Science of the Total Environment 615 (2018) 741–750

Contents lists available at ScienceDirect

Science of the Total Environment

journal homepage: www.elsevier.com/locate/scitotenv

Shifts of radiocesium vertical profiles in sediments and their modelling in Japanese lakes

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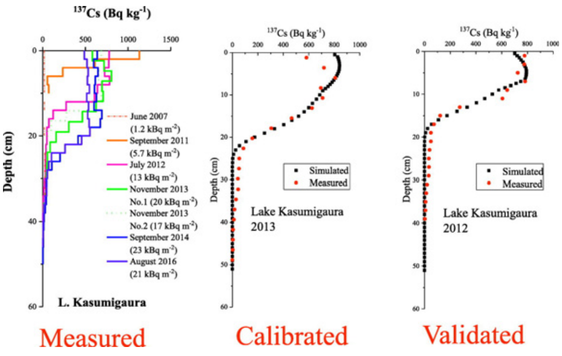
HIGHLIGHTS

- Influences of the 2011 Fukushima nuclear accident on lake sediments were traced.
- Shifts of radiocesium vertical profiles in sediments were compared in five lakes.
- Rapid penetration of radiocesium to a certain depth was observed in some lakes.
- A one-dimensional differential sediment model successfully described the shifts.
- Wind-induced stress and sediment porosity are the key parameters for the submodels.

GRAPHICAL ABSTRACT



ARTICLE INFO

Article history:
Received 14 June 2017
Received in revised form 25 September 2017
Accepted 26 September 2017
Available online 17 October 2017

Editor: D. Barcelo

Keywords:
Fukushima nuclear accident
Sediment core
Sedimentation
Mixing
Porosity

ABSTRACT

Vertical profiles of radiocesium concentrations were measured in sediment cores collected at various times after the 2011 Fukushima nuclear accident in five Japanese lakes (Hinuma, Kasumigaura, Kitaura, Onogawa and Sohara) with different morphological and trophic characteristics in order to investigate the sedimentation-diffusion processes. In lakes where sediments had high porosities and experienced considerable wave action due to shallowness, we observed rapid penetration of radiocesium to a certain depth just after the accident, followed by downward movement of the peak depths. In contrast, gradual downward transfers of distinct peaks were found in other types of lakes. A one-dimensional differential sediment model with water-sediments interaction processes was constructed to describe the vertical shift of radiocesium profiles. Our proposed submodels relating to the length scales of the mixing using wind-induced stress and porosity of sediments were constructed based on one measurement of the vertical distribution of radiocesium in three lakes (Hinuma, Kasumigaura and Sohara). This model was then validated using samples from those lakes in different years, as well as from two other lakes. Good agreement was obtained. We discuss our findings, the limits of model application, and future research targets.

1. Introduction

After the Great East Japan Earthquake and resulting tsunami on 11 March 2011, a serious accident occurred at the Fukushima Dai-ichi

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https://doi.org/10.1016/j.scitotenv.2017.09.286

Nuclear Power Plant (FDNPP). Radionuclides (including 15 PBq of ^{137}Cs) were released into the atmosphere and ocean, mainly on 15–16 March 2011 (TEPCO, 2011; Yasunari et al., 2011).

Accumulation of Chernobyl-derived ^{137}Cs was monitored in bottom sediments of several Finnish lakes over many years (1969, 1978, 1988, 1990, 2000 and 2003), and both increasing and decreasing tendencies were observed 14 years after the accident (Ilus and Saxen, 2005). Another study looked at yearly and spatial changes in ^{137}Cs in surface sediments (0–5 cm) in a cooling basin of the Ignalia Nuclear Power Plant (Marciulioniene et al., 2015). The results indicated complicated sedimentation features, which may have been affected by a number of natural and anthropogenic factors, resulting in mixing, resuspension, and remobilization of sediments and radionuclides. Seasonal variations of ^{137}Cs have been reported in Lake Juodis (Lithuania), due to its remobilization in the bottom water, but this process was considered site-specific (Tarasiuk et al., 2008). Kansanen et al. (1991) reported a tendency for radiocesium concentrations to increase with depth in the sediments of Lake Paijanne, Finland, and considered this a result of the focusing effect. However, limited investigation has been done on the temporal changes of vertical profiles of ^{137}Cs within the framework of sedimentation processes in lakes.

A number of processes have been considered for simulation of the vertical profiles of radiocesium in sediments in order to reproduce the measured profiles. These include bioturbation (Christensen and Bhunia, 1986), physical mixing (Smith and Comans, 1996), sediment resuspension (Luettich et al., 1990), non-local mixing (direct injection of part of the flux into deeper sediment layers) (Soetaert et al., 1996), initial deposition (penetration through connected pore spaces) (Abril and Gharbi, 2012), incomplete mixing zone (fast and homogeneous mixing of a mobile fraction, and sedimentation of an irreversibly bounded fraction) (Abril, 2003), buoyancy effect (disturbances in the thermohalinic stability of sediment interstitial liquids inducing their interfacial transfer) (Tarasiuk et al., 2010), and release from sediments (Tarasiuk et al., 2008). In terms of sorption to sediment solids, irreversible sorption (Smith and Comans, 1996; Meili and Worman, 1996; Liu et al., 2003; Putyrskaya and Klemt, 2007) and the influence of NH_4^+/K^+ on the partition coefficient (Comans et al., 1989; Davison et al., 1993) were taken into account.

