

Characterization of ^{241}Am and ^{134}Cs bioaccumulation in the king scallop *Pecten maximus*: investigation via three exposure pathways

Marc Metian^{a,b}, Michel Warnau^a, Jean-Louis Teysseyé^a, Paco Bustamante^{b,*}

^a International Atomic Energy Agency, Marine Environment Laboratories, 4, Quai Antoine 1er, MC-98000, Principality of Monaco, Monaco

^b Littoral Environnement et Sociétés (LIENSs), UMR 6250, CNRS-Université de La Rochelle, 2 rue Olympe de Gouges, F-17042 La Rochelle Cedex 01, France

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ABSTRACT

In order to understand the bioaccumulation of ^{241}Am and ^{134}Cs in scallops living in sediments, the uptake and depuration kinetics of these two elements were investigated in the king scallop *Pecten maximus* exposed via seawater, food, or sediment under laboratory conditions. Generally, ^{241}Am accumulation was higher and its retention was stronger than ^{134}Cs . This was especially obvious when considering whole animals exposed through seawater with whole-body concentration factors (CF_{7d}) of 62 vs. 1, absorption efficiencies (A_{01}) of 78 vs. 45 for seawater and biological half-lives ($T_{b1/2}$) of 892 d vs. 22 d for ^{241}Am and ^{134}Cs , respectively. In contrast, following a single feeding with radiolabelled phytoplankton, the assimilation efficiency (AE) and $T_{b1/2}$ of ^{134}Cs were higher than those of ^{241}Am (AE: 28% vs. 20%; $T_{b1/2}$: 14 d vs. 9 d). Among scallop tissues, the shells always contained the higher proportion of the total body burden of ^{241}Am whatever the exposure pathway. In contrast, the whole soft parts presented the major fraction of whole-body burden of ^{134}Cs , which was generally associated with muscular tissues. Our results showed that the two radionuclides have contrasting behaviors in scallops, in relation to their physico-chemical properties.

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1. Introduction

During the last sixty years, human activities have resulted in various degrees of contamination of the world's seas and oceans with anthropogenic radionuclides (Friedlander et al., 2005). Although this contamination has tended to decrease (e.g. Toshimichi et al., 2003), it is still a major concern in coastal areas receiving radioactive inputs mainly from industries, nuclear accidents and fallout from nuclear weapon testing and use. Consequently, monitoring programs were established worldwide to monitor the levels of those radionuclides in the marine environment (Nielsen et al., 2007). Generally, surveys are based on the analysis of seawater, sediments and biological samples (e.g. Fegan et al., 2010). Overall, biomonitoring programs present the advantages 1) to reveal the bioavailability of the considered contaminants and 2) to magnify their levels above the analytical detection limits.

The use of marine organisms for monitoring radionuclide contamination is well established (Valette-Silver and Lauenstein, 1995; Burger et al., 2007; Thébault et al., 2008). In order to

understand field measurements, the characterization of bioaccumulation parameters and/or investigations on the relative importance of the different exposure pathways has been carried out for several radionuclides (Ke et al., 2000; Wang et al., 2000; Baines et al., 2005; Borretzen and Salbu, 2009). Beside mussels, other bivalve species are used to a lesser extent in biomonitoring programs among which scallops appear of great interest as they accumulate trace elements strongly from their environment (Bryan, 1973; Bustamante and Miramand, 2005; Metian et al., 2008a, 2009a; Pan and Wang, 2008). Scallops have also been reported to efficiently concentrate natural and anthropogenic radionuclides such as ^{241}Am , ^{137}Cs , ^{210}Po , ^{238}Pu , ^{239}Pu and ^{90}Sr in their tissues (Miramand et al., 1991; Nonnis Marzano et al., 2000; Bustamante et al., 2002). As for some metals (Brooks and Rumsby, 1965; Chouvelon et al., 2009; Bustamante and Miramand, 2004), scallops sometimes display higher bioaccumulation capacity for ^{137}Cs than other filter-feeders such as oysters and mussels occurring in the same areas (JCAC, 2002). Within the Pectinid family, current knowledge on concentrations of anthropogenic radionuclides is limited to field measurements (Bustamante et al., 2002; Nonnis Marzano et al., 2000; Miramand and Germain, 1986), and to waterborne exposure experiments using Am (Miramand and Germain, 1986; Miramand et al., 1991). The importance of other contamination pathways in the bioaccumulation process of Am and

* Corresponding author. Tel.: +33 546 507 625; fax: +33 546 458 264.

E-mail addresses: metian@hawaii.edu (M. Metian), paco.bustamante@univ-lr.fr (P. Bustamante).

Cs in scallops is not known although sediment and food have been considered as possible sources of Am and Cs following analyses carried out on scallops from the field or after laboratory studies (Miramand and Germain, 1986; Nonnis Marzano et al., 2000) including a histo-autoradiography approach (Miramand et al., 1991). Overall, sediment is considered as a major vector of transuranic elements, such as Am, to biota (Miramand et al., 1982; Bustamante et al., 2006; Ryan, 2002). In comparison, Cs transfer from sediments appears relatively limited (Bustamante et al., 2006; Borretzen and Salbu, 2009). Recently, food has been demonstrated as a major pathway for metal bioaccumulation in scallops (Metian et al., 2009a,b). Therefore, it appears necessary to determine experimentally the bioaccumulation of radionuclides in scallops via seawater, food and sediments in order to better understand the relative contribution of these three pathways of exposure.

