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Review Article

A mini review on bioaccumulation of ^{210}Po by marine organisms

Alam, L. and *Mohamed, C.A.R.

Marine Ecosystem Research Centre (EKOMAR),
Faculty of Science & Technology, Universiti Kebangsaan Malaysia,
43600 Bangi, Selangor, Malaysia

Abstract: ^{210}Po, a naturally occurring radioisotope, is known to be enriched by various kinds of marine organisms. It can, however, have a toxic effect and therefore play an important role in humans, as among the natural radioactivity in human foodstuff ^{210}Po is the major contributor. Those seaweeds which are rich in protein can have a higher activity of ^{210}Po as all species display a comparatively higher ^{210}Po activity in muscles and storage organs than in their hard parts. ^{210}Po concentrations may, however, vary according to the sampling site and geographical location. Moreover, literature surveys reveal that ^{210}Po concentration in organisms can vary according to the species, their feeding habits, physiological processes, body size and seasonal changes. From the literature survey undertaken some conflict between the different research studies was found and it was also noted that some important aspects within the research were not well-documented and therefore comprehensive. This being the case, further research is essential.

Keywords: polonium, seafood, natural radionuclide, marine flora

Introduction

The bioaccumulation of Po-210 refers to a process by which Po-210 accumulates in various tissues of a living organism. Polonium, a naturally occurring radionuclide of the uranium series (Ivanovich and Harmon, 1992), was first discovered by Pierre and Marie Curie in 1898 in the course of research into the radioactivity of uranium and thorium minerals (Figgins, 1961). Polonium has 25 known radioactive isotopes with mass numbers of 192-218, of which only the 208, 209 and 210 isotopes have half-lives longer than 1 day (Connan et al., 2007). Among these three isotopes, ^{210}Po is of the most interest from a marine environmental impact viewpoint. This is because it is considered an important source of internal radiation dose to marine organisms (Cherry and Shannon, 1974; Cherry and Heyraud, 1981, 1982). ^{210}Po is a high energy alpha particle emitter in the uranium decay chain (Cherry and Shannon, 1974) and among natural radionuclides occurring in the ocean, alpha emitters are considered to be the most important (Hernandez et al., 2002), because of their high mass and charge, are more damaging and so are accorded a "radiation weighting factor" of 20.

^{210}Po can have a toxic effect even in small concentrations due to its high-energy alpha radiation (Moroz and Parfenov, 1972). Despite its toxic properties, the radionuclide ^{210}Po is readily assimilated by marine primary producers (Fisher et al., 1983) and is known as a major contributor (90%) of the natural radiation dose from alpha-emitting radionuclides, as received by most marine organisms (Cherry and Shannon, 1974; McDonald et al., 1991). Such that, ^{210}Po is commonly accumulated by marine organisms and transferred through the food chains (Heyraud and Cherry, 1979; Carvalho, 1988), with ingestion being the major route of entry (Carvalho and Fowler, 1994) it correspondingly plays an important role in human life. ^{210}Po enters into the human body through the ingestion of food and water, along with inhalation; in this case the ingestion of seafood is the most significant route (Chen et al., 2001). More than half of the internal radiation dose that man receives originates from this natural radionuclide as a result of seafood consumption (Aarkrog et al., 1997). It has recently been discovered that ^{210}Po is by far the major contributor, with the average dietary fish-product component containing 2 Bq kg−1 ^{210}Po (UNSCEAR, 2000). Through the ingestion pathway, ^{210}Pb and

*Corresponding author.
Email: *carmohd@ukm.my*
Tel: +60389214618, Fax: +60389214619.

^{210}Po deliver about 83% of the annual effective dose to humans (UNSCEAR, 2000). The affinity of ^{210}Po for protein enables it to pass through the food chain, and increased body burdens of ^{210}Po have been found where diets include protein-rich meat and seafood (Watson, 1985; Skwarzec et al., 2001).

