Experimental Evidence For ^{234}Th Bioaccumulation In Three Antarctic Crustaceans: Potential Implications For Particle Flux Studies

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Abstract

^{234}Th is considered a valuable and useful tracer of oceanic biogeochemical processes occurring over timescales of days to weeks. While the geochemical behaviour of this radionuclide in the marine environment is well known, relatively few studies have explored its interactions with biota. To better understand biologically related ^{234}Th dynamics, bioaccumulation of ^{234}Th from the dissolved phase and its subsequent retention in small Antarctic crustaceans (the isopod *Natatolana oculata* and the amphipods *Orchomenella ultima* and *Uristes stebbingi*) was determined under controlled laboratory conditions. Despite morphological and behavioural differences, all three species displayed comparable concentration factors (CF $\geq$ 80) and very long retention of ^{234}Th (biological half-life not significantly different from infinity). From 16% (isopod) to 49% (both amphipods) of accumulated ^{234}Th was associated with the animal soft parts, which is substantial when compared with reported values for other particle-reactive transuranic elements. The relevance of zooplankton as a potential modulator of ^{234}Th distribution in the water column is discussed in light of these findings. CF-based computations suggest that, for typical zooplankton biomass, biologically-mediated interactions with particle flux models can be neglected. In contrast, in waters with very high crustacean biomass, such as krill schools, ^{234}Th distribution in the water column would be largely determined by these organisms. In such waters the biological compartment should be addressed as it could confound the reliability of vertical particle flux assessment using ^{234}Th as a proxy.

Keywords: Zooplankton, Amphipods, Isopods, ^{234}Th, Particle Fluxes, Southern Ocean

1. Introduction

Over the past few decades, ^{234}Th has been increasingly used to assess particle fluxes, especially particulate organic carbon (POC) out of surface waters (Coale and Bruland, 1985; Eppley, 1989; Buesseler et al., 1992; Charette and Moran, 1999; Cochran et al., 2000). Given the interest in climate change and the potential role of the oceans in sequestering C, this issue has taken on renewed attention (Buesseler et al., 2004). Any oceanographic study that aims to investigate particle fluxes by means of a selected proxy requires the proxy's "biogeochemical" behaviour to be well known. The knowledge of its interactions with biological, geological and chemical compartments represents a *sine qua non* condition for a reliable application of the tracer methodology.

^{234}Th distribution in the water column has been studied throughout the world, from the poles to the tropics (Cochran et al., 2000; Benitez-Nelson et al., 2001; Rutgers van der Loeff et al., 2002), and its chemistry has been investigated extensively (Santschi et al., 1980; Burd et al., 2000; Dai and Benitez-Nelson, 2001; Quigley et al., 2001). In contrast, available information regarding ^{234}Th interactions with organisms is surprisingly sparse. With the exclusion of some studies that have indicated how sinking fecal pellets produced by zooplankton influence vertical flux rates of ^{234}Th (e.g., Krishnaswami et al., 1985; Fisher et al., 1987), only a few studies have dealt with ^{234}Th uptake by biota directly (Ishikawa et al., 2004). Among these, even fewer address aspects of its bioaccumulation (Santschi et al., 1983; Fisher et al., 1987; Ishikawa et al., 2004). Moreover, results of these different investigations vary. For instance, Dunne et al. (2000) suggest that ^{234}Th contamination due to zooplankton accidentally caught in sediment traps (swimmers) is negligible because the ^{234}Th activity:mass ratio measured in swimmers is always much lower than in trapped particles (Dunne et al., 2000). However, other studies have

measured activity values up to 560 dpm ^{234}Th g^{-1} dry wt in zooplankton (Krishnaswami et al., 1985; Coale, 1990), a significant activity:mass ratio when compared to that reported for sinking particles, which typically ranges from 1,000 - 4,000 dpm g^{-1} dry wt (e.g., Coale, 1990; Murray et al., 1996; Rodriguez y Baena et al., unpubl. data). This activity:mass ratio may be species dependent. Recently, significant values of natural ^{234}Th were also reported in different marine benthic organisms, reaching 17 dpm g^{-1} dry wt in barnacle soft parts, 26 dpm g^{-1} dry wt in ascidian liver (Ishikawa et al., 2004) and 284-462 dpm g^{-1} dry wt in red algae (CRIIRAD, 2004). ^{234}Th bioaccumulation is not surprising if one considers the body of literature of experimental studies reporting that related transuranic elements such as ^{237}Pu, ^{241}Am, and ^{252}Cf are accumulated in a wide range of organisms (e.g., Fisher et al., 1983; Carvalho and Fowler, 1985; Fowler et al., 1986; Warnau et al., 1996).

The different applications of ^{234}Th with regards to particle fluxes, and C export studies in particular (e.g., sediment trap calibration, assessment of sedimentation and bioturbation rates or of major component export), merely consider the chemical and geological properties of the radionuclide (Buesseler et al., 1994; DeMaster et al., 1994; Rutgers van der Loeff et al., 2002). These studies generally overlook the biological aspects, most likely due to the lack of available information (Benitez-Nelson and ^{234}Th-Group, 2004). However, when examining the few data reported on ^{234}Th activities in organisms, it is possible that they may contain a significant fraction of ^{234}Th within their tissues and hence play a role in the behaviour of ^{234}Th in the water column.

The objective of our study is to better understand ^{234}Th interactions with marine zooplankton, and then to discuss the relative importance of these interactions in the framework of particle flux studies. This work examines the bioaccumulation of ^{234}Th in

three different species of common, small-sized crustaceans from Antarctica, a region of concern due to its potential influence on climate change through C uptake and sequestration (Feely et al. 2001) and an area where ^{234}Th has been used to understand vertical C fluxes (e.g. Buesseler et al., 2004)

2. Methods

2.1. Organism Collection and Acclimation

During the *R/V Polarstern* expedition ANT XXI/2 (Nov. 17th 2003 – Jan. 18th 2004), baited fish traps were moored for 102 h (from Dec 10th to 14th) on the eastern Weddell Sea shelf (stations PS 65/103 FT and PS 65/104 FT, 70°48.86'S 10°40.81'W; 372 m depth). Among the 20 small-sized crustacean taxa collected (for a complete list, see De Broyer et al., 2005), the three most abundant species were selected for this study: the Cirolanidae isopod *Natatolana oculata* and the Lysianassidae amphipods *Orchomenella ultima* Bellan-Santini and *Uristes stebbingi* Walker. Thirty to fifty individuals of each species were isolated and maintained on-board for 4 weeks in closed circuit 50 L aquaria (natural Antarctic seawater constantly gently aerated; De Broyer et al., 2004).

