Radioactivity

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Acronyms and Abbreviations

ERICA	European project Environmental Risk from Ionising Contaminants: Assessment and Management
EW	Exempt waste
HLW	High level waste
IAEA	International Atomic Energy Association
ILW	Intermediate level waste
LLW	Low level waste
NOAA	National Oceanic and Atmospheric Association
NORM	Naturally Occurring Radioactive Materials
TENORM	Technologically Enhanced Naturally Occurring Radioactive Materials
UNSCEAR	United Nations Scientific Committee on the Effects of Atomic Radiation
USA	United States of America
VLLW	Very low level waste
VSLW	Very short lived waste

10.1 Introduction

Co-author Amanda shares an experience from her youth:

"*When I was in high school we had a soap box event in the court yard once a week. The soap box was essentially an up-side-down milk crate that you could stand on and talk about anything. This was mostly a student led activity but teachers would sometimes become involved. One of the common topics discussed week-in-week-out was the threat of nuclear war and a following nuclear winter. This threat felt very real to us and after organising a lunchtime viewing of the video The Day After, a fictional story about nuclear war and post war life, it felt even more real and more frightening. It seemed to our young minds at the time that war games being played by Mikhail Gorbachev, president of the Union of Soviet Socialist Republics (USSR), and Ronald Reagan, president of the United States of America (USA), threatened the existence of the world as we knew it. The idea of Cold War resulting in nuclear winter was both literally and figuratively chilling and confronting. Certainly, there were tensions, but the 1980s was also a time of considerable negotiation and over 1987-1988 the Intermediate-Range Nuclear Force Treaty was signed, approved and ratified.*"

In 2018 Donald Trump, president of the USA at the time, announced that the USA was withdrawing from the Intermediate-Range Nuclear Force Treaty, due to Russian non-compliance and amidst the continuing growth of China's missile forces. The USA formally withdrew from the Treaty on the 2nd of August 2019. Today, in 2023, we face the threat of nuclear force being used in the Russian invasion of Ukraine. With the dissolution of the Soviet Union, Ukraine became an independent nation and gave up its sizeable nuclear arsenal in return for security guarantees offered by the United States and Russia. Ukraine is now vulnerable if those guarantees are not kept. Are we at another crossroads?

The devastating impacts of nuclear warfare were realized with the detonation of two nuclear weapons over Hiroshima and Nagasaki, on August 6th and August 9th, 1945, respectively, in effect ending World War II. The first nuclear weapons test had only occurred in July 1945 in New Mexico, USA. Many tests that followed relied upon the remoteness of uninhabited islands and atolls such as Enewetak and Bikini Atolls in the Marshall Islands, Johnston Atoll near Hawaii, Kiritimati in Kiribati, the archipelago Novaya Zemlya in the Arctic Ocean, Montebello Islands off the northwest coast of Australia, Mururoa and Fangataufa Atolls in French Polynesia, and in the open Pacific and South Atlantic Oceans. Many other tests occurred in remote mountainous areas and underground. Today, nine sovereign states (political entities with one centralized government) are considered to have nuclear weapons capabilities.

Similar technologies are required to make both nuclear weapons and nuclear power. Nuclear fission reactions are slower in a power plant compared to a weapon; however, they both use plutonium-239 and uranium-235, both produce waste, and both have responsibilities for various accidents that have resulted in radioactive pollution in the marine environment. But if we are to gain a rational understanding of nuclear science we must also consider natural sources of radioactivity and other uses of nuclear chemistry, such as for scientific research and medical therapy and diagnosis, all of which result in radioactive waste. There is also the matter of managing waste generated from the nuclear industry. This chapter introduces nuclear chemistry and radioactive pollution in the marine environment from intentional and accidental human activities. It describes how radioactivity is measured, what it is, natural and anthropogenic sources, legacy waste and current waste management practices, and discusses the effects of radioactivity on marine biota.

10.2 Understanding Radioactivity and Units of Measurement

10.2.1 Radioactivity and Radioactive Decay

After the discovery of X-rays by Wilhelm Roentgen (1845–1923) a new field of science emerged. Henri Becquerel (1852–1908) became interested in substances that became luminous after exposure to sunlight. One of these substances was uranium ore and as a result Becquerel discovered another type of radiation. Following from this work, Marie Curie (1867–1934) and her husband Pierre Curie (1859–1906) discovered polonium and radium and named the radiation they produced **radioactivity**.

Radioactivity results from the degradation of unstable atoms to achieve a more stable form. All matter around us is made of atoms, each of which has a nucleus made up of protons and neutrons. Whereas the number of protons (atomic number) is what defines an element (e.g. all atoms of carbon have nuclei containing six protons), the number of neutrons within the atoms of a given element can vary. Some combinations of protons and neutrons result in a nucleus that is unstable, with excess energy stored within it. Different forms of a given element are called **isotopes**. Isotopes of any given element have the same number of protons, but different numbers of neutrons. Isotopes of an element that have a combination of neutrons and protons that is stable are called stable isotopes and do not decay. Isotopes that have an unstable combination of neutrons and protons are called radioactive isotopes, radionuclides, or **radioisotopes**. Over time, these radioisotopes spontaneously lose nuclear material and energy (protons, neutrons, and/or electrons) to achieve a more stable state. This emission of radiation is measured as **radioactivity**.

The loss of nuclear material associated with radiation emission is called **radioactive decay** and results in a new atom, which may be a different element (due to a change in the number of protons) or a different isotope of the same element (due to a change in atomic mass). Often this new atom will have a stable nucleus and no further decay will occur. However, depending on the initial radioisotope and the form of radioactive decay that occurs, the new atom may be another radioisotope. In this circumstance, the atom will undergo further radioactive decay until the nucleus reaches a stable state (i.e. the atom becomes a stable isotope).

10.2.2 Alpha, Beta and Gamma Decay

In the years following the discovery of radioactivity, researchers investigated the properties of radiation. Some of the early experiments were conducted by Ernest Rutherford (1871–1937). It was in 1898, while Rutherford was still a student, that he noted two forms of radioactive rays with different abilities to penetrate matter. He named these rays alpha (α) and beta (β) rays, after the first two letters of the Greek alphabet. By mid-1902, this naming scheme had been extended to include gamma (γ) rays, named after the third Greek letter. Alpha, beta and gamma radiation represent the three most common forms of radioactive decay and vary in their properties and characteristics ◘ Fig 10.1.

