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Differential bioaccumulation of ^{134}Cs in tropical marine organisms and the relative importance of exposure pathways

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ABSTRACT

Bioaccumulation of ^{134}Cs was determined in 5 tropical marine species: three bivalves (the oysters *Isognomon isognomum and Malleus regula*, and the clam *Gafrarium pectinatum*), one decapod (shrimp *Penaeus stylirostris*) and one alga (*Lobophora variegata*). Marine organisms were exposed to the radionuclides via different pathways: seawater (all of them), food (shrimp and bivalves) and sediment (bivalves). Our results indicate that the studied tropical species accumulate Cs similarly than species from temperate regions whereas retention capacities seems to be greater in the tropical species. Bioaccumulation capacities of the two oysters were similar for all the exposure pathways. The alga, and to a lesser extent the shrimp, concentrated dissolved Cs more efficiently than the bivalves (approx. 14 and 7 times higher, respectively). Assimilation efficiencies of Cs in bivalves and shrimp after a single feeding with radiolabelled food were comprised between 7.0 ± 0.4 and 40.7 ± 4.3%, with a variable retention time (half-life –$T_{b1/2}$– ranging from 16 ± 3 to 89 ± 55 d). Although the clam lives buried in the sediment, this exposure pathway resulted in low bioaccumulation efficiency for sediment-bound Cs (mean transfer factor: 0.020 ± 0.001) that was lower than the two oyster species, which are not used to live in this media (0.084 ± 0.003 and 0.080 ± 0.005). Nonetheless, Cs accumulated from sediment was similarly absorbed (61.6 ± 9.7 to 79.2 ± 2.3%) and retained ($T_{b1/2}$: 37 ± 2 to 58 ± 25 d) for the three bivalves species. Despite the poor transfer efficiency of Cs from food, the use of a global bioaccumulation model indicated that the trophic pathways was the main uptake route of Cs in the bivalves and shrimp. In shelled organisms, shells played a non-negligible role in Cs uptake, and their composition and structure might play a major role in this process. Indeed, most of the Cs taken up from seawater and sediment was principally located on the hard parts of the bivalves and shrimp, with the exception of *G. pectinatum*, where Cs was mainly distributed in the soft-parts.

1. Introduction

Since the 1950s, marine ecosystems were sporadically subjected to the release of radionuclides (such as ^{134}Cs) from industries, nuclear accidents and fallout from nuclear weapon testing (Friedlander et al., 2005). Although this radioactive contamination has tended to decrease (e.g. Ito et al., 2003), it is still a concern in coastal areas receiving radioactive inputs. Marine biota can be directly impacted by waterborne contamination. It was particularly true after the accident that occurred in the civilian nuclear power plant of Fukushima where an important amount of radioactive Cs was released in the marine environment (Bailly du Bois et al., 2012; Chino et al., 2011). After this accident, Cs isotope concentrations increased by up to 10–1000 times over prior levels in coastal waters off Japan (Buesseler et al., 2012).

Studies on Cs accumulation in marine biota have been subjected to many field investigations. These studies provided some clues of Cs accumulation capacities of bivalves and fish (e.g. Kawai et al., 2013; Rowan and Rasmussen, 1994) using metrics such as field concentration factors or ecological half-lives. However, until now,

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mechanisms explaining high accumulation capacities reported for certain marine organisms have yet to be unraveled (Hamada and Ogino, 2012; Buesseler, 2012). Hence, levels reported from the field and questions about contamination pathways or depuration capacities of contaminated organisms need to be investigated further.

Laboratory characterizations of bioaccumulation parameters under controlled conditions are key to better understand the significance of field measurements and some studies already highlighted the importance of some factors (e.g., salinity, temperature, individual size/age) that influence Cs bioaccumulation (Carlson and Erlandsson, 1991; Ke et al., 2000; Topcuoğlu, 2001) or the relative importance of the different exposure pathways (e.g. Metian et al., 2010). Indeed, laboratory studies allow (1) comparing the bioaccumulation capacities of different marine organisms in fairly comparable contamination conditions, (2) informing about food chain transfer, (3) delineating the major uptake pathway(s) through computation of the data, and (4) providing a clear insight of major biological mechanisms that are activated during pollution events. In the past, a series of experimental studies have investigated the bioaccumulation of radio-cesium in different phyla (e.g. Bustamante et al., 2006; Hattink et al., 2009; Warnau et al., 1996a) focusing on food and/or seawater exposures. Some of these works suggested that Cs might be potentially biomagnified through marine food chain (e.g. Mathews and Fisher, 2008; Mathews et al., 2008; Zhao et al., 2001). Although few studies have tested the contribution of sediment in the global accumulation (Børretzen and Salbu, 2009; Bustamante et al., 2006; Metian et al., 2011), limited knowledge about Cs transfer from sediments is available.

Due to the documented influence of temperature on the Cs uptake (Carlson and Erlandsson, 1991; Ke et al., 2000; Topcuoğlu, 2001), bivalves from tropical regions are expected to accumulate more Cs than those from temperate regions. However, limited information is available on radio-cesium bioaccumulation in tropical areas, although previous radioactive contamination events have occured there (e.g. Mittelstaedt et al., 1999). Furthermore, no information is available regarding the relative importance of the uptake pathways in these warmer areas.