On completion of the expedition, the organisms were shipped in a 150 L isothermic container to our laboratory in Monaco where they were acclimated for five weeks to laboratory conditions (constantly aerated open circuit 700 L aquarium, 5% water renewal hr^{-1}, salinity: 36 p.s.u., temperature: -1.5 ± 0.3°C, darkness). A subgroup of 10 similar sized individuals of each species (mean individual wet wt: 1.60 ± 0.32 g for *N. oculata*, 0.17 ± 0.04 g for *O. ultima* and 0.15 ± 0.05 g for *U. stebbingi*) was then acclimated for 10 d to experimental conditions (gently aerated, closed circuit 5 L glass aquarium, filtered seawater renewal: every second day, salinity: 36 p.s.u., temperature: -1.5 ± 0.3°C, darkness). In order to keep batches as homogeneous as possible, organisms were not fed

during the 10-d pre-exposure acclimation period nor during experiments. Stress due to starvation is minimal in these species. Indeed, their feeding metabolism has been observed to be very slow, with a gut clearance time longer than 2 weeks (De Broyer and Danis, pers. comm.). This was further confirmed in the batch of remaining, non-tested crustaceans, which survived 8 weeks without feeding and had no observable weight loss or mortality.

2.2. Experimental Procedure

Four days before starting the experiments, ^{234}Th ($T_{1/2}$ = 24.1 d; carrier free) was milked and purified from a ^{238}U/^{234}Th standard solution using two successive ionic exchange steps (Walton and Rocklin, 1990). The stock solution of purified ^{234}Th was kept in 0.1 N HNO_3 (372.2 dpm in 1 mL) to avoid possible loss to bottle walls.

Thirty crustaceans (10 individuals of each of the three species) were individually placed in a 50 mL cylindrical vial of which both the top and bottom were replaced with an 800 µm mesh net to enable free water circulation. Each vial was then placed for 7.5 d in the experimental glass aquarium containing 5 L of natural filtered seawater spiked with ^{234}Th. The seawater was changed and the radionuclide spike was renewed every second day. Radioactivity in the water was checked twice daily, as well as before and after each seawater renewal, in order to determine the time-integrated radionuclide activity (i.e. mean value of all measurements performed over the time period considered). The maximum decrease in seawater radioactivity between two successive seawater renewals was 31 ± 11% and averaged 16 ± 6%. For the entire experimental time course, the time-integrated ^{234}Th activity was 14.7 dpm L^{-1}.

At different times, each individual was carefully transferred to an isothermic counting vial (see below), where it was weighed (wet wt), γ-counted alive and then replaced in the experimental microcosm to determine the radionuclide uptake kinetics. At the end of the 7.5-d exposure period, three individuals from each species were placed for 2 min in 3 successive batches of 10 mL 6N HCl to leach and remove from the cuticle all externally-adsorbed ^{234}Th. The acid concentration and leaching duration were based upon preliminary tests indicating that these conditions allowed for digesting the outer cuticle without damaging the internal soft parts. The three 10 mL HCl solution were then combined into one 30-mL "cuticle-associated ^{234}Th" sample. The two resulting samples (soft tissue-associated ^{234}Th and cuticle-associated ^{234}Th) were directly γ-counted to determine the incorporated/adsorbed ^{234}Th partitioning in the three crustaceans.

The 7 remaining individuals of each species were then placed for 14 d in new, clean 50 mL cylindrical vials in clean flowing filtered seawater (gently aerated, open circuit 50 L aquarium; seawater renewal: 5% hr^{-1}; salinity: 36 p.s.u.; temperature: -1.5 ± 0.5°C, darkness). These individuals were removed at various time periods, γ-counted alive, and then replaced in the experimental microcosm to determine radionuclide loss kinetics. At the end of the depuration period, individuals were subjected to the HCl leach described above to determine the incorporated/adsorbed ^{234}Th partitioning.

^{234}Th radioactivity in seawater and organisms was determined using a high-resolution γ-spectrometry system consisting of three coaxial Germanium (N- or P-type) detectors (EGNC 33-195-R, Intertechnique; 40% efficiency) connected to a multi-channel analyzer and a personal computer employing spectral analysis software (Interwinner 4, Intertechnique). The radioactivity of the samples was determined at 63.3 keV by comparison with known standards of appropriate geometry and was corrected for

background and ^{234}Th radioactive decay. In addition, all radioanalyses were carried out on samples kept in darkness and at the experimental temperature (-1.5°C) in "isothermic counting vials". These counting vials were surrounded by an ice ring, the size of which was previously adapted to allow for maintaining the required temperature during a 90 min counting time. Counting times were adapted to obtain counting rates with relative propagated errors less than 5%, typically 10-30 min for whole organism radioanalysis and 30-90 min for seawater, soft tissue-associated ^{234}Th and cuticle-associated ^{234}Th samples.

2.3. Data Analysis

Uptake of ^{234}Th from seawater was expressed as a change in concentration factors (CF = dpm g^{-1} wet organism divided by the time-integrated dpm g^{-1} seawater) over time. Uptake kinetics in whole individuals were described by either using a single-component first-order kinetic model

$$CF_t = CF_{ss} (1-e^{-k_e t}),$$

where:

CF_t and CF_{ss} represent the concentration factors at time t (d) and at steady-state respectively, and

k_e is the biological depuration rate constant (d^{-1}) (Whicker and Schultz, 1982),

or, if individuals did not tend to reach a steady-state during the exposure time course, by a simple linear regression model

$$CF_t = k_u t,$$

where:

k_u is the regression slope (i.e. rate of increase in CF, d^{-1}).

Linearity of the uptake kinetics expressed as CF was tested using one-way analysis of variance for regression with replication (Zar, 1996).