Alpha rays, or alpha particles, are the most easily absorbed, with the lowest power to penetrate matter. Atoms undergo alpha decay through the loss of two protons and two neutrons from the nucleus. Alpha particles are therefore helium nuclei without electrons and are positively charged. Because they are relatively large, heavy, and strongly charged, alpha particles have a strong tendency to interact and collide with the molecules in matter. This results in their low penetrative power. Alpha rays travel only a few centimetres in air and can be stopped by a sheet of paper.

Beta rays, or beta particles, have a moderate ability to penetrate matter and can be produced through either negative beta decay or positive beta decay. Both of these beta decay processes result in the charge of an atom increasing or decreasing by one unit whilst the atomic mass number remains unchanged. In negative beta decay (electron emission), a neutron within the nucleus of an unstable atom decays into a proton and electron. Whereas the proton from this decay remains in the nucleus, the electron is emitted at high speed and is a negatively charged beta particle. In positive beta decay (positron emission), a proton in the nucleus decays into a neutron, which remains in the nucleus, and a positron is emitted. Positrons have similar properties to electrons, except that they have a positive charge. Beta particles are therefore high-energy, charged, fast-moving, and relatively small, with essentially no mass. These properties allow beta rays to travel some metres through air and mean that beta rays are able to pass through paper but can be absorbed and stopped by human tissue, or around a 0.5 mm sheet of aluminium.

Gamma rays have the greatest penetrative power but, unlike alpha and beta rays, do not consist of particles. Rather, gamma rays consist of photons, which are packets of high-frequency electromagnetic radiation that move in waves. Gamma rays have no mass and can travel indefinitely through air. Thick sheets of lead or metres of concrete are required to stop gamma rays (◘ Figure 10.1).

10.2.3 Developing a Measurable Unit

Many of the units for measuring radiation and radioactivity (◘ Table 10.1) are named after the pioneering scientists of the field—Wilhelm Roentgen (1845–1923),

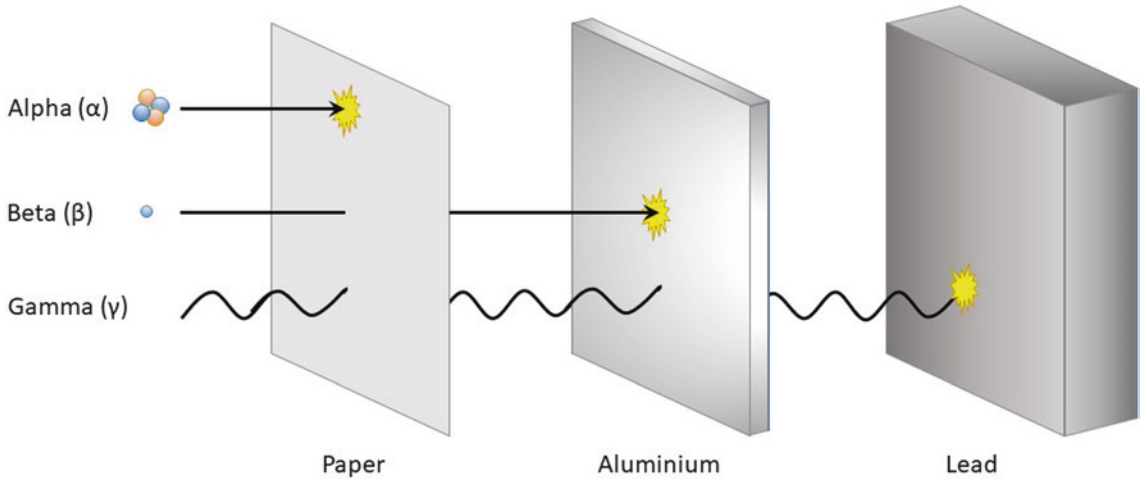
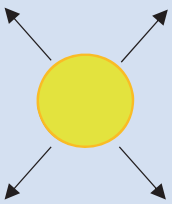
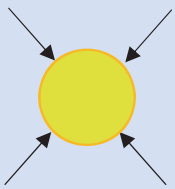


Figure 10.1 Penetrative power of alpha radiation (helium nuclei), beta radiation (high-energy electrons), and gamma rays (photons moving in waves). *Image*: J. Oakes

Table 10.1 Units of radiation and radioactivity

	Measure	Quantity	Unit
Radiation source 	Energy of ionising radiation	Radiation energy	Electron volts Joules Ergs
	Amount of radioactivity (number of particles or photons emitted per second)	Activity	Becquerel (Bq)[a] Rutherford (Rd) = 1.0×10^6 Bq Curie (Ci) = 3.7×10^{10} Bq
	Amount of radioactivity per unit mass of a radionuclide	Relative or specific radioactivity	e.g. Bq/mmol or Ci/mmol
Received radiation 	Ionisation in air	Exposure	Coulombs per kilogram (C/kg)[a] Roentgen (R)
	Absorbed energy per mass	Absorbed dose	Gray (Gy)[a] Radiation absorbed dose (rad)
	Absorbed dose weighted by type of radiation (measure of effective biological damage)	Equivalent dose	Sievert (Sv)[a] Roentgen equivalent man (rem)

[a] SI unit (unit specified by International System of Units)
Note: 1 Sv = 100 Roentgens = 100 rem, 1 Gy = 100 rad

Henri Becquerel (1852–1908), Marie Curie (1867–1934) and her husband Pierre Curie (1859–1906), and Ernest Rutherford (1871–1937).

The original unit for measuring the amount of radioactivity was the curie (Ci)–first defined to correspond to radioactive decay of one gram of radium-226 but more recently defined as:

$$1 \text{ curie} = 3.7 \times 10^{10} \text{ radioactive decays per second (exactly)}$$

The International System of Units (SI) has replaced the curie with the becquerel (Bq), where:

$$1 \text{ becquerel} = 1 \text{ radioactive decay per second} = 2.703 \times 10^{-11} \text{ Ci}$$

and is the number of nuclei that decay per unit time (Table 10.1). The specific radioactivity or relative radioactivity can be determined as the radioactivity per unit mass of a substance.

Ionising radiation (radiation with enough energy to ionise [or remove electrons from] other atoms) is measured using electron-volts, joules and ergs. The electron-volt (eV) is the energy gained by an electron when it moves from rest through a potential difference of

one volt (e.g. the energy an electron gains as it moves from a negative plate to a positive plate with a 1-V higher potential). Electron-volts are a useful unit for expressing very small amounts of energy. One joule (J) is equal to 6.242×10^{18} electron-volts and is equivalent to the amount of energy used by a one-watt light bulb lit for one second. The erg is a unit of energy equal to 6.242×10^{11} electron-volts or 1×10^{-7} J.

