Contrasting transfer of polonium-210 and lead-210 across three trophic levels in marine plankton

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ABSTRACT: The naturally occurring radionuclides ^{210}Po and ^{210}Pb can be used as geochemical tracers in marine systems, but their interactions with biota in surface waters need to be understood before oceanographic data can be interpreted unambiguously. We compared the food chain dynamics of these radionuclides in plankton assemblages by measuring the uptake and trophic transfer of ^{210}Po and ^{210}Pb from phytoplankton to brine shrimp *Artemia* sp. to euphausiids *Meganyctiphanes norvegica* under controlled laboratory conditions. The ratio of ^{210}Po:^{210}Pb within organisms increased 5- to 12-fold with each trophic level (phytoplankton to grazer to carnivore), reflecting a preferential bioaccumulation of ^{210}Po over ^{210}Pb. *M. norvegica* assimilated 44 % of the polonium ingested but only 3.5 % of the ^{210}Pb ingested. Because ^{210}Pb was unassimilated, the ratio of ^{210}Po:^{210}Pb was 1 to 2 orders of magnitude smaller in zooplankton fecal pellets than in the animals producing them. These results suggest that in surface waters ^{210}Po has the potential to build up in food chains and be biologically recycled, whereas ^{210}Pb would not build up in marine food chains and would display shorter residence times. Since euphausiids comprise an important link between small plankton and larger predatory animals in many marine ecosystems, they may serve as an important conduit of ^{210}Po to those predators consumed as seafood by humans.

KEY WORDS: Polonium · Lead · Trophic transfer · Plankton · Bioaccumulation · Residence time · *Artemia* · Krill

INTRODUCTION

For many elements that bioaccumulate in marine organisms, the greatest bioconcentration from water into tissues occurs at the bottom of the food chain (IAEA 2004). Once associated with phytoplankton, a metal or metalloid can be transferred to higher trophic levels, or the passage up the food chain can be blocked if key components of the food chain (e.g. herbivores) do not assimilate the ingested metal and instead package it into sinking fecal matter (Fisher & Reinfelder 1995). One element that has received increasing attention as both a prospective geochemical tracer and as a potential source of high levels of radioactivity to marine organisms and humans is polonium-210. ^{210}Po ($t_{1/2}$ = 138 d) is a naturally occurring radionuclide formed by the beta decay of its grandparent ^{210}Pb

($t_{1/2}$ = 22 yr) via ^{210}Bi. Unlike ^{210}Po, ^{210}Pb is not effectively accumulated in marine organisms (Carvalho & Fowler 1993), but is still used as a geochemical tracer. The geochemical cycling of both ^{210}Po and ^{210}Pb has been used to study particle scavenging, water mass mixing and vertical fluxes (Cherry et al. 1975, Cochran et al. 1983, Fowler & Knauer 1986, Nozaki et al. 1997, Masqué et al. 2002), although detailed studies that provide mechanistic explanations for their different behaviors are still largely lacking. Further, efforts to find a specific radiotracer of carbon flux in the oceanic water column have explored the application of ^{210}Po to complement work with ^{234}Th (Friedrich & Rutgers van der Loeff 2002).

The concentration of ^{210}Po in seafood has received much interest from the marine scientific community because it is a high-energy alpha-emitter that accounts for

most of the radiation dose received by marine organisms (Shannon & Cherry 1967, Shannon et al. 1970, Heyraud & Cherry 1979), even those exposed to anthropogenic radioactive releases into coastal waters (Aarkrog et al. 1997). Consumption of seafood could increase the radiation dose to humans by an order of magnitude beyond background radiation (Thomas et al. 2003). Bustamante et al. (2002), for example, found that activity from the ^{210}Po alpha particles consumed in 4 kg of scallops would effectively account for the current human annual exposure limit of 1 mSv.