In this context, the aim of the present study was to investigate the bioaccumulation of Cs in tropical marine organisms via different exposure pathways (seawater, food, and sediment): the bivalves *Gafrarium pectinatum* (previously called *Gafrarium tumidum*), *Isognomon isognomum* and *Malleus regula*, the Pacific blue shrimp *Penaeus stylirostris* and the brown alga *Lobophora variegata*. When more than one exposure pathways was investigated, we also assessed the relative contribution of these pathways.

2. Material and methods

2.1. Sampling and acclimation

All the organisms examined originated from New Caledonia. The Pacific blue shrimps *P. stylirostris* were obtained from the Ifremer experimental farm (Station d'Aquaculture Ifremer de Saint-Vincent, New Caledonia) in 2002. The clams *G. pectinatum* were collected by seashore fishing in Dumbéa Bay (22°11′2.50″S, 166°24′3.80″E), the two oysters *I. isognomum* and *M. regula* and the brown alga, *L. variegata* were collected by SCUBA diving in Maa Bay (22°12′0.29″ S, 166°19′0.42″ E) in 2002.

All the organisms were then shipped to IAEA-EL premises in Monaco, where they were acclimated to laboratory conditions (open circuit, 400–3000-L aquaria, water renewal: 30% h^{-1}; T°: 25 ± 0.5 °C; salinity: 36 p.s.u.; pH: 8.0 ± 0.1; light/dark cycle: 12 h/12 h) for 2 months prior to experiments. During this period, clams and oysters were fed daily a mixed algal diet (*Isochrysis galbana*, *Heterocapsa triquetra*, *Thalassiosira pseudonana*, *Emiliania huxleyi*; total cell density: 10^4 cells mL^{-1}) and shrimps were fed daily precooked mussels (*Mytilus edulis*).

2.2. Radiotracer and counting

Uptake and depuration kinetics of Cs in organisms were determined using a high-specific activity radiotracer purchased from Isotope Product Lab (^{134}Cs chloride – 0.1 N, $T_{1/2}$ = 2 years). Tracer was counted using a high-resolution γ-spectrometer system composed of 5 Germanium (N- or P-type) detectors (EGNC 33–195-R, Canberra® and Eurysis®) connected to a multi-channel analyzer and a computer equipped with a spectra analysis software (Interwinner® 6). The radioactivity was determined by comparison with standards of known activity and of appropriate geometry. Measurements were corrected for counting efficiency and physical radioactive decay. The counting time was adjusted to obtain a propagated counting error less than 5% (Rodriguez y Baena et al., 2006; Metian et al., 2008).

2.3. Experimental procedure

Independent experiments were carried out to investigate Cs bioaccumulation in three bivalve species, one decapod crustacean and one alga. Depending on the species, one to three different exposure pathways were studied: seawater, food and sediment. Details of the experimental conditions are provided in Table 1. In all experiments, for individual recognition, bivalves were tagged on the shell, alga and shrimp were kept individually in a cylindrical PVC container (160 mm height × 80 mm diameter) covered above and below with 300-μm mesh size net (to allow free water circulation).

2.3.1. Seawater experiments

For each species, ten to twenty two individuals were placed in 70-L glass aquaria (T°: 25 ± 0.5 °C; salinity: 36 p.s.u.; pH: 8.0 ± 0.1, light/dark cycle: 12 h/12 h). Organisms were exposed from 24 d (for the shrimps) to 28 d (for the other species) to the ^{134}Cs radiotracer with a nominal activity of 1–1.6 kBq L^{-1} (added radiotracer was dissolved in 0.45-μm filtered seawater according to the method described by Warnau et al., 1996b). Seawater exposures were realized in close-circuit aquaria constantly aerated. For each experiment, no change in pH was detectable after tracer addition. In order to keep radioactivity in seawater as constant as possible, seawater and spike were daily renewed during the first two weeks, then every second day. Activity of the radiotracer in seawater was checked daily, and before and after each spike renewal in order to calculate time-integrated activities (1.14 ± 0.19 Bq mL^{-1} to 1.62 ± 0.13 Bq mL^{-1}). Immediately before each renewal of seawater and spike, bivalves and shrimps were fed briefly (30 min) with mixed algal diet (10^4 cells L^{-1}) and mussels (*ad libitum*), respectively, in clean – unspiked – seawater. For each experiment, organisms were collected at different time intervals to be whole-body radio-analyzed. For bivalves, some organisms (n = 3) were sacrificed at the same time intervals for dissection and determination of ^{134}Cs distribution between soft-parts and shells. At the end of the exposure period, 3 to 5 individuals of bivalves and shrimps were sacrificed for fine dissection (up to 5 body compartments for shrimp). Each body compartment was then weighed and radio-analyzed and activities of these compartments were summed up in order to get compartments presented in Table 3 and to determine ^{134}Cs body distribution. The remaining animals (n = 7–10 per species) were then placed in non-contaminating conditions (open circuit, water renewal: 50 L h^{-1}, T°: 25 ± 0.5 °C; salinity: 36 p.s.u.; pH: 8.0 ± 0.1, light/dark cycle: 12 h/12 h) for 43 d (shrimp) to 59 d