There are also other interrelated ways to consider radiation based on the objective of a study. For example, **exposure** describes the amount of radiation traveling through the air and is used in monitoring exposure. The units for exposure are the roentgen (R) and coulomb/kilogram (C/kg). Sometimes we might be interested in the **absorbed dose,** which is the amount of radiation absorbed by a living organism or an object. The units for absorbed dose are the radiation absorbed dose (rad) and gray (Gy). If we are interested in **effective dose** or dose equivalent, we consider both the amount of radiation absorbed and the effect of that radiation. Units for dose equivalent are the roentgen equivalent man (rem) and sievert (Sv). Biological dose equivalents are commonly measured in 1/1000th of a rem (known as a millirem or mrem). They are influenced by the penetrating power of alpha, beta and gamma radiation.

10.2.4 Half-Lives

In addition to quantifying and describing radioactivity and dose, as described in ▶ Section 10.2.3, it is useful to be able to express how slowly or rapidly radioactive material decays.

All unstable atoms will undergo radioactive decay at some point, but this decay is a random event; it is impossible to predict at what point in time any given atom will decay. However, for a very large number of atoms, the number of nuclei that will decay in a given period of time is predictable. The proportion of atoms decaying in a given period of time remains constant, (i.e. the number of atoms of a given radioisotope remaining within a sample reduces exponentially over time). This radioactive decay is expressed in terms of **half-life**.

The half-life of a radioisotope is defined as the time taken for half of the radioactive nuclei within a sample of that isotope to decay or the time taken for the activity (the number of decays per unit of time) to halve (▶ Box 10.1). Each radioisotope has a specific half-life, which may be anywhere from microseconds to hundreds of years, or even longer. In fact, the half-life of some radioisotopes is so long that they have remained in their current state since before the Earth was formed, and some isotopes have half-lives that are longer than the age of the universe. For example, bismuth-209 has recently been found to have a half-life of 1.9×10^{19} years, whereas the universe is estimated to have an age of only 1.38×10^{10} years.

Knowing the **decay rate** of radioisotopes has a number of practical applications. For example, decay rates can allow us to determine how long an environment, plant, or animal contaminated by radioactive waste will remain hazardous and can allow us to determine the age of various materials including archaeological artefacts, sediment, etc. (e.g. ^{14}C dating; ▶ Box 10.2).

Box 10.1: Understanding Half-lives

The half-life of a radioisotope is the amount of time that it takes for one-half of the original number of atoms to undergo radioactive decay to form a new element. For example, lead-210 decays to Bi-210 according to the nuclear equation below.

$$^{210}_{82}\text{Pb} \longrightarrow {}^{210}_{83}\text{Bi} + {}^{0}_{-1}\text{e}$$

The half-life of lead-210 is 22.2 years so the radioactivity halves every 22.2 years, as shown in ◘ Figure 10.2. You can calculate the remaining radioactivity for any given time period using a simple equation, demonstrated here:

You have 150 g of lead-210. How much lead-210 remains after 92 years?

$$\frac{92}{22.2} = 4.14 \text{ half lives}$$

Fraction remaining after 4.14 half-lives:

$$\frac{1}{2^n} = \frac{1}{2^{4.14}} = \frac{1}{18}$$

n = number of half-lives

The amount of lead-210 remaining $\left(\frac{1}{18}\right)(150)$ g = 8.33 g

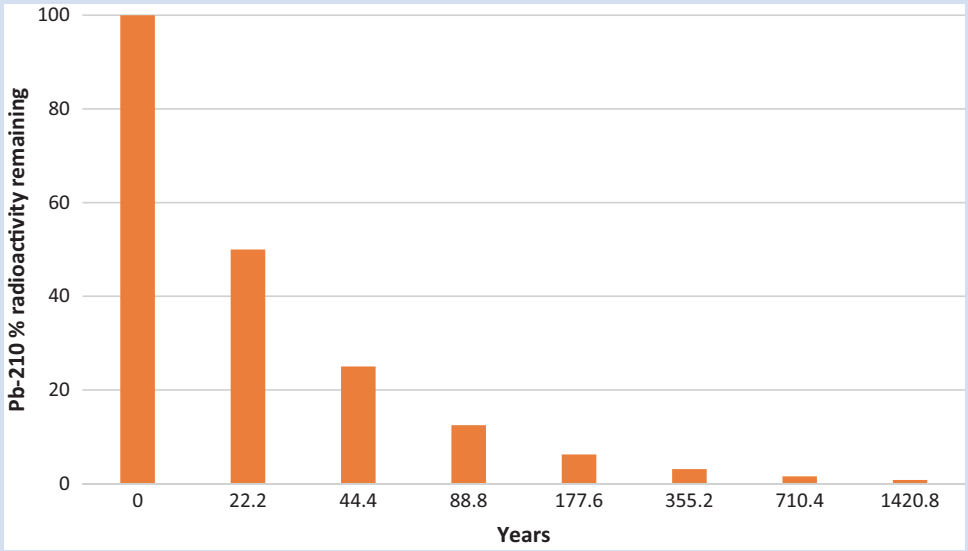


Figure 10.2 ▶ Box 10.1: The radioactive decay of Pb-210, which has a half-life of 22.2 years

10

10.3 Sources of Radioactivity

Radioactive substances occur naturally across the whole biosphere, and life has evolved in this radioactive environment. The natural background levels provide a reference for acceptable levels and are important to understand before we attempt to measure anthropogenic increases. Radioactive elements may be found in differing concentrations around the world as a result of natural and anthropogenic processes. To date, around 3000 natural and artificial radioisotopes have been identified.

10.3.1 Natural Radioactivity

Radioisotopes of naturally occurring elements comprise **Naturally Occurring Radioactive Materials (NORM)** and are ubiquitous in the environment, occurring in soil, sand, clay, rocks, air, water, and the tissues of plants and animals. These radioisotopes undergo radioactive decay that results in one or more types of radiation. Cosmic rays from the sun and outer space are referred to as ionising radiation and constantly bombard the Earth. Most naturally occurring radioactive substances (predominantly radium and radon) are the result of uranium and thorium decay. They may be mobilised, redistributed and concentrated by human activities such as fossil fuel mining and burning and fertiliser mining. When NORM are concentrated, or the potential for exposure has been enhanced, due to human activities they are termed **Technologically Enhanced Naturally Occurring Radioactive Materials (TENORM)** (Ojovan et al. 2019).