The aim of this study was thus to determine the kinetics of uptake and depuration of ^{241}Am and ^{134}Cs in a typical pectinid from European waters, the king scallop *Pecten maximus*, following its exposure to radiolabelled seawater, food or sediment. The radionuclides were selected for their contrasting characteristics in seawater (particle-reactive Am and soluble Cs). There is a particular interest to study a transuranic and radiocesium in *P. maximus* since the geographic distribution of this species (European North-Atlantic coasts) coincides with areas subject to direct inputs from the nuclear retreatment plants of Dounreay (Scotland – facility closed in 1996), La Hague (France) and Sellafield (England), which affect the Norway where *P. maximus* is cultured (Bergh and Strand, 2001).

2. Materials and methods

2.1. Sampling

In spring 2004 and 2005, seventy king scallops *P. maximus* were collected on the French Atlantic coast (Pertuis Breton, Charente-Maritime) by SCUBA diving. They were carefully transported to IAEA-MEL premises in Monaco and were acclimated to laboratory conditions for 4 weeks (constantly aerated, open-circuit 800 l aquarium; flux: 501 h^{-1} ; salinity: 36 p.s.u.; temperature: $17 \pm 0.5\text{ }^\circ\text{C}$; pH: 8.0 ± 0.1 ; light/dark cycle: 12 h/12 h) prior to experimentation. During this period, scallops were fed daily an algal mixed diet (*Isochrysis galbana*, *Skeletonema costatum*).

2.2. Radiotracer and counting

Uptake and depuration kinetics of ^{241}Am and ^{134}Cs in scallop were determined using high-specific activity radiotracers purchased from Isotope Product Lab (^{241}Am nitrate – 0.1 N, $T_{1/2} = 433$ years; ^{134}Cs chloride – 0.1 N, $T_{1/2} = 2$ years). Tracers were counted using a high-resolution γ -spectrometer system composed of four 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).

2.3. Seawater exposure

Twenty five *P. maximus* (average weight \pm SD: $208 \pm 46\text{ g}$) were placed in a 70-l glass aquarium (constantly aerated, closed-circuit aquarium; salinity: 36 p.s.u.; temperature: $17 \pm 0.5\text{ }^\circ\text{C}$; pH: 8.0 ± 0.1 ; light/dark cycle: 12 h/12 h) and simultaneously exposed for 7 d to

^{241}Am and ^{134}Cs dissolved in $0.45\text{ }\mu\text{m}$ filtrated seawater (0.3 and 1.4 kBq l^{-1} , respectively). No change in pH was detectable after the tracer addition. Spiked seawater was renewed twice a day during the first two days and then daily in order to keep radioactivity in seawater constant. The ^{241}Am and ^{134}Cs in seawater was checked before and after each spike renewal, yielding time-integrated activities of $0.13 \pm 0.09\text{ kBq l}^{-1}$ and $1.23 \pm 0.03\text{ kBq l}^{-1}$ respectively.

Nine tag-identified scallops were collected at different time intervals and were whole-body radioanalyzed alive (same identified individual each time). At the end of the 7 d exposure period, five scallops among the twenty-five were randomly selected, sacrificed and dissected. Shell, digestive gland, kidneys, gills, gonad, mantle, intestine, adductor muscle and the remaining soft tissues were separated and radioanalyzed in order to assess the radionuclide body distribution. The remaining scallops were then placed in uncontaminated conditions (constantly aerated, open-circuit aquarium; flux: 501 h^{-1} ; salinity: 36 p.s.u.; temperature: $17 \pm 0.5\text{ }^\circ\text{C}$; pH: 8.0 ± 0.1 ; light/dark cycle: 12 h/12 h) for 36 d and the nine tag-identified individuals were regularly radioanalyzed alive in order to follow the depuration of ^{241}Am and ^{134}Cs from the scallops. Three non-exposed individuals were introduced into the aquarium in order to control possible tracer recycling from the contaminated scallops. During the 36 d depuration period, scallops were fed daily with *S. costatum* and *I. galbana* ($5 \times 10^4\text{ cells ml}^{-1}$). At the end of the depuration period, four contaminated scallops were collected and dissected into several body compartments as previously described.

2.4. Food exposure

The Bacillariophyceae *S. costatum* was used to study the transfer of ^{241}Am and ^{134}Cs to scallops through their diet. Phytoplankton cells were exposed to $4.5\text{ kBq }^{241}\text{Am l}^{-1}$ and $7\text{ kBq }^{134}\text{Cs l}^{-1}$ during their exponential growing phase (10 d). After that period, the phytoplankton medium was filtered ($1\text{ }\mu\text{m}$ -mesh size; Osmonic filters), and the phytoplankton cells re-suspended in a 70 l aquarium (constantly aerated, closed-circuit aquarium; salinity: 36 p.s.u.; temperature: $17 \pm 0.5\text{ }^\circ\text{C}$; pH: 8.0 ± 0.1 ; light/dark cycle: 12 h/12 h) at a cell concentration of $5 \times 10^4\text{ cells ml}^{-1}$ to avoid pseudofeces production by the scallops. Nine *P. maximus* (average weight \pm SD: $199 \pm 32\text{ g}$) had been placed in the aquarium for one week before the feeding experiment. Scallops were then allowed to feed on radiolabelled *S. costatum* for 2 h. After the feeding period, all scallops were γ -counted and flowing seawater conditions (50 l h^{-1}) were restored in the aquarium. Individuals were then whole-body γ -counted alive at different time intervals to follow the depuration kinetics of both elements. Three non-exposed individuals were introduced into the aquarium in order to control possible tracer recycling from the contaminated scallops. During the 21 d depuration period, scallops were fed daily with *S. costatum* and *I. galbana* ($5 \times 10^4\text{ cells ml}^{-1}$). Four contaminated individuals were randomly collected after 21 d and dissected to determine the radionuclide distribution among the different body compartments (shell, digestive gland, kidneys, gonad, mantle, intestine, adductor muscle and the remaining soft tissues).

