Article

Radiocesium contamination of lake sediments and fish following the Fukushima nuclear accident and their partition coefficient

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Abstract

To evaluate the influences of the Fukushima nuclear accident in 2011 on lake sediments and fish and to understand the difference in their contamination levels, data on radiocesium concentrations (^{137}Cs and ^{134}Cs) were analyzed for sediments and 18 fish species (including one freshwater prawn) taken from 15 lakes in northeastern Japan. Temporal trends in radiocesium concentrations (referenced to radioactivity on 15 Mar 2011) were not clear during 2011–2013 except in a few species of fish. There was a decrease among topmouth gudgeon (*Pseudorasbora parva*), icefish (*Salangichthys microdon*), and pond smelt (*Hypomesus nipponensis*) but an increase among channel catfish (*Ictalurus punctatus*) and kokanee (*Oncorhynchus nerka*). Significant positive correlations between lake-averaged radiocesium concentrations of sediments and fish were observed for most fish species. The partition coefficients (PCs), defined as fish concentration divided by sediment concentration on a dry weight basis, clustered mostly in the range of 0.3–3 ([Bq/kg-dry]/[Bq/kg-dry]) and were high in large-sized types (e.g., channel catfish and brown trout [*Salmo trutta*]) but low in small-sized types (e.g., topmouth gudgeon and icefish). After normalization, the PCs of the respective lakes were analyzed together with lake characteristics related to water exchange and lake dimensions, water quality, and sediments and were found to be high in the lakes with high water retention time and vice versa, suggesting prolonged contact and equilibration between the water and the sediments. Finally, the efficacy and potential problems of using the PCs between sediments and fish are discussed.

Key words: fish, lake characteristics, radiocesium, partition coefficient, sediments

Introduction

After the Great East Japan Earthquake and resulting tsunami on 11 March 2011, a serious accident occurred at the Fukushima Dai-ichi Nuclear Power Plant. Huge amounts of radionuclides (e.g., 15 PBq of radiocesium ^{137}Cs) were released into the atmosphere and oceans, mainly during 15–16 March 2011 (TEPCO 2011, Yasunari et al. 2011). The Japanese government and prefectural governments carried out environmental surveillance initially on air, water, soil, sediments, animals, and food, among others (Hirose 2012). Two years after the accident, values of radioactivity exceeding the detection limit of ordinary monitoring were observed only for samples of sediments and fish in aquatic environments.

Following the similarly serious Chernobyl accident, efforts were made to monitor and model radiocesium contamination in lake environments, sometimes accompanied by laboratory experiments. For example, in-lake variations in sediments and fish (Håkanson 1999, Erlinger et al. 2008), long-term changes in sediments and fish (Kapala et al. 2008, Saxén and Ilus 2008), transfer between water and sediments (Smith et al. 2000, Konoplev et al. 2002), and fish contamination (Kryshev 1995, Kryshev et al. 1996), sometimes with modeling of in-lake dynamics (McDougall et al. 1991, Heling 1997, Monte et al. 2003, 2004, 2005, Kumblad et al. 2006) were investigated and discussed.

Putyrskaya et al. (2009) notably described the detailed behavior of ^{137}Cs in Lago Maggiore and other pre-alpine lakes based on the ^{137}Cs data in tributaries, lake water, and

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bottom sediments, revealing the role of ^{137}Cs as a marker of sedimentation processes; however, the relationship between sediments and other biotic components was not analyzed. Sundbom et al. (2003) analyzed time-series data of ^{137}Cs in fish from 3 Swedish lakes and quantified the effect of body size and trophic level on the concentrations, but the differences between lakes were not elucidated. Håkanson et al. (1996) proposed a simple but general model, VAMP, to make state-of-the art predictions of radiocesium in lake water and predatory fish. Their model was validated against an extensive dataset from 7 European lakes that covers a wide range of lake and catchment characteristics and requires many parameter values to be observed. Särkkä et al. (1996) reported the radiocesium concentrations in the surface layer of bottom sediments of 52 lakes of southern Finland and analyzed the relationships among those concentrations in sediments; tissues of perch, pike, and roach; lake water quality; and morphometric values. In addition to the small number of fish species, the influence of lake parameters on the partition of radiocesium between sediments and fish was not investigated. Consequently, the relationship between sediments and fish contamination, differences in radiocesium dynamics between lakes, and other interactions are not well understood.

In the present study, temporal changes and in-lake variations of radiocesium concentrations in sediments and fish are first analyzed using surveillance data in 15 lakes before their relationships are investigated. Subsequently, the partition coefficients (PCs) between sediments and fish are proposed to express the between-lake and between-fish differences in fish contamination. Finally, the characteristics of PCs are analyzed and discussed according to lake type and fish species.

Methods

Database

We used databases (through 31 March 2013) on radionuclides related to sediments from the Japanese Ministry of Environment (MOE 2013) and on aquatic products from the Ministry of Agriculture, Forestry and Fisheries (Fisheries Agency 2013) for 15 lakes located in northeastern Japan (Table 1; Fig. 1). Data from the database on aquatic products were used for analysis of 17 species of fish and 1 species of prawn (hereafter abbreviated as 18 species of fish), including eel (*Anguilla japonica*, Anguilliformes); carp (*Cyprinus carpio*), crucian carp (*Carassius cuvieri*), dace (*Tribolodon hakonensis*), silver crucian carp (*Carassius auratus langsdorfii*), and topmouth gudgeon (*Pseudorasbora parva*, Cypriniformes); channel catfish (*Ictalurus punctatus*, Siluriformes); brown trout (*Salmo trutta*), char (*Salvelinus leucomaenis*), cherry salmon (*Oncorhynchus masou*), kokanee (*Oncorhynchus nerka*), landlocked salmon (*Oncorhynchus masou masou*), and rainbow trout (*Oncorhynchus mykiss*, Salmoniformes); icefish (*Salangichthys microdon*) and pond smelt (*Hypomesus nipponensis*, Osmeriformes); largemouth bass (*Micropterus salmoides*) and smallmouth bass (*Micropterus dolomieu*, Perciformes); and freshwater prawn (*Macrobrachium nipponense*, Decapoda). In these lakes, 518 fish data as well as 220 sediment data were used for analysis (Table 1).