Naturally occurring radioisotopes have been used in the environmental sciences for over 150 years and enable the study of processes from a cellular level to broad oceanic scales (see ▶ Box 10.2). They can be applied both in field and laboratory studies (Table 10.2). The

Table 10.2 Selected naturally-occurring radioisotopes that are used in industry and science

Radioisotope	Half-life	Uses
Radon-222	3.82 days	Detecting and quantifying groundwater input to estuaries
Beryllium-7	53.22 days	Determining age of water and sediment
Lead-210	22.2 years	Dating layers of sand and soil laid down up to 80 years ago
Carbon-14	5700 years	Measuring the age of organic material up to 50,000 years old
Chlorine-36	301,000 years	Measuring sources of chloride and the age of water up to 2 million years old
Beryllium-10	1.39 mill years	Investigating soil formation and erosion rates, time of rock exposure (exposure dating), and dating of layers within ice cores
Uranium-235	704 mill years	Used in nuclear reactors, nuclear weapons, nuclear powered submarines

evolving field of **radioecology** has had a strong focus on marine research since the 1970s, and new applications are expanding the research scope (Cresswell et al. 2020). Studies using naturally occurring radioisotopes are useful for understanding the chronological formation of the Earth, sedimentology, contaminant behaviour, nutrient transport through food chains, global element cycles, defining natural and anthropogenic sources of nutrients, industry compliance, ecotoxicology, and remediation success, among others (e.g. Vandecasteele 2004; Call et al. 2015; Riekenberg et al. 2020; Cresswell et al. 2020). Indeed, the Journal of Environmental Radioactivity, established in 1984, is dedicated to this research field.

Box 10.2: Radioisotopes in Environmental Science: Nutrients Release 6000 Year Old Carbon from Coastal Sediment

The carbon-14 radioisotope is commonly used to date artefacts of biological origin from up to about 50,000 years ago. This technique, called radiocarbon dating or carbon-14 dating, can also be used to determine the age of organic matter in marine sediment. Radiocarbon dating uses the ratio of carbon-14 (^{14}C) to the common, stable form of carbon (^{12}C). In living plants and animals, which are constantly taking in new carbon, the ^{12}C:^{14}C ratio is relatively constant. However, once a plant or animal dies, and no new carbon is taken in, the amount of ^{14}C in its tissues begins to decline due to radioactive decay. Because the amount of ^{12}C remains unchanged, there is a shift in the ^{12}C:^{14}C ratio. Based on this shift, and the known half-life of ^{14}C (5700 years) it is possible to estimate how much time has elapsed since organic matter was part of a living thing.

Radiocarbon dating was used in a recent study looking at the impact on coastal systems of nutrients, which are increasing in coastal and marine systems globally due to human activities (Rockström et al. 2009). Riekenberg et al. (2020) observed that coastal sediments subjected to high concentrations of nutrients lost more carbon to the overlying water than unaffected sediments (◘ Figure 10.3). This is concerning, given that coastal sediments are increasingly recognised as important sites for storage of excess carbon. Release of stored carbon from coastal sediments could increase atmospheric carbon dioxide concentrations, contributing to climate change. However, the source of the extra carbon lost from the nutrient-impacted sediments was unknown; was it stored (old) carbon, or new carbon (e.g. produced in the sediment by microalgae)?

Radiocarbon dating showed that the carbon lost from nutrient-impacted sediment was around 6000 years old, confirming that increasing nutrient inputs to coastal systems may cause the loss of old, stored carbon from sediments. This use of radiocarbon dating highlighted the potential for coastal nutrient inputs to shift carbon budgets locally and, if nutrient inputs increase more broadly, possibly impact climate change by altering atmospheric carbon concentrations.

◘ **Figure 10.3** ▶ Box 10.2: Mud flat in the Richmond River, NSW, Australia, where radiocarbon dating showed that excess nutrients cause loss from the sediment of 6000 year old stored carbon (Riekenberg et al. 2020). *Photo*: J. Oakes

Mobilisation and Distribution from Agriculture

The agricultural industry is an important potential source of TENORM in the marine environment. Agricultural TENORM are associated with the production of phosphorus-containing fertilisers, which are applied to soil to enhance the growth and production of crops and pastures.

Phosphorus-containing fertilisers are derived from phosphate ore, which **naturally contains small amounts**

of radioisotopes, including uranium, radium, and thorium, and the radioisotopes produced through their decay. During the treatment of phosphate ore to produce fertiliser, some of these radioisotopes transfer to the fertiliser. Phosphorus-containing fertiliser added to fields can therefore increase the concentration of radioisotopes in soils (Pfister et al. 1976; Hameed et al. 2014), sometimes over many decades of application, although in some instances there is no or negligible increase in radioisotope concentration (e.g. Saueia et al. 2006). Radioisotopes in soil have the **potential to transfer to crops** that are consumed by humans, and the potential to enter adjacent waterways via erosion and/or groundwater and ultimately accumulate in the marine environment. Whether or not fertiliser application causes harmful levels of radioisotopes in the environment will depend on the radioactivity of the fertiliser used, its application rate, and biogeochemical characteristics of the soil and receiving environment.

A more significant source of TENORM associated with agriculture is phosphogypsum (hydrated calcium sulphate), which is a solid by-product of phosphorus fertiliser production. Around 100–280 megatonnes of phosphogypsum are produced globally per year (Yang et al. 2009; Parreira et al. 2003), with around 5 tonnes produced per 1 tonne of phosphorus fertiliser (Rutherford et al. 1994). Phosphogypsum has potential application in agriculture as a readily available source of gypsum, which adds calcium and sulfur to the soil, thereby enhancing root penetration (Nisti et al. 2015). However, during fertiliser production, up to 90% of the radioisotopes in phosphate ore, particularly radium (^{226}Ra), selectively transfer to phosphogypsum (Mazzilli et al. 2000). Due to this elevated radioactivity, the use of phosphogypsum in agriculture is restricted and phosphogypsum is typically treated as waste.

