from the deep underground reservoir rock. The radionuclides are primarily ^{226}Ra, ^{228}Ra and ^{210}Pb and appear in sludge and scales and in the produced water. Scales and sludge containing NORM represent an increasing amount of waste. There are different national regulations for handling this type of waste. In Norway, for example, waste containing radioactivity above 10 Bq g^{-1} is stored on land in a place specially designed for this purpose. However, there are reasons to believe that a major part of radioactive contaminated scales and sludge from the worldwide offshore oil and gas production are discharged to the sea.

Reported NORM values in scales are in the ranges of 0.6–57.2 Bq g^{-1} ^{226}Ra + ^{228}Ra (Norway), 0.4–3700 Bq g^{-1} (USA) and 1–1000 Bq g^{-1} ^{226}Ra (UK).

Scales are an operational hindrance in oil and gas production. Frequent use of scale-inhibitors reduce the scaling process but radioactive components are released to the production water adding to its already elevated radioactivity. More than 90% of the radioactivity in produced water is due to ^{226}Ra and ^{228}Ra having a concentration 100–1000 times higher than normal for seawater.

The discharge of produced water is a continuous process and the amount of water discharged is considerable and increases with the age of the production wells. As an example, the estimated amount of produced water discharged to the North Sea in 1998 was 340 million m^3 and multiplying by an average value of 5 Bq^{-1} of ^{226}Ra in produced water, the total input of ^{226}Ra to the North Sea in 1998 was 1.7 TBq.

Discussion

The total input of anthropogenic radioactivity to the world's oceans is not known exactly, but a very rough estimate gives the following amounts: 85 PBq dumped, 100 PBq discharged from reprocessing and 1500 PBq from fallout. Some of the radionuclides have very long half-lives and will persist in the ocean, for example ^{99}Tc has a half-life of 2.1×10^5 years, $^{239, 240}$Pu, 2.4×10^4 years and ^{226}Ra, 1600 years. ^{137}Cs, ^{90}Sr and ^{228}Ra with half-lives of 30 years, 29 years and 5.75 years, respectively, will slowly decrease depending on the amount of new releases.

In an oceanographic context it is worth mentioning the differences in denomination between radioactivity and other elements in the ocean. The old denomination for radioactivity was named after Curie (Ci) and 1 g radium was defined to have a radioactivity of 1 Ci; 1 Ci = 3.7×10^{10} Bq and 1 PBq = 27 000 Ci. Therefore released radioactivity of 1 PBq can be compared to the radioactivity of 27 kg radium.

The common denominations for major and minor elements in seawater are given in weight per volume. For comparison if 1 PBq or 27 kg radium were diluted in 1 km^3 of seawater, this would give a radium concentration of 0.027 µg l^{-1} or 1000 Bq l^{-1}. Calculations like this clearly visualize the sensitivity of the analytical methods used for measuring radioactivity. In the Atlantic Ocean for example radium (^{228}Ra) has a concentration of 0.017–3.40 mBq l^{-1}, whereas ^{99}Tc measured in surface waters off the southwest coast of Norway is in the range of 0.9–6.5 mBq l^{-1}.

Measured in weight the total amount of radionuclides do not represent a huge amount compared to the presence of nonradioactive components in seawater. The radioisotopes of cesium and strontium are both important in a radioecological context since they have chemical behavior resembling potassium and calcium, respectively. Cesium follows potassium in and out of the soft tissue cells whereas strontium follows calcium into bone cells and stays. Since uptake and release in organisms is due to the chemical characteristics and rarely if the element is radioactive or not, radionuclides such as ^{137}Cs and ^{90}Sr have to compete with the nonradioactive isotopes of cesium and strontium.