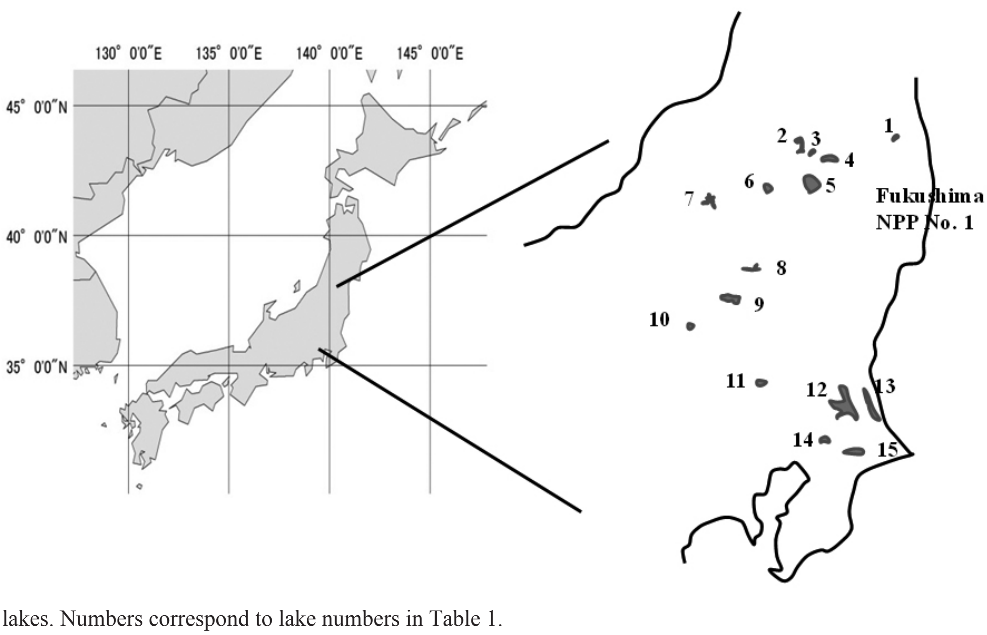

Fig. 1. Surveyed lakes. Numbers correspond to lake numbers in Table 1.

Table 1. Surveyed 15 Japanese lakes, morphometric parameters, and numbers of samples for sediments and fish.

No.	Name of lake	Lake area (km^2)	Averaged depth (m)	retention time (y)	No. of sediment sampling points	No. of sediment sampling	No. of fish samples
1	L. Hayama	1.8	19.9	0.48	1	17	8
2	L. Hibara	10.4	12.0	0.83	1	6	23
3	L. Akimoto	3.9	9.9	0.26	1	6	31
4	L. Onogawa	1.4	7.9	0.058	1	6	12
5	L. Inawashiro	104.8	37.0	5.0	12	6	12
6	L. Numazawa	3.1	62.4	0.56	1	8	12
7	L. Tagokura	8.8	56.2	0.31	1	3	13
8	L. Kawamata	2.6	33.8	0.42	1	5	7
9	L. Chuzenji	11.5	94.6	6.5	1	5	24
10	L. Akagi-onuma	0.9	9.1	1.7	1	4	25
11	L. Yanaka	4.5	5.9	0.50	1	4	9
12	L. Kasumigaura	171.0	3.8	0.56	4	6	140
13	L. Kitaura	34.0	5.0	0.56	2	6	128
14	L. Teganuma	6.5	0.9	0.042	4	6	19
15	L. Inbanuma	11.6	1.7	0.045	4	6	55

In the MOE (2013) database survey, 3 sediment subsamples up to 5–10 cm depth were taken using bottom grab samplers (Ekman-Birge type or Smith-Mcintyre type) at each location. The subsamples were mixed to create one composite sample for the given location. The radiocesium activity (^{137}Cs and ^{134}Cs) was measured using a Germanium (Ge) detector for the wet sample. The concentration was calculated on a dry weight basis using the water content of the sample; the detection limit was usually 10 Bq/kg-dry.

In the survey databases of the Fisheries Agency (2013), a fish sample of 5–10 kg was obtained for each species and the edible portion (muscle) was used for measurement. Because muscle to whole-body ratios of ^{137}Cs in freshwater fish were reported slightly higher than unity (~1.25 for largemouth bass; Peters and Newman 1999; and 1.20 for channel catfish; Peters et al. 1999), radiocesium concentrations in whole-body fish were expected to be close to but less than the reported values. The radiocesium concentration in fish was calculated on a dry weight basis in a similar manner to that described for sediments, except that the water content reported in the food composition database in the Japanese Ministry of Education, Culture, Sports, Science and Technology (MEXT 2013) was used. The detection limit was usually <5 Bq/kg-wet. For lakes Hayama, Akimoto, and Inawashiro, the results obtained by MOE (2013) were also used because a method similar to that of the Fisheries Agency (2013) was applied.

