



# Article Artificial Mussels: A New Tool for Monitoring Radionuclides in Aquatic Environments

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**Abstract:** Existing methods for monitoring radionuclides in aquatic environments require frequent sampling of a large volume of water, followed by tedious concentration and analytical procedures, which often make it impractical. Mussels have also been commonly employed to monitor radionuclides but bioaccumulation is significantly affected by physical and biological factors. This study explored the feasibility of using the 'Artificial Mussel' (AM) as a new tool for monitoring radionuclides in marine environments. We showed that (a) the uptake and accumulation of <sup>238</sup>U, <sup>88</sup>Sr, and <sup>133</sup>Cs by AMs are directly related to their concentration in water, and equilibrium could be reached within 7 to 8 weeks with high concentration factors. Our results suggest that AMs can serve as an effective and practical tool for monitoring radionuclides in the aquatic environment and overcoming the difficulties faced by existing methods in radionuclide monitoring.

Keywords: radionuclides; U; Cs and Sr; environmental monitoring; passive sampler; artificial mussel



**1. Introduction** 

In the last few decades, anthropogenic activities such as mining, processing, and discharge from nuclear power plants have released considerable amounts of radioactive compounds into the environment [1] As a result, artificial radionuclides are widely found in aquatic environments nowadays [2]. The Fukushima nuclear power plant accident in 2011, for example, released a huge amount of radioactivity (340–800 PBq) into the marine environment [3], leading to heavy contamination of <sup>131</sup>I, <sup>134</sup>Cs, <sup>137</sup>Cs, and <sup>239</sup>Pu beyond 20 km, which attracted global concern [4].

The ingestion of radionuclides through drinking water and food are the two major pathways for human radiation exposure [5]. Many radioactive compounds not only have long physical half-lives (https://www.nndc.bnl.gov/nudat3/, accessed on 26 June 2023) (e.g., the half-lives of  $^{235}$ U,  $^{137}$ Cs, and  $^{90}$ Sr are 7.04 × 10<sup>8</sup> years, 30.08 years, and 28.9 years, respectively) but can also be bioaccumulated along the food chain [6–9] and hence pose a long-lasting threat to both environmental and human health. For example, the bioaccumulation of  $^{137}$ Cs has been demonstrated in the pelagic food webs in the Norwegian and Barents Seas [8,10]. Likewise, high concentrations of  $^{134}$ Cs and  $^{137}$ Cs were found in demersal fish harvested from coastal waters after the Fukushima accident [11].

Given that radionuclide contamination poses severe and long-lasting environmental and public health risks, routine monitoring is deemed necessary. For example, a 14-year monitoring program of man-made radionuclides (<sup>137</sup>Cs and <sup>106</sup>Ru) was implemented along the French Mediterranean coast [12], and the monitoring of both natural radionuclides (<sup>7</sup>Be, <sup>40</sup>K, <sup>232</sup>Th, <sup>226</sup>Ra, and <sup>238</sup>U) and man-made radionuclide (<sup>137</sup>Cs) was carried out in

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**Copyright:** © 2023 by the authors. Licensee MDPI, Basel, Switzerland. This article is an open access article distributed under the terms and conditions of the Creative Commons Attribution (CC BY) license (https:// creativecommons.org/licenses/by/ 4.0/). Croatian Adriatic coastal waters [13]. Likewise, a monitoring program for radionuclides in marine sediment, seawater, and marine animals and plants was implemented on both the east and west coasts of Canada [14], and radionuclides (<sup>131</sup>I, <sup>134</sup>Cs, and <sup>137</sup>Cs) in fish were monitored regularly following the Fukushima accident [11].