Radiolabelled *S. costatum* did not provide significant contamination of the scallops with ^{134}Cs . Therefore, another phytoplankton species was used to study the trophic transfer of this radionuclide. To this end, the Haptophyceae *I. galbana* was used following the same method as previously described for *S. costatum* except that phytoplankton cells were exposed to ^{134}Cs over 7 d (growing phase of *I. galbana*). Six *P. maximus* (average weight \pm SD: $127 \pm 14\text{ g}$) were exposed and whole-body γ -counted alive at different time of the depuration experiment (21 d). Four individuals were dissected at the end of the depuration period to determine the

^{134}Cs distribution among the different body compartments (as described above).

2.5. Sediment exposure

Since *P. maximus* lives buried into the sediment, ^{241}Am and ^{134}Cs exposure through sediment was assayed. Sediment was collected in Wimereux (North-Atlantic coast of France). Sediment grain size distribution was measured on a Mastersizer micro and the evaluation of the dry/wet weight ratio was calculated after freeze drying in a LABCONCO Freezone18. Aerated sediment (9 kg) was placed in a plastic container, spiked with ^{241}Am (8 kBq) and ^{134}Cs (13 kBq) for 6 d with constant agitation, then used to form a homogeneous sediment layer of 4 cm height in a 20 l aquarium. Weakly bound radioisotopes were allowed to leach overnight under flowing seawater (50 l h^{-1}). Ten *P. maximus* (average weight \pm SD: $118 \pm 5\text{ g}$) were then placed for 13 d in the aquarium (open-circuit; parameters as previously described) and six tag-identified individuals were regularly whole-body radioanalyzed alive. Sediment samples were also regularly collected and γ -counted to verify that the radiotracer activities in sediment remained constant. Activity of ^{241}Am and ^{134}Cs in sediment was constant across the exposure period (8.2 ± 0.8 and $13.1 \pm 3.0\text{ Bq g}^{-1}$ wet wt, respectively). At the end of the exposure period, four scallops were collected, dissected (shell, digestive gland, kidneys, gills, gonad, mantle, intestine, adductor muscle and the

remaining soft tissues), weighed and radioanalyzed to determine the radionuclide body distribution. The remaining six scallops were placed in non-contaminating depuration conditions for 31 d (in a new 20 l glass aquarium with clean sediment under flowing seawater, 50 l h^{-1} , daily feeding on *S. costatum* and *I. galbana* at $5 \times 10^4\text{ cells ml}^{-1}$) and regularly γ -counted. Radioactivity in sediment was regularly checked in order to ensure that no tracer recycling occurred in the sediment. Although no radioactivity was detected, the whole sediment was renewed after 1 week. After 31 d of depuration, four scallops were collected and dissected as described above to determine body distribution of ^{241}Am and ^{134}Cs .

2.6. Data analysis

Uptake of the radioisotope was expressed in terms of concentration factors (CF: ratio between the radioisotope activity in scallops – Bq g^{-1} wet wt and time-integrated activity in the seawater – Bq g^{-1}) over time for the seawater exposure and in terms of transfer factors (TF: ratio between the radioisotopes activity in scallops – Bq g^{-1} wet wt and time-integrated activity in the sediment – Bq g^{-1} wet wt-) over time for the sediment exposure of *P. maximus*. Uptake kinetics of ^{241}Am and ^{134}Cs in whole-body scallops were fitted (Statistica® 6) using a simple exponential kinetic model (Eq. 1) or using a linear model (Eq. 2):