In order to protect the health of seafood consumers, many countries have set guidelines for the maximum permissible levels of toxic pollutants in seafood. These guidelines are essential to protect public health. Other problems, however, arise when guidelines are not met and these may lead to dramatic economic impacts on the people and companies making their living from such sea resources. It is therefore vital that the levels of radionuclides, specifically ^{210}Po, are monitored in the coastal zones, where most fishery and farming activities occur. This paper focuses on the concentrations and distribution of the naturally occurring radionuclide, ^{210}Po, in marine organisms. Moreover, this review will make recommendations for the conducting of future research.

Sources of ^{210}Po

Radioactive substances are found in the atmosphere, but are predominantly present in the lithosphere. Of principal importance are the ores of uranium and thorium, but also potassium salts which contribute to natural radioactivity. The uranium-radium decay chain starts with ^{238}U, in the course of which ^{226}Ra, ^{222}Rn, ^{210}Pb, and ^{210}Po are produced. Figure 1 displays this decay chain. From the mother nuclide of all the members of the uranium family, ^{238}U (4.468 × 10^9 years), two short-lived isotopes of thorium and protactinium are formed by one alpha and one beta decay. Another uranium isotope, ^{234}U with a half-life of 2.455 × 10^5 years, is formed by beta decay from protactinium. Further alpha decays subsequently produce ^{230}Th (7.54 × 10^4 years), ^{226}Ra (1600 years) and the noble gas ^{222}Rn (3.825 days). Its decay products include polonium, bismuth and lead isotopes up until ^{214}Po, all short-lived (less than 30 minutes). The next isotope ^{210}Pb has a half-life of 22.3 years. By beta decay, ^{210}Bi (5.013 days) is formed then by another beta decathe alpha emitter ^{210}Po (138.38 days) is formed, until the stable ^{206}Pb is reached (Johansson, 2008).

Mining activities (uranium, coal and mineral sands) are considered to be the main source of technically enhanced natural radioactivity. Moreover, the combustion of fossil fuels and other energy production, such as geothermal energy and the use of phosphate rock, also contribute to environmental radioactivity. In regions of excessive mining, contamination is caused by dissolved radionuclides from the ^{238}U and ^{232}Th decay chains which are brought to the surface by the drainage of mine water and then discharged into the nearby water systems. Additionally, contamination occurs when mine waste deposited in close proximity to the mines is exposed to weathering thus leading to the leaching of radionuclides with water. This technically enhanced natural radioactivity also includes ^{210}Po, which is thought to result in elevated radiation exposure among those people living in these areas.

^{210}Po accumulation in marine flora

Biota constitutes one of the most important living and renewable resources in the marine environment. Measurement of ^{210}Po concentrations in biota (Shannon and Orren, 1970; Folsom and Beasley, 1973; Germain and Simon, 1995) can be used in marine food chains. ^{210}Po is accumulated by phytoplankton during the dissolved phase and the uptake of ^{210}Po is unaffected by light or temperature (Stewart and Fisher, 2003). For this reason it appears that the passive adsorption on the surface of cells control the uptake of this element (Stewart and Fisher, 2003), as is shown for many other metals (Fisher and Wante, 1993; Fisher and Reinfelder, 1995). It has been demonstrated that ^{210}Po concentrations vary in marine phytoplankton in a manner which is dependent on both the size and protein content of the cells (Stewart and Fisher, 2003).

Laboratory studies have also investigated the accumulation of ^{210}Po in bacteria and phytoplankton, and related its cytological distribution to that of protein (Fisher et al., 1983; Cherrier et al., 1995). This is consistent with observations that ^{210}Po is associated with the animal tissue protein (Heyraud et al., 1976; Carvalho and Fowler, 1994; Durand et al., 1999). Moreover, as brown algae are rich in protein, a higher activity of ^{210}Po has been found in seaweeds (Suriyanarayanan et al., 2007). The activity concentration of ^{210}Po in seaweed was shown to be higher during the autumn/winter months and lower during the spring/summer period (Connan et al., 2007). This may be the reason for the bioavailability of ^{210}Po, in relation with the physico-chemical modifications of the environment.