The aim of this work was (1) to compare the changes in radiocesium vertical profiles in five Japanese lakes from the viewpoint of mixing and (2) to see if the profiles could be reproduced using a one-dimensional (1-D) advection-diffusion equation with a wave-induced mixing process and partitioning of ^{137}Cs between dissolved and particulate components. We focused on the profiles collected at the centers of the respective lakes, and the horizontal transfer of radiocesium as a function of direct fallout was considered the primary input.

2. Methods

2.1. Characteristics of the lakes

Information about and characteristics of the five lakes and their watersheds are shown in Table 1 and Fig. 1. According to the criteria on lake phosphorus concentration proposed by the Organization for Economic Co-operation and Development (OECD, 1982), Lakes Kasumigaura and Kitaura are hypereutrophic, Lake Hinuma is eutrophic, and Lakes Onogawa and Sohara are oligotrophic.

Lake Hinuma is a brackish-water lake near Mito City. This lake is connected to the Pacific Ocean through the Hinuma and Naka Rivers, and is affected by tidal movement. The concentration of chlorine ion in lake water varied from 3 to 8500 mg l^{-1} (mean: 1690 mg l^{-1}) in a survey over 1965–2002 (Taba et al., 2006). Land use in the watershed is ~32% forest, 17% paddy fields, 25% ploughed fields, and 25% residential and other. Lakes Kasumigaura, Kitaura and Hinuma are so shallow that vertical stratification is easily destroyed by a strong wind, and are thus considered polymictic lakes.

Lake Kasumigaura, the second largest lake in Japan, is located in the eastern part of Japan's Kanto Plain. Land use in the watershed is ~44% forest, 25% paddy fields, 14% ploughed fields, and 17% residential and other.

Lake Kitaura is connected to Lake Kasumigaura through the Sotonasakaura River, which is located downstream of the two lakes. There is a possibility of water exchange between the lakes (Fukushima and Arai, 2015). Land use in the watershed is ~44% forest, 16% paddy fields, 23% ploughed fields, and 18% residential and other.

Lakes Onogawa and Sohara are dimictic dammed lakes produced by the eruption of Mt. Bandai in 1888 and located about 800 m above sea level in the Urabanndai Plateau in Bandai-Asahi National Park. The outflow of Lake Sohara flows into Lake Onogawa. Their watersheds are covered by forest. Some of the water from Lake Onogawa is used for electric power generation.

2.2. Sampling of sediments

Sampling was done 17 times at the centers of the lakes as shown in Fig. 1 and the Supplementary Information 1, except for the downstream site in Lake Onogawa. Scuba divers in general used acrylic tubes (10 cm in diameter and 100 cm long) during the sediment sampling campaigns for Lakes Hinuma, Kasumigaura, and Kitaura, except for the samplings in Aug. 2011 in Lake Hinuma, Sep. 2011 in Lake Kasumigaura, and Jul. 2011 and Jan. 2012 in Lake Kitaura, when a KB-type gravity core sampler (an acrylic tube 7 cm in diameter and 50 cm long) was used (Supplementary information 1). An HR-type gravity core sampler (an acrylic tube 11 cm in diameter and 50 cm long) was deployed in Lakes Onogawa and Sohara. Because the vertical profiles and inventories of radiocesium taken by the HR-type core sampler were quite similar to those taken by scuba divers in Lakes Kasumigaura and Nakanuma (area: 0.01 km^2; maximum depth: 13 m) (Arai et al., 2017), it was assumed that the difference in the sediment sampling methods had negligible influence on the results. In addition, Arai et al. (2017) measured radiocesium concentrations at 2-cm intervals to the depth of 10 cm for the sediment samples taken by scuba divers (4 replicates) and the HR-type core sampler (3 replicates) in Lake Kasumigaura, and reported that the variation coefficients (standard deviation divided by the mean) were 5–18% for scuba divers and 2–14% for the core sampler, respectively.

2.3. Analysis of sediments

Core samples from the campaigns were sliced at 2-cm intervals in less than a few hours after the sampling. The changes in sediment

Table 1
Characteristics of the investigated lakes.

Lake	Altitude m	Lake area km^2	Mean water depth m	Maximum water depth m	Water retention time y	Watershed area km^2 *1	Watershed area/lake area	^{137}Cs deposition kBq/m^2 *2
Hinuma	0	9.4	2.1	6.5	0.14	439	47	6.5
Kasumigaura	0	172	4.0	7.3	0.56	1440	8	14.6
Kitaura	0	36	4.8	7.8	0.56	390	11	9.4
Onogawa	797	1.4	7.9	22	0.06	140	100	28.2
Sohara	830	0.35	5.1	12	0.53	2.6	7	33.0

*1: excluding the lake area, *2: averaged for the area within square of the lake area from the sampling point.