Should we measure plutonium concentrations in marine sediments near Fukushima?

R. Periáñez · Kyung-Suk Suh · Byung-Il Min

Abstract Pu contamination originating from Fukushima accident has not been detected in marine sediment samples collected outside a 30 km circle around the plant. It is shown, by means of numerical modelling, that if any impact from the accident has occurred, this would remain in a very close area to Fukushima because of the low Pu mobility in the marine environment. Since the situation inside the 30 km zone remains unknown, further studies on the determination of Pu isotopes in sediments within this area are required.

Keywords Plutonium · Marine dispersion · Fukushima · Numerical modelling · Sediments

Introduction

A number of reports on ^{137}Cs distributions in the marine environment, resulting after the March 2011 Fukushima accident, have been published [1, 2]. Also, several modelling studies on the dispersion of this radionuclide have been reported. As an example, a comparison of the performances of some of these models may be seen in [3]. All models consider Cs as a perfectly conservative radionuclide, neglecting water/sediment interactions. Recently, the ^{137}Cs contamination of marine sediments off Fuskushima has been successfully simulated considering uptake/release reactions between water and sediments [4].

R. Periáñez (✉)
Dpt Física Aplicada I, ETSIA Universidad de Sevilla,
Ctra Utrera km 1, 41013 Sevilla, Spain
e-mail: rperianez@us.es

K.-S. Suh · B.-I. Min
KAERI, Daedeok-Daero 989-111, Yuseong-Gu, Daejeon,
Republic of Korea

Much less information is available in the case of plutonium isotopes. Trace amounts of Pu isotopes originating from the accident have been identified in soil samples [5, 6]. While it is know that atmospheric releases of Pu were several orders of magnitude lower than those from Chernobyl accident [6], no information on Pu isotopes in the liquid direct releases to the sea is available [7]. Pu isotopes have been measured in marine sediments outside a 30 km radius circle around Fukushima. Results do not show any contamination due to the accident [7]. Instead, Pu isotopes here detected are attributed to global fallout. However, the situation inside the 30 km zone remains unknown. It could be possible that Pu isotopes entered this coastal area from the direct release of contaminated water in early April 2011. The objective of this work consists of showing, by means of numerical modelling, that, if Pu contamination originating from the accident would be present in sediments of the close area to Fukushima, contamination would not reach areas far from the plant. Contamination would be restricted to the close area because of the low mobility of Pu. Thus, it would not be detected if samples are not collected there. Consequently, further studies on the determination of Pu isotopes in seawater and sediments within the 30 km zone would be required.

The used model, which has already been applied to ^{137}Cs [4], is described briefly in the next section since details may be seen in the indicated references. Results are discussed in section 3.

Methods

Daily averaged three dimensional wind and density driven currents have been obtained from the JCOPE2 (Japan Coastal Ocean Predictability Experiment 2) model in the

time frame from March 12th to June 30th. The area studied extends from 140.46° to 142.04° longitude and from 35.96° to 39.54° latitude. There are 23 vertical levels and spatial resolution is 5 min of arc. As an example, surface currents corresponding to March 12th may be seen in Fig. 1. The Kuroshio current -flowing to the NE- is clearly appreciated. Currents are weak in the area of Fukushima and flow southwards. To the south of Fukushima an anticyclonic eddy may be seen.

The dispersion model consists of a 3D advection/diffusion equation with terms describing the adsorption/desorption reactions between the deepest water layer, in contact with the seabed, and bed sediments. These processes are formulated in a dynamic way, in terms of kinetic transfer coefficients. A detailed description of such formulation may be seen elsewhere [8–10]. There has been evidence to suggest that uptake takes place in two stages: fast surface adsorption followed by slow migration of ions to pores and interlattice spacings [11–14]. Thus, a 2-step model has been used. It considers that exchanges are governed by two consecutive reversible reactions: surface adsorption is followed by another process that may be a slow diffusion of ions into pores and interlattice spacings, inner complex formation or a transformation such as an oxidation. k_1 and k_2 are forward and backward rates for the first reaction; and k_3 and k_4 are the corresponding rates for the second one (Fig. 2). Thus, sediments are divided in two phases: a reversible and a slowly reversible fraction. It has been shown that the 2-step model reproduces both the adsorption and release kinetics of plutonium in the Irish Sea, where it is released from Sellafield nuclear fuel reprocessing plant [15]. It has also been used in our previous ^{137}Cs dispersion simulations [4] for Fukushima. A detailed formulation of the kinetic model may be seen elsewhere [15, 16], thus it will not be repeated here.