^{234}Th loss kinetics were expressed in terms of the percentage of remaining radioactivity, i.e. radioactivity at time t divided by initial radioactivity measured in the organisms at the beginning of the depuration period. The kinetics were described by a double-component exponential model

$$A_t = A_{0s}\, e^{-k_{es}\, t} + A_{0l}\, e^{-k_{el}\, t},$$

where

A_t and A_0 are the remaining activities (%) at time t (d) and 0, respectively, and

k_e is the biological depuration rate constant (d^{-1}).

k_e allows the calculation of the radiotracer biological half-life ($T_{b1/2}=\ln 2/k_e$). The "s" subscript refers to a short-lived component (loss of the fraction of radiotracer pool that is weakly associated to the organism) while the "l" subscript refers to a long-lived component (loss of the fraction of the radiotracer pool that is tightly bound in the organism) (Whicker and Schultz, 1982; Warnau et al., 1996).

Constants of the uptake and loss equations and their statistics were estimated by iterative adjustment of the models and Hessian matrix computation, respectively, using the nonlinear curve-fitting routines in the STATISTICA© 5.1 software (StatSoft Inc.; Tulsa, Oklahoma, USA).

3. Results

The three species tested readily accumulated dissolved ^{234}Th from seawater, reaching mean wet wt whole-body CF of 90-140 after a 7.5-d exposure period (Fig. 1). Both

amphipod species were found to initially concentrate ^{234}Th much faster than the isopod, and their uptake rapidly reached a plateau (steady-state equilibrium at CF = 80 for *U. stebbingi* and 101 for *O. ultima*) (Fig.1, Table 1A). In contrast, ^{234}Th uptake in the isopod followed first order linear kinetics over the time course of the experiment (Fig. 1, Table 1A), indicating that uptake from water could take more than several weeks to reach steady-state under natural conditions, and that the mean steady-state CF for *N. oculata* is much higher than the CF = 140 determined at the end of the 7.5-d observation period.

Distribution of the ^{234}Th whole-body activity between exoskeleton and internal soft tissues was determined at the end of the exposure period (Table 2). Both amphipod species were found to incorporate almost 50% of the whole-body ^{234}Th within their soft tissues, whereas the isopod contained a much lower fraction of assimilated ^{234}Th, 16% of the whole-body activity. Assuming that the quasi-constant mean fraction of assimilated ^{234}Th in both species of amphipods (48 and 49% of the whole-body activity) corresponds to a steady-state situation, the lower fraction measured in the isopod may not be indicative of a lower internalization capacity, but rather a slower assimilative process that has not yet reached equilibrium partitioning.

At the end of the exposure period, the organisms were maintained for 14 d in flowing seawater to follow radionuclide loss kinetics (Fig. 2). Due to unexplained mortality in the *O. ultima* amphipod group (6 out of the seven individuals) after 5 d in flowing seawater conditions, loss kinetics could not be established for this species. In the two other groups, ^{234}Th was released following double exponential loss kinetics (Fig. 2, Table 1B). Global R^2 were relatively low (0.20). This was mainly due to the somewhat poor description of the short-lived exponential component of the depuration biokinetics: data acquisition every 24h did not allow making a reliable fit of this component characterized by a very

short estimated biological half-life, $T_{b1/2s}$ = 0.3-1.3 d. In contrast, the long-lived component of the loss kinetics represented the major fraction of the total ^{234}Th radioactivity in whole crustaceans (84% in *U. stebbingi* and 93% in *N. oculata*) and displayed depuration rate constants (k_{el}) which were not significantly different from zero, thus resulting in an extremely long $T_{b1/2l}$ ($\approx \infty$) (Table 1B). This indicates that 84% and 93% of the bioaccumulated ^{234}Th would be very strongly bound to *U. stebbingi* and *N. oculata*, respectively. Although the strong retention capacity of ^{234}Th by both amphipods and isopods is clearly shown, the depuration experiment was conducted for only 14 d, and extrapolation to longer periods of time requires experiments of longer duration.

Body distribution of ^{234}Th was determined in the organisms at the end of the depuration period, including the only surviving *O. ultima* amphipod (Table 2). In contrast to the *O. ultima* individual, which displayed a somewhat higher fraction of incorporated ^{234}Th at the end of the depuration period (65 vs. 48 ± 12%), *U. stebbingi* and *N. oculata* showed very similar (not significantly different) ^{234}Th partitioning between exoskeleton and internal soft tissues at the end of the exposure and depuration periods (Table 2). Overall, these results suggest that both the exoskeleton-adsorbed and soft tissue-incorporated fractions of ^{234}Th are retained with similar efficiencies.

4. Discussion

In a closed system, all the isotopes of a given decay series reach secular equilibrium whereas, in a natural open system, reactivity and transport cause a separation between parent and daughter nuclides. The extent of the resulting disequilibrium may be used to identify and assess the processes responsible for the difference (Rutgers van der Loeff, 2001).

^{234}Th-based particle fluxes are estimated by examining the disequilibrium between this particle-reactive radionuclide and its soluble and conservative parent ^{238}U: the larger the depth integrated deficit of ^{234}Th with respect to ^{238}U, the greater the particle flux necessary to maintain the observed water column activity balance (Cochran et al., 2000; Benitez-Nelson et al., 2001; Rutgers van der Loeff et al., 2002). ^{234}Th export models rely on the premise that the observed water column ^{234}Th/^{238}U disequilibrium is essentially due to particle dynamics (sedimentation/remineralization), corrected for hydrodynamic biases (e.g. Savoye et al., 2005). The aim of our discussion is to critically evaluate whether particle export and hydrodynamic constraints alone are always sufficient to account for the observed imbalance, or whether ^{234}Th removed from, or released to the system by living organisms, could possibly represent a non-negligible feature (Fig. 3).