Oceanic water has a cesium content of about 0.5 l µg^{-1} and a strontium content of about 8000 µg l^{-1}. Uptake in a marine organism is most likely to be in proportion to the abundance of the radioactive and the nonradioactive isotopes of the actual element. This can be illustrated by the

following example. The sunken nuclear submarine *Komsomolets* contained an estimated (lowest) amount of 1.55 PBq ^{90}Sr (about 300 g) and 2.03 PBq ^{137}Cs (about 630 g). If all this was released at once and diluted in the immediate surrounding 1 km^3 of water the radioactive concentration would have been 1550 Bq l^{-1} for ^{90}Sr and 2030 Bq l^{-1} for ^{137}Cs, the concentration in weight per volume would have been 0.000 3 µg l^{-1} ^{90}Sr and 0.000 63 µg l^{-1} ^{137}Cs. This means that even if the radioactive material was kept in the extremely small volume of 1 km^3, compared to the volume of the deep water of the Norwegian Sea available for a primary dilution, the proportion of radioactive to nonradioactive isotopes of strontium and cesium, available for uptake in marine organisms, would have been about one part per 2.7×10^6 and one part per 7.9×10^{-5}, respectively.

From the examples above it can be seen that if uptake, and thereby impact, in marine organisms follows regular chemical–physiological rules there is a 'competition' in seawater in favor of the non-radioactive isotopes for elements normally present in seawater. Measurable amount of radionuclides of cesium and strontium are detected in marine organisms but at levels far below the concentrations in freshwater fish. Average concentrations of ^{137}Cs in fish from the Barents Sea during the period with the most intensive nuclear weapon tests in that area, 1962–63, never exceeded 90 Bq kg^{-1} fresh weight, whereas fallout from Chernobyl resulted in concentrations in freshwater fish in some mountain lakes in Norway far exceeding 10 000 Bq kg^{-1}.

For radionuclides like technetium and plutonium, which will persist in the marine environment, uptake will be based only on the actual concentrations in seawater of radionuclide. The levels of ^{99}Tc, for example, increased in seaweed (*Fucus vesiculosus*) from 70 Bq per kg dry weight (December 1997) to 124 Bq kg^{-1} in January 1998 in northern Norway which reflected the increased concentration in the water as the peak of the technetium plume from Sellafield reached this area.

Previously the effects of anthropogenic radioactivity have been based on the possible dose effect to humans. Most of the modeling work has been concentrated on assessing the dose to critical population groups eating fish and other marine organisms. But even if the radiation from anthropogenic radionuclides to marine organisms is small compared to natural radiation from radionuclides like potassium, ^{40}K, the presence of additional radiation may give a chronic exposure with possible effects, at least on individual marine organisms.

The input of radioactivity, NORM, from the offshore oil and gas production may also give reason for concern. The input will increase as it is a continuous part of the production. Even if radium as the main radionuclide is not likely to be taken up by marine organisms the use of chemicals like scale inhibitors may change this making radium more available for marine organisms.

Conclusion

The sea began receiving radioactive waste from anthropogenic sources in 1946, in a rather unregulated way in the first decades. Both national and international regulations controlling disposals have now slowly come into force. Considerable amounts are still discharged regularly from nuclear industries and the practice of using the sea as a suitable wastebasket is likely to continue for ever. In 1994 an international total prohibition on radioactive waste disposal at sea came into force, but the approximately 85 PBq of solid radioactive waste that has already been dumped will sooner or later be gradually released to the water masses.

Compared to other wastes disposed of at sea the amount of radioactive waste by weight is rather diminutive. However, contrary to most of the 'ordinary' wastes in the sea, detectable amounts of anthropogenic radioactivity are found in all parts of the world oceans and will continue to contaminate the sea for many thousands of years to come. This means that anthropogenic radioactive material has become an extra chronic radiation burden for marine organisms. In addition, the release of natural occurring radionuclides from offshore oil and gas production will gradually increase the levels of radium, in particular, with a possible, at present unknown, effect.

However, marine food is not, and probably never will be, contaminated at a level that represents any danger to consumers. The ocean has always received debris from human activities and has a potential for receiving much more and thereby help to solve the waste disposal problems of humans. But as soon as a waste product is released and diluted in the sea it is almost impossible to retrieve. Therefore, in principal, no waste should be disposed of in the sea without clear documentation that it will never create any damage to the marine environment and its living resources. This means that with present knowledge no radioactive wastes should be allowed to be released into the sea.