The uptake of ^{210}Po has been examined from the dissolved phase to marine phytoplankton (Fisher et al. 1983a, Stewart & Fisher 2003a) and from phytoplankton to zooplankton grazers (Stewart & Fisher 2003b). We are not aware of controlled experiments that examined the trophic transfer of this element from herbivores to carnivorous plankton, although it can be inferred from field studies (Heyraud et al. 1976) that it is likely that such transfer occurs in plankton assemblages. It has been assumed that assimilation of metals in herbivore grazers controls the ultimate fate of metals in food chains and is sufficient to predict bioaccumulation in higher animals (Fisher & Reinfelder 1995), but this has not been carefully examined under controlled conditions. To explore this issue further, we conducted experiments to measure the transfer of ^{210}Po from herbivorous zooplankton (brine shrimp) to the euphausiid *Meganyctiphanes norvegica*, or 'northern krill,' feeding as a carnivore. We also compared the uptake of ^{210}Po with that of ^{210}Pb in these plankton assemblages. Many studies have confirmed that marine animals can take up metals and radionuclides from the dissolved phase and from food (Reinfelder et al. 1998, Wang & Fisher 1999). Because the uptake of ^{210}Po from water has been shown to be a non-significant (<3%) route of exposure in crustaceans (Carvalho & Fowler 1993, 1994, Stewart & Fisher 2003b), we focused solely on uptake through food in these experiments.

Euphausiids (Crustacea: Euphausiacea) are the second most common net zooplankton (>20 μm) globally after copepods, and form a 'bridge' between the plankton and larger predators such as fishes, cephalopods and mammals. They can be grazers, filter-feeders, predators, or detritivores (Rupert & Barnes 1994), but enzyme activity and fatty acid biomarkers indicate that *Meganyctiphanes norvegica* is primarily omnivorous (Mayzaud et al. 1999, Buchholz & Saborowski 2000, Virtue et al. 2000). Euphausiids are important prey for many commercially valuable fishes and invertebrates including blue whiting, hake, mackerel (Bozzano et al. 1997, Cabral & Murta 2002) and the Norway lobster *Nephrops norvegicus* (Cristo & Cartes 1998). Further, krill are not only an important part of the marine food chain, they also comprise a large fishery in themselves

for both human consumption and animal feed (Kils & Klages 1979).

The concentrations of trace metals (Fowler 1977, Stoeppler & Brandt 1979, Locarini & Presley 1995, Nygard et al. 2001), anthropogenic radionuclides (Antezana & Fowler 1972, Higgo et al. 1977, Marzano et al. 2000, Heldal et al. 2003) and ^{210}Po (Cherry et al. 1975, Heyraud & Cherry 1979) have been measured in euphausiids from the Antarctic, Pacific, Mediterranean and Atlantic. In all these studies, *Meganyctiphanes norvegica*, the Antarctic krill *Euphausia superba*, and other euphausiid species provided a clear link between the plankton and larger organisms and were shown to play an important role in the transfer of these elements in marine ecosystems.

It is necessary to assess the assimilation and retention of metals, including ^{210}Po and ^{210}Pb, in marine zooplankton in order to evaluate the build-up of these metals in marine food chains. This information is required for understanding both the biogeochemical cycling of these elements and their potential for exerting toxic effects, including those to humans. ^{210}Pb is used as a tracer of particle flux in marine systems and ^{210}Po has the potential to be used as a specific tracer of particulate organic carbon, and yet very few comparative assimilation and trophic transfer studies have been performed with these elements. We therefore conducted a study to assess these processes in simple marine food chains to provide a mechanistic basis for understanding oceanographic observations on the distributions of these radionuclides.

MATERIALS AND METHODS

The diatom *Thalassiosira pseudonana* and the prymnesiophyte *Isochrysis galbana* were grown in seawater enriched with *f*/2 nutrients (Guillard & Ryther 1962) until the stationary phase, after which they were radiolabeled for 48 h with ^{210}Po and ^{210}Pb; this is sufficient time to uniformly label these cells (Stewart & Fisher 2003a). Microliter additions of these radioisotopes were added to the seawater; the isotopes were in radioactive equilibrium and came from a stock solution in 1 N HNO_3. Microliter volumes of NaOH immediately following the radioisotope addition ensured that the pH of the seawater was not affected by the radiolabeling. The activity of the radionuclide solution was 19.8 kBq l^{-1}.

