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Effects of Arctic Temperatures on Distribution and Retention of the Nuclear Waste Radionuclides ^{241}Am, ^{57}Co, and ^{137}Cs in the Bioindicator Bivalve *Macoma balthica*

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ABSTRACT

The disposal of radioactive wastes in Arctic seas has made it important to understand the processes affecting the accumulation of radionuclides in food webs in coldwater ecosystems. We examined the effects of temperature on radionuclide assimilation and retention by the bioindicator bivalve Macoma balthica *using three representative nuclear waste components, ^{241}Am, ^{57}Co, and ^{137}Cs. Experiments were designed to determine the kinetics of processes that control uptake from food and water, as well as kinetic constants of loss. ^{137}Cs was not accumulated in soft tissue from water during short exposures, and was rapidly lost from shell with no thermal dependence. No effects of temperature on ^{57}Co assimilation or retention from food were observed. The only substantial effect of polar temperatures was that on the assimilation efficiency of ^{241}Am from food, where 10% was assimilated at 2°C and 26% at 12°C. For all three radionuclides, body distributions were correlated with source, with most radioactivity obtained from water found in the shell and food in the soft tissues. These results suggest that in general Arctic conditions had relatively small effects on the biological processes which influence the bioaccumulation of radioactive wastes, and bivalve concentration factors may not be appreciably different between polar and temperate waters.* © *1998 Elsevier Science Ltd. All rights reserved*

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MATERIALS AND METHODS

Macoma balthica individuals were obtained from intertidal mudflats in South San Francisco Bay, California and acclimated to experimental temperatures (2 or 12°C) and salinity (35‰) for several weeks, during which they were fed unlabeled diatom cultures (*Thalassiosira pseudonana*, clone 3H). Clams were blotted dry and live weights for each individual were determined before use in experiments. Because contaminant uptake and retention differ with body weight in this species (Strong and Luoma, 1981), we used medium-sized animals of relatively uniform weights (0.7–1.5 g).

Exposure to isotopes was examined from both food and dissolved (solute) phases. Uptake from solution, and depuration from both dissolved and food sources were monitored non-destructively with live animals using a large well NaI(Tl) gamma detector interfaced to a Canberra Series 35+ multichannel analyzer. Activities were standardized using geometrically appropriate standards, and counting times were adjusted to yield propagated counting errors of $< 5\%$. Gamma activities of isotopes were measured at 60 keV (^{241}Am), 122 keV (^{57}Co), and 662 keV (^{137}Cs) and were corrected for counting efficiencies, energy spillover between detection windows, and radioactive decay for ^{57}Co ($t_{1/2} = 272$ d). Isotope additions were as microliter quantities in 0.1 N HCl (^{57}Co, ^{137}Cs) or 3 N HNO_3 (^{241}Am), and were neutralized with appropriate quantities of Suprapur NaOH.

For food exposure experiments, cultures of the centric diatom *T. pseudonana* were grown in modified f/2 medium (no EDTA, Cu or Zn) at 16°C using a 14 h photoperiod under cool-white fluorescent lights (170 μEin m^{-1} s^{-1}). Log-phase cultures were harvested by gentle filtration and resuspended (10^5 cells ml^{-1}) in radiolabeling medium containing ^{241}Am (123 kBq l^{-1}), ^{57}Co (123 kBq l^{-1}) and ^{137}Cs (185 kBq l^{-1}). Uptake of the three isotopes by diatoms was followed using the methods described in Fisher *et al.* (1983).

For dissolved exposure experiments, six groups of 6–10 clams were exposed to radiolabeled water (^{241}Am, ^{57}Co, and ^{137}Cs added at the same concentrations as in the diatom labeling medium) for 12 h. The goal of the short exposures was to determine unidirectional influx of the radionuclides into the animals by measuring concentrations at the earliest point that significant uptake was detectable (Neame and Richards, 1972; Luoma, 1977). Measurements of influx kinetics by initial-rate transfer are based on the assumption that radionuclide concentrations in the animal at the time of sampling are small enough for outward transfer to be ignored. This assumption is typically reasonable for trace elements where turnover rates are relatively slow. Unidirectional influx from solution can be a critical kinetic parameter controlling net bioaccumulation (Wang *et al.*, 1996; Luoma and Fisher, 1997). Each group of clams was exposed at the appropriate temperature in 50 ml 0.2 μm-filtered seawater with aeration. The groups of animals were then transferred into unlabeled circulating seawater held in an aerated 20-l aquarium at the appropriate temperatures, and loss of all three isotopes was followed over a 6-week period. These experiments were designed to have negligible recycling of each radionuclide and thus determine rate constants describing loss (efflux). Water was changed every 2–3 days in the depuration containers to reduce isotope recycling, and the clams were fed unlabeled diatom cultures. Food exposure experiments used the same loss protocol following a 4 h feeding of radiolabeled diatoms. For the feeding experiments, six groups of six clams were fed in 100 ml 0.2 μm-filtered aerated seawater in the dark, with radiolabeled diatoms added at 7.7×10^6 cells ml^{-1}.