Data treatment

Data values below the detection limit were not included in the analysis (<2% of all measurements for sediments and fish). Decay correction was performed for all measurements from 15 March 2011 because the influence of natural attenuation could be neglected. When only sums of ^{137}Cs and ^{134}Cs were measured (<20% of the observations for fish), their average ratio over other periods at the same lake was used to estimate the respective counts of ^{137}Cs and ^{134}Cs. Averages of at least 2 measurements in the respective lakes were employed for all other analyses. The $^{134}Cs/^{137}Cs$ activity ratios corrected to 15 March 2011 were 0.93 (SD 0.05) for sediments and 1.05 (SD 0.12) for fish, indicating close agreement with the ratio of the Fukushima-derived radionuclides (nearly 1; Hirose 2012). Because the results obtained for ^{134}Cs were fairly similar to those of ^{137}Cs, only values for ^{137}Cs are subsequently reported, except in special cases.

Statistical analysis

Correlation analysis was used to analyze the relationships (e.g., radiocesium concentrations) between sediments and fish. Two-way analysis of variance was applied to the sediment radiocesium concentrations in the respective lakes to compare the spatial and temporal variations. A statistical t-test (Excel-statistics for Windows 2012) was

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used to check the significance of the difference in the means of 2 groups (e.g., radiocesium concentrations in 2 periods). We applied >10 models (exponential, polynomial, logarithmic, and others) to the relationships between lake characteristics and partition coefficient of radiocesium concentrations between sediments and fish using Origin Pro 9J.

Results

^{137}Cs concentrations in surficial sediment varied to a large extent (23–26 000 Bq/kg-dry; Fig. 2), and coefficients of variation in the respective lakes ranged from 0.24 in Lake Inbanuma to 1.36 in Lake Inawashiro. The results of 2-way analysis of variance applied for the lakes having at least 3 sampling points indicated that spatial factor was significant in lakes Inawashiro and Teganuma but insignificant in lakes Kasumigaura and Inbanuma, suggesting that deposition of radiocesium in some of the investigated lakes had been spatially inhomogeneous. In contrast, temporal variations were small (judged as insignificant by 2-way analysis of variance for the 4 lakes) and did not show increasing or decreasing trends in any of the lakes ($p > 0.05$ by t-test for the differences between the periods before and after 31 Mar 2012; i.e., during the first and second year in lakes having at least 2 samples in both periods).

For fish ^{137}Cs concentration, significant decreasing tendencies were observed for topmouth gudgeon in lakes Teganuma and Inbanuma, icefish in Lake Kasumigaura, and for pond smelt in lakes Hibara, Onogawa, Akagi-onuma, Kasumigaura, and Kitaura (Fig. 3a; $p < 0.05$ by t-test for the differences between the periods before and after 31 Mar 2012 in lakes having at least 2 samples in both periods). Significant increases were detected for channel catfish in Lake Kasumigaura and for kokanee in Lake Numazawa ($p < 0.05$; Fig. 3b). The other 8 cases showed insignificant changes. In quantitative terms, ^{137}Cs concentrations in pond smelt decreased in an approximately logarithmic manner (Fig. 3a; $r^2 = 0.39$–0.91, $p < 0.01$) with estimated half-lives without natural attenuation ranging from 139 d in Lake Hibara to 456 d in Lake Kasumigaura. In general, small fish such as topmouth gudgeon, icefish, and pond smelt

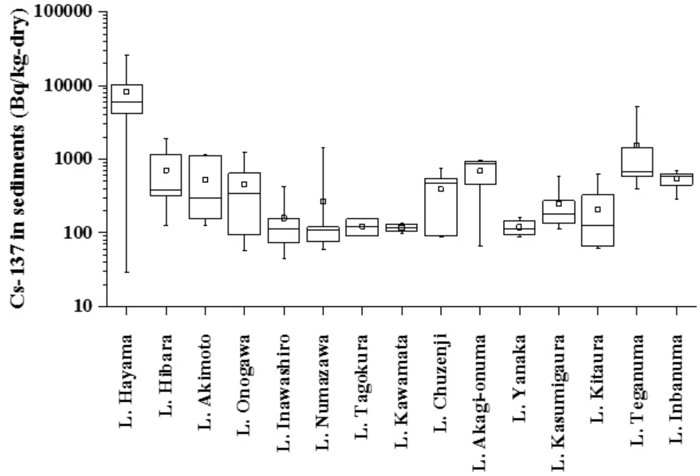

Fig. 2. Box and whisker plots of ^{137}Cs concentrations in the surficial sediments of 15 Japanese lakes. The box is determined by the 25th, 50th, and 75th percentiles; whiskers are the 5th and 95th percentiles; and small squares indicate the mean. Minimum and maximum are also shown.