Despite morphological, size, feeding, and behavioural dissimilarities (De Broyer et al., 2004), all three crustacean species studied in this work exhibited a significant, rapid, and persistent bioaccumulation of ^{234}Th from seawater. Previous experimental studies have also reported that other, more commonly studied particle-reactive transuranic elements were bioaccumulated in a similar manner in different small crustacean species. For example, the pelagic euphausiid *Meganyctiphanes norvegica* and the bentho-pelagic isopod *Cirolana borealis* displayed mean concentration factors (CFs) for ^{237}Pu of 70 and 50, respectively, and 130 and 170 for ^{241}Am (Fowler et al., 1976; Fisher et al., 1983; Carvalho and Fowler, 1985). Likewise, *C. borealis*, *M. norvegica* and the bentho-pelagic shrimp *Lysmata seticaudata* bioaccumulated ^{252}Cf in a relatively narrow range of mean CFs, i.e. 170, 200 and 220, respectively (Aston and Fowler, 1983; Carvalho and Fowler, 1985; Fowler et al., 1986). In addition, similar comparisons led Fisher et al. (1983) to conclude that chitinous surfaces of amphipods and euphausiids have comparable affinities

for ^{241}Am. Although the chemistry of these particle-reactive transuranic elements may differ from that of ^{234}Th, these results provide additional evidence that zooplanktonic crustaceans may display a similar affinity for dissolved ^{234}Th. In contrast, field measurements of the soluble ^{234}Th-parent nuclide, ^{238}U, invariably show ^{238}U activities in biota that are 2 orders of magnitude lower than ^{234}Th (e.g., Fisher et al., 1987; Krishnaswami et al., 1985; Ishikawa et al., 2004). The fact that ^{234}Th and ^{238}U are selectively taken up indicates that any influence of zooplanktonic crustaceans on ^{234}Th distribution in the water column will impact the ^{234}Th/^{238}U disequilibrium and, ultimately, C export estimation.

In order to test the role of the biotic component in ^{234}Th-based particle export models, we coupled our information on ^{234}Th bioaccumulation with previously published data concerning densities and distribution of various crustacean zooplankton. Considering that the accumulation rate is high and the elimination rate is very low relative to the ^{234}Th 24.1-d half-life, the impact of crustacean zooplankton on the distribution of any ^{234}Th activity entering a given parcel of seawater (e.g., input from ^{238}U decay, remineralization) may be approximated by multiplying a CF of 80 (lower estimate between the three calculated ones) by the planktonic crustacean wet biomass within the water body.

In order to evaluate the relevance of the biotic component in ^{234}Th-based particle dynamic models, we still need to clearly define the basis on which its significance may be defined. In this respect, a comparison with the analytical uncertainty on ^{234}Th measurements represents an appropriate and objective criterion. Given that recent ^{234}Th activity measurements have reached a precision of about 5% (Rutgers van der Loeff et al., 2005 and references therein), any bias smaller than this 5% threshold could be considered negligible. As can be seen from Fig. 4, crustacean zooplankton abundance in the water

must reach values of at least 650 mg L^{-1} wet wt in order to significantly impact the ^{234}Th water column distribution. An overview of the related literature indicates that reported biomasses of crustacean plankton in oceans are generally in the range of a few μg L^{-1} wet wt, reaching maxima in the range of tens of μg L^{-1} wet wt during high grazing pressure periods (e.g., Nicol, 1986; Bradford-Grieve et al., 2001; Ashjian et al., 2004). A CF-based computation indicates that these typical biomasses result in a negligible impact of the biotic component on total ^{234}Th distribution (<< 5%).

However, Antarctic krill *Euphausia superba*, described by several scientists as one of the most abundant multicellular species of the planet, reaches large-scale biomass values on the order of mg L^{-1} wet wt (e.g., Gutt and Siegel, 1994; Hewitt et al., 2004). The behaviour of krill further leads them to reach much higher biomass per given area as they aggregate into huge schools (often improperly named "swarms"; Hamner and Hamner, 2000) that may extend over several square kilometers (e.g., Watkins, 2000). Due to nychtemeral vertical migrations, schools remain at depth most of the time during daylight hours and then rise to the surface at night. They can be so concentrated that local densities may reach 25,000 to 64,000 individuals m^{-3}, turning the water red (Hamner et al., 1983; Higginbottom and Hosie, 1989; Hamner and Hamner, 2000; Watkins, 2000). In a water-body characterized by such a high, though rather common, demographic pressure of *E. superba,* up to 64 individuals per liter of seawater may be found. For large individuals, this would represent a wet wt biomass of up to more than 100 g L^{-1}. Consequently, taking into account our ^{234}Th-CF estimate in a water body sustaining such a huge biomass, nuclide bioaccumulation could result in up to 89% of total water-column ^{234}Th being associated with krill. Considering that this euphausiid normally lives in the productive upper 200 m of the water column (Watkins, 2000), one can easily visualize how strongly

the presence of dense krill schools could influence ^{234}Th export models. The analysis of the interactions among different simultaneous dynamic processes (e.g., ^{234}Th uptake in and loss from the organisms, radioactive decay of water- and animal-associated ^{234}Th, ^{234}Th production from ^{238}U decay, export of particle-associated ^{234}Th, etc.) is complex and would require intense mathematical modelling beyond the scope of this paper. Nevertheless, a simple overview of such a situation indicates that in a water body sustaining high krill biomass, a significant part of the dissolved ^{234}Th produced from ^{238}U decay would be taken up by krill (ratio 80:1 relative to seawater), just to replace any decaying animal-associated ^{234}Th. Since krill are not representatively captured by conventional particle collection devices typically used for ^{234}Th studies (i.e. Niskin bottles, *in-situ* pumps, sediment traps) due to their avoidance behaviour (Hamner et al., 1983; Hamner and Hamner, 2000), this uptake would result in an apparent ^{234}Th deficit relative to ^{238}U that could be wrongly interpreted as a *vertical* ^{234}Th export, thereby artificially increasing the export which actually occurs through sinking of particle-associated ^{234}Th.

The impact of krill on ^{234}Th distributions could be considerably cyclically enhanced during certain times of the year. Krill undergo ecdysis regularly throughout their life cycle (Daly, 1990) and ecdysis appears to be synchronised within a krill school (Hamner et al., 1983). Hence, ecdysis would result in the rapid loss of exoskeleton-associated ^{234}Th (roughly 50% of total ^{234}Th body burden), and in the rapid re-adsorption of an equivalent amount of ^{234}Th from seawater on the new cuticle. The combination of these behavioural and kinetic characteristics (ecdysis synchronism and fast ^{234}Th uptake rate) suggests that moulting events would result in a limited (both in time and space), but significant impact on total ^{234}Th distribution in the water column. For instance, for a typical equilibrium

^{234}Th activity in seawater of 2.5 dpm L^{-1}, the presence of a krill school of 15 m thickness, 35 g L^{-1} wet wt density, undergoing a synchronised moulting event, would remove ^{234}Th via new cuticle adsorption to such an extent that it would correspond to an additional ^{234}Th flux of ca. 1500 dpm m^{-2} d^{-1} over the upper 200 m (the mean depth to which krill schools migrate). In such a situation, regardless if a steady-state or a non- steady-state model were used (e.g., Coale and Bruland, 1985; Buesseler et al., 1992), any measured ^{234}Th deficit with respect to ^{238}U would be related to ^{234}Th scavenging onto sinking particles, whereas a large fraction of this apparent flux (up to 87% for a measured ^{234}Th deficit of 0.3 dpm L^{-1} averaged over 200 m) would be due to biosorption on krill. This would result in an overestimate of ^{234}Th export by almost one order of magnitude.