^{210}Po concentration in Crustaceans

The high bioaccumulation of ^{210}Po in particular marine organisms has been acknowledged for many years (Hoffman et al., 1974; Heyraud and Cherry, 1979). Since the food chain is clearly the main transfer pathway for polonium in marine crustacean, ^{210}Po concentration in organisms should reflect the ^{210}Po content of their prey to a degree, dependent

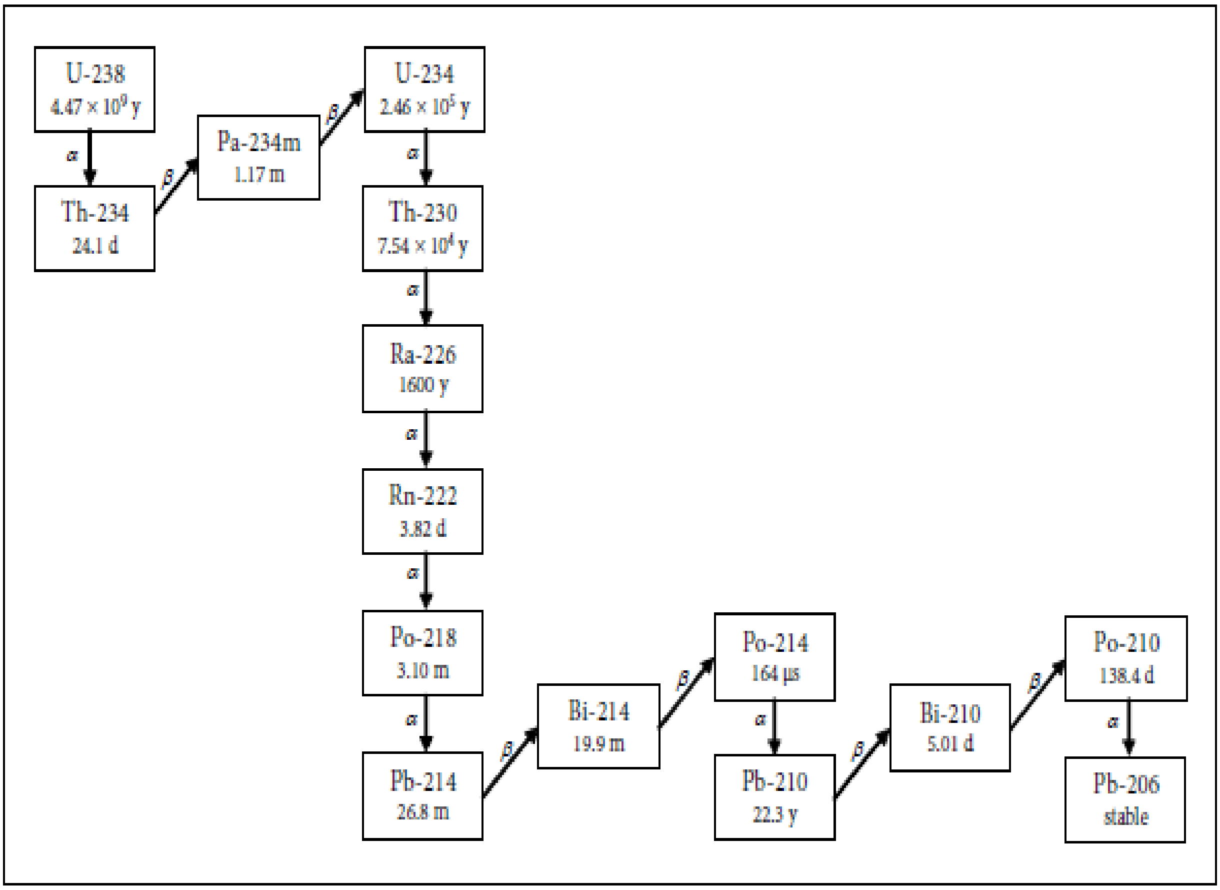

Figure 1. The ^{238}U decay chain

upon food assimilation efficiency (Carvalho and Fowler, 1993). Therefore, the use of ^{210}Po as a natural tracer in diet studies of marine organisms, as proposed by Heyraud et al. (1988) and Cherry et al. (1989), is well justified. In the hepatopancreas of marine invertebrates, the radiation dose from ^{210}Po ranges from 13000 to 280000 mremyr^{-1} (Heyraud and Cherry, 1979; Cherry and Heyraud, 1981, 1982), in contrast, the human organ which receives the highest natural dose of radiation is the lung, with a typical dose from all sources of about 400 mremyr^{-1} (UNSCEAR, 1977).

