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Allometric relationship in the bioaccumulation of radionuclides (^{134}Cs & ^{241}Am) and delineation of contamination pathways (food and seawater) in bloody cockle *Anadara senilis* using radiotracer techniques

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ABSTRACT

The uptake and depuration kinetics of ^{134}Cs and ^{241}Am were investigated in the bloody cockle *Anadara senilis* exposed via seawater and food in controlled conditions, using animals of different weight groups in order to assess how their bioaccumulation is affected by allometry and, hence, the individual's age. This study is one of the few experiments investigating bioaccumulation capacities of radionuclides in a West-African bivalve. Results showed that allometric relationships were mainly dependent on the exposure pathway considered. Significant relationships with body weight of bloody cockles were found during the uptake from dissolved phase for both radionuclides; they followed inverse power functions: smaller cockles concentrated both radionuclides more than larger ones. In contrast, radionuclide absorption and assimilation efficiencies from water and food, respectively, did not show any significant relationship with weight: only slight variation was observed between small and large organisms for the retention of ^{241}Am accumulated from food. A bioaccumulation model was used to assess the contribution of each pathway of exposure (food vs. water) in organisms grouped in small and large individuals. We found that, regardless of the size, ^{134}Cs was mainly bioaccumulated through the dietary pathway. In the case of ^{241}Am, the relative contribution of each pathway is weight-dependent: major contribution of dissolved pathway in smaller organisms and the major dietary contribution in larger organisms.

1. Introduction

Radionuclides of cesium and americium are waste products from nuclear-related activities such as nuclear power generation in nuclear power plants or research reactors (Al-Trabulsy et al., 2011; El Mamoney and Khater, 2004). Although nuclear industries in Western Africa are underdeveloped, Ghana operates a 30-kW miniature neutron source reactor (MNSR) and is currently prospecting for the introduction of nuclear power as supplement to its energy mix to boost industrialization and national development (Dampare et al., 2005; Diawuo and Kaminski, 2017). At present time, operational safety performances of nuclear facilities worldwide are trustful. However, the occurrence of accidental releases of radionuclides cannot be disregarded since it may impact the public and the environment, including aquatic systems. Such possibilities are part of the preparedness and emergencies scenarios that are examined prior to the installation of any nuclear premise. Prior and during operations, surveys of environmental occurrence of radionuclides are also required to assess initial situation and to allow detecting and identifying any radionuclide release and contamination.

Among the common approaches carried out to study environmental contamination, the use of bioindicator species has proven to be a relevant and informative approach (e.g. Nielsen et al., 2007). The criteria for selecting an organism to be used as a bioindicator have been described by many authors (e.g. Jones and Kaly, 1996; Phillips, 1977; Rainbow, 1995). The main criterion is to bioaccumulate the target contaminants. Bioindicators should also be preferentially sessile or sedentary organisms, widely distributed and easy to collect all year round. In addition, after selection of a candidate bioindicator, influence of biotic and abiotic factors (such as temperature, salinity or body size) that are likely to affect its ability to bioaccumulate the target contaminants should be characterized (Ke et al., 2000; Wang and Fisher, 1998; Warnau and Bustamante, 2007). Among these factors, organism

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body size, which for bivalves is correlated with age and weight, is known to be of primary importance (Boyden, 1974; Hédouin et al., 2006; Strong and Luoma, 1981; Tang et al., 2017).

Since they fulfill the criteria of relevant bioindicators, bivalves are widely used in biomonitoring programs (Fernández-Tajes et al., 2011; Rainbow, 1995; Yusof et al., 2004; Zuykov et al., 2013). In Ghana, the bloody cockle *Anadara senilis*, a seafood locally consumed due to high nutrient content (Gabriel et al., 2011), is a good candidate bioindicator since it is long-lived, abundant and easy to identify in the field (Mirsadeghi et al., 2013; Sarkar et al., 1994, 2008; Theng et al., 2004). Some studies have already highlighted the ability of this species to bioaccumulate trace elements, including radionuclides such as ^{210}Po (Theng et al., 2004), which can potentially expose human consumers to contamination by ingestion.

The main objectives of the present study were to (1) assess the bioaccumulation capacity of the bloody cockles *Anadara senilis* for two radionuclides (^{134}Cs and ^{241}Am) when exposed via seawater and through the food, (2) assess the relative contribution of dissolved vs. trophic exposure pathways to the whole-body radionuclide bioaccumulation, and (3) describe the possible influence of the body weight of the bloody cockles *A. senilis* on the bioaccumulation of these radionuclides in order to select the most appropriate individual weight range to be used in biomonitoring studies.

2. Materials and methods

2.1. Origin and acclimation of bloody cockles

Bloody cockles of different sizes and weights (shell length: 21–63 mm, whole-body wet weight: 5–75 g) were hand-picked from the Narkwa lagoon located in the Central region of Ghana. The cockles were transported to the premises of the Environment Laboratories of the IAEA in the Principality of Monaco, where experimental work was carried out. Upon reaching the laboratory, cockles were cleansed of adhering sediments, sorted into different weight batches and held in a 140-L aquarium containing 0.45 µm-filtered seawater. The cockles were then acclimated to experimental conditions in a constantly aerated, open-circuit aquarium (salinity: 38 p. s.u.; water renewal rate: 50 L h^{-1}; temperature: 27 ± 0.5 °C; pH: 8.0 ± 0.1; light/dark cycle: 12 h/12 h depicting natural light conditions using fluorescent tubes) for two months prior to experiments. During the acclimation period, the bloody cockles were fed daily a phytoplankton diet (*Isochrysis galbana* and *Skeletonema costatum*). Observed mortality was limited to few individuals.

