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General Information on Marine Radioecology and on Risk Assessment

1.1. General information on radioecology

Radioecology has three objectives: the first is to detect the presence of radionuclides in the environment and to investigate their origins; the second is to understand their transfer and concentration processes in ecosystems and the third is to evaluate the impact of natural and anthropogenic radioactivity on the environment (radioecological impact) and on the exposure of the human population (dosimetric impact). These studies follow the same approach as ecotoxicology, which is concerned with stable chemical pollutants such as metals (cadmium, mercury, etc.), nitrates or persistent organic pollutants, whereas radioecology is concerned only with unstable isotopes of elements or radionuclides. Given the presence of radionuclides in all ecosystems and the complexity of transfers, radioecologists work on the three main environmental media: the atmosphere, the hydrosphere (marine and aquatic environments) and the lithosphere (terrestrial environment). In this book, only the marine environment will be discussed.

The term "radioecology" appeared in 1935 but did not become widespread until the 1950s. It results from the fusion of the words "radioactivity" and "ecology". The first work on radioecology was made public in 1955 in Geneva, during the first international meeting on the peaceful use of nuclear energy. The first international congress on radioecology was held in the United States (1961) and was edited by Schultz and Klement [SCH 63]. Many other radioecology congresses followed, such as the one organized by the IAEA in Vienna in 1966, the first one organized in France in 1969 (Cadarache) and the one coordinated by the National Research Council of the National Academy of Science in 1967–1970 [NAS 71].

Radioecological studies are carried out using two main approaches, field (in situ) and experimental approaches [CAQ 01]. The former are the most realistic, but they do not allow for the determination of transfer mechanisms. The objective of the experiments is to build radionuclide transfer models [THI 99] that can be used to quickly predict the impact of radioactive contamination on a territory, based on a limited number of measurements of the radioactivity of sediment samples taken in the field.

In the field, radioecologists choose physical environments that fix radionuclides, such as sediments, or species called bioaccumulators that concentrate radionuclides, such as mosses, lichens, algae or mollusks (mussels or oysters). In the laboratory, radioecologists reconstitute simplified ecosystems such as, for example, the contamination of crops by various radionuclides by varying the nature of the soil and climatic conditions. This work leads to the development of models that simulate the transfer of radionuclides in the various environmental media.

1.1.1. *History of radioecology*

Initially, radioecological studies were oriented to provide data from a health point of view. The corollary (introduced in particular by the ICRP in 1977, ICPR26) that prevailed was that Man was the most sensitive species to the effects of ionizing radiation and that by protecting Man, one protected all living organisms. Therefore, the first "environmental" studies were aimed at monitoring food sources (of wild or anthropogenic origin). In the environment, the measures concerned mainly edible species and very often only the consumed parts of wild species such as fish muscles. Since environmental contamination in the 1950s and 1960s was mainly due to atmospheric fallout from atomic weapons testing, studies were devoted mainly to the two relatively long-lived radionuclides resulting from these explosions, namely cesium-137 and strontium-90, and to a lesser extent to the associated transuranics (notably ^{239,240}Pu).

In the 1970s, with the decrease in atmospheric fallout, interest focused on fission products that were released into the environment around spent fuel reprocessing plants, such as cobalt-60, ruthenium-106, antimony-125, silver-110m and manganese-54. Following the Chernobyl accident (1986), radioecological studies were essentially focused on cesium-137 and secondarily on some transuranics. It was also at this time that the number of studies on the effects of radiation on wildlife increased. In the 2000s, the European Union launched two important programs for radioecology. The first one being "Environmental Protection for Ionizing Contaminants in the Arctic" where the sentinel species are lichens, soil

invertebrates, as well as mammals (lemmings, reindeer) and birds (marine species are in the minority). The second program is called FASSET (Framework for ASSessment of Environmental ImpacT), and it is responsible for assessing the effects of radiation in particular on mortality, morbidity, its impact on reproduction and cellular damage in various species [STO 02]. After the war in Kosovo, several studies were interested in the effects of depleted uranium used in military equipment (in particular "shaped charge" rockets).