Crustaceans display comparatively higher ^{210}Po activity in muscle than in their exoskeleton (Suriyanarayanan et al., 2007). Polonium, which is stable in seawater as divalent species Po^{2+} (Brookins, 1988), is mostly likely adsorbed onto shrimp exoskeleton through chelation by functional groups on organic surfaces, as is the case for other divalent metals (Stumm, 1992). On the other hand, an analysis of tissue, demonstrated that the experimental uptake of ^{210}Po from water was predominantly due to adsorption onto shrimp exoskeleton which is more than 90% of the total body burden, whereas absorption into shrimp internal organs, although noticeable, was comparatively low (Carvalho and Fowler, 1993). Whilst limited data is available on ^{210}Po sub cellular distribution in storage organs such as the digestive gland or liver, those that exist show a great heterogeneity. Thus in crustacean, about 60% of the radionuclide was found in the cytosolic fraction of the digestive gland from the species *Saduria entomon* (Stepnowski and Skwarzec, 2000b), while in the South African lobster *Jasus lalandii* species, 52% of the ^{210}Po was found in the microsomal fraction of the digestive cells and 22% was in the cytosolic fraction (Heyraud et al., 1987). Research undertaken by Carvalho and Fowler (1993) found that ^{210}Po dissolved in sea water and is not the main mechanism for the accumulation of high ^{210}Po concentrations measured in organisms.

The extent of ^{210}Po concentration in the digestive organ varies between different species. For example, the ^{210}Po level in the shrimp hepatopancreas are two to three order of magnitude greater than that found in tail muscle (Swift et al., 1994). Such that although the shrimp hepatopancreas only constitute a minor fraction, about 5% of the whole body weight, it does however contain a large fraction of the total ^{210}Po inventory (Young et al., 2002). Accumulation of ^{210}Po from seawater into the internal tissue is closely related to the intake of water for osmotic regulation (Carvalho and Fowler, 1993). It has been suggested that net water absorption in intermolt crustaceans occurs through the hepatopancreas and gut wall, and before ecdysis, through the integument (Mantel and Farmer, 1983).

^{210}Po concentrations in shrimp may vary significantly according to the different sites (Young et al., 2002). In a study of the content of ^{210}Po in the marine shrimp, Cherry and Heyraud (1981) noted a steady increase in levels on going from estuarine to coastal and pelagic to deep sea species. The extent of the increase between the estuarine and coastal environment was in the order of 3-fold. A plausible explanation to account for the increase in ^{210}Po concentrations between estuarine and coastal environments was provided in terms of changes in the chemical form of dissolved ^{210}Po (Cherry and Heyraud, 1981). Crab and shrimp are opportunistic feeders and indiscriminately take food from the benthic zone (Warner, 1977). Indeed, it has been reported that there appears to be a clear correlation between diet and ^{210}Po concentration in penaeid and carid shrimps from the north-eastern Atlantic (Heyraud et al., 1988). ^{210}Po concentrations in crabs from along the eastern coastline appear to be lower compared with those from Wales and the English Channel (Young et al., 2002). This disparity may relate to the fact that edible crabs show strong geographical and seasonal differences in size composition (Bannister, 1999). Variability in ^{210}Po concentration is large, even among individuals of the same habitat. For example, Heyraud and Cherry (1979) found considerably higher levels in *Sergestes* spp. compared to *Pasiphaea* spp. although these two shrimps live at a similar depth and occupy the same niche.