The extent to which bioaccumulation of ^{234}Th by krill will impact the budget of ^{234}Th depends on the kinetics and extent of ^{234}Th adsorption. Indeed, if krill bioaccumulation follows the kinetic pattern observed in amphipods, the equilibrium between dissolved and organism-associated ^{234}Th is reached rapidly (within 2 d), and is stable due to the high ^{234}Th retention capacity in these small crustaceans. Therefore, any impact on total ^{234}Th distribution in the water column would be rather limited in time. In contrast, if krill bioaccumulation kinetics of ^{234}Th follow those observed in the isopod, the bias in ^{234}Th export could be much greater. Indeed, this species accumulated ^{234}Th linearly over a time period (> 7.5 d) that is not negligible compared to the ^{234}Th half-life (24.1 d). In addition, the CF that could be reached in this situation would be much higher than the lower estimate used in CF-based computation (mean ^{234}Th-CF in isopods was already 140 after 7.5 d).

To the best of our knowledge, the effect of a krill school on ^{234}Th deficits has never been observed directly. However, it cannot be in principle excluded that krill school-

related events (such as synchronic moulting) be co-responsible for the important deficit-derived ^{234}Th fluxes (up to 3500 dpm m^{-2} day^{-1}; Buesseler et al., 2001) often reported in the Southern Ocean and commonly ascribed to sinking of biogenic particles.

Although it is clear that biota may impact ^{234}Th fluxes under specific circumstances, one may argue that it may not play a significant role in the estimation of C export. Indeed, the latter is derived by multiplying the ^{234}Th flux by a POC/^{234}Th ratio measured on sinking particles (Buesseler et al., 2005). This ratio should presumably co-vary (increase or decrease) with ^{234}Th distribution in the water column and thus reflect ^{234}Th removal by biota and ^{234}Th reintroduction in the vertical fluxes when animal are dying, thereby compensating for possible biases in ^{234}Th deficit due to biota interactions. However, in the case of krill, which has a lifespan of several years, any uptake of ^{234}Th will remain biota-associated for a long period. This ^{234}Th will not be taken into account in the POC/^{234}Th ratio measurement except, after moulting events (sinking exuviae will partly become a component of the particulate flux). In other words, krill-associated ^{234}Th would generate a systematic positive export bias in depth integrated ^{234}Th profiles.

Although far from being exhaustive, these considerations further indicate that in the occurrence of high density krill schools, ^{234}Th in organisms should be measured as the POC/^{234}Th ratio does not automatically compensate for rapid ^{234}Th-biota interactions. Therefore, organisms and especially data on their biomass should be collected at the same time as the seawater samples for ^{234}Th measurements. However, since krill are not easily caught using conventional systems such as Niskin bottles or plankton nets (avoidance behaviour; see e.g. Hamner et al., 1983; Hamner and Hamner, 2000), the easiest way to meet these requirements would probably be to recommend using *in situ* camera for biomass assessment (e.g., Gutt and Siegel, 1994). In addition, a higher vertical resolution

in POC/^{234}Th ratio determination (viz. at several depths in the euphotic layer) could also be recommended if high zooplankton biomass is suspected in order to correct any increase/decrease in POC/^{234}Th ratio due to variations in available ^{234}Th in upper water column.

It is worth noting that isopods and amphipods displayed very efficient retention of ^{234}Th. Although we do not have direct evidence, this observation is most probably due to the tightness of ^{234}Th adsorption onto crustacean cuticle as well as to intracellular trapping within animal soft parts. Indeed, amino-polysaccharides (e.g., chitin) and associated glycoproteins, which are major constituents of the crustacean cuticle contain many carboxyl, hydroxyl, nitrogen and sulphur residues that are strong ligand-binding sites for cationic metals (e.g., O'Brien et al., 1991; Keteles and Fleeger, 2001; Compère et al., 2002). In particular, Th^{4+} and other actinides such as U^{6+}, Pu^{4+} and Am^{3+} are known to bind very strongly to biogenic chelators such as polysaccharides (e.g. Hirose and Tanoue, 2001; Hirose, 2004, Santschi et al., 2003). On the other hand, dissolved ^{234}Th-related transuranic elements (^{239}Pu and ^{241}Am) and even the much less particle-reactive actinide U (both ^{235}U and ^{238}U), were shown to precipitate as insoluble phosphate in intracellular sphaerocrystal and lysosomes of crabs and crayfish (e.g., Chassard-Bouchaud, 1996).

So far, our estimates of possible biologically-mediated impacts on total ^{234}Th distribution only take into account the bioaccumulation of *dissolved* ^{234}Th, and it should be noted that trophic transfer may also contribute to ^{234}Th uptake in these crustaceans. Even though available information on closely-related, particle-reactive transuranic elements such as ^{237}Pu, ^{241}Am or ^{252}Cf suggest that the contribution of the trophic pathway is likely small (e.g. Fowler et al., 1976; Fisher et al., 1983; Carvalho and Fowler, 1985; Warnau et al., 1996), this contribution should not be excluded without further study

(Fowler and Fisher, 2004). For example, unexpectedly high absorption efficiencies (up to 60%) have been found in at least two crustacean species, viz. the crabs *Carcinus maenas* and *Cancer pagurus*, fed food previously labelled with ^{237}Pu (Fowler and Guary, 1977).