See also

International Organizations. Nuclear Fuel Reprocessing and Related Discharges. Single Compound

Radiocarbon Measurements. Uranium–Thorium Decay Series in the Oceans Overview.

Further Reading

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RADIOCARBON

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Introduction

In 1934 F.N.D. Kurie at Yale University obtained the first evidence for existence of radiocarbon (carbon-14, ^{14}C). Over the next 20 years most of the details for measuring ^{14}C and for its application to dating were worked out by W.F. Libby and co-workers. Libby received the 1960 Nobel Prize in chemistry for this research.

The primary application of ^{14}C is to date objects or to determine various environmental process rates. The ^{14}C method is based on the assumption of a constant atmospheric formation rate. Once produced, atmospheric ^{14}C reacts to form ^{14}CO$_2$, which participates in the global carbon cycle processes of photosynthesis and respiration as well as the physical processes of dissolution, particulate deposition, evaporation, precipitation, transport, etc. Atmospheric radiocarbon is transferred to the ocean primarily by air–sea gas exchange of ^{14}CO$_2$. Once in the ocean, ^{14}CO$_2$ is subject to the same physical, chemical, and biological processes that affect CO$_2$. While alive, biota establish an equilibrium concentration of radiocarbon with their surroundings; that is, ^{14}C lost by decay is replaced by uptake from the environment. Once the tissue dies or is removed from an environment that contains ^{14}C, the decay is no longer compensated. The loss of ^{14}C by decay can then be used to determine the time of death or removal from the original ^{14}C source. After death or removal of the organism, it is generally assumed that no exchange occurs between the tissue and its surroundings; that is, the system is assumed to be closed. As a result of the ^{14}C decay rate, the various

reservoir sizes involved in the carbon cycle, and exchange rates between the reservoirs, the ocean contains approximately 50 times as much natural radiocarbon as does the atmosphere.

Carbon-14 is one of three naturally occurring carbon isotopes; ^{14}C is radioactive, has a half-life of 5730 years and decays by emitting a β-particle with an energy of about 156 keV. On the surface of the earth, the abundance of natural ^{14}C relative to the two stable naturally occurring carbon isotopes is ^{12}C$:^{13}$C$:^{14}$C $= 98.9\% : 1.1\% : 1.2 \times 10^{-10}\%$. Natural radiocarbon is produced in the atmosphere, primarily by the collision of cosmic ray produced neutrons with nitrogen according to the reaction [I].

$$_{0}^{1}\text{n} + _{7}^{14}\text{N} \Rightarrow _{6}^{14}\text{C} + _{1}^{1}\text{H} \qquad [\text{I}]$$
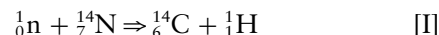
where n is a neutron and H is the proton emitted by the product nucleus. Similarly, the decay of ^{14}C takes place by emission of a β-particle and leads to stable nitrogen according to reaction [II],

$$_{6}^{14}\text{C} \Rightarrow _{7}^{14}\text{N} + \beta^{-} + \bar{v} + Q \qquad [\text{II}]$$
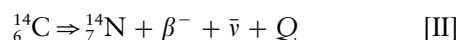
where $\bar{v}$ is an antineutrino and Q is the decay energy.

The atmospheric production rate varies somewhat and is influenced by changes in the solar wind and in the earth's geomagnetic field intensity. A mean of 1.57 atom cm^{-2} s^{-1} is estimated based on the long-term record preserved in tree rings and a carbon reservoir model. This long-term production rate yields a global natural ^{14}C inventory of approximately 50 t (1 t $= 10^6$ g). Production estimates based on the more recent record of neutron flux measurements tend to be higher, with values approaching 2 atom cm^{-2} s^{-1}. **Figure 1** shows the atmospheric history of ^{14}C from AD 1511 to AD 1954 measured by Minze Stuiver (University of Washington) using tree growth rings. The strong decrease that occurs