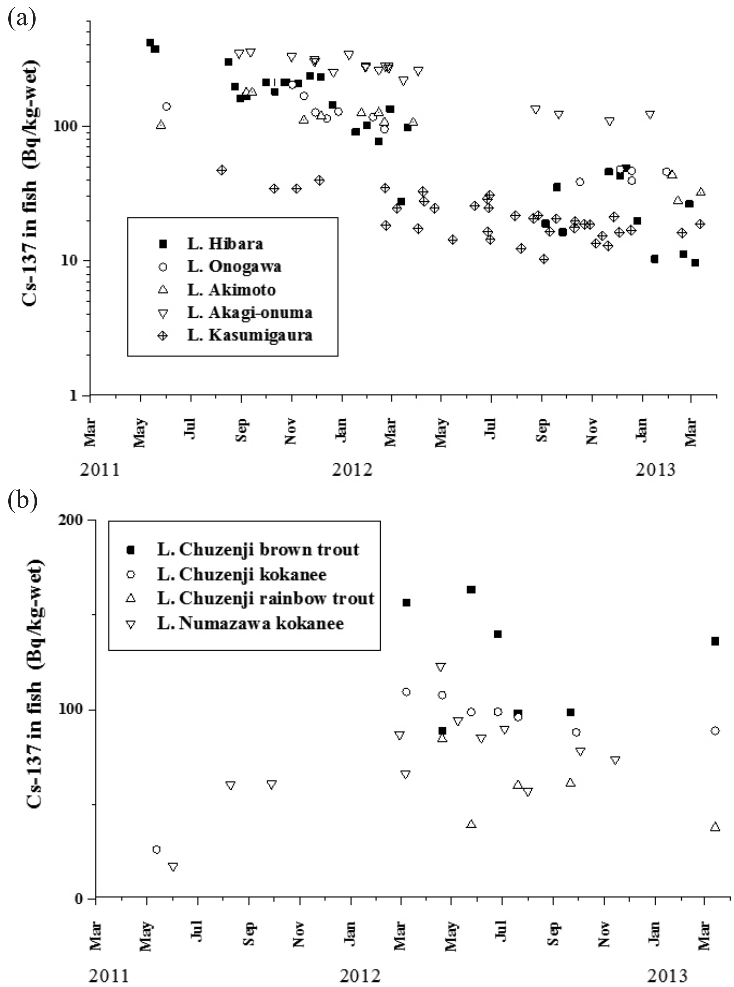

Fig. 3. Temporal changes in fish ^{137}Cs concentrations. (a) Logarithm of ^{137}Cs concentrations of pond smelt in 5 lakes vs. time; (b) ^{137}Cs concentrations of brown trout, kokanee, and rainbow trout vs. time.

showed decreasing trends, and increasing trends were observed in relatively larger-sized fish such as channel catfish and kokanee.

Almost all comparisons between sediments and fish revealed positive correlations, of which 5 were significant ($p < 0.05$ for 8 species of fish observed in at least 4 lakes; Fig. 4). For ^{134}Cs, the same levels of correlation as ^{137}Cs were observed (silver crucian carp: $r^2 = 0.987$**, pond smelt: 0.805**, dace: 0.487, char: 0.953*, freshwater prawn: 0.459, channel catfish: 0.999**, crucian carp: 0.00, carp: 0.928; single * indicates $p < 0.05$, double ** indicates $p < 0.01$). When we excluded Lake Hayama, where radiocesium concentrations in both sediments and fish were overwhelmingly higher than in other lakes, similar levels of significance were obtained, indicating scientifically meaningful relationships.