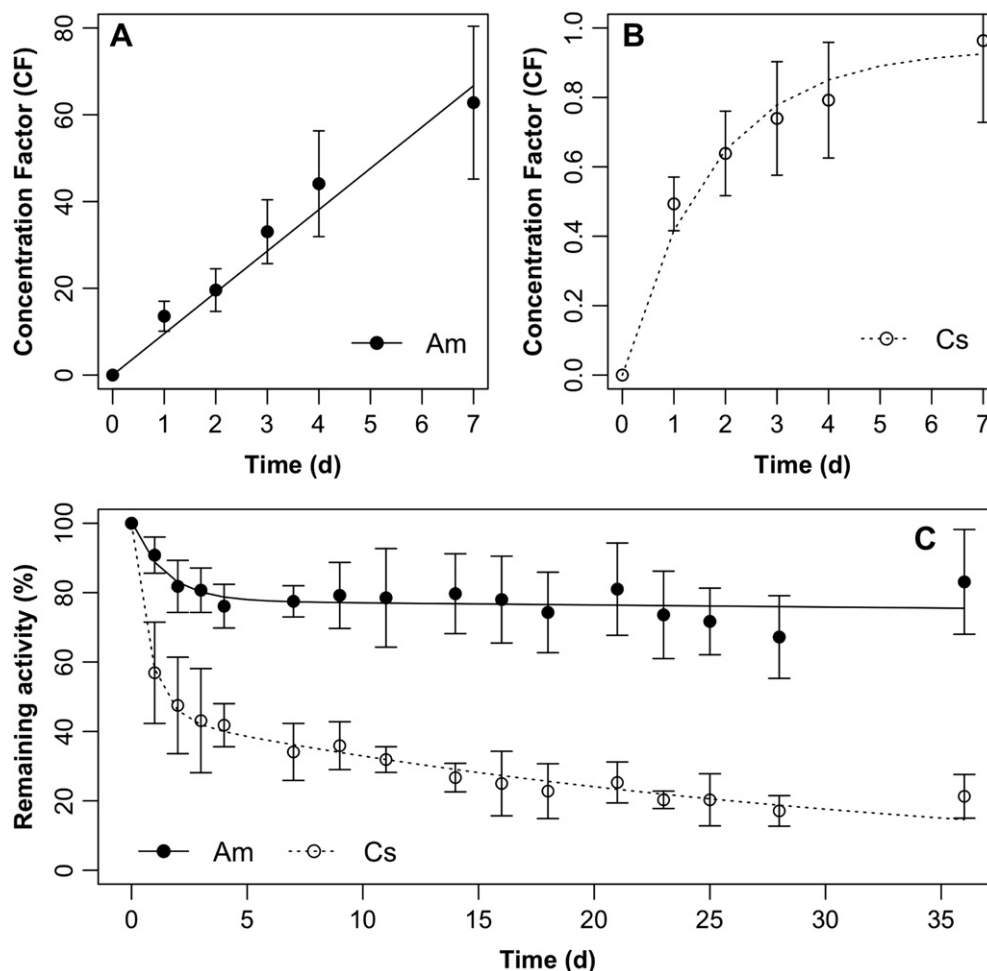


Fig. 1. Uptake kinetics of (A) ^{241}Am and (B) ^{134}Cs in *P. maximus* exposed for 7 d to dissolved radiotracers ($n = 9$) and their following 36 d of depuration kinetics (C) ($n = 9$). All values are mean \pm SD.

Table 1
Whole-body uptake and depuration kinetic parameters of ^{241}Am and ^{134}Cs in *P. maximus* following different exposure experiments: 1) exposed for 7 d to waterborne radionuclides (n = 9) followed by 36 d of depuration (n = 9); 2) after a 2 h feeding on radiolabelled *S. costatum* for ^{241}Am followed by 21 d of depuration (n = 9) and *I. galbana* for ^{134}Cs followed by 16 d of depuration (n = 6) 3) exposed for 13 d via the radiolabelled sediments (n = 6) and then maintained for 31 d in clean sediment and running seawater (n = 6).

Experiment	Radionuclide	Uptake			Depuration				
		CF _{SS} ± ASE	k _u ± ASE	R ²	A _{0s} ± ASE	T _{b1/2s} ± ASE	A _{0l} ± ASE	T _{b1/2l} ± ASE	R ²
1) Seawater	^{241}Am	–	9.53 ± 0.35 ^d	0.80	22.76 ± 4.34 ^d	1.0 ± 0.4 ^a	77.64 ± 2.95 ^d	892	0.35
	^{134}Cs	0.94 ± 0.05 ^d	0.55 ± 0.07 ^d	0.79	54.87 ± 3.88 ^d	0.5 ± 0.1 ^d	45.02 ± 2.92 ^d	22 ± 3 ^d	0.89
2) Feeding	^{241}Am	–	–	–	79.72 ± 4.46 ^d	0.4 ± 0.1 ^d	20.29 ± 3.80 ^d	9 ± 2 ^c	0.93
	^{134}Cs	–	–	–	71.93 ± 6.30 ^d	0.1	28.07 ± 4.37 ^d	14 ± 7 ^a	0.88
3) Sediment	^{241}Am	–	n.a.	–	21.89 ± 8.74 ^a	0.7	78.22 ± 6.24 ^d	79 ± 42	0.35
	^{134}Cs	–	n.a.	–	32.01 ± 11.63 ^b	0.2	67.99 ± 6.54 ^d	74 ± 49	0.24

Uptake parameters: CF_{SS}: concentration factors at steady state; k_u: uptake rate constant (d⁻¹). Depuration parameters: A_{0s} and A_{0l}: remaining activity (%) according to the short- and the long-lived exponential component, respectively; T_{b1/2}: biological half-life (d). ASE: asymptotic standard error; r²: determination coefficient of the uptake or depuration kinetics.

n.a.: information not available.

^a Probability of the model adjustment: p < 0.05.

^b Probability of the model adjustment: p < 0.01.

^c Probability of the model adjustment: p < 0.001.

^d Probability of the model adjustment: p < 0.0001.

$$\text{CF}_t = \text{CF}_{\text{SS}}(1 - e^{-k_e t}) \quad (1)$$

$$\text{CF}_t = k_u t \quad (2)$$

where CF_t and CF_{SS} (CF_{SS} = k_u/k_e) are the concentration factors at time t (d) and at steady state, respectively; k_u and k_e are the uptake and depuration rate constants (d⁻¹), respectively (Whicker and Schultz, 1982).

Depuration of ^{241}Am and ^{134}Cs (seawater, food and sediment experiments) were expressed in terms of percentage of remaining

radioactivity (radioactivity at time t divided by initial radioactivity measured in scallops at the beginning of the decontamination period × 100). The percentages of remaining activity were plotted against time and depuration kinetics were described by a double-component exponential model (Eq. 3):

$$A_t = A_{0s}e^{-k_{es}t} + A_{0l}e^{-k_{el}t} \quad (3)$$

where A_t and A₀ are the remaining activities (%) at time t (d) and 0, respectively; k_e is the depuration rate constant (d⁻¹); 's' and 'l' are the subscripts for the 'short-lived' and 'long-lived' components. For each exponential component (s and l), a biological half-life can be calculated (T_{b1/2s} and T_{b1/2l}) from the corresponding depuration rate constant (k_{es} and k_{el}, respectively) according to the relation T_{b1/2} = ln2/k_e (Warnau et al., 1996). Regarding feeding experiments, the 'long-lived' exponential term describes the fraction of the radiotracer ingested with food that is actually absorbed by the organism (Warnau et al., 1996). The corresponding A_{0l} represents the assimilation efficiency (AE) of the considered radiotracer. The best fitting regression models were selected according to highest determination coefficient and examination of residuals. The level of significance for statistical analysis was always set at α = 0.05.

3. Results

No mortality of scallops was recorded during the acclimation period nor during the different experiments.