Traditional methods for determining radionuclide concentrations in water require the sampling of a large volume of water, followed by evaporation, to form a concentrated water sample for measurements of gamma activities [15], or to form a dry sample for measurements of alpha and beta activities [16]. Despite the baromembrane (based on reverse osmosis) having been developed to reduce the time and effort spent concentrating radionuclides in water [17] and the range of analytical techniques developed for the determination of radionuclides (e.g., inductively coupled plasma mass spectrometry, laser-induced kinetic phosphorimetry, solid phase extraction, electrochemical approaches), a large volume of water, tedious sample pre-treatment, and stringent conditions are still typically required [1,18]. Empore Caesium Rad Disks [19,20], onsite flow filtration/adsorption [21], and nonwoven fabric cartridge filters impregnated with potassium zinc ferrocyanide [22] have also been employed to monitor radionuclides in aquatic environments. However, the results are often affected by competitive adsorption, and a long time period is required for sample pretreatment, chemical separation, and measurement to achieve the detection limit. Currently, the most sensitive method for measuring radionuclides in ultra-low environmental concentrations is accelerator mass spectrometry, but the sampling and analytical procedures remained very tedious. Given that both spatial and temporal variations in radionuclides in aquatic environments are typically large, the frequent collection of a large number of water samples representing different sites and times, each with a large volume, followed by tedious concentration and laborious analytical procedures, is apparently impractical. Due to the high variability in the toxicity and bioaccumulation potential of different radionuclides (variability in the uptake of different radionuclides may be up to 100,000 times), the non-selective measurement of total radioactivity is inadequate, and the monitoring of individual radionuclides is deemed necessary [23]. Obviously, monitoring different types and levels of radionuclides would be very difficult, if not impractical.

Mussels have a remarkable ability to accumulate radionuclides from both water and planktonic food and therefore have been commonly employed to monitor radionuclides in aquatic environments [24–30]. However, it is well established that both the uptake and retention of radionuclides in mussels are significantly affected by the prevailing physical factors in the environment (e.g., salinity, temperature, food availability) and biological factors (e.g., seasonal growth and reproductive conditions) [31]. For example, Guendouzi et al. found no seasonal variation of radionuclide concentrations in seawater along the Atlantic coast of Portugal, and yet, marked seasonal changes in <sup>210</sup>Po and <sup>210</sup>Pb were clearly evident in mussels during the same period, which was attributable to changes in body weight and the concentrations of lipophilic compounds in the mussels [27]. These confounding factors make it very difficult, if not impossible, to compare the levels of radionuclides in mussels under different hydrographic conditions over time. More importantly, the limited distribution of mussel species in the natural environment often prevents comparisons over large areas. Clearly, rapid and cost-effective methods are urgently required for monitoring radionuclides in the environment.

The passive sampler 'Artificial Mussel' (AM) (with Chelex 100) developed by Wu et al. can provide a time-integrated estimate of metal concentrations in marine and freshwater environments [32]. Extensive laboratory and field studies in the last two decades demonstrated that AMs can provide a reliable time-integrated estimate of a variety of metals over large biogeographic areas with very different hydrological conditions and therefore overcome the longstanding problems of monitoring metals in water, sediment, and biomonitors [33–42]. Recent field studies in the Gulf of California [43] and Turkey [44] further indicated that AMs can also take up U from water, alongside other metal species, despite U not being detected in native mussels deployed at the same site. For monitoring radionuclides in aquatic environments, AMs must fulfill the following three criteria: (a) able to concentrate radionuclides at environmentally relevant concentrations from the environment; (b) the accumulation, uptake, and release of radionuclides are directly related to the concentration of radionuclides in the water; and (c) both the uptake and release of any individual radionuclides are not significantly affected by the presence of other radionuclides in the environment.

U, Sr, and Cs are three radionuclides commonly produced by nuclear power plants. U is an alpha emitter with a very long half-life ( $7.4 \times 10^8$  years), while Cs and Sr are beta emitters with physical half-lives of 30.08 and 28.9 years, respectively. In this study, a series of laboratory experiments was conducted systematically to examine and compare the uptake and release of U, Sr, and Cs by AMs under different conditions, to examine if AM can meet the above three criteria, and to explore the feasibility of using AMs as a new tool for the practical monitoring of radionuclides in aquatic environments.