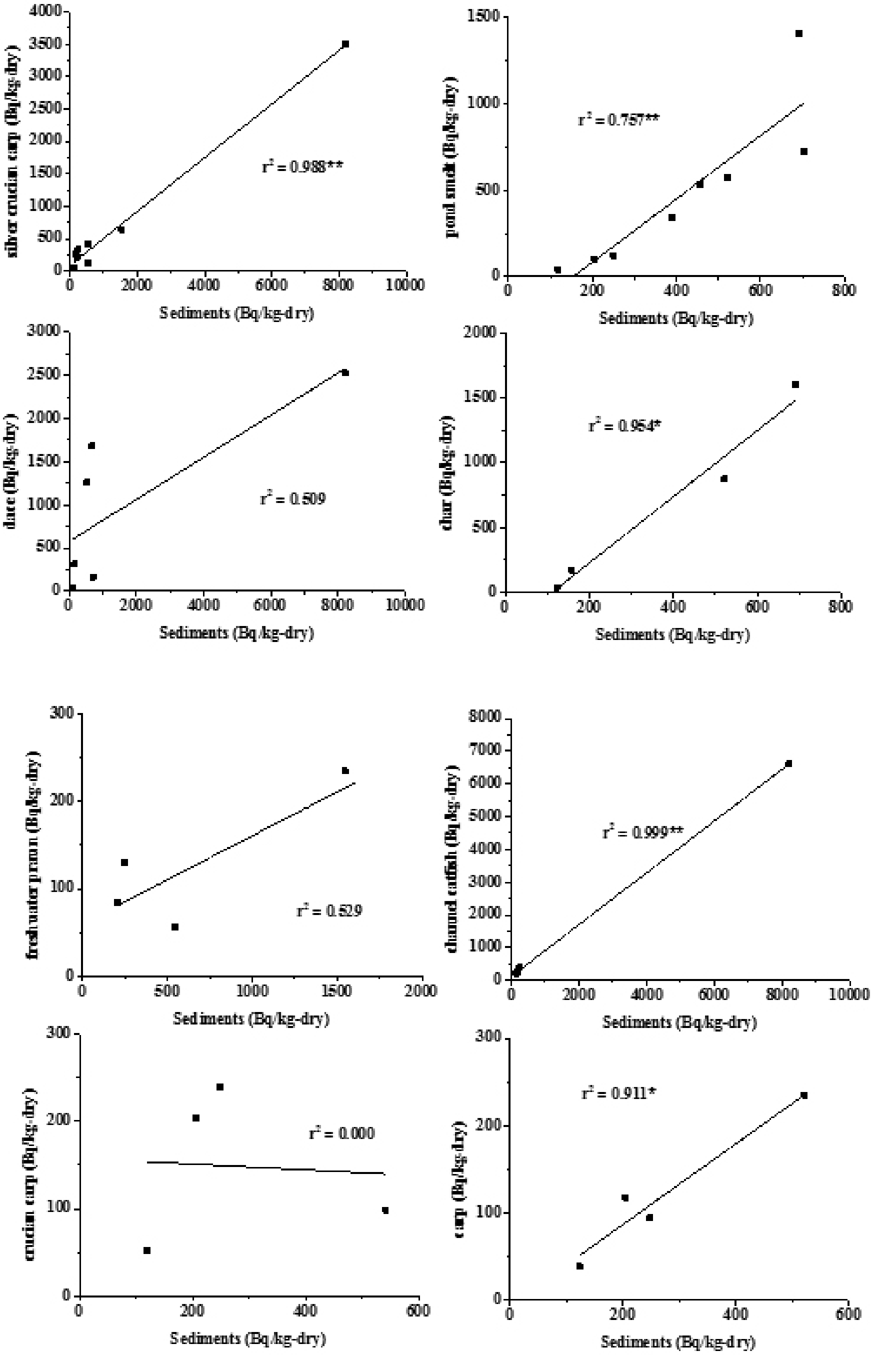

Fig. 4. ^{137}Cs concentrations in the surficial sediments vs. ^{137}Cs concentrations in 7 species of fish and a freshwater prawn. * indicates $p < 0.05$; ** indicates $p < 0.01$.

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Based on these linear relationships, the ratio of fish to sediment radiocesium concentrations was used to calculate the PC between them. PC values were distributed from 0.1 to 7 ([Bq/kg-dry]/[Bq/kg-dry]), mostly concentrated in the range of 0.3–3 (>83% of 65 values; Table 2; Fig. 5; for ^{134}Cs, >87% of 65 values). The PCs in lakes Inawashiro and Akagi-onuma were high, whereas those in lakes Teganuma and Inbanuma were low. Lower PCs were observed in carp, topmouth gudgeon, icefish, and freshwater prawn. In contrast, channel catfish, brown trout, char, cherry salmon, landlocked salmon, largemouth bass, and smallmouth bass had higher PCs.

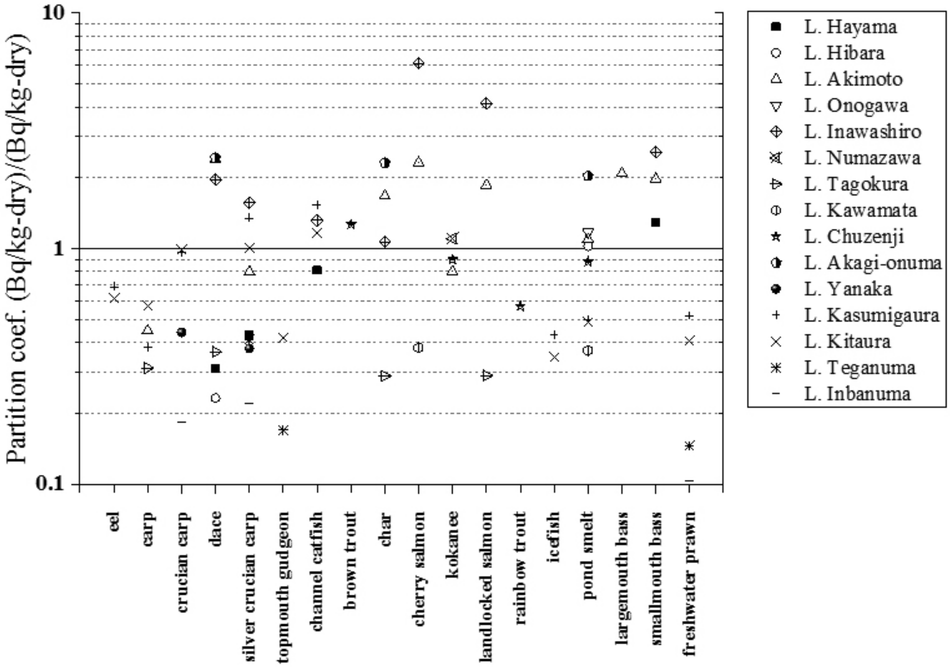

Fig. 5. Partition coefficients (PCs) between sediment ^{137}Cs concentrations and fish ^{137}Cs concentrations in 15 lakes.