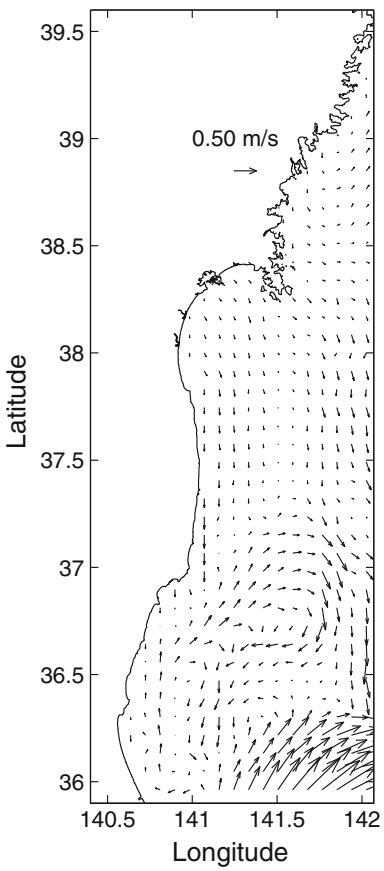

Fig. 1 Surface currents produced by the JCOPE2 model for March 12th

Results

Kinetic rates already used for plutonium have been applied [15]. Although it is true that these parameters are site-specific, there is not information about them for Japan coastal waters. On the other hand, this approximation is enough for our purposes since we do not want to compare model predictions with observations, but only assess the behavior of plutonium in these coastal waters. Details on the other parameters involved in the model may be seen in [4]. Also, Pu may be present in the marine environment in different oxidation states. The reduced Pu [Pu(III) and Pu(IV)] is highly particle reactive and has been shown to possess a k_d that is approximately two orders of magnitude higher than that of the more soluble oxidized Pu [Pu(V) and Pu(VI)]. Different rates should be used for oxidized and reduced Pu. However, we will use effective rates representing the potential mixture of oxidation states, as done in previous works [8, 17], due to the lack of data. Redox reactions could be simulated if information were available [15].

Atmospheric deposition has not been considered since Pu fallout from Fukushima is characterized by a high ^{241}Pu/$^{239+240}$Pu activity ratio. Since ^{241}Pu is not detectable in sediments offshore Fukushima, it was concluded that the atmospheric deposition of Pu from the accident is negligible in marine sediments [7], at least in a scale of the order of 100 km.

For direct liquid releases to the ocean, the same timing and magnitude as in the ^{137}Cs simulations have been used [4]. It must be taken into account that the magnitude of

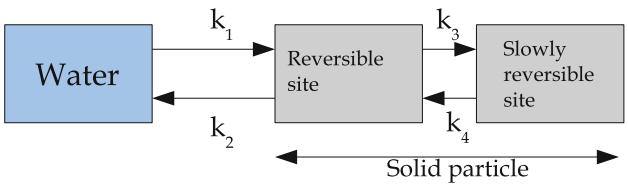

Fig. 2 Scheme representing the applied kinetic model for water/sediment interactions

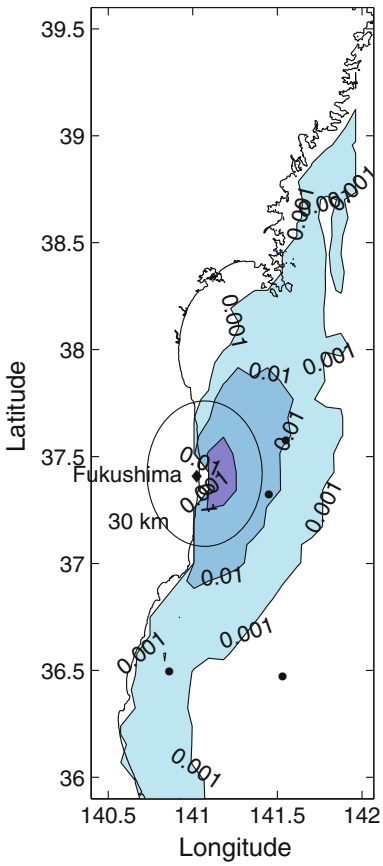

Fig. 3 Calculated concentrations of plutonium in bed sediments for June 30th, normalized to the maximum value in the domain. The 30 km circumference around Fukushima is shown. *Black dots* indicate points were Pu concentrations have been measured in bed sediments [7]