Cs concentration factors which are ≤ 1, further suggesting that clams would not accumulate this element efficiently from either food or water (I. Stupakoff, C. Gagnon and N. Fisher, unpublished data). The high levels of K^+ and Na^+ present in seawater competitively inhibit Cs^+ uptake by phytoplankton (Avery *et al.*, 1992; Hutchins *et al.*, 1996*a*), preventing substantial buildup of Cs radioisotopes in Arctic marine biota. Even in freshwater, Cs^+ uptake by diatoms is inhibited by low levels of alkali earth metal ions, but in the absence of these competing ions Cs concentration factors in diatoms can reach 10^4 (Fisher, unpublished data). Thus, Cs generally does not accumulate efficiently in marine food chains. This element does, however, display considerable trophic transfer in terrestrial ecosystems of the Arctic through lichen–caribou– human transfer (Hanson, 1967).

Uptake and loss kinetics of isotopes of both soft tissues and shell are important because many predators (e.g. walrus) ingest both parts of this soft-shelled bivalve. However, Arctic temperatures appear to have only minor effects on the individual processes that govern such kinetics. Among the effects of Arctic temperatures, the only substantial effect was that colder temperatures reduced the assimilation efficiency, and therefore the uptake, of ^{241}Am by *M. balthica* from diatom food. Relatively high assimilation efficiencies (AE) have been reported previously for *M. balthica* fed ^{241}Am-labeled diatoms (33–41%, Luoma *et al.*, 1992; Reinfelder *et al.*, in press), especially compared to assimilation by mussels (Wang and Fisher, 1996). The AE at 12°C reported here (26%) for *M. balthica* is similar to the values observed in the earlier experiments at warmer temperatures, while reducing the temperature to 2°C resulted in less than one-half the assimilation. Nearly all of the ^{241}Am ingested at low temperatures was egested; in contrast, temperature had no appreciable effect on assimilation of ^{57}Co.

The present experiments employed relatively short exposures, as well as relatively short depuration periods. Proportionation of elements among assimilated kinetic compartments in marine organisms is dependent upon exposure time (Van Weers, 1973; Cutshall, 1974); therefore the proportion of ingested ^{57}Co in CII, for example, was probably affected by the exposure regime. However, the rate constants of loss should not be affected by exposure time, provided time is sufficient for isotope exchange into all physiological compartments. The physiological turnover rates in these experiments are similar to those observed in many previous studies (Wang *et al.*, 1996) for a number of bivalve species under temperate conditions.

These results suggest that *Macoma* would be less effective as a bioindicator of trophic exposure to highly particle-reactive nuclear waste isotopes such as ^{241}Am (and possibly chemically similar elements as well) than some other common Arctic benthic organisms such as the macroalga *Fucus* (Boisson *et al.*, in press) or sea stars (asteroid echinoderms) which retain as much as 57% of ^{241}Am ingested with food (Hutchins *et al.*, 1996*a*). The somewhat higher AE for ^{57}Co suggests that this species would be a more effective bioindicator of exposure to radioactive activation products (most of which are transition metals) and heavy metals, and this bivalve has been used extensively as a sentinel of metal exposure in temperate habitats (Thomson *et al.*, 1984; Luoma *et al.*, 1985).

In these relatively short-term experiments radioisotope tissue distributions were tightly correlated with source. If differential tissue partitioning after long-term exposures from food and water is also the case, dissections of field-collected *Macoma* could provide valuable information about the importance of dissolved and food pathways in contaminated ecosystems. Our dissection results support earlier work suggesting that isotope concentrations found mostly in the visceral mass can be taken as evidence that the