The European project ERICA (Environmental Risk for Ionising Contaminants: Assessment and Management) of the 6th PCRD-Euratom launched in March 2004, was completed in February 2007. It allowed for the updating of the database concerning the effects of ionizing radiation on non-human organisms, the exploitation of this database to define criteria for the protection of ecosystems and the design of a method that enables the characterization of ecological risk by analyzing the exposure of fauna and flora to ionizing radiation and the effects of this exposure.

1.1.2. *The main marine radioecology laboratories*

For about 40 years, the vast majority of laboratories working in the field of marine radioecology were state laboratories. For example, in the United States, these were USAEC laboratories, and in the Soviet Union, laboratories dependent on the Academy of Sciences such as MMBI (Murmansk Marine Biological Institute of the Russian Academy of Sciences) in Murmansk or the Sevastopol Biological Station in Sevastopol in Crimea (USSR). In the United Kingdom, the main laboratory at Lowestoft (CEFAS) is dependent on the Ministry of Agriculture, Fisheries and Food (MAFF) [HUN 98]. In France, the marine radioecology laboratory was under the authority of the *Commissariat à l'énergie atomique* (CEA, Central Commission for Nuclear Energy), which itself was originally directly attached to the French Prime Minister. This laboratory was located on the site of the spent fuel reprocessing plant at La Hague. As a result, public information was highly controlled. With its independence, it moved to Cherbourg-Octeville.

In addition, in the United States, various academic institutions have worked in the field of marine radioecology such as the School of Marine Sciences at Stony Brook University, the School of Oceanography at Oregon State or the Department of Fisheries at the University of Washington (Seattle, WA).

At the international level, the IAEA created a laboratory called MEL (Marine Environment Laboratory) located in Monaco [FOW 88]. This laboratory has provided valuable data on the measurement of contaminants in the sea.

After the Chernobyl accident and especially after the Fukushima accident, a large number of academic teams became interested in marine radioecology topics, renewing and diversifying the studied themes.

1.1.3. *The triad of radioecology: exposure, bioaccumulation and adverse effect*

As with most pollutants, the impact of radionuclides on living beings depends on an interaction at three levels, exposure, bioaccumulation and adverse effect.

1.1.3.1. *Transfers from physical environments to organisms*

Radionuclides in the marine environment have multiple origins, whether they are natural or anthropogenic radionuclides (Chapter 2). Their dispersal in all marine environments leads to an exposure of living organisms to radioactive elements. However, the exposure will not systematically induce an accumulation of environmental contaminants within organisms because different barriers will limit their intake[1] (Figure 1.1). Even though the circulation of contaminants is a worldwide phenomenon, the more distant a site is from the source of contamination, the greater the dilution in the water. This contamination gradient will be the first factor to control the intake. Under the influence of environmental conditions, many radionuclides, and the molecules that accompany them in physical media, are likely to be transformed: chelation[2], hydrolysis, photodegradation, biodegradation, etc. Though the majority of radionuclides are likely to cross biological membranes, these can however play a barrier role, for example, by altering the speciation and the bioavailability of a radioactive metal by the secretion of chemical modifiers in the external medium or by precipitating it on the cell surface in the form of metal sulfide microcrystals. Many organisms (bacteria, animals, plants) secrete mucopolysaccharides of highly variable composition, and subtle changes in charges and reactive groups are likely to interfere with contaminant intake. Another way to limit intake is the existence of impervious extracellular barriers such as carapaces, integuments, tests, shells and scales that help to reduce the surface area of exposed living epithelium available for cutaneous transport [MAS 95a].