5. Conclusions

In general, our findings suggest that the bias in ^{234}Th flux measurements due to biological interactions is negligible in most environmental situations since the typical zooplankton biomasses reported do not result in significant ^{234}Th removal from the dissolved phase. However, in waters sustaining very high biomass, such as in the Southern Ocean, biological compartmentalization should be addressed as it may confound the reliability of vertical flux assessments using ^{234}Th. Furthermore, possible biologically-mediated interferences with ^{234}Th-flux computations should not be automatically neglected, regardless of the geographic zone being investigated, as high densities of pelagic crustacean populations are not restricted to *E. superba* in the Southern Ocean. In fact, large-scale, aggregative behaviour leading to very high biomasses also occurs in other macrozooplanktonic species such as the northern krill *Meganyctiphanes norvegica* (Nicol, 1986) or the tropical tuna-crab *Pleuroncodes planipes* (Robinson and Gómez, 1998).

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Captions to Figures

Fig. 1. Uptake kinetics of ^{234}Th from seawater in whole crustaceans (mean CF ± SD, *n*=10). Parameters of the equations fitting the data are given in Table 1A.

Fig. 2. Loss kinetics of ^{234}Th in whole crustaceans previously exposed for 7.5 d in seawater (mean % remaining activity ± SD, *n*=7). Parameters of the equations fitting the data are given in Table 1B.

Fig. 3. Diagrammatic representation of a typical vertical profile of ^{234}Th (black dots) vs ^{238}U (white dots) in a 1000 m depth water column, indicating the possible causes for deficit and excess of ^{234}Th relative to its parent nuclide ^{238}U.

Fig. 4. CF-based computation representing the fraction (%) of total ^{234}Th in the water column associated with different biomasses of crustacean zooplankton (g L^{-1} wet wt). The arrow indicates the biomass threshold above which zooplankton is considered to be potentially interfering with particle flux assessment using ^{234}Th as a proxy.

Table 1

Parameters and statistics of the equations best fitting the uptake and loss kinetics of ^{234}Th in whole amphipods and isopods during seawater exposures.

CF_t, CF_{ss}: concentration factor at time t (d) and at steady state, respectively; k_u, k_e: uptake and loss rate constant (d^{-1}), respectively; ASE: asymptotic standard error; R^2: determination coefficient; A_t, A_0: remaining activities (%) at time t (d) and time 0, respectively; s, l subscripts: short- and long-lived component, respectively; $T_{b1/2}$: biological half-life (d). *: $p = 0.02$; **: $p = 0.0005$; ***: $p < 0.00001$

A. Uptake kinetics

Species	Equation	CF_{ss} (ASE)	k (ASE)	R^2
Uristes stebbingi	$CF_t = CF_{ss} (1-e^{-k_e t})$	79.2 (4.8)***	1.977 (0.846)*	0.54
Orchomenella ultima	$CF_t = CF_{ss} (1-e^{-k_e t})$	101.2 (5.1)***	2.111 (0.572)**	0.67
Natatolana oculata	$CF_t = k_u t$		21.18 (0.73)***	0.83

B. Loss kinetics

Equation: $A_t = A_{0s} e^{-k_{es} t} + A_{0l} e^{-k_{el} t}$

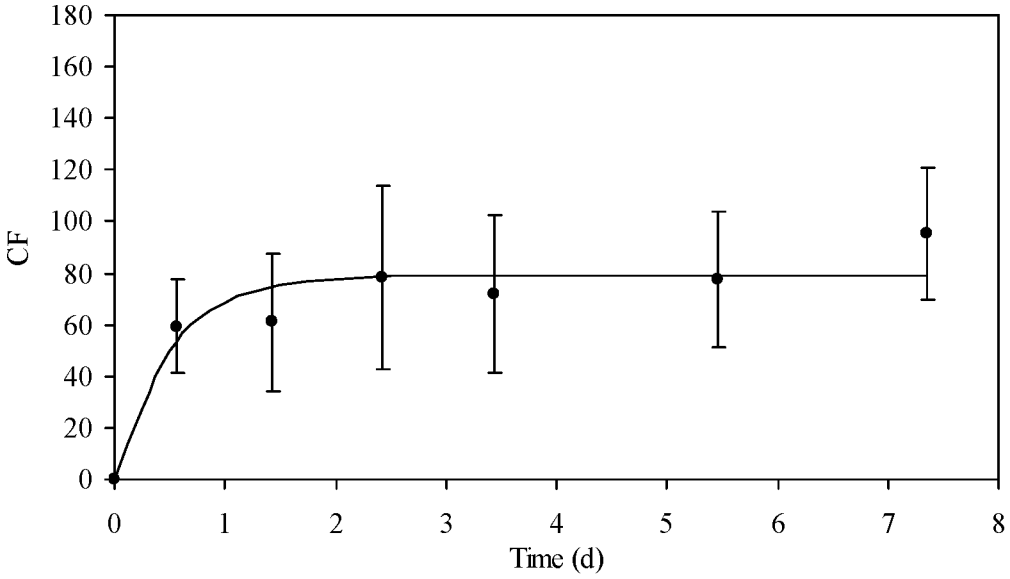
Species	A_{0s} (ASE)	k_{es} (ASE)	$T_{b1/2s}$	A_{0l} (ASE)	k_{el} (ASE)	$T_{b1/2l}$	R^2
Uristes stebbingi	15.9 (11.4)	2.14 (2.55)	0.32	84.1 (9.7)***	0.0042 (0.019)	165	0.20
Natatolana oculata	7.1 (5.8)	0.53 (0.82)	1.31	93.1 (7.4)***	0.00044 (0.0081)	$1.6\ 10^3$	0.20