Although the immediate source of ^{210}Po in shrimp was suggested to be the food they consume, the ultimate source of ^{210}Po in food was that found in the water. Given that the variation in the ^{210}Po content of seawater is relatively small, it was further suggested that the ultimate availability of ^{210}Po to the food chain depends on its form in seawater (Young et al., 2002). The direct intake of seawater alone can account for the increased concentration of ^{210}Po measured in the muscle of shrimp exposed to the radionuclides in seawater (Carvalho and Fowler, 1993). Results from a study of ^{210}Po concentrations in the common shrimp and crab from the North Sea indicated that although levels were significantly variable, it was impossible to correlate the changes with time (Swift et al., 1994).

Influence of ^{210}Po on Molluscs

It has been found that the ^{210}Po/^{210}Pb activity ratio is about 0.1 in the atmospheric particles which fall into the ocean, 0.5 in sea water, 2 in zooplankton fecal pellets, 20 in whole zooplankton and 100 in the hepatopancheas of many marine invertebrates (Cherry and Shannon, 1974; Cherry and Heyraud, 1982). Bivalve molluscs, specifically the different species of the genus *Mytilus*, have an ability to accumulate and concentrate heavy metals to various orders of magnitude with respect to the levels found in their environment (Boisson et al., 1998; Nasci et al., 1998; Da Ros et al., 2000). These organisms accumulate most of the contaminants at much higher levels that those found in the water column and they are representative of the pollution of an area, for that reason they are ideal markers for monitoring the quality of coastal water (Uğur et al., 2002). Several studies have demonstrated that the natural alpha emitting radionuclide ^{210}Po is accumulated to an exceptionally high level in the tissue of a variety of marine organisms, well above the levels of the parent radionuclide ^{210}Pb (Carvalho and Fowler, 1994; Stepnowski and Skwarzec, 1999), the hepatopancreas of marine invertebrates together with the pyloric caecum of fish (Folsom et al., 1972; Heyraud and Cherry, 1979; Cherry and Heyraud, 1982) appear to constitute the highest known natural dose domain in our biosphere. In the United Kingdom, much attention has historically been focused upon fish and shellfish consumers in the vicinity of Whitehaven (Cumbria), where there are known anthropogenic inputs of natural radionuclides. Concentrations of ^{210}Pb, ^{226}Ra, U and Th radionuclides in the edible fractions were much lower in those than for ^{210}Po, suggesting that the dose to man resulting from seafood consumption is extremely sensitive to variation in diet (Young et al., 2002).

As mussels and cockles are filter and suspension feeders, respectively and graze on phytoplankton and other suspended matter in the near bottom water (Boyle, 1981; McLusky, 1981), the variation rate of ^{210}Po activity in the mussels is high. The activities in grazing mollusk, i.e., limpets, are much lower than in mussels and oysters, which are filter-feeding molluscs. ^{210}Po levels were investigated in the scallop *Chlamys varia* from the French Atlantic coast in order to determine the levels and distributions among the tissues (Bustamante et al., 2002). Surprisingly, the highest ^{210}Po concentrations were found in the scallops from Ré Island, France, along with mussels, however, there is no evidence of a source of ^{210}Po in this site as it remote from industrial activities

Connan et al., (2007) found that the highest ^{210}Po activities were observed during the winter period and the lowest during the hot period. In this case, it can be assumed that during the warm period mussels accumulate less ^{210}Po than during the cold period. The winter period generally corresponds, for mussels, to the gametogenesis period. Since the reproduction period runs through the spring and summer (Seed, 1969), so it is possible that a link exists between the storage of ^{210}Po in winter and the physiological variation associated with the sexual and biochemical cycles, as has been reported for metals (Bryan, 1976; Cossa, 1989). ^{210}Po concentrations can also vary between different sites. In a pilot study in which spot samples were collected from six coastal sites between Scotland, England, France and Monaco, it was found that levels of ^{210}Po ranged from 111-459 Bqkg^{-1} dry weight in soft tissue (McDonald et al., 1986) and varied according to their body size. Ryan et al., (1999) noted that at several sampling sites, the smaller mussels had higher ^{210}Po concentrations than the larger ones. Similarly, Bustamante et al., (2002) discovered that small scallops also displayed the highest ^{210}Po concentrations in the whole soft parts and in the digestive glad. In contrast to Connan et al., (2007) this proved that mussels have the highest polonium activity and that differences might be explained by age and metabolic rates.