3.1. Seawater exposure

Uptake of ^{241}Am in whole-body *P. maximus* displayed linear kinetics (R² = 0.80; Fig. 1A and Table 1) whereas the uptake of ^{134}Cs displayed exponential kinetics reaching a steady state (R² = 0.79; Fig. 1B and Table 1). The values estimated for the kinetic parameters and their associated statistics are shown in Table 1. The concentration factors measured at the end of the exposure period (CF_{7d}) of ^{241}Am and ^{134}Cs were 63 ± 18 and 1.0 ± 0.2 in whole-body scallops, respectively. In the case of ^{134}Cs , the estimated steady state CF calculated by the model (CF_{SS}) reached 0.94 ± 0.05 (Fig. 1B and Table 1).

Calculated CF_{7d} for the different compartments and organs are shown in Table 2. ^{241}Am is systematically more concentrated than ^{134}Cs when considering the same compartment of the scallops (by up to 2 orders of magnitude). The shells of the scallops displayed higher capacities of ^{241}Am bioconcentration than their whole soft parts (CFs: 130 vs. 30) whereas the opposite was observed for ^{134}Cs

Table 2
Concentration Factors (mean CF ± SD) and body distribution (mean % ± SD) of ^{241}Am and ^{134}Cs in *P. maximus* during seawater (after 7 d of exposure and after 36 d of depuration) and feeding experiments (21 d after feeding with *S. costatum* for ^{241}Am and 16 d after feeding with *I. galbana* for ^{134}Cs).

Radionuclides Compartments	Seawater contamination		Food contamination	
	Uptake (7 d, n = 5)		Loss (21 or 16 d, n = 4)	
	Concentration Factor	Distribution (%)	Distribution (%)	Distribution (%)
^{241}Am				
Digestive gland	140 ± 51	23 ± 3	43 ± 6	46 ± 11
Gills	53 ± 30	18 ± 3	9 ± 1	7 ± 2
Kidneys	40 ± 14	2 ± 0	3 ± 1	3 ± 1
Intestine	109 ± 75	2 ± 1	<1	5 ± 4
Gonad	23 ± 12	5 ± 2	9 ± 2	4 ± 2
Foot	44 ± 23	2 ± 1	<1	2 ± 2
Mantle	30 ± 10	34 ± 2	19 ± 2	27 ± 7
Adductor muscle	8 ± 3	10 ± 2	7 ± 3	1 ± 1
Remaining tissues	71 ± 14	6 ± 4	8 ± 4	2 ± 2
Whole soft part	30 ± 8	7 ± 3	5 ± 0	12 ± 7
Shell	130 ± 21	93 ± 3	95 ± 0	88 ± 7
^{134}Cs				
Digestive gland	6 ± 2	10 ± 5	14 ± 2	27 ± 7
Gills	3 ± 1	11 ± 7	1 ± 1	4 ± 2
Kidneys	8 ± 1	3 ± 1	2 ± 0	16 ± 10
Intestine	2 ± 0	<1	<1	1 ± 1
Gonad	3 ± 0	6 ± 3	1 ± 0	5 ± 1
Foot	4 ± 1	2 ± 0	<1	3 ± 1
Mantle	2 ± 0	24 ± 5	5 ± 0	16 ± 16
Adductor muscle	3 ± 1	41 ± 13	76 ± 3	14 ± 10
Remaining tissues	4 ± 0	2 ± 1	<1	11 ± 11
Whole soft part	3 ± 1	76 ± 6	92 ± 2	n.a.
Shell	<1	24 ± 6	8 ± 2	n.a.

n.a.: information not available.

(CFs: 1 vs. 3). Among the soft tissues, the digestive gland and the kidneys presented the highest CF of ^{241}Am and ^{134}Cs , respectively (Table 2).

At the end of the uptake experiment, the highest ^{241}Am load was in the shell (more than 90% of the total body load) and that of ^{134}Cs was in whole soft parts (more than 70% of the total load; Table 2). Among soft tissues, ^{241}Am was mainly contained in the mantle, digestive gland and gills (34, 23 and 18% of total body load, respectively; Table 2) whereas ^{134}Cs was mainly present in the adductor muscle and the mantle (41 and 24% of total body load, respectively; Table 2).

When non-contaminating conditions were restored, the whole-body depuration kinetics of both ^{241}Am and ^{134}Cs were best described by a two-component exponential model (Fig. 1C and Table 1). The majority of the bioaccumulated ^{241}Am was efficiently absorbed (A_{01} : 78%) whereas only 45% of the bioaccumulated ^{134}Cs was absorbed in *P. maximus*. The estimated depuration rate constant of the long-lived components (k_{e1}) for ^{134}Cs was $0.031 \pm 0.004 \text{ d}^{-1}$ and, consequently, the derived biological half-life was $22 \pm 3 \text{ d}$ (Table 1). In the case of ^{241}Am , the depuration rate constant was not significantly different from 0 ($p < 0.05$), thus the corresponding $T_{b1/2}$ may be considered as infinite. However, an estimation of $T_{b1/2}$ based on the mean value of k_{e1} is shown in Table 1 (892 d).

After 36 d of depuration, the distribution of both radionuclides between the shell and the whole soft parts remained similar to that observed at the end of the exposure period: ^{241}Am was mainly found in the shell and ^{134}Cs in the soft tissues (Table 2). Within soft tissues, radionuclide distribution displayed a different pattern than the one observed at the end of the exposure period (Table 2).

Indeed, the digestive gland contained most of the total ^{241}Am load (43%) while the adductor muscle was the main storage organ for ^{134}Cs (76%). Nevertheless, the activities of both radionuclides in all the compartments of *P. maximus* decreased over the depuration phase (data not shown).