Table 2

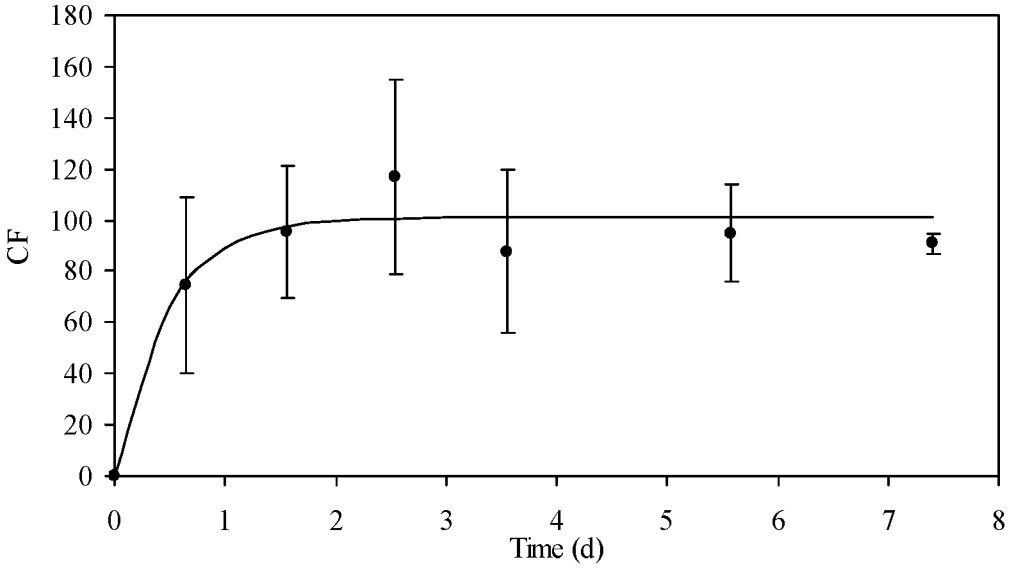
Proportion (mean % ± SD, n = 3) of accumulated ^{234}Th associated with soft tissues of crustaceans exposed via seawater

Species	End of exposure period (7.5 d)	End of depuration period (14 d)
Uristes stebbingi	49 ± 9	49 ± 20
Orchomenella ultima	48 ± 12	65*
Natatolana oculata	16 ± 5	13 ± 4

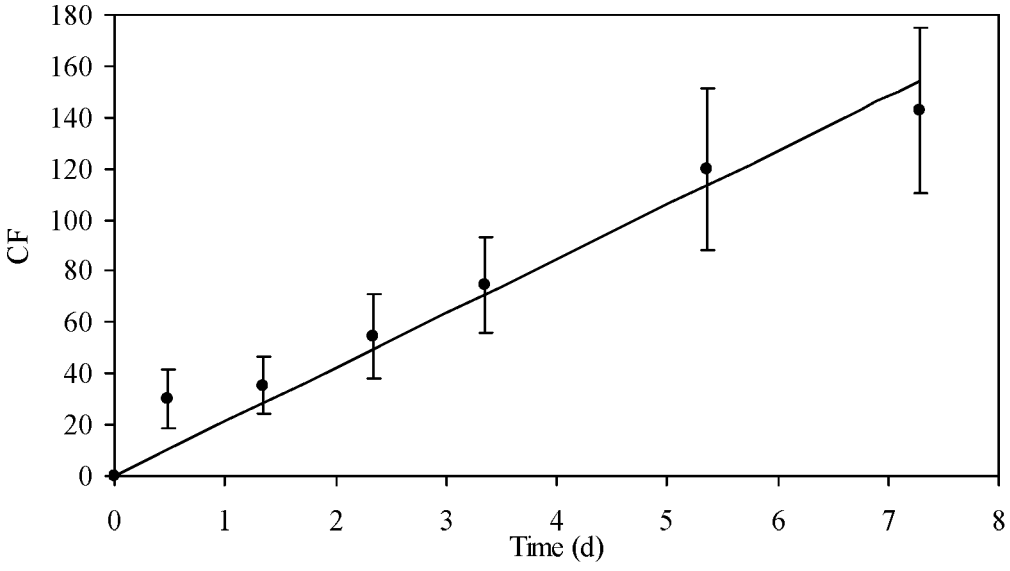
* n=1 (see text: 3. Results)