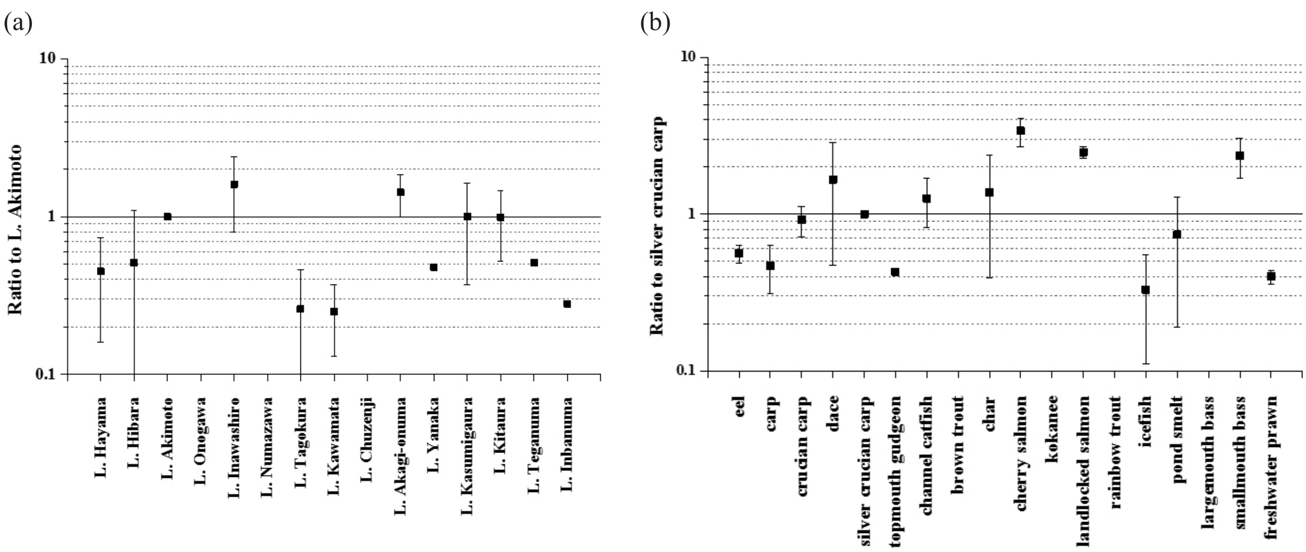

Fig. 6. Partition coefficients (PCs) between sediment ^{137}Cs concentrations and fish ^{137}Cs concentrations. (a) Ratio of lake-normalized PC normalized to Lake Akimoto; (b) ratio of fish-normalized PC normalized to silver crucian carp.

Table 2. Partition coefficients (PCs) between sediment ^{137}Cs concentrations (Bq/kg-dry) and fish ^{137}Cs concentrations (Bq/kg-dry) in 15 Japanese lakes.

	Anguilliformes	Cypriniformes					Siluriformes	Salmoniformes						Osmeriformes		Perciformes		Decapoda
Lake name	**eel**	**carp**	**crucian carp**	**dace**	**silver crucian carp**	**top-mouth gudgeon**	**channel catfish**	**brown trout**	**char**	**cherry salmon**	**kokanee**	**land-locked salmon**	**rainbow trout**	**icefish**	**pond smelt**	**large-mouth bass**	**small-mouth bass**	**fresh-water prawn**
L. Hayama	–	—	—	0.31	0.43	—	0.81	—	—	—	—	—	—	—	—	—	1.29	—
L. Hibara	—	—	—	0.23	—	—	—	—	—	—	—	—	—	—	1.02	—	—	—
L. Akimoto	—	0.45	—	2.40	0.80	—	—	—	1.67	2.32	0.80	1.86	—	—	1.10	2.09	1.98	—
L. Onogawa	—	—	—	—	—	—	—	—	—	—	—	—	—	—	1.17	—	—	—
L. Inawashiro	—	—	—	1.97	1.57	—	1.32	—	1.07	6.12	—	4.14	—	—	—	—	2.58	—
L. Numazawa	—	—	—	—	—	—	—	—	—	—	1.10	—	—	—	—	—	—	—
L. Tagokura	—	0.31	—	0.36	—	—	—	—	0.29	—	—	0.29	—	—	—	—	—	—
L. Kawamata	—	—	—	—	—	—	—	—	—	0.38	—	—	—	—	0.37	—	—	—
L. Chuzenji	—	—	—	—	—	—	—	1.27	—	—	0.90	—	0.57	—	0.88	—	—	—
L. Akagi-onuma	—	—	—	2.44	—	—	—	—	2.32	—	—	—	—	—	2.03	—	—	—
L. Yanaka	—	—	0.44	—	0.38	—	—	—	—	—	—	—	—	—	—	—	—	—
L. Kasumigaura	0.69	0.38	0.96	—	1.35	—	1.54	—	—	—	—	—	—	0.43	0.50	—	—	0.52
L. Kitaura	0.62	0.57	0.99	—	1.01	0.42	1.17	—	—	—	—	—	—	0.35	0.49	—	—	0.41
L. Teganuma	—	—	—	—	0.41	0.17	—	—	—	—	—	—	—	—	—	—	—	0.15
L. Inbanuma	—	—	0.18	—	0.22	0.10	—	—	—	—	—	—	—	—	—	—	—	0.10

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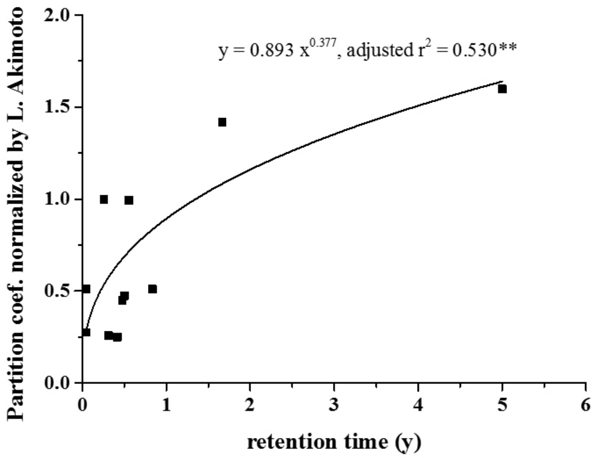
(a)



(b)

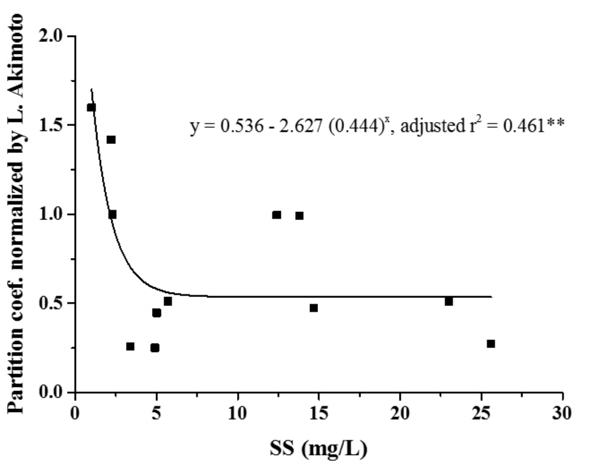


Fig. 7. Lake characteristics vs. partition coefficient (PC). (a) Retention time; (b) suspended solids (SS) concentration in lake water. ** indicates p (significance of adjusted r^2) < 0.01.