3.2. Dietary exposure

The depuration kinetics of the radionuclides ingested with food from the whole-body *P. maximus* were best fitted by a double exponential model (Fig. 2A and Table 1). ^{241}Am and ^{134}Cs displayed similar assimilation efficiencies ($20\% < AE < 30\%$) and close depuration rate constants, k_{e1} , respectively 0.08 ± 0.02 and 0.05 ± 0.02 , which give close $T_{b1/2}$ ($9 \pm 2 \text{ d}$ and $14 \pm 7 \text{ d}$, respectively). At the end of the depuration period, the ^{241}Am load was essentially in the shell (88% of the total body load); Table 2. Among the soft tissues, the digestive gland contained most of the ^{241}Am (46%) whereas ^{134}Cs was mainly distributed between the digestive gland, the kidney, the mantle, the adductor muscle and the remaining tissues, with the digestive gland presenting the higher average load, i.e., 27% (Table 2).

3.3. Sediment exposure

Bioaccumulation of sediment-bound radionuclides was measured in *P. maximus*. However, their whole-body uptake kinetics could not be fitted by a model having a biological meaning (Fig. 2B and Table 3). At the end of the exposure period, the highest transfer factor (TF)

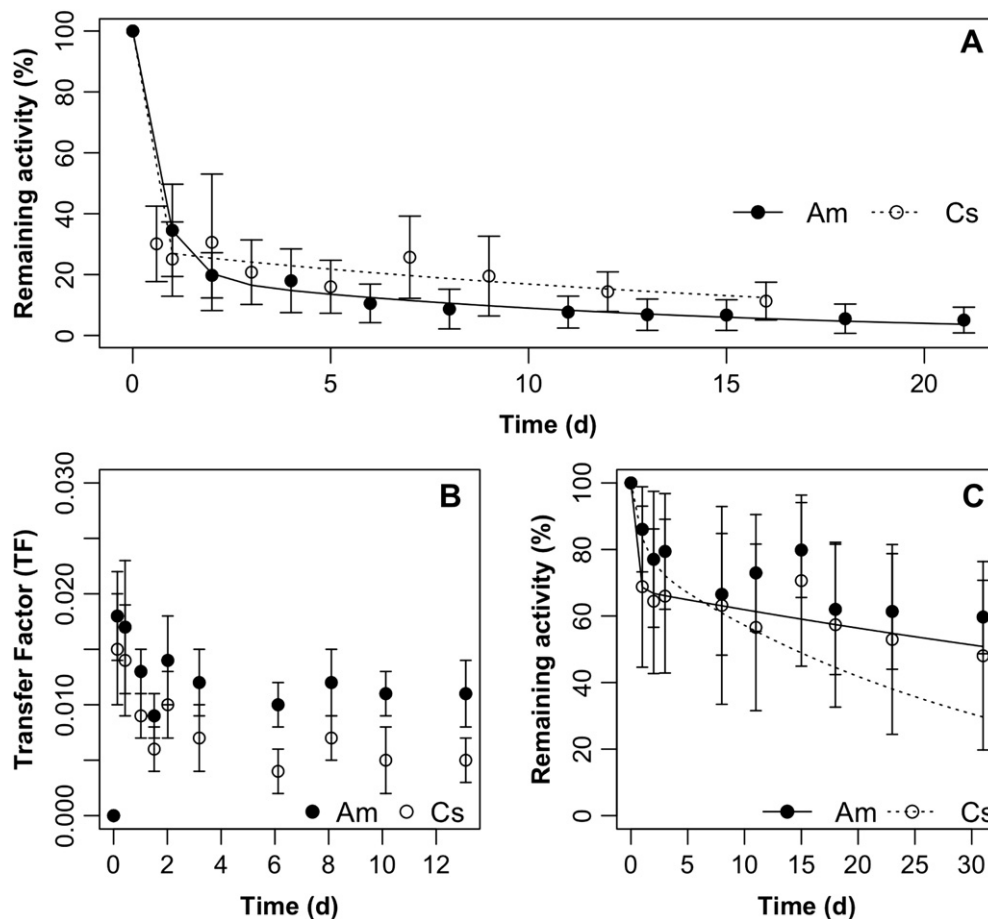


Fig. 2. Kinetics of ^{241}Am and ^{134}Cs in *P. maximus* (A) Depuration after a 2 h feeding on radiolabelled *S. costatum* for ^{241}Am followed by 21 d of depuration ($n = 9$) and *I. galbana* for ^{134}Cs followed by 16 d of depuration ($n = 6$); Uptake and depuration kinetics of ^{241}Am and ^{134}Cs in *P. maximus* (B) exposed for 13 d to the radiolabelled sediments ($n = 6$) and (C) then maintained for 31 d in clean sediment and seawater ($n = 6$). All values are mean \pm SD.

Table 3

Transfer Factors (mean TF \pm SD) of ^{241}Am and ^{134}Cs in *P. maximus* after 13 d of exposure via sediment and body distribution (mean \pm SD) of ^{241}Am and ^{134}Cs at the end of the 13 d exposure (n = 4) and 31 d depuration periods (n = 4).