Aliquots of each of the phytoplankton cell suspensions for a total of 5×10^4 cells ml^{-1} (or about 1 mg dry wt l^{-1}) were then presented to 1000 adult brine shrimp *Artemia* sp. for 56 h. Thus the *Artemia* sp. were presented with a mixed algal diet of contrasting cell types, one a naked flagellate, the other with a siliceous test. The exposure time was sufficient to attain enough

radioactivity in the brine shrimp to follow the subsequent transfer to euphausiids. Because the algal cells were not removed from their radioactive media, *Artemia* sp. were exposed to the radioisotopes in both particulate (13% of total radioactivity) and dissolved (87% of total radioactivity) forms.

Euphausiids *Meganyctiphanes norvegica* were collected using a 200 μm plankton net at 20 m depth at the French JGOFS time series DYFAMED site (43° 25' N, 7° 52' E) in the NW Mediterranean. They were kept in 2 l of unfiltered seawater at 15°C in the dark for 36 h until arrival at the laboratory, at which time each individual was transferred into 400 ml of 0.2 μm filtered seawater, that was changed daily. The 13 experimental euphausiids were starved for 1 d prior to the beginning of the experiment.

Before feeding *Artemia* sp. to *Meganyctiphanes norvegica*, 50 radiolabeled *Artemia* sp. were gamma-counted and alpha-counted for ^{210}Pb and ^{210}Po activity, respectively. ^{210}Pb measurements were performed at 46 keV using a Pharmacia-Wallac LKB gamma spectrometer equipped with a well-type NaI detector. ^{210}Po was plated onto silver discs (Flynn 1968) in the presence of ^{209}Po as a yield monitor, and ^{210}Po activities were determined by alpha spectrometry. The fecal pellets were also collected from the radiolabeled *Artemia* sp. and analyzed for ^{210}Pb and ^{210}Pb activities. Then, batches of 15 *Artemia* sp. were gamma-counted and presented to each euphausiid in separate flasks. At the same time, 15 *Artemia* sp. were placed in filtered seawater under the same conditions, as a control to evaluate the loss of ^{210}Po and ^{210}Pb from the brine shrimp to the dissolved phase during the euphausiid feeding period. The euphausiids fed for 5 to 7 h in the dark at 15°C.

After feeding, the number of *Artemia* sp. remaining in each flask was counted to determine how many had been eaten. The euphausiid was removed, gamma-counted for ^{210}Pb, and placed in another flask with filtered seawater and 5 non-radioactive *Artemia* sp. overnight to purge their guts of radiolabeled food (Fisher et al. 1983b). When the individual euphausiids had finished feeding on the *Artemia* sp., the activities of ^{210}Po and ^{210}Pb were determined for the seawater, the fecal material, and the remaining *Artemia* sp. in each feeding flask (including the *Artemia* sp. control). After gut evacuation, the euphausiids were collected for gamma- and alpha-counting and their total fecal material was also collected. The activity of ^{210}Po and ^{210}Pb in the fecal pellets of the control *Artemia* sp. was normalized to how many *Artemia* sp. survived the feeding and deducted from the total radioactivity in each euphausiid's fecal pellets. The very small (<5% in all cases) amount of ^{210}Po and ^{210}Pb radioactivity on the euphausiids that did not eat *Artemia* sp. was subtracted from the euphausiid counts to exclude uptake from the dissolved phase.

RESULTS

The activity of ^{210}Po and ^{210}Pb in the cellular suspension was measured at the end of labeling and the activity was within 5% of the original activity, indicating that there was no significant loss of radionuclides to volatilization or sorption to flask walls. The uptake and cellular distribution of polonium and lead in the phytoplankton was assumed to follow the established trends previously described by Fisher et al. (1983a) and Stewart & Fisher (2003a). Before being fed to *Artemia* sp., the phytoplankton cells had a ^{210}Po:^{210}Pb activity ratio of 1:1.