#### 2. Materials and Methods

The AMs used in this study were prepared following the protocol described by Wu et al. [32]. In brief, 200 mg Chelex-100 (50–100 mesh; from Bio-Rad, CA, USA) was suspended in 8 mL artificial seawater (a standardized mixture of dissolved mineral salts that simulates the chemical composition of natural seawater, AQUA CLOVER, Hong Kong) inside a non-permeable Perspex tube (length: 6 cm; diameter 2.5 cm) with both ends capped by a layer of polyacrylamide gel (thickness: 1 cm). The following experiments were carried out to test whether AMs can be used for monitoring <sup>238</sup>U, <sup>88</sup>Sr, and <sup>133</sup>Cs in the aquatic environment.

Experiment 1 was carried out to determine the individual uptake of <sup>238</sup>U, <sup>88</sup>Sr, and <sup>133</sup>Cs by AMs under three different concentrations. The reported environmentally relevant concentrations of <sup>238</sup>U, <sup>88</sup>Sr, and <sup>133</sup>Cs are 3 µg/L, 10 mg/L, and 0.3 µg/L, respectively [45-47]. In this experiment, solutions with low, medium, and high concentrations of  $^{238}$ U (3 µg/L, 15 µg/L, 30 µg/L),  $^{88}$ Sr (10 mg/L, 50 mg/L, 100 mg/L), and  $^{133}$ Cs  $(0.3 \ \mu g/L, 1.5 \ \mu g/L, 30 \ \mu g/L)$  were prepared for the experiments (equivalent to 1 times, 5 times, and 10 times the reported environmentally relevant concentrations of  $^{238}$ U and  $^{88}$ Sr, and 1 times, 5 times, and 100 times the environmentally relevant concentrations of  $^{133}$ Cs). Radionuclides of U ( $^{238}$ U) and stable isotopes of Sr and Cs ( $^{88}$ Sr and  $^{133}$ Cs), with purity >90.9%, radioactivity <0.078 Gbq/kg (Electron Microscopy Sciences, PA, USA), and purity >99.99% (Sigma Aldrich, MO, USA), were used for the experiments. For each of the above treatments, pre-cleaned plastic tanks were set up at room temperature (25 °C, pH 7.8, salinity 25‰), and each tank contained 45 AMs submerged in 4 L of artificial seawater (AQUA CLOVER, Hong Kong) spiked with an appropriate amount of <sup>238</sup>U, <sup>88</sup>Sr, and <sup>133</sup>Cs. Tanks without <sup>238</sup>U, <sup>88</sup>Sr, and <sup>133</sup>Cs were set up in parallel and served as a control. Three AMs were randomly sampled from each tank weekly, and the concentrations of respective radionuclides accumulated by each AM were determined following the procedures described in the Chemical Analysis section below. Equilibrium was assumed if there was no further increase in <sup>238</sup>U, <sup>88</sup>Sr, and <sup>133</sup>Cs for two consecutive weeks (ANOVA, p < 0.05). Seawater was also sampled from each tank every week, and concentrations of <sup>238</sup>U, <sup>88</sup>Sr, and <sup>133</sup>Cs were determined to provide an estimate of the measured background concentration and to ensure that the variation was <20% of the nominal concentration. Concentration factors of <sup>238</sup>U, <sup>88</sup>Sr, and <sup>133</sup>Cs (concentration in Chelex/concentration in water) upon exposure to different concentrations at different time intervals were calculated and compared.

**Experiment 2** was carried out to determine the release of <sup>238</sup>U, <sup>88</sup>Sr, and <sup>133</sup>Cs accumulated by AMs at equilibrium upon returning them to clean seawater. The remaining AMs in each treatment tank of Experiment 1 were returned to clean artificial seawater after Experiment 1. Three AMs were sampled weekly from each treatment tank from Week 8 (or Week 9) to Week 13, and the concentrations of the respective radionuclides

accumulated by each AM were determined following the procedures described in Chemical Analysis, below.

**Experiment 3** was designed to determine the uptake and release of <sup>238</sup>U, <sup>133</sup>Cs, and <sup>88</sup>Sr in AM upon exposure to mixtures of the three radionuclides at low, medium, and high concentrations (see Table 1 below) at set time intervals to test the hypothesis that both the uptake and release of each individual radionuclides would not be significantly interfered with by the presence of other radionuclides.