Radionuclides compartments	Sediment contamination		
	Uptake (13 d, n = 4)		Loss (31 d, n = 4)
	Transfer factor	Distribution (%)	Distribution (%)
^{241}Am			
Digestive gland	0.18 \pm 0.07	47 \pm 17	49 \pm 7
Gills	0.02 \pm 0.02	10 \pm 10	8 \pm 1
Kidneys	0.04 \pm 0.04	3 \pm 3	3 \pm 1
Intestine	0.08 \pm 0.07	2 \pm 2	9 \pm 9
Gonad	0.31 \pm 0.31	14 \pm 14	4 \pm 2
Foot	0.03 \pm 0.01	2 \pm 1	6 \pm 2
Mantle	<0.01	14 \pm 9	3 \pm 1
Adductor muscle	<0.01	3 \pm 3	16 \pm 2
Remaining tissues	0.04 \pm 0.02	4 \pm 4	1 \pm 1
Whole soft part	0.02 \pm 0.02	19 \pm 11	8 \pm 2
Shell	0.03 \pm 0.01	81 \pm 11	92 \pm 2
^{134}Cs			
Digestive gland	0.02 \pm 0.02	16 \pm 11	19 \pm 13
Gills	< 0.01	11 \pm 5	16 \pm 18
Kidneys	0.02 \pm 0.01	3 \pm 1	31 \pm 40
Intestine	0.06 \pm 0.06	4 \pm 4	3 \pm 3
Gonad	0.04 \pm 0.04	12 \pm 6	12 \pm 13
Foot	0.08 \pm 0.08	10 \pm 10	3 \pm 1
Mantle	<0.01	18 \pm 8	3 \pm 2
Adductor muscle	<0.01	41 \pm 13	11 \pm 11
Remaining tissues	0.02 \pm 0.02	5 \pm 3	2 \pm 1
Whole soft part	0.005 \pm 0.003	25 \pm 1	24 \pm 16
Shell	0.006 \pm 0.002	75 \pm 1	76 \pm 16

measured in for whole scallops was 0.011 ± 0.003 for ^{241}Am and 0.005 ± 0.002 for ^{134}Cs . Overall, the different body compartments displayed low TF_{13d} with elevated standard deviations (Table 3). Consequently, none of the tissues could be identified as the main bioaccumulation organ using TF. However, in terms of body distribution, the shell displayed the main part of both radionuclides (>75%). Among soft tissues, the digestive gland and the adductor muscle contained the major part of ^{241}Am ($47 \pm 17\%$) and ^{134}Cs ($41 \pm 13\%$), respectively (Table 3).

The whole-body depuration kinetics of both ^{241}Am and ^{134}Cs after exposure to spiked sediment were best described by a two-compartment exponential equation (Fig. 2C and Table 1). The results indicated that 78% of ^{241}Am and 68% of ^{134}Cs previously bioaccumulated were efficiently retained, with biological half-lives of 79 ± 42 d and 74 ± 49 d, respectively. At the end of the 31 d depuration period, both radionuclides were mainly associated to the shell (Table 3). The distribution of ^{241}Am showed that in soft tissues, the major part of the radionuclide was retained in the digestive gland with $49 \pm 7\%$ of the total ^{241}Am load.

4. Discussion

Biomonitoring programs using scallops (Class Bivalvia, family Pectinidae) already exist (Fegan et al., 2010; JCAC, 2002). Interestingly, scallops show a high accumulation capacity for radionuclides (Miramand et al., 1991; Nonnis Marzano et al., 2000; Bustamante et al., 2002), more so than in other bivalve species such as mussels (JCAC, 2002). However, little is known about the behavior of radionuclides in scallops and their mode of uptake in comparison to other bivalve families such as Ostreidae or Mytilidae (Ryan, 2002). To the best of our knowledge, experimental investigations have been limited to describing the uptake of waterborne ^{241}Am in *P. maximus* and to the localization of this transuranic element in digestive gland cells (Miramand and Germain, 1986; Miramand et al., 1991). Therefore, there is a lack of information on the

uptake and retention of Am by scallops following other natural exposure pathways (i.e., food and sediment) although food and/or sediment were shown to constitute the main pathway of accumulation for Ag, Cd, Co, Pb, Zn (Metian et al., 2007, 2008b, 2009a,b,c). Concerning Cs, no information on its bioaccumulation by scallops is currently available in the literature.

Using realistic activities of dissolved ^{241}Am (i.e., within the range of environmental levels), our study confirmed that ^{241}Am was efficiently accumulated in hard and soft tissues of the scallops, reaching a whole-body CF of 63 after 7 d of exposure. This CF value is relatively high for bivalves. For example, CFs were 10–30 after 5 d of exposure in *Mytilus edulis* (Bjerregaard et al., 1985), 230 after 28 d of exposure in *Cerastoderma edule* and 140 after 31 d of exposure in *Scrobicularia plana* (Miramand et al., 1987). Our CF was higher than in other marine invertebrates such as the sea urchin *Paracentrotus lividus* over comparable exposure times (i.e., approx. 30; Warnau et al., 1996). For *P. maximus*, Miramand and Germain (1986) showed a CF of 80 but that was after a longer exposure period to radiolabelled seawater (i.e., 38 d). Based on the linear model describing the uptake kinetics, the scallops from the present experiment could accumulate ^{241}Am up to a CF of 360 after 38 d. This difference may be due to the size/weight difference of the studied organisms since the scallops of Miramand and Germain (1986) were half-lighter than our organisms. Indeed, previous works have shown that this factor affects metal bioaccumulation (Boyden, 1974, 1977; Warnau et al., 1995; Hédouin et al., 2006).

