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Radionuclide concentration processes in marine organisms: A comprehensive review

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ABSTRACT

The first measurements made of artificial radionuclides released into the marine environment did reveal that radionuclides are concentrated by marine biological species. The need to report radionuclide accumulation in biota in different conditions and geographical areas prompted the use of concentration factors as a convenient way to describe the accumulation of radionuclides in biota relative to radionuclide concentrations in seawater. Later, concentration factors became a tool in modelling radionuclide distribution and transfer in aquatic environments and to predicting radioactivity in organisms. Many environmental parameters can modify the biokinetics of accumulation and elimination of radionuclides in marine biota, but concentration factors remained a convenient way to describe concentration processes of radioactive and stable isotopes in aquatic organisms. Revision of CF values is periodically undertaken by international organizations, such as the International Atomic Energy Agency (IAEA), to make updated information available to the international community. A brief commented review of radionuclide concentration processes and concentration factors in marine organisms is presented for key groups of radionuclides such as fission products, activation products, transuranium elements, and naturally-occurring radionuclides.

1. Introduction

Accumulation of radionuclides in marine organisms, and in aquatic organisms in general, was noticed through the first measurements of radioactivity made in marine biota in the mid of 20th century (Polikarpov, 1966; Shannon and Cherry, 1967). Using radiation detection instruments that are largely outdated today, researchers succeeded to measure artificial radionuclides such as caesium-137 and cobalt-60 from nuclear weapons fallout in fish, mollusc and crustacean and naturally occurring polonium-210 in marine plankton (Polikarpov, 1966). When the 1st International Conference on Radioecological Concentration Processes was held in 1966, a large number of scientific reports on radioactivity in marine biota had been published already, and the conference enabled to consolidate at the highest international level such recently acquired knowledge (Aberg and Hungate, 1967).

Since then, the data base of radioactivity measurements and the concepts relevant to the understanding of chemical and physiological processes regulating accumulation of radionuclides in marine biota have continuously expanded (Cherry and Shannon, 1974; Eisenbud and Gesell, 1997; Nevissi and Schell, 1975b; Pentreath, 1980). This knowledge was the basis for more recent attempts to model bio concentration and transfer of radionuclides in the environment in general, and in the marine environment in particular (e.g., Strand et al., 2000; Tateda et al., 2013). The development of such radioecological models is crucial to predict the concentration of radionuclides by marine biota and the transfer and dispersal of radionuclides in the environment in order to provide a sound basis for radiological risk assessment to man and to other biological species, as needed from enhanced radioactivity levels in the environment. This need has been highlighted in several circumstances and, especially, following the nuclear accident of Fukushima and subsequent radioactivity releases into the sea (Tagami and Uchida, 2016) and significant advances has been made with identification of models' limitations and discrepancies (Vives I Batlle et al., 2016).

The correct understanding of bio concentration mechanisms and the harmonized use of concepts and terminology is very important to capture in computational models the parameters that underpin concentration and transfer of radionuclides in the environment. The better the radionuclide concentration mechanisms are understood, the better the radioecological models will be. With the purpose of better understanding the processes underlying biological concentration of radionuclides in the marine environment, some of the key concentration mechanisms in marine organisms are reviewed and summarized herein.

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2. The concept of concentration factor

The Concentration Factor (CF), understood as the ratio of concentration of a contaminant substance in biota and the concentration of the same substance in surrounding water, was first introduced in aquatic toxicology (Rand, 1995). This concept was found useful to describe the uptake and accumulation of radionuclides in aquatic biota and it was adopted in environmental radioactivity and radioecology reports since the 1950s (Ancellin et al., 1979; Eisenbud and Gesell, 1997). However, in the 1st Radioecological Conference held 50 years ago, the expression CF was still in use side by side with other expressions and it was not of generalized use yet (Aberg and Hungate, 1967). Gradually, in the next decades the CF concept became of overwhelming use in radioecology (Coughtrey et al., 1984). This factor is usually represented in terms of a concentration in biota relative to that in ambient sea water. If both biological material and seawater concentrations are derived per unit mass, this term is dimensionless (IAEA, 2004). Then, CF (dimensionless) = Concentration per unit mass of organism (kg/kg or Bq/kg wet weight)/Concentration per unit mass of sea water (kg/kg or Bq/kg).

In data compilations, usually CF values originated from laboratory experiments and from field measurements are pooled (IAEA, 2004). These data compilations may display a considerable disagreement of CF values for the same species that occurs because in many laboratory experiments test organisms were exposed to contaminants dissolved in water only, while in the environment the accumulation of contaminants occurred, inter alia, through combined uptake from water and from food with a significant effect on the CF value (Ancellin et al., 1979; Pentreath, 1980; Carvalho and Fowler, 1994). Because of this difference some authors introduced a different terminology to distinguish values from laboratory experiments (Bio Concentration Factors, or BCFs) from values obtained from field studies (Bio Accumulation Factors, or BAFs). There is some value in this distinction, although some of the international data compilations, including those of the IAEA, do not make use of them (Arnot and Gobas, 2006).