Discussion

Because spatial heterogeneity of sediment ^{137}Cs concentrations possibly brought about a slight error in calculating PCs, multiple sampling points in the respective lake were needed. For spatial heterogeneity, the relationship between proportion of fine particles and ^{137}Cs concentrations in sediments was analyzed in each of lakes having at least 3 sampling stations (lakes Inawashiro, Kasumigaura, Teganuma, and Inbanuma). Insignificant correlations in all lakes ($p > 0.05$) were indicated, suggesting spatial heterogeneity of radiocesium supply rather than coupling of particle size effect on adsorption of ^{137}Cs by sediments (He and Walling 1996) and particle-size specific sediment transport.

Two factors, fish species and lake characteristic, are suggested to strongly influence PC values; however, the PC matrix (Table 2) is sparse, so that simple averages of PC values possibly introduce biased results when a relationship exists between fish species and lake characteristics. To overcome this problem, normalization procedures were adopted. Raw PCs values were divided by the PCs of the fish species measured in the greatest number of lakes (i.e., most popular fish species measured in these lakes) or by the PC of the lake having the greatest number of fish species. Silver crucian carp and Lake Akimoto were chosen for the normalization standards of fish species and lake, respectively. The following equations were used to normalize the PC values:

$PC\ (i, j)_{\text{normalized by silver crucian carp}}$ = PC (i, j)/PC (i = silver crucian carp, j), if both PC (i, j) and PC (i = silver crucian carp, j) are not blank; (1)

$PC\ (i, j)_{\text{normalized by silver crucian carp}}$ = blank, if PC (i, j) or PC (i = silver crucial carp, j) is blank; (2)

$PC\ (i, j)_{\text{normalized by Lake Akimoto}}$ = PC (i, j) / PC (i, j = Lake Akimoto) if both PC (i, j) and PC (i, j = Lake Akimoto) are not blank; and (3)

$PC\ (i, j)_{\text{normalized by Lake Akimoto}}$ = blank, if PC (i, j) or PC (i, j = Lake Akimoto) is blank, (4)

where PC (i, j) is PC (i = fish, j = lake); i = 1 to m (number of fish species); and j = 1 to n (number of lake). Subsequently, the averages of $PC\ (i, j)_{\text{normalized by silver crucian carp}}$ in all the lakes (j = 1 to n) except $PC\ (i, j)_{\text{normalized by silver crucian carp}}$ = blank were calculated to yield a fish-normalized PC (Fig. 6a). Similarly, the averages of $PC\ (i, j)_{\text{normalized by Lake Akimoto}}$ (i = 1 to m) were calculated to yield a lake-normalized PC (Fig. 6b).

Higher values of $PC\ (i, j)_{\text{normalized by silver crucian carp}}$ were found in cherry salmon, landlocked salmon, and smallmouth bass, and lower ones were associated with icefish, freshwater prawn, and topmouth gudgeon. Sundbom et al. (2003) proposed 4 parameters to express the changes in fish ^{137}Cs concentrations: the timing (t_{max}) and level (Cs_{max}) of the fish peak concentrations, and the near steady-state level (Cs_{base}) and the long-term decline rate (λ). They reported as follows: (1) t_{max} ranged from 56 to 806 days after the fallout, (2) t_{max} increased with body size, and (3) Cs_{max} increased with fish size but was highest at intermediate trophic levels.

Our results on temporal trends in fish radiocesium concentrations agreed well with Sundbom et al. (2003). Taking into account the temporal changes in fish and sediment ^{137}Cs concentrations, our PC (i, j) values were close to the maximum for each fish species and each lake because the samples were taken within 2 years after the fallout. In addition, the characteristics in fish-normalized PC (i, j) were in close agreement with their results.

The order of PC (i, j) was nearly the same as that from analysis following the Chernobyl accident. For example, the PC values calculated by Kryshev (1995) for ^{137}Cs concentrations in sediments and several species of fish in the Chernobyl NPP Cooling Pond were 0.7–4.2 in 1986, 0.3–1.0 in 1988, and 0.2–2.3 in 1990 for carp, silver bream, silver carp, perch, and pike-perch based on wet-weight. The PC values on a dry basis are close to those on a wet basis because sediments and fish usually have similar water contents. Kryshev et al. (1996) also reported PC values of 0.24 in Kiev Reservoir in 1986, 0.24 in Kanev Reservoir in 1986, and 0.75 in Lake Kozhanovskoe in 1991–1993.