After 56 h the ^{210}Pb activity in the *Artemia* sp. appeared stabilized at 22 ± 2.6 Bq individual^{-1} (Fig. 1). ^{210}Po activity was not assessed due to the destructive nature of alpha-sampling, but partitioning between dissolved and particulate phases (*Artemia* sp. in this case) was assumed to be in equilibrium based on uptake kinetics (described in detail in Stewart & Fisher 2003a,b). After feeding, some ^{210}Pb desorbed from the radiolabeled *Artemia* sp., resulting in a ^{210}Pb activity of 12.8 ± 3.2 Bq individual^{-1}. The activity of ^{210}Po on *Artemia* sp. at the end of labeling was 66.5 ± 4.4 Bq individual^{-1}. Thus, the ^{210}Po:^{210}Pb activity ratio in the brine shrimp at the end of the experiment was about 5.2, a value within the range of such ratios measured in natural zooplankton samples.

Of the 13 experimental euphausiids, 4 did not eat *Artemia* sp. during the feeding period. These krill allowed us to calculate the uptake by the euphausiids of radioactivity that desorbed from the brine shrimp into the dissolved phase during feeding. The very small amount of activity (<2% of total activity in each flask) measured in the non-eating euphausiids was subtracted from the total activity of the feeding
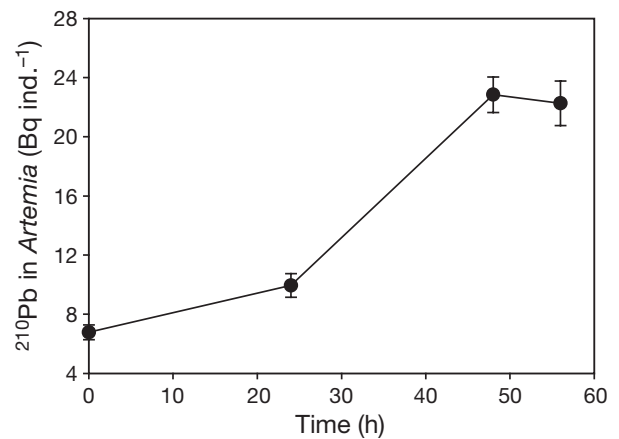

Fig. 1. *Artemia* sp. Uptake of ^{210}Pb (Bq individual^{-1}) over time in *Artemia* sp. feeding on *Thalassiosira pseudonana* and *Isochrysis galbana*

euphausiids to determine the radionuclides taken up by ingestion only. There was no statistical correlation between the size of the euphausiid individuals and the number of *Artemia* sp. eaten. When normalized to weight, the average final ^{210}Po activity was 30.8 ± 0.9 Bq mg^{-1} in the large individuals (300 mg wet wt) and 32.9 ± 0.5 Bq mg^{-1} in the small euphausiids (100 mg wet wt). The ^{210}Po content of *Meganyctiphanes norvegica* increased linearly with the number of *Artemia* sp. that were consumed, in contrast to the minimal accumulation of ^{210}Pb in the euphausiids, regardless of the number of *Artemia* sp. eaten (Fig. 2).

The total activities of ^{210}Po and ^{210}Pb ingested by each euphausiid were calculated by multiplying the number of brine shrimp eaten by the activities of ^{210}Po and ^{210}Pb in each *Artemia* sp. individual. These activities should be taken as upper limits of the exposure to *Meganyctiphanes norvegica*, as it is assumed that there was no loss of polonium or lead from the transfer of *Artemia* sp. to the euphausiids via 'sloppy feeding.' After gut clearance in the euphausiids, the total activity of each *M. norvegica* and its fecal material was calculated and compared to the total ^{210}Po and ^{210}Pb ingested. The percentages of total ingested ^{210}Po and ^{210}Pb remaining in the euphausiids and found in the feces are shown in Fig. 3, and accounted for >70% of the total activity ingested. The remainder (typically 20 to 30%) was assumed to be in the dissolved phase following either desorption from food or soluble excretion from the euphausiids, but was below detection limits due to dilution or incomplete collection of fecal material.