Where phosphogypsum is treated as waste, it may be directly discharged to the marine environment (El Kateb et al. 2018; Belahbib et al. 2021) and can cause substantial radioisotope contamination (e.g. Martínez-Aguirre and García-León 1994; Villa et al. 2009). However, this practise has become less common in recent times. Instead, vast quantities of phosphogypsum are stored in large stacks around the world, including in Europe, China, and the USA. These stacks are often near or in the coastal zone (Papaslioti et al. 2020) and leaching from the stacks has the potential to contaminate groundwater, transferring radionuclides to coastal and marine sediment and water (Tayibi et al. 2009). This is particularly the case for older stacks that were constructed and operational in the 1990s and earlier, before practises were improved to minimise environmental contamination. Even in these older stacks, however, leached radionuclides can be rapidly attenuated within the underlying sediment due to reactions with, and adsorption to, reactive coastal sediment (e.g. within 50 cm; Guerrero et al. 2020). Accordingly, for ^{226}Ra in phosphogypsum stacks there is often an initial pulse to the environment, with the remaining ^{226}Ra only slowly dissolved thereafter (Haridasan et al. 2002).

For radioisotopes that enter the marine environment, their distribution and impact, and whether they reach a level that is harmful to humans and ecological communities, is determined by their interaction with salinity and tidal movement (Martínez-Aguirre and García-León 1994), as well as redox conditions and the presence or absence of ion exchangers within the sediment.

10.3.2 Anthropogenic Radioactivity

Of more than 3000 known radioactive isotopes, only around 84 occur naturally. Most radioactive isotopes are artificially produced in reactors and accelerators for the purposes of research, energy generation, and/or medical treatments and diagnosis, or result from radioactive decay of these isotopes (Table 10.3). Anthropogenic emissions of radioactive isotopes add to the natural background levels of radioactivity. Much research has explored the risk to the marine environment from the production and distribution of anthropogenic radioactivity throughout the world (see review by Livingston and Povinec 2000), including in the Barents Sea and Arctic Ocean (e.g. Klungsøyr et al. 1995; Macdonald and Brewers 1996), the Western Sea on the Swedish west coast (Lindahl et al. 2003), the Pacific Ocean (e.g. Eigl et al. 2017; Buesseler et al. 2018), the North Atlantic Ocean (Villa-Alfageme et al. 2018), and the Flores Sea and Lombok Strait (Suseno and Wahono 2018). These studies vary not only in location but also in the source of the radioactive risk.

Nuclear Weapons

There are currently nine sovereign states considered to have nuclear capabilities: Russia, USA, France, China, United Kingdom, Israel, Pakistan, India and North Korea. Weapons testing is the predominant form of intentional nuclear emissions, including that arising from their use in war. There are around 13,000 nuclear weapons in the world, primarily in Russia (6255) and the USA (5550), with as few as 40–50 in North Korea (SIPRI 2021). Both the USA and Russia also have the highest stock piles of enriched uranium and separated plutonium (SIPRI 2021). The main nuclear weapon test sites that have resulted in marine contamination are in Novaya Zemlya, the Marshall Islands, Christmas Island, French Polynesia, and Lop Nop (Livingston and Povinec 2000).

Nuclear Energy

The International Atomic Energy Association (IAEA) is an international organization with 171 member states, founded in July 1957, that seeks to promote the

Table 10.3 Selected artificially produced radioisotopes that are used in industry and science

Radioisotope	Half-life	Uses
Technetium-99 m	6.01 h	Studying sewage and liquid waste movements. Also used in medical imaging. Produced in 'generators' from the decay of molybdenum-99, which is in turn produced in reactors
Gold-198	2.70 days	Tracing sand movement in river beds and on ocean floors, and studying coastal erosion. Also used to trace factory waste causing ocean pollution, and to study sewage and liquid waste movements
Chromium-51	27.7 days	Tracing sand to study coastal erosion
Ytterbium-169	32.03 days	Used in gamma radiography
Iridium-192	73.83 days	Used in gamma radiography. Also used to trace sand to study coastal erosion
Zinc-65	243.66 days	Predicting the behaviour of heavy metal components in effluents from mining waste water
Manganese-54	312.12 days	Predicting the behaviour of heavy metal components in effluents from mining waste water
Cobalt-60	5.27 years	Used in gamma radiography, gauging, commercial medical equipment sterilisation, and cancer treatment. Also used to irradiate fruit fly larvae in order to contain and eradicate outbreaks, as an alternative to the use of toxic pesticides. Used to irradiate some foods to extend shelf-life
Hydrogen-3 (tritium)	12.32 years	Used as a tracer in tritiated water to study sewage and liquid wastes, animal metabolism, and in biochemical research. Also used for luminous (glow in the dark) dials
Cesium-137	30.08 years	Radiotracer to identify sources of soil erosion and depositing, and also used for thickness gauging. Also a marker for sediment deposited in the mid-1960s (which had high Cs-137 levels due to nuclear bomb fallout) contributing to dating of sediment layers and quantification of subsequent rates of sedimentation
Americium-241	432.5 years	Used in neutron gauging and smoke detectors
Sodium-24	15 h	Detection of leaks in pipes
Sulphur-35	87.5 days	Determining sulphate reduction rates in coastal sediments
Fluorine-18	109.7 min	Used in medical imaging as a positron source for positron emission tomography (PET) scans
Calcium-47	4.5 days	Investigating bone metabolism
Californium-252	2.6 years	Used in cancer treatment, detection of gold and silver ore, portable metal detectors, detection of metal fatigue and stress
Iodine-131	8.04 days	Treatment of overactive thyroid and thyroid cancer. Also used in diagnostic imaging. Also used as an industrial tracer
Gadolinium-153	241.6 days	Used as a contrast agent in Magnetic Resonance Imaging (MRI)

peaceful use of nuclear energy, and to inhibit its use for any military purpose, including nuclear weapons. According to the IAEA (2021) Annual Report there were 437 operational nuclear power reactors in the world. The global use of nuclear energy is growing with 56 reactors currently under construction. The IAEA (2019) predicted that nuclear power capacity will increase by 12–25% by 2030 and up to 80% by 2050. Still, there is not a commonly agreed solution to the growing nuclear waste problem (Choudri and Baawain 2016). Furthermore, in the context of the marine environment **nuclear reactors require large volumes of water in their cooling towers** and ocean water is often used as a cheap and suitable source, avoiding the consumption of freshwater resources. A consequence of this is that many reactors are located on coastlines; there is even some discussion in the literature regarding floating nuclear power reactors (Srandring et al. 2009).