The disparity in the distribution of ^{210}Po between mussel tissues is less marked. The digestive gland of a mussel accounts for approximately 10% of the total soft tissue weight but contains between 15% and 36% of the ^{210}Po soft tissue inventory (Wildgust et al., 1998). Wildgust et al., (1998) suggested that environmental fluctuations of ^{210}Po are better reflected in the digestive gland than the whole soft tissue because this organ is the major entry point of particle-bound ^{210}Po. For *Chlamys vaira*, the highest ^{210}Po concentrations were found in the digestive gland (Bustamante et al., 2002). High ^{210}Po concentrations have also been measured in the digestive gland of several seashell species such as the mussel, *M Trossulus*, the scallop *P yessoensis* and the bivalve *B Cornutus* (Yamamoto et al., 1994; Stepnowski and Skwarzec, 2000a). Similarly, Connan et al., (2007) undertook research on abalones, an edible species, which has rarely been researched in the marine environment. The muscle, the part consumed by humans, presents very low activity, lower even than the shell. Distribution among the various organs shows comparable activities in the innards, digestive gland, gonads which are different from those which have been observed in other species.

In *Chlamys islandicus*, the highest ^{210}Po concentrations were found in the gills which appear to constitute the main ^{210}Po entrance for this species (Bustamante et al., 2002). Connan et al., (2007) noted from investigation that in oysters, the organs with the highest concentration of ^{210}Po are the digestive gland, the gills and the mantle. The concentrations in the soft part of the scallop *Chlamys varia* were 2-3 times higher than those measured in the common mussel *Mytilus edulis*. Mussels are considered as sentinel of indicator species for contaminants but values reported for whole Mytilidae are always lower than those from *Chlamys varia* (Bustamante et al., 2002). The availability of food and the variation of the suspended matter in the water column are also important parameters in the bioaccumulation of metals (Connan et al., 2007). Wildgust et al., (1998) observed a significant variation of about five fold in levels of ^{210}Po in the mussel digestive gland over a 12-month period. Levels of ^{210}Po in the digestive gland have been shown to be strongly correlated with changes in levels of suspended particulate materials, so it is suggested that the majority of ^{210}Po in this organism is derived from leaching from sedimentary particles (Young et al., 2002). Germain et al. (1992) and Ryan et al. (1997) were unable to find any consistent temporal changes in the level of ^{210}Po from the whole soft tissues in mussels.

Impact of ^{210}Po on fish

It is well-known that ^{210}Po is responsible for a large share of the radiation doses received by humans from the consumption of seafood product. The ^{210}Po in marine organisms consumed by humans would therefore be the source of exposure between 5 and 20 μSvyr^{-1} per person and could contribute up to 80% of the collective dose received by a population consuming marine animals (CEC, 1989; Aarkrog et al., 1997; Pollard et al., 1998). Thus fish also plays a significant role in the transfer of radionuclides to human. The sulphur-analog properties of polonium (Moroz and Parfenov, 1972; Heyraud et al., 1987) may explain its longer turnover time in marine organisms, through binding and cycling with amino acids and sulphur-containing proteins.

^{210}Po accumulation in fish can vary according to habitat, feeding behavior and species. Heyraud and Cherry (1979) deduced that the ingestion of food should play a major role in the accumulation of ^{210}Po. Later, Cherry et al. (1989) reported the results of an experimental study on ^{210}Po ingestion by anchovy and pilchard which demonstrated that the ^{210}Po content of fish flesh reflects to some degree the concentrations of

^{210}Po in food. Variation levels of ^{210}Po were observed between the individual fish species as a result of a differing diet. A higher accumulation of ^{210}Po was recorded in the muscle of carnivore feeders than plankton feeders (Suriyanarayanan et al., 2007). More generally, Shannon (1973) reported that pelagic fish, i.e., mackerel, contain roughly five times more ^{210}Po than demarsal species such as plaice. The potential use of this radionuclide as a natural tracer of the diet of marine organisms has been suggested (Carvalho, 1988; Heyraud et al., 1988; Cherry et al., 1989; Miquel et al., 1993). Connan et al. (2007) found that Sparus, which predominantly feeds on small bivalve molluscs and crustacean, is rich in polonium in comparison to Solea which feeds on very tiny seabed organisms. On the other hand, Lazorenko et al. (2002) deduced that the highest concentration of ^{210}Po were specific to planktivorous pelagic fishes and the lowest to bottom predator species. The intermediate position was occupied by demersal species. The role of plankton in this process is significant because this food source is the primary source of uptake of this radionuclide among fish.