Once the radionuclide is incorporated (Figure 1.1), it will either be stored within the tissues or excreted. Storage in intra- or extracellular compartments will not automatically result in an adverse effect on the body. Thus, with respect to transition metals and their radioactive isotopes, detoxification processes are present in many organisms. This can be the synthesis of particular metalloproteins, for example,

1 The term “intake” means incorporation of the radionuclide into the body.

2 Chelation is a physico-chemical process during which a complex is formed between a ligand and a radionuclide.

metallothioneins in animals and phytochelatins/metallothioneins in plants, which sequester the metals with their thiol groups, preventing them from interfering with the enzymes whose activity they would otherwise disrupt. This is the most common defense process used in vertebrates. It also exists in most taxa [AMI 06], but in invertebrates, the preferred pathway of detoxification is the biomineralization of metals in various inclusions [MAR 02]. However, detoxification by storage is not effective for radionuclides as it is for their stable counterpart for the ionizing radiation remains within the tissues.

Unlike most other pollutants, the rule "no effect without bioaccumulation and no bioaccumulation without exposure" does not apply to radionuclides. Because of the production of various types of ionizing radiation, an organism externally exposed to a radionuclide will experience external irradiation that may cause adverse effects. Bioaccumulation leads to internal irradiation.

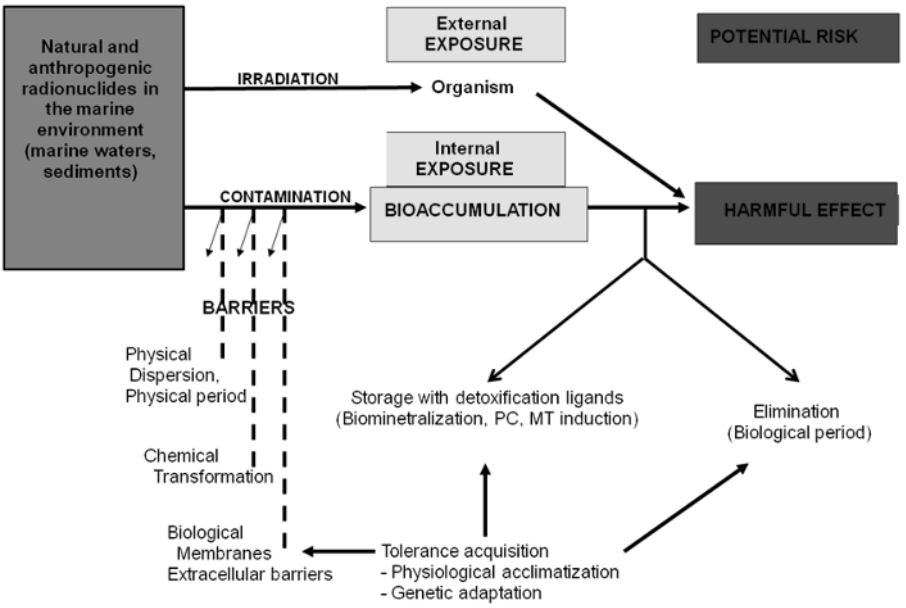

Figure 1.1. *The triad of marine radioecology. For a color version of this figure, see www.iste.co.uk/amiard/radioecology.zip*

1.1.3.2. *Contamination and irradiation, specificities of radionuclides*

Transfers of radionuclides to marine organisms can be direct from the various physical environments (marine waters and sediments) or indirect through food chains.

In living organisms, radioactive contamination can be external or internal. This contamination will remain external if the radionuclide adsorbs onto the external parts of the organism such as its skin, its gills or its lungs. It will be considered internal if the radionuclide is absorbed, crossing the biological barriers that are the skin, the gills or lungs, the digestive tract. In the same way, the ingestion of a contaminated prey item (or food), its digestion and the passage through the wall of the digestive tract of part of its contents will lead to a more or less intense internal contamination and irradiation (Figure 1.2).