2.2. Experimental procedures

2.2.1. Radiotracers and counting

Uptake and depuration kinetics of radionuclides in bloody cockles were determined using high-specific activity radionuclides (^{134}Cs as chloride in H_20, $t_{1/2}$ = 2 years; ^{241}Am as nitrate in 1 M HNO_3, $t_{1/2}$ = 433 years) purchased from Czech Metrological Institute and Isotope Products. The radioactivity of Cs and Am in water, cockles and phytoplankton were measured using a high-resolution γ-spectrometry system equipped with five Germanium - N or P type - detectors (EGNC 33–195-R, Canberra® and Eurysis®) connected to a multi-channel analyzer and a computer employing spectral analysis software (Interwinner 6, Intertechnique). To assure quality of radioanalyses, standards were used. These standards were consisting of "phantoms" for each of the types of sample used (liquids and three sizes of bloody cockles), spiked with known radionnuclide activities in order to get the appropriate density and geometry (Cresswell et al., 2017). To briefly describe the preparation of the "phantoms" of bloody cockles, three different size standards were done, covering the whole range of sizes, using empty shells filled with imbibed (2 M HCL) paper towel, spiked with known concentrations of radiotracers and then closed and placed in a counting box identical to the one used to count the living bloody cockles. The position of "phantoms", defined for the calibration of our detectors, and living bloody cockles was kept the same during the experiment in order to ensure a constant geometry. Corrections were made for background and physical radioactive decay. The counting time was adjusted to obtain a propagated counting error less than 5% (Rodriguez y Baena et al., 2006).

2.2.2. Seawater exposure

Bloody cockles of different weights/sizes (7.51–74.17 g, 23–63 mm of length, 16–40 mm of width, 20–52 mm of height; n = 43) were placed in a 70-L aquarium (constantly aerated, closed-circuit; salinity: 38 p. s.u.; temperature: 27 ± 0.5 °C; pH: 8.0 ± 0.1; light/dark cycle: 12 h/12 h) and exposed for 28 d to dissolved radiotracers in 0.45-mm filtered seawater (1 Bq ^{134}Cs mL^{-1} and 0.2 Bq ^{241}Am mL^{-1}). Seawater and spikes were renewed daily for 5 days, then every second day in order to keep exposure activities as constant as possible. The radioactivity in seawater was checked before and after each spike renewal to yield a time integrated activity of 0.94 ± 0.09 Bq mL^{-1} and 0.13 ± 0.08 Bq mL^{-1} respectively for ^{134}Cs and ^{241}Am. Prior to the renewal of seawater and spike, the cockles were fed for 1 h on a mixed diet of *I. galbana* and *S. costatum* (5 10^4 cells mL^{-1}) in clean seawater in order to avoid ingestion of radiotracer via the food. Twenty tag-identified bloody cockles from two weight classes were collected at different time intervals. Class "small" had an average weight of 9.8 ± 2.2 g (Mean ± SD; with a length of 26.5 ± 2.9 mm, width of 20.6 ± 2.3 mm, height of 25.0 ± 2.4 mm, n = 10) and class "big" had an average weight of 36.3 ± 6.7 g (with a length of 43.4 ± 3.3 mm, width of 31.0 ± 2.6 mm, height of 38.6 ± 2.8 mm; n = 10) were collected at different time intervals and were whole-body radioanalysed alive in order to follow the uptake kinetics of the radiotracers. Prior to radioanalysis, radiolabelled cockles were rinsed in uncontaminated seawater for 2 min to remove loosely bound radionuclides. At the end of the 28-d exposure period, all the bloody cockles were radioanalysed and then placed in clean water in the same conditions for 35 d and 10 tag-identified individuals were regularly radioanalysed alive in order to follow the depuration kinetics of the radiotracers. During the 35-d depuration period, non-exposed individuals were used as controls of possible tracer recycling through seawater. Bloody cockles were fed daily with non-radiolabeled *I. galbana* and *S. costatum* (5 10^4 cells mL^{-1}).

2.2.3. Dietary exposure

Transfer of radionuclides (^{134}Cs and ^{241}Am) to bloody cockles through their diet was studied using the Haptophyceae *I. galbana*. Phytoplankton cells were exposed to 10 Bq ^{134}Cs mL^{-1} and 2 Bq ^{241}Am mL^{-1} during their exponential growth phase (5 d). After that period, the phytoplankton medium was filtered (1-µm mesh size; Osmonic® filters) and filters were rinsed with non-contaminated seawater. Since the ingestion rate (IR) is dependent of the weight of the organisms, bloody cockles of two weight classes had been placed into two different 20-L aquaria (closed-circuit; same parameters as previously described), before the dietary experiment. Class "small" had an average weight of 12.20 ± 1.96 g (Mean ± SD; with a length of 29.8 ± 2.9 mm, width of 22.3 ± 2.1 mm, height of 27.1 ± 2.9 mm; n = 9) and class "big" had an average weight of 28.77 ± 5.07 g (with a length of 40.9 ± 2.3 mm, width of 29.9 ± 2.3 mm, height of 38.0 ± 4.9 mm; n = 7). Bloody cockles were single-fed with radiolabeled phytoplankton for 2 h in closed-circuit conditions by resuspension of the filtered cells to a density of 5 10^4 cells mL^{-1} to avoid pseudofeces production by the bivalves and thus to avoid the loss of a part of the ingested radionuclides prior to the first analysis (Kuranchie-Mensah et al., 2016; Pouil et al., 2015). After the feeding period, the radiolabelled cockles were rinsed in a non-radioactive free flowing seawater aquarium for 2 min to remove possibly adhering radiolabeled phytoplankton, pat dry with absorbent paper and subsequently γ-counted