There was a striking difference between the partitioning of the ^{210}Pb and ^{210}Po in the euphausiids. Only
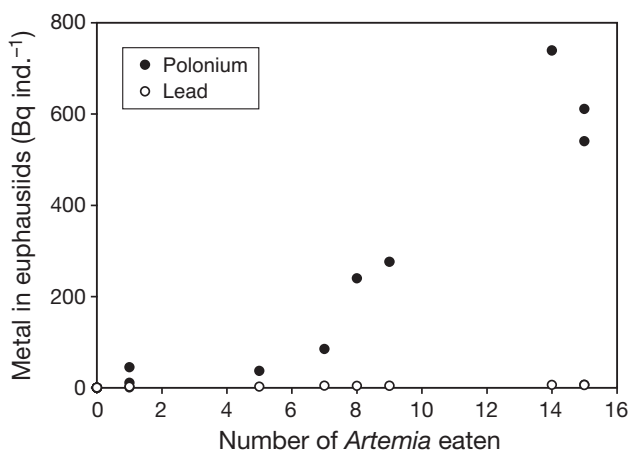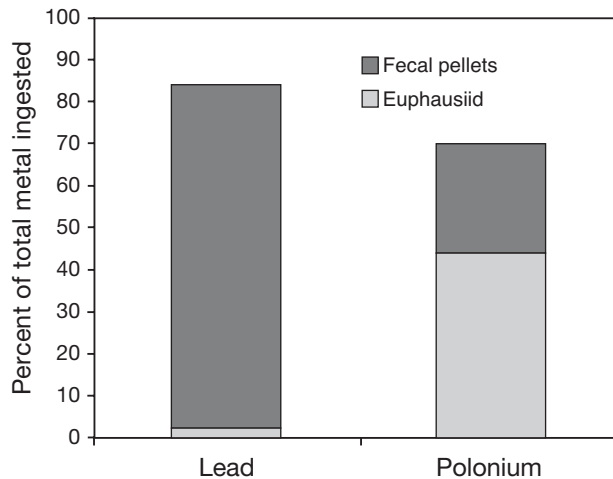

Fig. 3. *Meganyctiphanes norvegica*. ^{210}Po and ^{210}Pb in euphausiids and their fecal pellets as a percent of total radionuclides ingested with labeled *Artemia* sp.

3.5 ± 2.5% of the ^{210}Pb ingested remained in the euphausiids, compared to 84 ± 15% in the fecal pellets. In contrast, approximately 44 ± 19% of the ingested ^{210}Po remained in the euphausiids after gut clearance, with only 26 ± 11% in the fecal pellets. Thus, the assimilation efficiency (AE) of ^{210}Po (calculated as the percent of ingested activity that remained in the euphausiid after gut evacuation) was 12.6 times that of ^{210}Pb in the euphausiids. Given the ^{210}Po:^{210}Pb activity ratio in the *Artemia* sp. after feeding (see next paragraph), we can surmise that the ^{210}Po AE was approximately 5 times higher than the ^{210}Pb AE in *Artemia* sp.

The activity ratio of ^{210}Po:^{210}Pb was calculated in each stage of the experiment, and is shown in Fig. 4. The ratio in the phytoplankton cells was not much higher than the equilibrium solution (1:1) in which they were labeled. The preferential assimilation of ^{210}Po
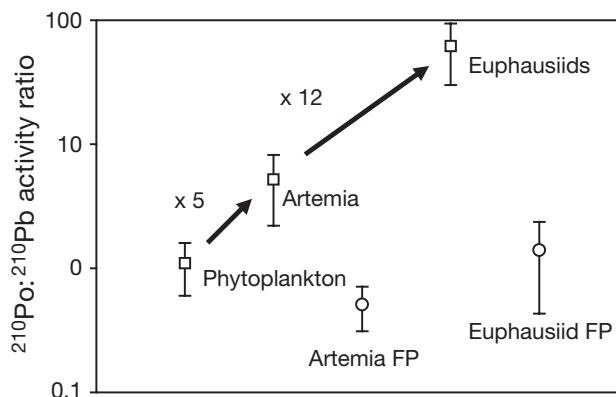