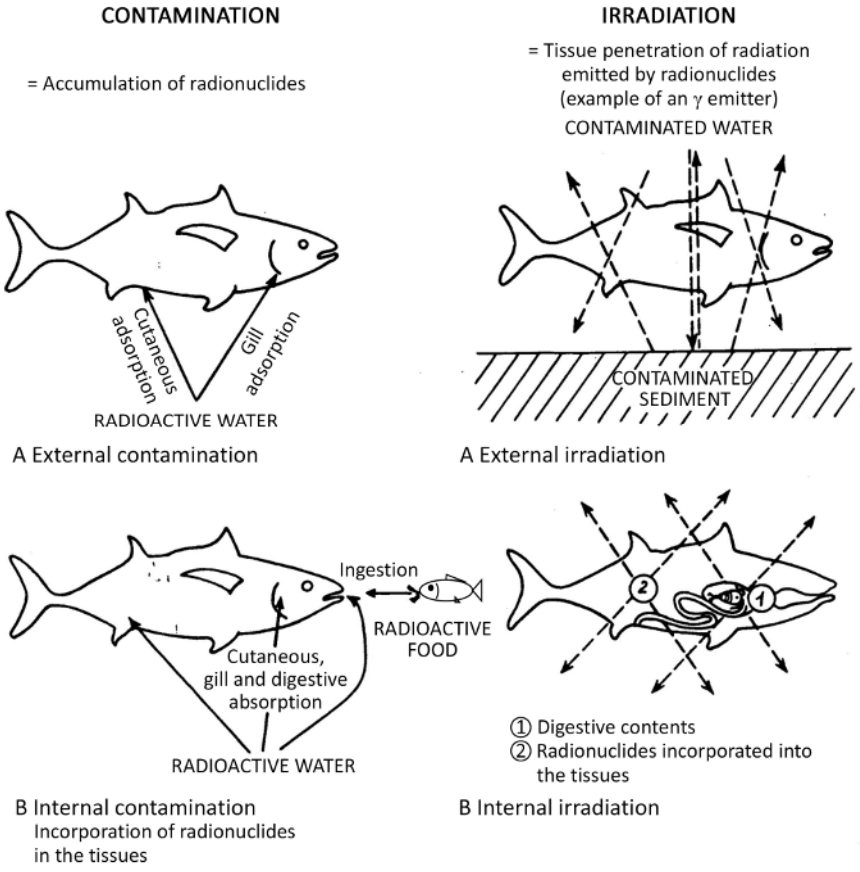

Figure 1.2. *Contamination and irradiation of organisms (from [AMI 80])*

The contamination of the physical environment (air, water, soil, sediment), external to the organism, induces an external irradiation. Conversely, internal contamination of the organism induces internal irradiation. By extension, the radioactive air inhaled during breathing, the absorbed water and contaminated food ingested during feeding will be considered as internal irradiation.

1.2. The principle of risk assessment

Living things are subject to three major classes of hazards: physical stresses, chemical stresses and biological stresses [AMI 17a]. Frequently the notions of hazard and risk are confused by the public.

A risk is the probability that a person will suffer harm or adverse health effects from exposure to a hazard. The risk is therefore the probability of the hazard occurring. This risk depends on two probability distributions, that of the hazard and that of the exposure. In the case of ionizing radiation, the hazard is a function of the dose (dose–response function). On the other hand, the probability of exposure is an inverse function of the dose. Indeed, the probability of suffering a high dose is lower than that of suffering a low dose. As a result, the risk zone is located at the overlap p(D) x p(E/D). The risk is therefore the sum of the hazard and the exposure. This reasoning can be applied in a similar way when considering an organism exposed to a potentially toxic compound in its environment.

Risk analysis involves three steps: hazard assessment, risk management and risk communication. Risk assessment involves four steps: hazard analysis, dose–response assessment, exposure assessment and risk characterization (Figure 1.3). These four steps will be detailed in this book for radioactive risk assessment of marine organisms.

The general principles of environmental risk assessment are shown in Figure 1.4. In the end, risk characterization consists of comparing the PEC (Predicted Effect Concentration) to the PNEC (Predicted No Effect Concentration) and risk is present when the ratio is equal to or greater than one.