Nuclear energy provides a carbon free energy source and countries with the lowest carbon emissions are those that have a higher dependence on nuclear energy. But this type of energy does not come without its own risks. Unintentional release of radioactive materials can occur because of human and mechanical error (e.g. 1979—Three Mile Island, USA and 1986 -Chernobyl, Ukraine), and due to extreme natural events (e.g. 2011—damage caused by a tsunami generated from an earthquake at Fukushima, Japan ; see ▶ Box 10.3).

Figure 10.4 Anti-nuclear protests after the Fukushima nuclear accident in Japan (January 2012, Shibuya, Tokyo). *Photo*: A. Reichelt-Brushett

The production of nuclear energy is controversial and accidents create emotive responses from the public (Figure 10.4). Of course, there is much to be concerned about with the long-term global effects of nuclear accidents, global distribution of fall out and impacts on marine and terrestrial food chains. This concern has resulted in long-term research studies related to accident sites. However sometimes there is a misrepresentation of facts in the media, which leads to heightened public concern and enhanced public anxiety. One outstanding example of this misrepresentation is how the image in Figure 10.5 was promoted in the media and widely used to show the radiation leakage from the Fukushima accident. However, the National Oceanic and Atmospheric Association (NOAA) actually produced this map to show the maximum wave heights of the tsunami generated by the Japan earthquake on March 11, 2011. That being said, research studies have shown enhanced levels of radioactivity derived from the Fukushima accident in locally sourced seafood (Buesseler et al. 2012), and seafood from the North Pacific (e.g. Azouz and Dulai 2017), with some radioactivity transported via fish migration (Madigan et al. 2012). The levels detected have predominantly been below various limits of concern (e.g. Buesseler et al. 2012; Fisher et al. 2013; Azouz and Dulai 2017).

Box 10.3: Radioactive Pollution in the Marine Environment from the Fukushima Dai-ichi Nuclear Power Plant Accident

An earthquake generated tsunami wave seriously damaged the reactors at the Dai-ichi Nuclear Power Plant, Fukushima, Japan, on the 11th March 2011. Like other reactors, the Fukushima reactors have many radioactive elements, but three radioactive isotopes were of particular concern for marine ecosystems after the accident: iodine-131, cesium-137, and cesium-134. Iodine-131 has a half-life of 8 days, which means it is highly radioactive in the short term and was of concern immediately after the accident. Cesium-137 and -134 were released in the largest amounts. Levels 50 million times higher than before the accident were recorded in the ocean, posing a direct threat to marine life at the site. Levels dropped sharply after the first month but ongoing leaks have been indicated (e.g. Inoue 2018). Cesium-137 has a

relatively long half-life (30.08 years), but is present in the ocean due to nuclear weapons testing in the 1950s and 1960s. Cesium-134 is much shorter-lived (2.06 years) and therefore, if present in seawater samples, it most likely comes from Fukushima. Tritium-2 (12.3 year half-life) was also measured throughout the western North Pacific at very low concentrations (Kaizer et al. 2018). Although of relatively low concern in regards to health impacts, it is found in stored water at the site even after decontamination processes—management considerations continue.

Most Japanese fisheries were unaffected by the accident, but coastal fisheries nearest the reactors were closed because of concern that some species, particularly those that are benthic and sessile, would be exposed. Biota testing to date still occurs on a regular basis and is compared against Japan's limits for radiation in seafood (which are more stringent than USA regulations). If seafood exceeds these regulations it cannot be sold. Fortunately, the contamination is very localised but, in light of the high consumption of seafood in the Japanese diet, there has been much concern raised within Japan about seafood safety as a result of the accident. A questionnaire, exploring factors affecting consumer behaviour towards seafood from regions near the accident with uncertain risks, highlighted that the consumer class perceiving the highest risk and greatest negativity towards this seafood were parents of young children and of higher academic achievement. Interestingly, environmental awareness and higher age range categories showed a more positive response to seafood from this location indicating that the desire to support the economic recovery of the seafood industry outweighed the risk concerns (Aruga and Wakamatsu 2018). No studies have been published that show consumption of seafood from the impacted area causes serious human health risks.

10.3.3 Radioactive Waste Management

The first sea dumping of radioactive waste took place in 1946 at a site in the North East Pacific Ocean, about 80 km off the coast of California (Calmet 1989). Dumping of low level waste (LLW) continued for 36 years and occurred as late as 1982, at a site about 550 km off the European continental shelf in the Atlantic Ocean (Calmet 1989). An estimated 63 PBq (1.7 MCi) of radioactive waste coming from research, med-