Fig. 2. *Meganyctiphanes norvegica*. Assimilated ^{210}Po and ^{210}Pb in euphausiids in relation to number of *Artemia* sp. consumed during feeding. Relationship between ^{210}Po concentration in euphausiids and number of *Artemia* sp. eaten can be modeled by linear equation with a slope of 40 and a y-intercept of −39.7 (r^2 = 0.88)

Fig. 4. Mean (±1 SD) ^{210}Po:^{210}Pb ratio in phytoplankton, brine shrimp *Artemia* sp., and euphausiids *Meganyctiphanes norvegica* and in *Artemia* sp. and euphausiid fecal pellets (FP). *Y*-axis is in log-scale

over ^{210}Pb resulted in an activity ratio close to 5 for *Artemia* sp., and a ratio of 62 for *Meganyctiphanes norvegica*. The ^{210}Po:^{210}Pb activity ratio in the euphausiid fecal pellets was close to 1, whereas the ratio in the brine shrimp fecal pellets was 0.51. Thus, *Artemia* sp. were about 10 times more enriched in ^{210}Po than in ^{210}Pb relative to their fecal pellets, and *M. norvegica* were about 60 times more enriched in ^{210}Po than in ^{210}Pb.

DISCUSSION

Although a few experiments have demonstrated that ^{210}Po can be transferred efficiently from marine phytoplankton to herbivores (Wildgust et al. 2000, Stewart & Fisher 2003b), we believe that this study is the first to evaluate the trophic transfer of this important radionuclide to carnivorous plankton. We expected that the assimilation efficiency (AE) of ^{210}Po through predation would be as high as, or higher than, the AE via herbivory. Twining & Fisher (2004) found that AEs of some trace metals were higher in copepods feeding on protozoans than in copepods feeding on phytoplankton due to the cellular distribution of the elements in the various cells used in the feeding studies. Wallace & Lopez (1996) determined that changing the metal exposure pathway in prey could affect assimilation in predators, with only metals in certain fractions of prey organisms being available to higher trophic levels. Whereas the assimilation of ^{210}Po by herbivores (Stewart & Fisher 2003b), like other metals (Reinfelder & Fisher 1991), is strongly correlated with its cytoplasmic distribution in phytoplankton food cells, it can be inferred from our euphausiid results that a greater fraction of total ^{210}Po than of total ^{210}Pb was in a labile pool in the *Artemia* sp. prey.

Assimilation efficiencies of ingested radionuclides in our experiments were comparable to values found in crustaceans by other authors. The AE in prawns fed ^{210}Po-labeled food was 35% (Carvalho & Fowler 1994), and the AE was estimated to be at least 5 times higher for Po than Pb in shrimp (Carvalho & Fowler 1993). Furthermore, the turnover of ^{210}Po was much slower than that of ^{210}Pb in shrimp tissue during extended depuration experiments (Carvalho & Fowler 1993). The same results were found in herbivorous copepods, where the AE ranged from 20 to 55% depending on the algal diet (Stewart & Fisher 2003b). The high AEs and low excretion rates of ^{210}Po and the low AEs of ^{210}Pb generally seen in marine zooplankton (Fisher et al. 1995, and this study) explain the high ^{210}Po:^{210}Pb activity ratios (10 to 1000) in invertebrate tissues (Carvalho & Fowler 1994).