In the environment, the assessment of adverse effects begins with the identification of hazards based on laboratory toxicity data for a single species tested and/or with data from different trophic levels, but the ecosystems are simplified as much as possible. The tests are standardized at the international level. Predicted No Effect Concentrations (PNEC) are estimated mainly by two methods (Figure 1.5): the scaling factor method and the statistical scaling method [AMI 17a]. The determination of the threshold value (PNEC) by extrapolation factors is based on toxicity data obtained by standardized laboratory tests. This method has two serious

shortcomings, the extrapolation of laboratory data to the real impact in the field with in particular the problem of interactions (additivity, synergistic and antagonistic effects) and the extrapolation from the short-term to the long-term. The uncertainties on the threshold value obtained are mainly due to intra- and interspecific variations (biological variability) and to intra- and inter-laboratory variations of toxicity data. In order to take into account the different uncertainties, an extrapolation factor is applied. This extrapolation factor applied will be inversely proportional to the knowledge of the toxicity of the substance. In the marine environment, due to its high biodiversity and the small number of standardized ecotoxicity tests, the extrapolation factors applied to freshwater aquatic species are systematically increased by a factor of 10. Thus, if the PNEC is obtained with a single short-term lethal concentration value for three freshwater or saltwater species representative of three taxonomic groups (algae, crustaceans, fish) from three trophic levels, the maximum extrapolation factor is 10,000 [EC 96, AMI 17a].

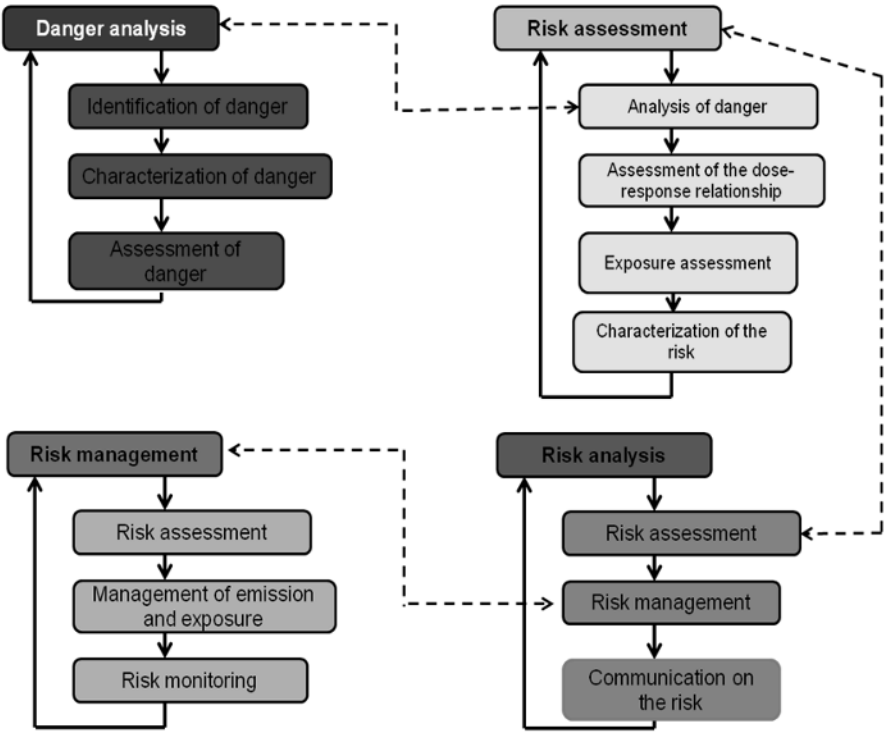

Figure 1.3. *Principle of risk analysis (modified from [DUF 01]). For a color version of this figure, seewww.iste.co.uk/amiard/radioecology.zip*