Heyraud and Cherry (1979), Skwarzec (1988), Swift et al. (1994), Wildgust et al. (1998) have all reported that ^{210}Po is non-uniformly distributed in fish and benthic organisms. The highest ^{210}Po concentration occurs in organs involved in digestion and metabolism, such as the hepatopancreas of marine invertebrates (Swift et al., 1994) and the intestine, stomach, spleen and pyloric caecal of fish (Skwarzec, 1988). Iyenger et al. (1980) has shown that the concentration of ^{210}Po in the muscles of fish is always higher than in the bones. A lower concentration of ^{210}Po in muscle is important because humans predominantly consume this part of the fish, which accounts for a low radiation dose. Moreover, Dahlgaard (1996) found a large individual variation of the ^{210}Po concentration in fish meat. Skawarzec (1988) observed that fish from the Baltic Sea showed high levels of activity in the intestine and liver.

Levels of ^{210}Po found in the digestive organs of fish tend to correlate with the degree of stomach repletion and thus decrease if food is scarce (Skwarzec, 1988). The residence time of ^{210}Po within the digestive system of fish was short thus resulting in a rapid decrease in 210Po content in the liver and intestine when the stomach was empty. After uptake, ^{210}Po is distributed internally in the fish and the muscular tissue in the following order: entrails ≥ liver > skeleton > muscles (Lazorenko et al., 2002). Data is available on the high concentration of this radionuclide in fish kidneys and spleens (Cherry and Shannon, 1974). Connan et al. (2007) investigated two species, Solea and Sparus, and observed that low activities of ^{210}Po were found in the gills, muscles, skins and bones. The highest activities were observed in the livers and intestine whilst the muscles presented the lowest activity. It can be seen that the distribution of ^{210}Po among the various organs in these two species varies according to their lifestyle. As Solea is a benthic sediment burrowing flatfish while Sparus is an open sea fish, the ^{210}Po activities are higher in Sparus. Cherry et al. (1994) observed that the family clupeidae is high in polonium. Hernandez et al. (2002) determined the ^{210}Po concentration in fish, molluscs and crustacean collected in Cienfuegos Bay. The highest values of ^{210}Po concentrations were found in crustaceans but a significant accumulation was also observed in fish and molluscs. Carvalho and Fowler (1994) observed that the ^{210}Po absorption efficiency for prawn and fish was approximately 0.33 and 0.05, respectively, and roughly corresponded to the assimilation efficiencies of protein from food.

Remarks and Recommendations

This paper provides a baseline view on ^{210}Po accumulation in marine organisms. It has been found that all marine organisms accumulate this radionuclide in a high concentration because of its solubility in seawater and affinity to organic matter. Thereby it serves as the major contributor of the natural radiation dose received by most marine organisms. Furthermore ^{210}Po has been found to be concentrated at a higher degree in the soft tissues of organisms that are consumed by humans. As a result, human consumers of seafood receive the largest dose of natural radiation. Additionally, the enhancement of the natural levels of ^{210}Po in the marine environment from various industries can increase the potential risk of their transfer to humans through the consumption of seafood. Unfortunately, there is limited data available on the accumulation pattern of ^{210}Po in marine flora, particularly in microorganisms such as bacteria and virus. Correspondingly, the study of ^{210}Po accumulations transferring to humans through small organisms in the food chain is far from adequate. Therefore, it is strongly recommended that further and more complete research is undertaken to study the bioaccumulation of ^{210}Po in the marine environment and the safety dose uptake level of seafood, which is a notable source of ^{210}Po in humans.

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