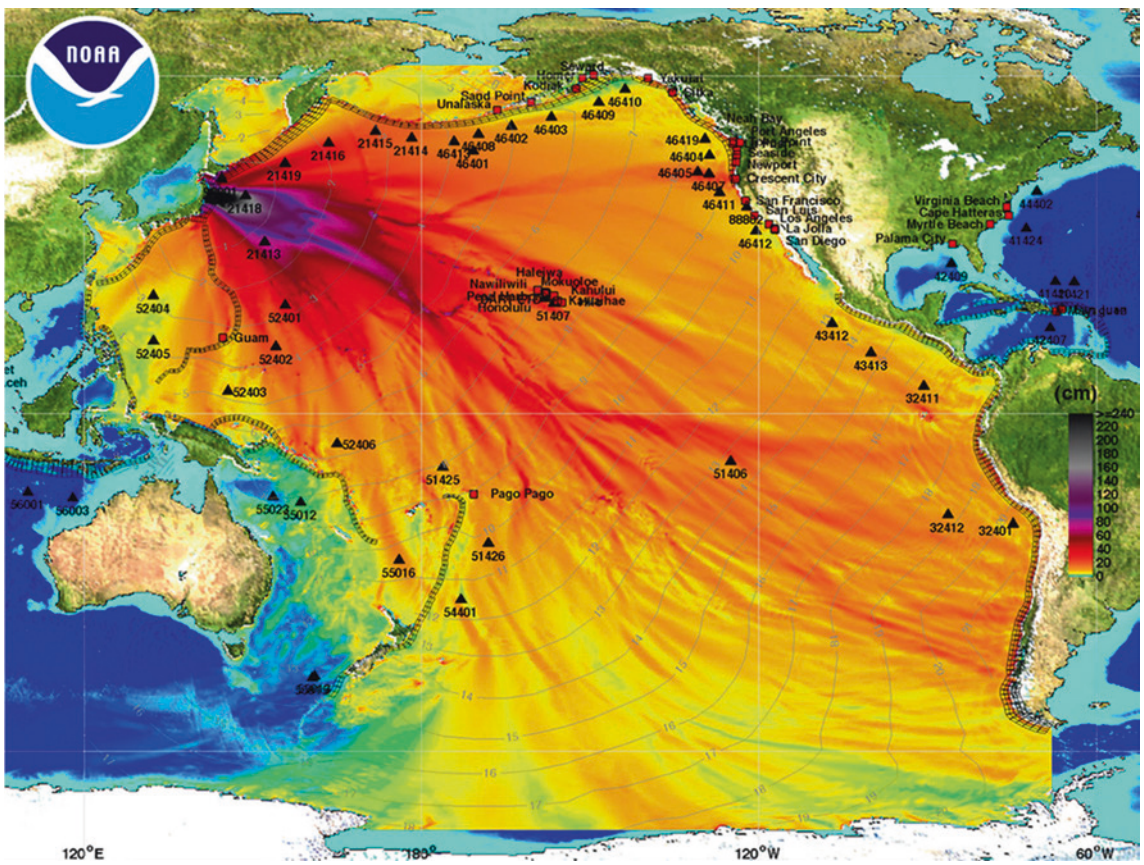


Figure 10.5 This is not radioactive leakage from the Fukushima nuclear accident spreading across the Pacific Ocean. *Image:* created by NOAA's Center for Tsunami Research and graphically shows maximum wave heights of the tsunami generated by the Japan earthquake on March 11, 2011

Table 10.4 Classification of radioactive waste according to the IAEA (2009)

Classification code	General criteria
Exempt waste (EW)	Classification explained in IAEA (2004)
Very short lived waste (VSLW)	Contains only radionuclides with very short half-lives, can be stored until the activity has fallen below the levels for clearance[a]
Very low level waste (VLLW)	Waste arising from decommissioning of nuclear facilities with levels only slightly above specified levels, other waste containing naturally occurring radionuclides[a]
Low level waste (LLW)	Radioactive waste that does not need shielding during normal handling, suitable for near surface disposal[a]
Intermediate level waste (ILW)	Contains long lived radionuclides in quantities that need a greater degree of containment and isolation from the biosphere than provided in near surface disposal[a]
High level waste (HLW)	Contains high concentrations of both short and long lived radionuclides where long term safety needs to be ensured. These are heat generating wastes arising from spent fuel from nuclear reactors. Requires deep geologic disposal[a]

[a] Further details in IAEA (2009)

icine, and other nuclear industry activities were packaged, usually in metal drums lined with a concrete and bitumen matrix, and disposed of at sea (Calmet 1989). Over 50 dump sites are recorded across the northern Atlantic and Pacific Oceans (Calmet 1989).

All human uses of radioactive material generate waste, and as humans are inclined to do, we collect our waste and attempt to manage it. Pritchard (1960) highlighted the production of radioactive waste as an unavoidable consequence of utilising atomic energy. He recognised the responsibilities to assure that the atomic energy industry did not endanger humans or the resources of the sea. After considering the analysis he suggested permissible concentrations and disposal conditions for radioisotope disposal to the sea. Similarly, Vilks (1976) provided insight into a workshop held at Woods Hole in 1976 to discuss the disposal of high level waste (HLW) in oceans, showing that concern was there but serious consideration was not yet being given to ocean disposal. Due to the concerns raised, novel approaches were proposed to manage some forms of radioactive waste. Krutenat (1978) proposed that plutonium-239 waste, with a half-life of 25,000 years, should:

» "*Be disposed of in the basement rock of an oceanic plate at the edge of its subduction zone [to] allow the crustal movement to carry the waste to the centre of the earth*".

The IAEA was no doubt considering the results and recommendations of these and many similar studies at the time. Yet, even with this engaging and long-lasting discussion, there has been considerable disposal of nuclear wastes to the oceans over the years which remains as legacy waste in ageing storage containments. **Today** radioactive waste **has been classified by the IAEA (2009)** (Table 10.4; Figure 10.6). This classification scheme considers the type of waste based on half-life as well as its state (solid, aqueous, organic, liquid) and provides detailed direction for appropriate disposal.

Site specific **legacy nuclear waste and radioactivity** from both intentional activities (e.g. waste dumping, weapons testings) and accidents (e.g. power plants, nuclear submarines) will need to be managed long into the future. Even though serious consideration was given to disposing of nuclear waste in the sea in the 1960s and 1970s (e.g. Pritchard 1960; Vilks 1976) it is now not considered an option. There are, however, current sources of anthropogenically derived radioactivity that enter the marine environment, including global fall out, and low level release from nuclear power plants and nuclear fuel reprocessing plants as part of normal operating procedures (Livingston and Povinec 2000). Potential for accidents exists wherever radioactive material is used or when it is transported. Most LLW can be managed safely on land in most places and there are now dedicated facilities for reprocessing and/or storage of intermediate and high level nuclear waste. Interestingly, there is a trade in nuclear waste management and this results in radioactive material being transported from the site of production to the site of disposal, usually by shipping transport.

10.4 Effects on Marine Biota

The enrichment of radioactive material in the marine environment causes risks to marine organisms and to human populations that consume these organisms. Radiation causes changes in living cells as it interferes with normal chemical processes within and between cells. Water within cells can be transformed to hydrogen peroxide. This is particularly the case for white blood cells and impacts an organism's ability to fight infec-

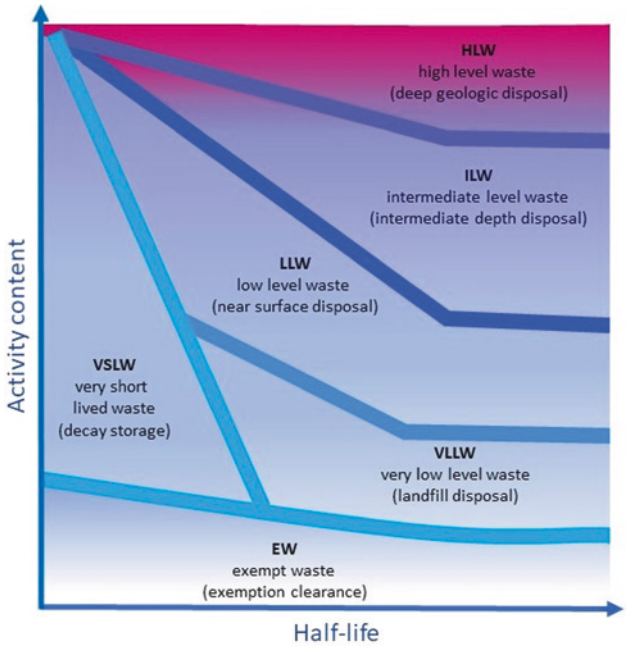


Figure 10.6 Conceptual diagram of the radioactive waste classification scheme of the International Atomic Energy Agency. Adapted from IAEA 2009 by A. Reichelt-Brushett

tion. Radiation has also been shown to induce cancer-like diseases like leukaemia in blood forming organs. It may also cause mutations that impact on heredity. Interestingly, the global background radiation may have influenced the current genetic evolution of species.