Currently, CFs remain basic operational values which have been useful to enable comparison of the concentrations of several radionuclides in the same biological species, and comparison of the ability of different species to concentrating the same radionuclide from marine environment. The clear advantage of using CFs resides in allowing rapid identification of species that will concentrate more the radionuclides from sea water, identifying those capable to providing early detection of radionuclides in the marine environment and, thus, the best to be used as sentinel organisms in environmental monitoring programmes. Once known the radionuclide CF value for a given species, the monitoring through periodic measurements in that species allows to surveying radionuclide concentrations in the marine environment confidently. Furthermore, in a given marine area, the use of a selected biological species will enable to identify the radionuclides of major concern in the environment (critical radionuclides). This bio monitoring approach was introduced near 50 years ago through the concept of Mussel Watch (Goldberg, 1975; Farrington et al., 2016). The Mussel Watch is used nowadays in many countries for monitoring radioactivity and other pollutants in coastal areas and allows monitoring the current status of contamination as well as temporal and spatial pollution trends (Carvalho and Civili, 2001; Villeneuve et al., 1999; Carvalho et al., 2011a, 2011b; NOAA, 2017).

3. Adsorption and absorption of radionuclides

For radionuclides in the marine environment two basic mechanisms have definite influence in their concentration by marine species: adsorption (i.e., uptake from sea water by physical-chemical processes onto the organism's external surfaces) and absorption (i.e., uptake of radionuclides from ingested food through the gut wall into the internal tissues and organs of organisms; absorption may also include respiratory uptake of radionuclides through the gills). Both adsorption and absorption may be very relevant in radionuclide concentration processes and, thus, in the build-up of CF values in marine organisms.

Adsorption is the key mechanism that intervenes in the concentration of transuranic radionuclides from sea water, particularly in organisms with hard and calcified external body surfaces, such as crustacean (e.g., crab, shrimp, lobster) and echinoderms (e.g., sea urchins, sea stars) (Fowler and Carvalho, 1985). The basic physical-chemical mechanism of adsorption is the attraction and binding of radionuclides by ionic charges on external body surfaces. Experimental results demonstrated that the uptake of plutonium, americium, californium and other transuranic elements was mainly due to radionuclide adsorption from sea water onto the external surfaces of organisms. For example, ^{237}Pu, ^{241}Am and ^{252}Cf concentrated in a marine isopod from sea water were mostly attached to the external body surface (exoskeleton) and were gradually loss by desorption when these crustacean isopods were transferred into non contaminated sea water (Carvalho and Fowler, 1985a; Fowler and Carvalho, 1985; Fowler et al., 1986). In experiments with transuranics and marine macrophytic algae, accumulation of radionuclides in algae was shown to be due to external sorption on algae blades. This adsorption was related to piston velocity across the blade boundary layer, and varied amongst radionuclides such as ^{241}Am, ^{237}Pu, and ^{210}Po (Carvalho and Fowler, 1985b). In microscopic planktonic algae, Pu and Am from sea water were mainly adsorbed onto the cell wall as well (Fisher et al., 1983a).

Absorption is the key mechanism in concentrating radionuclides from ingested food. This mechanism is particularly important when radionuclides are highly concentrated in the diet and when the gut transfer factor of radionuclides is high in the consumer species. Laboratory experiments have shown that for some radionuclides gut absorption may be low, while for other radionuclides it may be very high. For example, crustacean and fish fed with food contaminated with transuranium elements generally showed little absorption of these elements (a few percent only of the ingested dose, i.e., $< 5\%$) through gut walls (Pentreath, 1980; Carvalho et al., 1983; Carvalho and Fowler, 1985a). Contrasting with this, absorption of some elements from food by marine biota, such as polonium (^{210}Po), is very efficient and account for most of radionuclide concentrated from the environment being more important than ^{210}Po adsorption from sea water (Carvalho and Fowler, 1993, 1994). Experimental research on crustacean and fish using three polonium isotopes (^{208}Po, ^{209}Po, ^{210}Po) demonstrated through an elegant technique that Po from sea water practically did not contribute to ^{210}Po accumulated in internal tissues, while Po in ingested food explained most ($> 97\%$) of Po concentrated in internal organs (Carvalho and Fowler, 1994).

Marine organisms, such as for example those living in the epipelagic layer and in the deep layer of the oceans, are exposed to similar concentrations of ^{210}Po dissolved in seawater, about 1 mBq/L (Carvalho, 1990). Notwithstanding, in both environments there are fish species displaying a wide range of ^{210}Po concentrations as shown by several authors (e.g., Cherry and Shannon, 1974; Carvalho, 1988). Analyses of ^{210}Po in organisms from all layers of the ocean have documented this variation and ^{210}Po concentration in organisms was related to the energy transfer (mostly protein) in marine food chains and trophic levels (Carvalho, 1988, 1990, 2011). As a consequence, ^{210}Po CFs in marine organisms may vary, and have a reason to vary widely among species living in the same environment. Furthermore, even closely related biological species, i.e., belonging to the same taxon, as for example “Teleost Fish” (bony fish), may display very different CFs for ^{210}Po according to their trophic level (Fig. 1). In current CF data compilations, this effect and differences in CF for “Teleost Fish” (and other taxa) have not been taken into account yet.

CF values do not elucidate the concentration mechanisms and do not inform about the sites of radionuclide accumulation in organisms. The sites and type of binding (external adsorption by weak chemical bonds such as with transuranium elements, or internally accumulated