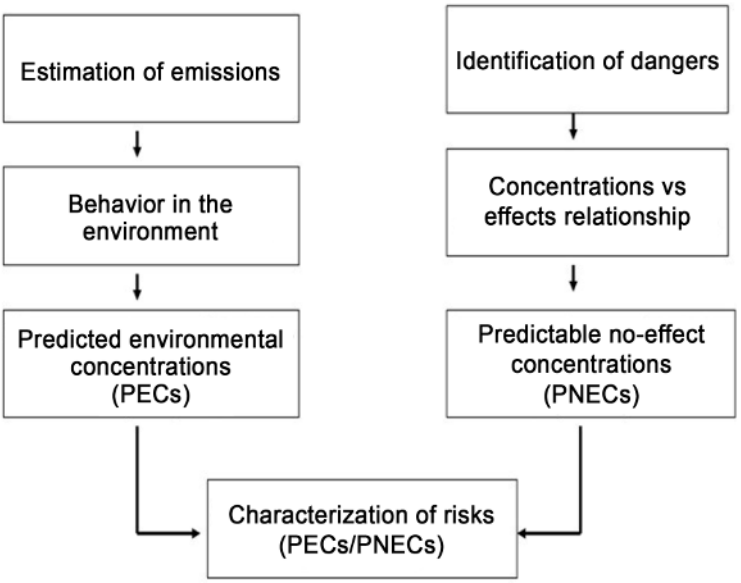

Figure 1.4. *Various steps of the risk assessment principle [AMI 17a]*

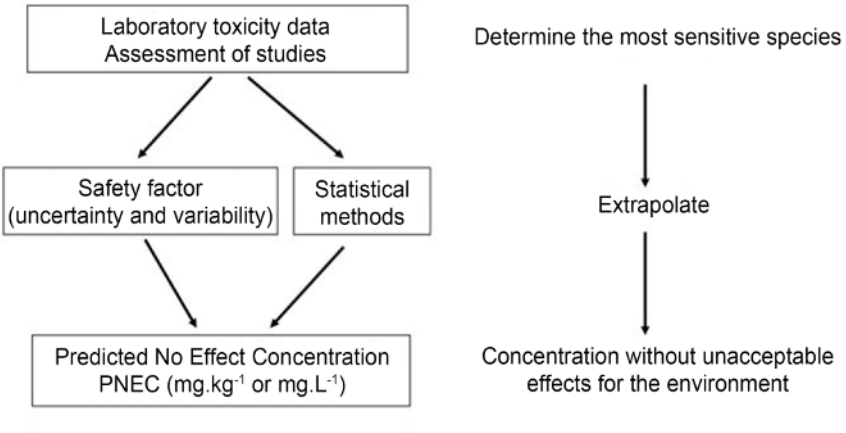

Figure 1.5. *Assessment of adverse effects (from [AMI 17a])*

The statistical extrapolation method is based on the species sensitivity distribution (SSD). The underlying assumptions [OEC 92] are that a species sensitivity distribution follows a known theoretical distribution function and the

group of species tested in the laboratory is a random sample of this distribution. Depending on the percentage p (often 5%) of sensitive species that can be considered negligible, we estimate the corresponding PNEC (Figure 1.6).

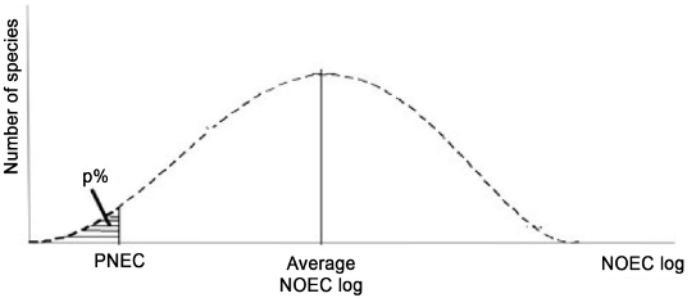

Figure 1.6. *Responses of various species of organisms to the NOEL for estimating a PNEC*