Table 1. Low, medium, and high concentrations of <sup>238</sup>U, <sup>88</sup>Sr, and <sup>133</sup>Cs in mixture solutions.

	<sup>238</sup> U	<sup>88</sup> Sr	<sup>133</sup> Cs
Low-concentration mixture	3 μg/L	10 mg/L	0.3 μg/L
Medium-concentration mixture	15 μg/L	50 mg/L	1.5 μg/L
High-concentration mixture	30 μg/L	100 mg/L	30 μg/L

For each of the above treatments, tanks were set up and each tank contained 40 AMs submerged in 4 L of artificial seawater spiked with an appropriate amount of <sup>238</sup>U, <sup>88</sup>Sr, and <sup>133</sup>Cs at equivalent concentrations.

Three AMs were randomly sampled from each tank until equilibrium was reached, and the concentrations of the respective radionuclides accumulated by each AM were determined. The remaining AMs in each treatment tank were then returned to clean artificial seawater, and three AMs were sampled from each tank weekly from Week 7 (or Week 8) to Week 13, and the concentrations of the respective radionuclides accumulated by each AM were determined following the procedures described in the Chemical Analysis section below.

**Experiment 4:** The uptake and release of <sup>133</sup>Cs by AMs (Chelex 100) did not meet the specified criteria (see results below), probably because the chelating moieties of Chelex 100 are iminodiacetates and their binding affinities towards monovalent metal ions are weak and may therefore limit its affinity with group I metals and other monocationic metal ions. Zeolites have been shown to have high cation-exchange capacity and selectivity for Cs, Sr, and Ba ions [48–50] and may therefore be considered an alternative exchanger for monovalent metal ions [51] to enhance the uptake of monovalent Cs<sup>+</sup> ions. As such, a new type of AM with the same amount ( $0.2 \pm 0.05$  g) of finely ground molecular sieve 5 Å (Cas: 69912-79-4, Dieckmann) instead of Chelex 100 was prepared to enable the uptake of Cs<sup>+</sup> ions. This new type of AM was tested following the same design and procedures described in Experiments 1–3 above.

## 3. Chemical Analysis

The content of each individual AM was emptied into a sintered glass filter and eluted after rinsing three times with 12.5 mL 6 M HNO<sub>3</sub> (analytical grade). The artificial seawater sample and the elutriate solution were made up to a known volume with deionized double-distilled water, and concentrations of <sup>238</sup>U, <sup>88</sup>Sr, and <sup>133</sup>Cs in the elutriate solution were determined using an Optima 8000 ICP-OES and NexION 2000 ICP-MS (Plasma flow: 15 L/min; auxiliary flow: 0.3 L/min; nebulizer flow: 0.8 L/min; RF power: 1300 W, pump rate: 1.0 mL/min) after calibration using a standard solution (1000 mg/mL in 2% HNO<sub>3</sub>) [32]. The detection limit was 0.1 µg/g for <sup>88</sup>Sr, <sup>133</sup>Cs, and <sup>238</sup>U. The accuracy of the analysis was evaluated by measuring replicate samples and determined by standard calibration curves with 1, 10, 25, 50, 100, 500, and 1000 µg/L. Concentrations of <sup>238</sup>U, <sup>88</sup>Sr, and <sup>133</sup>Cs in AMs were expressed as µg/g Chelex 100 or µg/g molecular sieve 5 Å.

### 4. Statistical Analysis

The concentrations of <sup>238</sup>U, <sup>88</sup>Sr, and <sup>133</sup>Cs accumulated by AMs exposed to different concentrations and conditions, as well as the temporal changes in each metal, were compared using one-way ANOVA and followed by Tukey's test if significant differences were

found. Data normality was tested using the Shapiro–Wilk test. Statistical analyses were conducted using Sigma plot 14.0.3 and the significance level was set at p < 0.05 for all tests.