Following waterborne exposure, ^{241}Am was more efficiently concentrated than ^{134}Cs in *P. maximus* (~ 20 times higher, Table 1). This difference is often found when marine organisms are exposed to these two radionuclides through seawater (Warnau et al., 1996; Bustamante et al., 2006). It could be related to physico-chemicals properties of each radionuclide: as a transuranic radionuclide, americium (III) is strongly particulate reactive (Ryan, 2002) while Cs is soluble and not reacting with particles. Such reactive properties of Am would lead to direct adsorption onto shells. It is therefore not surprising that ca. 95% of the accumulated ^{241}Am was found on the shell of the scallops (Table 2). After they have been accumulated from seawater, the radionuclides were depurated with very different rates with a much shorter biological half-life for Cs (22 d) than for Am (892 d; Table 1). Such a very long biological half-life for Am might result from its retention on the shell. ^{134}Cs was mainly present in the soft tissues, with 65% in the adductor muscle and the mantle (Table 2). This specific accumulation pattern in muscular tissues is related to the analogous behavior of Cs⁺ for K⁺ (Ke et al., 2000; Smith et al., 2002; Lacoue-Labarthe et al., 2010). The predominant distribution of ^{241}Am in the calcitic skeleton/endoskeleton has been shown by several authors (Grillo et al., 1981; Guary et al., 1982; Fowler and Carvalho, 1985). Fowler and Carvalho (1985) demonstrated a positive correlation between the ^{241}Am CF in different echinoderm species and the proportion of calcitic endoskeleton in the body wall of those species. Recently, Zuykov et al. (2009) showed a preferential accumulation of ^{241}Am in the organic periostracum of bivalve's shell. In *P. maximus*, it is noteworthy that the shell contained most of ^{241}Am (up to 95%) whatever the exposure pathway (seawater, food or sediment, Tables 2 and 3). According to our results of dietary exposure, it is apparent that, beside a direct adsorption of dissolved ^{241}Am onto the shell, the radionuclide is also translocated from soft tissues (e.g. the digestive gland) to the shell. It is important to note that the relative affinity of ^{241}Am with the shell of *P. maximus* has been previously observed in the field (Miramand and Germain, 1986). A good perspective for obtaining a temporal record of ^{241}Am in the shell would be by the use of ICP-MS coupled to laser ablation that has been already used in scallops for a chronological survey of other elements (Thébault et al., 2009).

When scallops were fed radiolabelled phytoplankton, dietary ^{241}Am and ^{134}Cs were relatively poorly assimilated (AE: 28 and 20%, respectively; Table 1). Furthermore, both radionuclides were rapidly depurated, resulting in relatively short biological half-lives (between 1 and 2 weeks). These AEs are much lower than in echinoderms and cephalopods which vary from 33 to 90% and from 30 to 60%, respectively (Ryan, 2002; Bustamante et al., 2006). However, our results for ^{241}Am are consistent with data reported for other bivalves, which displayed low ^{241}Am assimilation. For example, the AE of ^{241}Am ranged between 2 and 13% in mussels of the genus *Mytilus* (Baines et al., 2005). Such low AE by mussels could be related to the radionuclide association with mineral fractions of phytoplankton (e.g. diatom shells) or on algal cell surfaces (e.g. Chlorophyceae such as *Dunaliella tertiolecta*) due to the particle-reactive properties of ^{241}Am (Fisher et al., 1983; Fisher and Teyssié, 1986).

The scarcity of data on ^{134}Cs assimilation in marine invertebrates is conspicuous, especially compared to ^{241}Am data on the subject. Nevertheless, ^{134}Cs is not well bioaccumulated by phytoplankton. Heldal et al. (2001) working on ^{134}Cs uptake of five species (three prymnesiophytes and two diatoms) have shown that phytoplankton is unlikely to influence the Cs build-up in marine food webs and Cs flux to deep waters. Bivalves generally showed low AEs for Cs with values ranging between 0.4 and 10% in the green mussel *Perna viridis* (Wang et al., 2000). In predators, reported AEs for Cs are higher, ranging between 44 and 58% in the gastropod *Babylonia formosae habei* and between 23 and 29% in the cuttlefish *Sepia officinalis* (Wang et al., 2000; Bustamante et al., 2006).

Regarding their way of living and their nutrition, scallops are in direct contact with bottom sediments and that predisposes scallops to filter and ingest contaminated particles. For a species filtering large quantities of sediment particles, the TFs obtained at the end of the exposure were quite low (TF_{13d} of ^{241}Am and ^{134}Cs < 0.011 with a maximum TF < 0.05 over the whole exposure period) but consistent with other sediment exposure studies (Miramand et al. 1982; Bustamante et al., 2006; Borretzen and Salbu, 2009).

In scallops from the field, radionuclide activity provides an integrated value of the bioaccumulation process. However, experimental approaches are compulsory to quantify the different physiological parameters of element bioaccumulation and to determine the relative importance of the different exposure pathways (Warnau and Bustamante, 2007). According to the bioaccumulation kinetics of these radionuclides, Am seems to be mainly accumulated from the dissolved phase since exposure to particle-associated Am (food and sediment) resulted in quite poor absorption and retention, compared to ^{241}Am -dissolved bioaccumulation (efficient uptake and strong retention). In the case of the Cs, the kinetic analyses did not clearly reveal a major uptake pathway, even though the low amount of Cs bioaccumulated through sediment exposure (whole-body TF < 0.01) was highly absorbed (A_{01} of $68 \pm 7\%$) and strongly retained ($T_{b1/2} = 74$ d). The use of a bioaccumulation model would allow further exploration of the importance of each exposure pathway *sensu* Thomann et al., (1995) and their use has been already developed and applied on scallops in previous studies (Metian et al., 2007, 2008b, 2009b). However, variability of the kinetic parameters obtained during the sediment experiment was high because the exposure was relatively short. Thus, a bioaccumulation model with 3 exposure pathways could not be run in the present study. Experiments with longer exposure periods will be necessary to better specify these parameters.

5. Conclusion

The present study provided new information about the different bioaccumulation pathways of ^{241}Am and ^{134}Cs that scallops are facing in the field. In this context, our data showed that the shell

and the adductor muscle appeared to be the best scallop compartments for respectively monitoring ^{241}Am and ^{134}Cs in the marine environment.

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