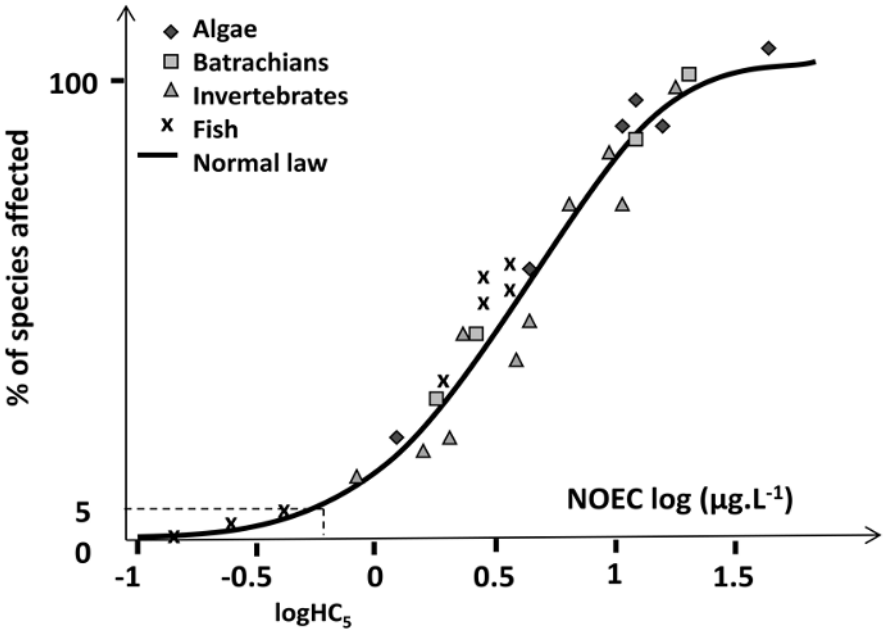

Figure 1.7. *Long-term toxicity of cadmium to freshwater organisms (HC_5 = 0.5 µg.L^{-1}, IC90 % = [0.27; 0.79], PNEC = HC_5/FE and 1 ≤ FE ≤ 5). For a color version of this figure, see www.iste.co.uk/amiard/radioecology.zip*

In practice, this method can only be applied when a large number of data are available. The transformation of long-term toxicity data (NOEC, No Observed Effect Concentration) is carried out in log-logistic or log-normal form and at least eight species are required; the extrapolation method requires at least 10 NOECs, preferably 15 for at least eight taxonomic groups; the choice of outcome is usually the 5th percentile (HC5, Hazardous Concentration 5%) and the assessment factor (AF) from 1 to 5 to reflect the identified uncertainties. The formula is therefore PNEC = HC5/AF. An example is provided in Figure 1.7 for long-term ecotoxicity to freshwater organisms contaminated with cadmium [AMI 17a].

1.3. The particular case of the risk linked to ionizing radiation

When applying the characterization of the radioactive risk related to the presence of radionuclides in the marine environment and in marine organisms, the first step, the identification of the hazard, is obvious; radioactivity is dangerous for all forms of life. The hazard of a radionuclide depends primarily on three criteria: its radiotoxicity, its radiation type(s) and its half-life. The hazard of radionuclides can be related to their chemical toxicity or to their ionizing radiation, called radiotoxicity. For all radionuclides, with the exception of uranium[3], radiotoxicity is clearly more dangerous than chemical toxicity. Radiotoxicity varies greatly depending on the radionuclide.

Radioactivity corresponds to an unstable nucleus which, to regain its stability, spontaneously emits one or more particles. Radiation is mainly of three types: α radioactivity, where a helium nucleus is emitted; β radioactivity, where either an electron and an electron antineutrino (β-) or a positron and an electron neutrino (β+) are emitted; and γ radioactivity by which a nucleus loses its energy by high-energy electromagnetic radiation. To this must be added X-rays, which are of the same photonic nature as γ-emitters, but which are produced by electronic transitions, and neutrons, which are present during chain reactions in nuclear reactors or during the explosion of atomic bombs.

The two main properties of radiation are the power of penetration in matter and the ionizing power.

In a radioactive sample, the number of decays per unit of time is proportional to the number of unstable nuclei of the radionuclide, present in the sample. As a result, for a given sample, the number of unstable nuclei decreases progressively with time.

3 From an enrichment in 235U equal to 7%, it is the radioactive toxicity that becomes higher.

The period during which half of the unstable nuclei disappear is called the physical period (Tp) or physical half-life. It is a characteristic and unchangeable quantity of each radionuclide. Thus, radionuclides made up of very unstable nuclei will have a physical half-life of a few fractions of a second, while those made up of very stable nuclei will have a physical half-life of thousands of years. Thus, after four physical half-lives, the radioactivity of a sample is decreased by a factor of 16.