Uristes stebbingi

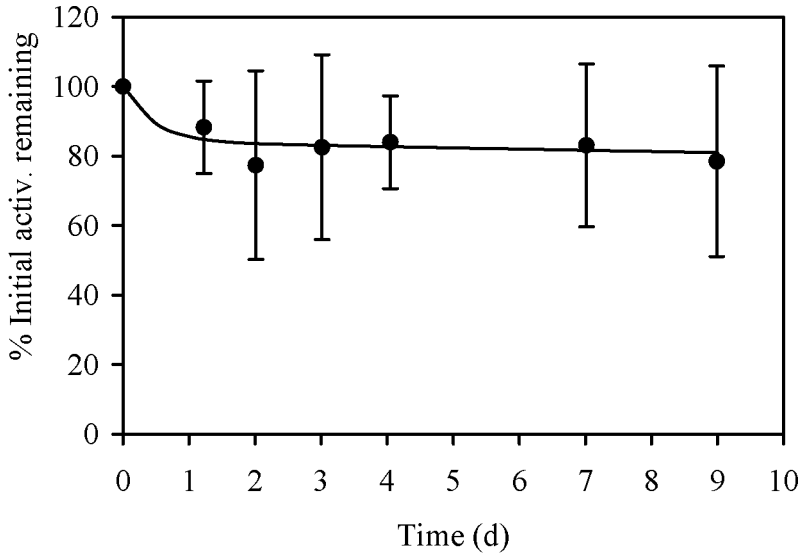


Orchomenella ultima

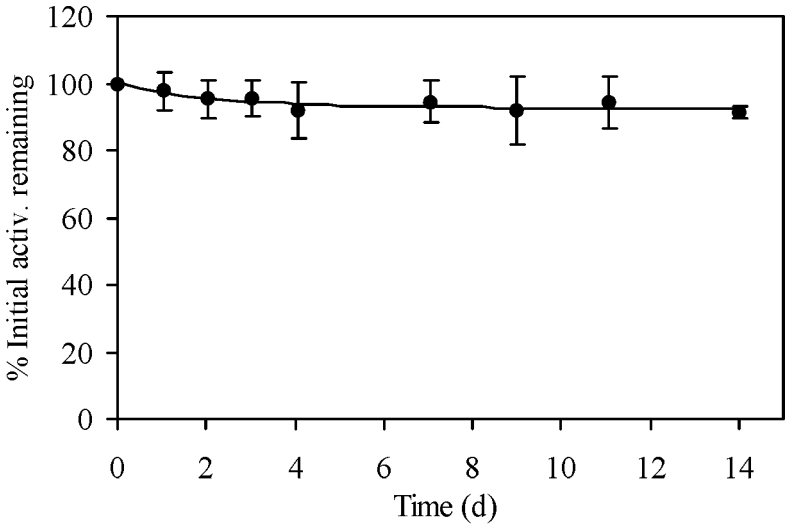


Natatolana oculata

Fig. 1



Uristes stebbingi



Natatolana oculata

Fig 2

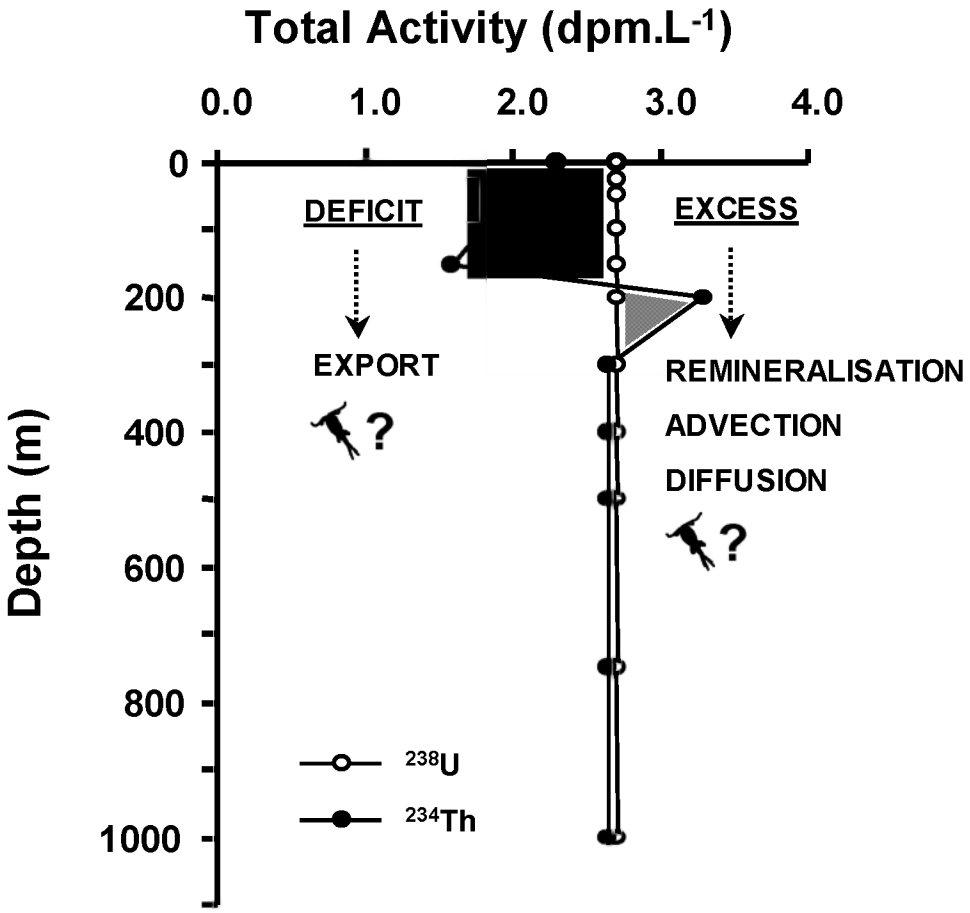

Total Activity (dpm.L-1)
0.0
1.0
2.0
3.0
4.0
0
200
400
600
800
1000
Depth (m)
DEFICIT
EXCESS
EXPORT
?
REMINERALISATION
ADVECTION
DIFFUSION
?
238U
234Th

Fig. 3

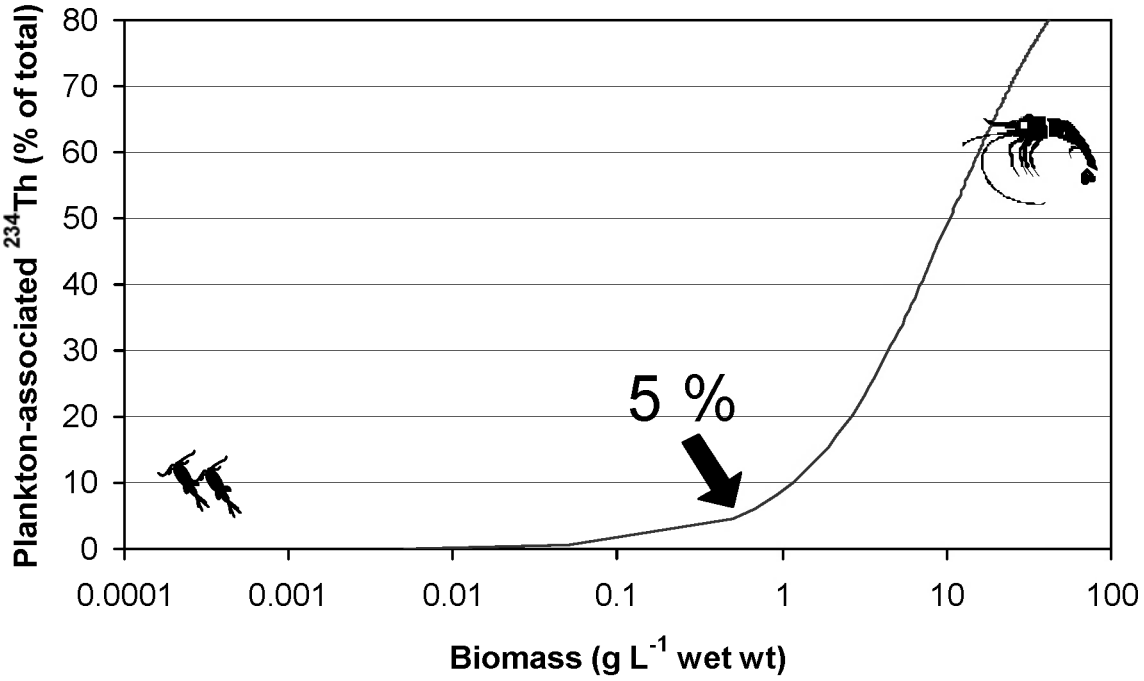

Plankton-associated ^{234}Th (% of total)
80
70
60
50
40
30
20
10
0
0.0001
0.001
0.01
0.1
1
10
100
5 %
Biomass (g L^{-1} wet wt)

Fig. 4