The calculated PC values change according to the reference fish species or lake chosen for normalization. For example, PC calculation was tried using pond smelt and Lake Kitaura (having the second greatest numbers of lakes and fish species, respectively) as the references. The correlation between the averages of PC $(i,j)_{\text{normalized by silver crucial carp}}$ (j = 1 to n) and those of PC $(i,j)_{\text{normalized by pond smelt}}$ (j = 1 to n), and that between averages of PC $(i,j)_{\text{normalized by Lake Akimoto}}$ (i = 1 to m) and those of PC $(i,j)_{\text{normalized by Lake Kitaura}}$ (i = 1 to m) were significant ($r^2 = 0.349$, $p < 0.05$; and $r^2 = 0.358$, $p < 0.05$, respectively), indicating that similar tendencies on PCs would be obtained regardless of the reference fish species or lake (also similarity confirmed for the following analysis on the relationship between lake-normalized PCs and lake characteristics).

The lake-normalized PCs were then analyzed with lake characteristics related to lake dimensions and water exchange (mean depth and water retention time), water quality (suspended solid and electric conductivity), and sediments features (proportion of fine particles and specific density of soil particles). Water retention time had a positive correlation with lake-normalized PC (r = 0.73**). In contrast, suspended solids had a negative correlation (r = −0.40), but not significant ($p > 0.05$). So, several nonlinear models were applied to fit the significant relationships (Figs. 7a and b). For ^{134}Cs, similar significant models were also obtained (retention time: adjusted $r^2 = 0.620$**, suspended solids concentration in lake water: adjusted $r^2 = 0.545$**).

Based on the surveys in 52 lakes of southern Finland in 1988 and 1992, Särkkä et al. (1996) indicated that ^{137}Cs in fish (perch and roach) was positively correlated with that in sediments, and that the perch ^{137}Cs concentration was high when the water retention time of the lake was long. Rask et al. (2012) concluded that the transfer of ^{137}Cs into fish increased with lower electrolyte concentrations, longer water retention time, and lower sedimentation rate based on data from small forest lakes in southern Finland. These results support the relationship (Fig. 7a), suggesting that longer water retention time results in prolonged contact and equilibration between the water and the sediments. Another possibility is that PCs decrease through fast sedimentation of particles with high radiocesium concentration carried during rainfall events, during which proportions of total inputs are expected to be larger in lakes with shorter retention time.

The unclear relationship between lake-normalized PCs and electrical conductivity observed in this study ($r^2 = 0.0001$) may be understandable on the following basis. Several studies (e.g., Putyrskaya et al. 2009) reported that desorption of radiocesium from sediments was induced by the enhanced concentration of competing ions, possibly related to electrolyte concentration. Thus, the increase in electrolyte concentration would bring about an increase of radiocesium in lake water while decreasing its transfer to fish, resulting in an unclear trend in PC with electric conductivity. The negative relationship between PCs and suspended solids in lake water would indicate negligible influence of detritus feeding on PCs. The high correlation between the reciprocal of water retention time and suspended solids concentration (r = 0.82, $p < 0.05$) and/or the suppressing influence of suspended solids on lake water radiocesium concentration would cause the negative relationship, but further investigation is warranted. As to the influence of pH, Poinssot at al. (1999) reported that Cs sorption on one type of clay minerals (illite) decreased at low pH but was nearly constant in the range of pH = 6–9 at low electrolyte concentrations. In our survey campaign, pH was not measured together with radiocesium concentration, but assuming negligible influence of pH on fish uptake of Cs, the effect of pH on PCs seemed not influential because the values of pH observed in these lakes (National Institute for Environmental Studies 2013) distributed mostly within the 7 to 9 pH range.

Finally, the efficacy and problems associated with the use of the partition coefficient between sediments and fish should be noted. Most important is the relative simplicity of determining this partition coefficient because the partition coefficient of radiocesium between fish and lake water and that between sediments and lake water are usually difficult to measure. The difficulty arises due to the low radiocesium concentration in lake water long after the fallout, thus facilitating the predicted fish radiocesium concentrations (need great labor) using sediment radiocesium concentration and this partition coefficient. Two potentially problematic points are noteworthy; the first is a question as to the direct relationship between sediments and fish, probably reflecting their relationships with water concentration; the second is the possibility of long-term changes in the PC values. Based on the data reported by Särkkä et al. (1996), the changes

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in fish ^{137}Cs from 1988 to 1992 (perch 26%, pike 28%, roach 39%; the ratio of 1992 to 1988) were similar to those in sediments (27% estimated by the ^{137}Cs inventory). Based on the long-term trend in ^{137}Cs concentrations in brown trout, Brittain and Gjerseth (2010) suggested a dynamic equilibrium between catchment input of ^{137}Cs and possible remobilization from lake sediments, lake outputs, and concentrations in fish; however, Kryshev (1995) reported PC values between sediments and fish of 0.18–1.06 in 1986, 0.25–1.00 in 1988, and 0.06–0.57 in 1990 (on a wet-weight basis), indicating a decreasing tendency. Thus, further surveys are needed on long-term change in PCs.

Conclusions

The database on radiocesium concentrations in sediments and 18 species of fish measured in 15 lakes in northeastern Japan were analyzed to evaluate the influences of the 2011 Fukushima nuclear accident on lake sediments and fish and to understand differences in their contamination. The temporal trends in fish radiocesium concentrations from 2011 to 2013 were fish-size dependent. Positive correlations between lake-averaged radiocesium concentrations of sediments and fish were observed for almost all species of fish, and on this basis normalized partition coefficients between sediments and fish concentrations were proposed. Results showed that the values of PC clustered in a range from 0.3 to 3, and their relations to fish types and lake characteristics were statistically analyzed. More investigations are needed on the long-term stability of lake- and fish-normalized coefficients and their future incorporation in modeling studies.

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