such plutonium direct release is totally unknown. As commented above for kinetic rates, this arbitrary source magnitude does not affect the conclusions of this study.

The computed distribution of plutonium in bed sediments for June 30th may be seen in Fig. 3. These concentrations are normalized to the maximum calculated value since the release magnitude is arbitrary (Cs value). Pu concentrations in sediments decrease by more than one order of magnitude within a 30 km circle around Fukushima. Measured sediment concentrations of $^{239+240}$Pu in points indicated in Fig. 3 are in the range 0.48–3.53 mBq/g and are attributed to nuclear weapon test fallout [7]. We cannot assess if some of this plutonium is coming from Fukushima, but it is clear that Pu concentrations in sediments within the 30 km circle would be more than two orders of magnitude larger than those eventually detected in the indicated sampling points. The impact of Fukushima releases, with respect to Pu, is restricted to a small zone well within a 30 km radius around the power plant. Pu measurements in this area would be required to assess if any impact has been produced.

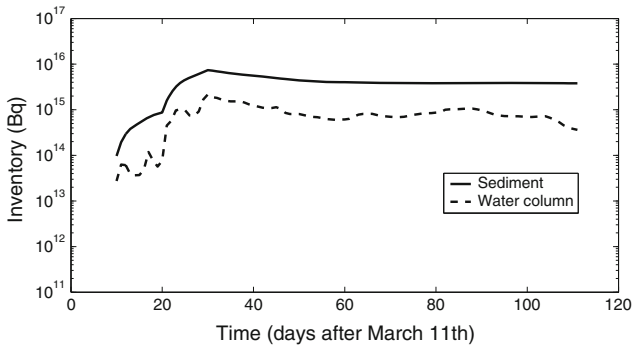

Fig. 4 Computed Pu inventory in the model domain for the water column and the bed sediment

Finally, the flushing time of plutonium has also been evaluated. The Pu inventory in the water column has been calculated each time step. The temporal evolution of this inventory may be described as an exponential function [18]:

$$I(t) = I_0 e^{-t/\tau} \quad (1)$$

where I_0 is the initial inventory and τ is flushing time. From numerical fitting of the temporal evolution of the inventory (once it has started to decrease, i.e., some 40 days after March 12th) to an exponential decay function, it was obtained that flushing time is 246 days. This value is significantly longer than that previously estimated for ^{137}Cs from the shelf [18], which is 43 $\pm$ 16 days. This again reveals the longer persistence of Pu contamination in comparison with Cs and the relevance of marine sediment as a long-term delayed source of previously radionuclides. Such an effect has been occurring in the Irish Sea with Sellafield releases. Computed inventories in the water column and the bed sediment are shown in Fig. 4. After the inventory peaks, they start to decrease very slowly because of the low Pu mobility in the marine environment.

Conclusions

Numerical modelling indicates that Pu contamination originating from the accident could be present in marine sediments near Fukushima, although it has not been detected in samples collected outside a 30 km radius from the plant (opposed to the case of ^{137}Cs, radionuclide for which contamination was measured). To confirm if any impact from the accident has occurred, Pu isotopes should be measured in a close area to Fukushima, since they would mostly remain in this zone because of the low mobility of Pu in the marine environment.

Flushing time of Pu from the model domain has been evaluated, resulting 246 days, significantly longer than that of ^{137}Cs.

Acknowledgements The authors are indebted to Dr Masanao Nakano (JAEA), who supplied JCOPE2 current data. KAERI was partially supported by a nuclear research and development program funded by the Ministry of Education, Science and Technology in Korea.