### 5. Results

The uptake and release of <sup>238</sup>U, <sup>88</sup>Sr, and <sup>133</sup>Cs by AMs (Chelex 100) under three different concentrations and two conditions (i.e., A: solution with <sup>238</sup>U or <sup>88</sup>Sr or <sup>133</sup>Cs only; and B: a mixture of <sup>238</sup>U and <sup>88</sup>Sr and <sup>133</sup>Cs) are shown in Figures 1–3, respectively. The normal distribution of data was confirmed by the Shapiro–Wilk test.



**Figure 1.** Uptake and release of <sup>238</sup>U by AMs (with Chelex 100) under different concentrations and conditions ((**A**): exposed to a solution with different concentrations of  $^{238}$ U only; (**B**): exposed to mixtures with different concentrations of  $^{238}$ U,  $^{88}$ Sr, and  $^{133}$ Cs, as stated in Table 1).

The uptake and release of <sup>238</sup>U by AM (with Chelex 100) under different concentrations and conditions are presented in Figure 1.

Upon exposure to a solution with <sup>238</sup>U only, 0.03, 0.74, and 1.59  $\mu$ g <sup>238</sup>U/g Chelex 100 were accumulated in the treatment of 3, 15, and 30 ug <sup>238</sup>U/L, respectively (Figure 1A). Uptake increased steadily from Week 1 to Week 8 in a concentration-dependent manner, and reached 0.94, 13.73, and 20.70 ug <sup>238</sup>U/g Chelex 100, respectively, in Week 8. No significant increase in uptake was observed between Week 7 and Week 8 (ANOVA, *p* > 0.05), suggesting that equilibrium was reached at Week 8. In the release phase (Week 9–Week 13), the <sup>238</sup>U accumulated by the AMs progressively decreased, demonstrating that <sup>238</sup>U accumulated by AMs can be released in response to lower environmental concentrations.

Both the uptake and release of  $^{238}$ U in a mixture with  $^{133}$ Cs and  $^{88}$ Sr exhibited a similar pattern as those revealed in the experiment with  $^{238}$ U only (Figure 1B). Despite the uptake of  $^{238}$ U in Week 1 being comparatively lower, uptake was also clearly concentration-dependent. The accumulation of  $^{238}$ U from the mixture solutions in Week 8 (0.60, 13.84, and 24.32 µg  $^{238}$ U/g Chelex 100) was comparable to the respective values when exposed to a solution with  $^{238}$ U only, showing that the binding and release of  $^{238}$ U are not significantly affected by the presence of  $^{88}$ Sr and  $^{133}$ Cs.

The uptake and release of <sup>88</sup>Sr by AMs (with Chelex 100) under different concentrations and conditions are presented in Figure 2.

A rapid decrease in <sup>88</sup>Sr accumulated by AMs in all treatments was found when the AMs were transferred to clean seawater, showing that the <sup>88</sup>Sr taken up by AMs can be released when the concentration becomes lower in the external medium.

The uptake and release of <sup>133</sup>Cs by AMs (with Chelex 100) under different concentrations and conditions are presented in Figure 3.



**Figure 2.** Uptake and release of <sup>88</sup>Sr by AMs (with Chelex 100) under different concentrations and conditions ((**A**): exposed to a solution with different concentrations of <sup>88</sup>Sr only; (**B**): exposed to mixtures with different concentrations of <sup>238</sup>U, <sup>88</sup>Sr, and <sup>133</sup>Cs, as stated in Table 1). Rapid and concentration-dependent uptake of 88Sr was clearly evident from Week 1 to Week 4, regardless of whether AMs were exposed to a solution with <sup>88</sup>Sr only (**A**) or to a mixture with <sup>238</sup>U and <sup>133</sup>Cs (**B**). No significant increase in the uptake of <sup>88</sup>Sr was observable between Week 6 and Week 7 (ANOVA; p < 0.05), suggesting that equilibrium was reached. In Week 7, the <sup>88</sup>Sr accumulated by AMs exposed to a solution with <sup>88</sup>Sr only (45.1, 191.7, and 451.3 ug <sup>88</sup>Sr/g Chelex 100, upon exposure to 10, 50, and 100 mg Sr/L) and to a mixture solution (with respective values of 44.2, 178.7, and 464.2 ug <sup>88</sup>Sr/g Chelex 100) was generally similar, showing that the accumulation of <sup>88</sup>Sr is not affected by the presence of <sup>238</sup>U and <sup>133</sup>Cs.