The fate of radionuclides depends largely on ocean geochemistry, physical processes and biological uptake, and these characterise exposure in marine environments. Radioactive elements bioaccumulate in a similar manner to other pollutants that contain similar chemical characteristics. The accumulation of radioactive elements is dependent upon chemical behaviour, physical chemistry, and how organisms interact with their environment. As with many metals and organic compounds some contaminant will disperse, but much will bind to particulates, accumulate in benthic environments, and have the potential for remobilisation (Batlle et al. 2011; Buesseler et al. 2017).

The study of in situ exposure is challenging (Buesseler et al. 2017), and exposure doses are highly variable given dispersal by currents and dilution (Batlle et al. 2014). There are increasing numbers of studies that document concentrations of radionuclides in marine species but few investigate effects; rather they rely on recommended values. Recent studies by Men et al. (2020a) showed that ^{134}Cs, ^{137}Cs and ^{110m}Ag accumulated in dolphin fishes after the Fukushima Daiichi Nuclear Power Plant Accident, but decreased with time. It was concluded from these radiation dose assessments that the released radiation would not have caused harm to dolphin fishes in the open ocean of the Northwest Pacific. Similar results were found for neon flying squid (Men et al. 2020b).

The European project Environmental Risk from Ionising Contaminants: Assessment and Management (ERICA) proposed a benchmark at the ecosystem level of 10 μGy/h. Another benchmark from the United Nations Scientific Committee on the Effects of Atomic Radiation (UNSCEAR 1996) concluded that dose rates of up to 400 μGy/h to a small proportion of individuals in aquatic populations would not result in adverse effects at the population level.

There is limited evidence of radioactivity having caused impacts to marine biota (e.g. Batlle 2011; Hosseini et al. 2012). The biological impacts of radiation on chronically exposed organisms are limited to a few laboratory studies, and it is suggested that there is a broad range of species' sensitivities (Batlle et al. 2014) (see ▶ Box 10.4). As with all environmental pollution studies, there is a multitude of endpoints that could be considered to indicate biological impacts of radiation. The selection of an endpoint is usually considered in respect to the likely or possible response to the contaminant of interest. In the case of radionuclides, cellular dysfunction and abnormalities are probable choices. There has been limited targeted analysis to determine genotoxic effects in studies on the impacts of radiation on marine biota. Jha et al. (2005) completed laboratory assays to assess the genotoxic effects of tritium

on the adult life stage of the mussel *Mytilus edulis*; they found a dose dependent response in micronuclei, and DNA single strand breaks (Comet assay), highlighting genetic damage. This study further suggested that the generic dose limits recommended by the IAEA for the protection of aquatic biota may not be applicable to all aquatic organisms.

Batlle et al. (2018) have made recommendations for the field of marine radioecology through the development of models to better predict radionuclide transfer to biota in non-equilibrium situations. Such models would be enhanced by an increased understanding of biogeochemical processes and their influence on radionuclide dispersion along with antagonistic and synergistic interactions related to uptake. Furthermore, Batlle et al. (2018) recommended a more integrated approach to marine radioecology that includes oceanography, radiochemistry, ecology, ecotoxicology and climate science to bring more ecological thinking into the discipline, with further focus on food chains and ecosystem processes.

Box 10.4: Bikini Atoll Five Decades On

A vast number of nuclear weapons tests occurred in waters and coral reef areas of the Marshall Islands and specifically Bikini Atoll, which was physically decimated by 23 surface and subsurface thermonuclear experiments (Figure 10.7). Five decades later, Bikini Atoll was shown to be flourishing with coral reefs and plentiful fish (Richards et al. 2008). Richards et al. (2008) determined that overall species richness before nuclear testing and 50 years after testing was approximately the same, but the species mix was different, suggesting that 28 species were genuine losses, predominantly from the lagoon habitat. The presumed initial losses were mainly attributed to physical impacts, shock waves, temperature rises, and sediment and nutrient suspension.

Figure 10.7 ▶ Box 10.4: Operation crossroads, test Baker as seen from Bikini Atoll, July 25, 1946. *Photo*: x-ray delta one licensed under CC BY-SA 2.0

10.5 Summary

Radioactivity results from the degradation of unstable atoms to achieve a more stable form. The units it is measured in are unique, and understanding these provides for an enhanced understanding of the topic of radioactivity.

There are both natural and artificial sources of radioactivity. Human uses of both are wide ranging; it is a relatively common source of power, it is used as a weapon, and also in life saving medical science and other scientific investigations. It behaves in the environment in a similar way to some other contaminants that bioaccumulate. The fate and behaviour of radioiso-

topes is influenced by biogeochemical and physical processes, and the degradation of radioisotopes is dependent on their half-life.

The world's oceans have been exposed to anthropogenic radioactivity as a result of nuclear accidents, weapons testing and waste disposal, and they are considered slightly contaminated by anthropogenic radionuclides. Currently, global fallout and authorised release of low-level waste from nuclear reprocessing facilities and power plants are the main sources of radionuclides to the ocean. There are now global guidelines and restrictions for the management of radioactive waste and recommended safe exposure levels for humans and ecosystems. However, there are few studies that have investigated concentration and effect relationships of radiation on marine biota. A more integrated approach to marine radioecology would help by bringing more ecological thinking into the discipline.

10.6 Study Questions And Activities

1. Describe alpha, beta, and gamma radiation.
2. Using ▶ Box 10.1 determine how much lead-210 would remain after 135 years.
3. Does your home country need to manage nuclear waste from energy generation? If so, see if you can investigate how that waste is managed.
4. Find a journal article that explores the impact of radioactivity on a marine species. Report how the effect is being measured.
5. Explore the IAEA website and record two new facts that you learn.

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