References

1. Bailly du BP, Laguionie P, Boust D, Korsakissok I, Didier D, Fiévet B (2012) Estimation of marine source term following Fukushima Dai-ichi accident. J Environ Radioact 114:2–9
2. Buesseler KO, Jayneb SR, Fisherc NS, Rypinab II, Baumannc H, Baumannc Z, Breiera CF, Douglassb EM, Georgec J, Macdonaldb AM, Miyamotod H, Nishikawad J, Pikea SM, Yoshidab S (2012) Fukushima-deriver radionuclides in the ocean and biota off Japan. PNAS, doi:10.1073/pnas.1120794109
3. Masumoto Y, Miyazawa Y, Tsumune D, Tsubono T, Kobayashi T, Kawamura H, Estournel C, Marseleix P, Lanerolle L, Mehra A, Garraffo ZD (2012) Oceanic dispersion simulations of 137-Cs released from the Fukushima Daiichi nuclear power plant. Elements 8:207–212
4. Periáñez R, Kyung-Suk S, Byung-Il M (2012) Local scale marine modelling of Fukushima releases. Assessment of water and sediment contamination and sensitivity to water circulation description. Mar Pollut Bull 64:2333–2339
5. Yamamoto M, Takada T, Nagao S, Koike T, Shimada K, Hoshi M, Zhumadilov K, Shima T, Fukuoka M, Imanaka T, Endo S, Sakaguchi A, Kimura S (2012) An early survey of the radioactive contamination of soil due to the Fukushima Dai-ichi Nuclear Power Plant accident, with emphasis on plutonium analysis. Geochem J 46:341–353
6. Zheng J, Tagami K, Watanabe Y, Uchida S, Aono t, Ishii N, Yoshida S, Kubota Y, Fuma S, Ihara S (2012) Isotopic evidence of plutonium release into the environment from the Fukushima DNPP accident. Sci Rep 2:304. doi:10.138/srep00304
7. Zheng J, Aono T, Uchida S, Zhang J, Honda MC (2012) Distribution of Pu isotopes in marine sediments in the Pacific 30 km off Fukushima after the Fukushima Daiichi nuclear power plant accident. Geochem J 46:361–369
8. Periáñez R (2008) A modelling study on ^{137}Cs and ^{239,240}Pu behaviour in the Alborán Sea, western Mediterranean. J Environ Radioact 99:694–715
9. Periáñez R (2009) Environmental modelling in the Gulf of Cadiz: heavy metal distributions in water and sediments. Sci Total Environ 407:3392–3406
10. Periáñez R (2012) Modelling the environmental behavior of pollutants in Algeciras Bay (south Spain). Mar Pollut Bull 64: 221–232
11. Nyffeler UP, Li YH, Santschi PH (1984) A kinetic approach to describe trace element distribution between particles and solution in natural aquatic systems. Geochim Cosmochim Acta 48: 1513–1522
12. Turner A, Millward GE (1994) Partitioning of trace metals in a macrotidal estuary. Implications for contaminant transport models. Estuar Coast Shelf Sci 39:45–58
13. Ciffroy P, Garnier JM, Pham MK (2001) Kinetics of the adsorption and desorption of radionuclides of Co, Mn, Cs, Fe, Ag and Cd in freshwater systems: experimental and modelling approaches. J Environ Radioact 55:71–91
14. El Mrabet R, Abril JM, Manjón G, García-Tenorio R (2001) Experimental and modelling study of plutonium uptake by suspended matter in aquatic environments from southern Spain. Water Res 35:4184–4190
15. Periáñez R (2003) Kinetic modelling of the dispersion of plutonium in the eastern Irish Sea: two approaches. J Mar Syst 38:259–275
16. Periáñez R (2004) Testing the behaviour of different kinetic models for uptake-release of radionuclides between water and sediments when implemented on a marine dispersion model. J Environ Radioact 71:243–259
17. Periáñez R (1999) Three dimensional modelling of the tidal dispersion of non conservative radionuclides in the marine environment. Application to ^{239,240}Pu dispersion in the eastern Irish Sea. J Mar Syst 22:37–51
18. Dietze H, Kriest I (2012) ^{137}Cs off Fukushima Dai-ichi, Japan. Model based estimates of dilution and fate. Ocean Sci 8: 319–332