**Figure 3.** Uptake and release of <sup>133</sup>Cs by AMs (with Chelex 100) under different concentrations and conditions ((**A**): exposed to a solution with different concentrations of <sup>133</sup>Cs only; (**B**): exposed to mixtures with different concentrations of  $^{238}$ U, <sup>88</sup>Sr, and <sup>133</sup>Cs, as stated in Table 1).

Unlike <sup>238</sup>U and <sup>88</sup>Sr, AMs (with Chelex 100) are not able to take up or release <sup>133</sup>Cs in a concentration-dependent manner. Moreover, no consistent pattern of temporal changes can be found in either the uptake or release phases.

Correspondingly, another series of experiments was then carried out by replacing Chelex 100 in the AMs with the same amount of finely ground molecular sieve 5 Å (made up of Zeolite with pores of ca. 5 Å sufficient for the penetration of the Cs ions) with high cation-exchange capacity to enhance the uptake of monovalent Cs+ ions. The experiments were repeated to determine the uptake and release of  $^{133}$ Cs by AMs (molecular sieve 5 Å) exposed to different concentrations and different conditions.



The uptake and release of <sup>133</sup>Cs by AMs (with molecular sieve 5 Å) under different concentrations and conditions are presented in Figure 4.

**Figure 4.** Uptake and release of <sup>133</sup>Cs by AMs (with molecular sieve 5 Å) under different concentrations and conditions ((**A**): exposed to a solution with different concentrations of <sup>133</sup>Cs only; (**B**): exposed to mixtures with different concentrations of  $^{238}$ U,  $^{88}$ Sr, and  $^{133}$ Cs, as stated in Table 1).

The rapid and concentration-dependent uptake of <sup>133</sup>Cs was clearly evident from Week 1 to Week 3, regardless of whether AMs were exposed to a solution with <sup>133</sup>Cs only or to a mixture solution. No significant increase in the uptake of <sup>133</sup>Cs was observable between Week 6 and Week 7 in the solutions with <sup>133</sup>Cs only, nor in the mixture solutions with <sup>238</sup>U and <sup>88</sup>Sr (ANOVA; *p* < 0.05), suggesting that equilibrium was reached. In Week 7, the <sup>133</sup>Cs accumulated by AMs exposed to the solutions with <sup>133</sup>Cs only (1.7, 2.4, and 17.6 ug <sup>133</sup>Cs/g molecular sieve 5 Å upon exposure to 0.3, 1.5, and 30 µg <sup>133</sup>Cs/L, respectively) and to the mixture solutions (with respective values of 1.5, 1.8 and 18.0 ug <sup>133</sup>Cs/g molecular sieve 5 Å) was generally similar, showing that the accumulation of <sup>133</sup>Cs is not affected by the presence of <sup>238</sup>U and <sup>88</sup>Sr.

The concentration of <sup>133</sup>Cs in AMs showed a progressive decrease in all treatments when the AMs were transferred to clean seawater from Week 8 to Week 13, showing that the <sup>133</sup>Cs taken up by AMs could be released in response to a lower concentration of <sup>133</sup>Cs in the external medium.