The assimilation of ^{210}Po and ^{210}Pb observed in our work should be considered in the context of previous field and biochemical investigations of these radio-isotopes in other marine crustaceans. The uptake of ^{210}Po and ^{210}Pb into larger crustaceans such as shrimp (Cherry & Heyraud 1982, Heyraud et al. 1988, Carvalho & Fowler 1993, 1994), isopods (Stepnowski & Skwarzec 2000) and lobsters (Heyraud et al. 1987, Durand et al. 2002) can be explained by the higher assimilation of ^{210}Po than ^{210}Pb that we found. Some of the highest ^{210}Po concentrations recorded in natural samples are found in the hepatopancreas of these crustaceans (Cherry & Heyraud 1982), possibly due to sulfur-rich, metal-binding proteins found in high concentrations in these organs. Durand et al. (2002) showed that hemocyanin and ferritin from lobsters had extremely high affinities for ^{210}Po, and Stepnowski & Skwarzec (2000) found the highest ^{210}Po activities (>540 Bq kg^{-1} dry wt) and highest ^{210}Po:^{210}Pb activity ratios in the hepatopancreas of Baltic crustaceans. In particular, they found that 60% of the total ^{210}Po in these crustaceans was bound to the cytosolic fraction of the hepatopancreas. These findings are consistent with and provide a mechanistic explanation for our assimilation results.

Further, the contrast that we observed between ^{210}Po and ^{210}Pb in interactions with marine plankton can help explain oceanographic observations on the divergent bioaccumulation patterns and residence times of these radionuclides. The mean ^{210}Po:^{210}Pb activity ratios in *Artemia* sp. (5) and euphausiids (62) are within the range of values measured in various other studies, and can possibly be used as diagnostic tools to evaluate the trophic structure of other pelagic animals. For example, net zooplankton off the Cape of Good Hope had activity ratios between 9 and 25 (Shannon et al. 1970), suggesting that the animals were an assemblage of herbivores and carnivores, or that the animals caught were omnivores feeding opportunistically on a mixed diet. Cherry & Heyraud (1981) reported an average ^{210}Po:^{210}Pb activity ratio of 77 for more than 30 different species of pelagic shrimps from various water bodies, and Carvalho & Fowler (1993) found that the Mediterranean shrimp *Lysmata seticaudata* had an activity ratio of 47. For other invertebrates, ^{210}Po:^{210}Pb activity ratios ranging from 3 to over 100 have been reported (Somasundaran & Hameed 2000, Stepnowski & Skwarzec 2000, Ugur et al. 2002). Our study provides an ecological framework within which ^{210}Po:^{210}Pb ratios can be interpreted.

In considering the implications of our assimilation results, in which ^{210}Po displayed significantly greater assimilation and longer retention than ^{210}Pb in zooplankton, it is expected that ^{210}Po would be recycled biologically in surface waters to a much greater extent

than ^{210}Pb, consistent with oceanographic observations (Nozaki et al. 1997). Further, it would be expected that the residence time of ^{210}Pb in surface waters would be relatively short (Cochran et al. 1983, Masqué et al. 2002), especially in waters in which the zooplankton biomass is high (Fisher & Fowler 1987), as sinking fecal pellets enriched in this isotope retain it for periods long enough to transport it to deep waters (Fisher et al. 1987).

The bioaccumulation of [210]Po in *Meganyctiphanes norvegica* is of interest in part because krill can be an integral part of the marine food chain in the Atlantic, Antarctic and Mediterranean. For example, Tarling et al. (2001) reported that euphausiids dominated the fall plankton community in the Ligurian Sea, where our euphausiids were collected. In addition, as found in our study, 2 distinct cohorts of *M. norvegica* are normally present in the Ligurian Sea: a born-in-the-year group (smaller individuals) and an older (1 yr +) cohort (larger individuals) (Labat & Cuzin-Roudy 1996). Because of the omnivorous nature of euphausiids, this species directly connects the phytoplankton, which are the first point of entry for [210]Po into marine food webs, with larger invertebrates and vertebrates, and forms a link between copepods and other small grazers with higher trophic levels. Thus, while the bioaccumulation of [210]Po by euphausiids is an important step in its cycling in marine systems, it also may influence the extent to which this radionuclide builds up in marine organisms (including krill themselves) that are harvested for human consumption or for animal feed.

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