## 6. Discussion

Overall, the results of this study showed that:

- 1. The uptake and accumulation of <sup>238</sup>U and <sup>88</sup>Sr by AM (with Chelex 100), as well as the uptake and accumulation of <sup>133</sup>Cs by AM (with molecular sieve 5 Å), are directly related to their respective concentrations in the external medium.
- 2. The equilibrium of <sup>238</sup>U could be reached within 8 weeks, and the equilibrium of <sup>88</sup>Sr and <sup>133</sup>Cs could be reached within 7 weeks.
- 3. High concentration factors were found for <sup>238</sup>U (1771), <sup>88</sup>Sr (6710), and <sup>133</sup>Cs (3675) upon exposure to their respective environmentally realistic concentrations, indicating that AMs with Chelex 100 can take up <sup>238</sup>U and <sup>88</sup>Sr, and AMs with molecular sieve 5 Å can take up <sup>133</sup>Cs efficiently at low, environmentally realistic concentrations.
- 4. <sup>238</sup>U and <sup>88</sup>Sr taken up by AMs (with Chelex 100) and <sup>133</sup>Cs taken up by AMs (with molecular sieve 5 Å) can be released when their respective concentrations in the external medium become lower.
- 5. The binding and release of <sup>238</sup>U, <sup>88</sup>Sr, and <sup>133</sup>Cs were not significantly affected by the presence of the other two radionuclides in the external medium at all concentrations.

6. Compared with <sup>133</sup>Cs and <sup>88</sup>Sr, <sup>238</sup>U exhibited a relatively higher uptake rate upon exposure to both single and mixed solutions and also a longer time to reach equilibrium. The release of accumulated <sup>238</sup>U was also much slower than that of <sup>88</sup>Sr and <sup>133</sup>Cs.

The above results suggest that AMs with Chelex 100 can serve as an effective tool for monitoring <sup>238</sup>U and <sup>88</sup>Sr, while AMs with molecular sieve 5 Å can be used for monitoring <sup>133</sup>Cs in aquatic environments. Indeed, field studies in the Gulf of California [43] and Turkey [44] reported that AMs (with Chelex 100) were able to take up U from aquatic environments (despite U not being found in native mussels and oysters deployed side by side), thus offering further evidence that AMs can serve as a promising tool for monitoring radionuclides in the natural environment.

Due to technical difficulties and laboratory safety, the radioisotope of U ( $^{238}$ U) and stable isotopes of Sr and Cs ( $^{88}$ Sr and  $^{133}$ Cs) were used for the experiments in this study. However, this does not in any way affect the conclusions drawn in the present study, nor the efficiency and reliability of using AMs in the field monitoring of radionuclides. Arguably, isotopic differences may affect binding affinity, since there may be slight differences in the ionic radii and atomic weight between different isotopes. However, given that the equilibrium constants for ion-exchange reactions are highly similar between different metal-ion isotopes (with a difference of <0.001%) [52], the difference in the uptake of metal-ion isotopes by AMs, if any, would be negligible from the perspective of monitoring metal radionuclides such as Cs, Sr, and U. The insignificant isotope effects on the binding affinity can be further supported by the very low isotope enrichments using ion exchangers (e.g.,  ${}^{1}\text{H}/{}^{2}\text{H}$ : 3%;  ${}^{6}\text{Li}/{}^{7}\text{Li}$ : (0.2–4.3%);  ${}^{7}\text{Be}/{}^{9}\text{Be}$ : 0.18%  ${}^{22}\text{Na}/{}^{24}\text{Na}$ : 0.014%;  ${}^{39}\text{K}/{}^{40}\text{K}$ : (-0.074-0%);  ${}^{25}\text{Mg}/{}^{26}\text{Mg}$ : 0.016%;  ${}^{59}\text{Co}/{}^{60}\text{Co}$ : (0.005–0.017%);  ${}^{84}\text{Sr}/{}^{88}\text{Sr}$ : 0.0097%;  ${}^{235}\text{U}/{}^{238}\text{U}$ : (-0.034%-0.007%)) [52].

Theoretically, other radioactive metal ions such as those of Ra, Pu, and Co should also behave like U, Sr, and Cs and other metals (e.g., Cd, Cr, Cu, Pb, and Zn), which can be taken up by binding to the chelating groups and released by AMs. As such, their levels in aquatic environments may also be monitored by using AMs containing Chelex 100, molecular sieve 5 Å, or other ion exchangers with suitable chelating functional groups to achieve effective uptake. Further laboratory studies should be extended to other radionuclides, and field studies should be carried out to further validate the laboratory findings of the present study.

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