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Seasonal dynamics of the activities of dissolved ^{137}Cs and the ^{137}Cs of fish in a shallow, hypereutrophic lake: Links to bottom-water oxygen concentrations

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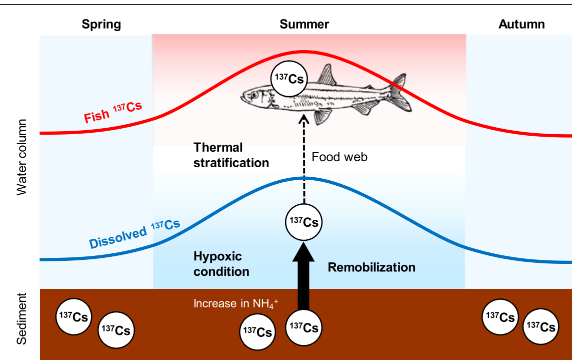
HIGHLIGHTS

- Intermittent hypoxic events may enhance ^{137}Cs remobilization in shallow lakes.
- We studied a lake where DO changes in response to meteorological conditions.
- Bottom dissolved oxygen concentrations determined dissolved ^{137}Cs dynamics.
- Dissolved ^{137}Cs and ^{137}Cs in fish showed a seasonal pattern with a summer peak.
- ^{137}Cs remobilized from sediments influenced seasonal changes of the ^{137}Cs in fish.

GRAPHICAL ABSTRACT



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ABSTRACT

Remobilization of radiocesium from anoxic sediments can be an important mechanism responsible for long-term contaminations of lakes. However, it is unclear whether such remobilization occurs in shallow lakes, where concentrations of dissolved oxygen in the hypolimnion (bottom DO) change temporally in response to meteorological conditions, and whether remobilized radiocesium influences the activity in fish. We examined the seasonal dynamics of the activities of dissolved ^{137}Cs and ^{137}Cs in fish (pond smelt and crucian carp) from Lake Kasumigaura, a shallow, hypereutrophic lake, five years after the Fukushima Daiichi Nuclear Power Plant accident. The activities of both dissolved ^{137}Cs and ^{137}Cs in fish declined during that time, but the declines showed a clear seasonal pattern that included a summer peak of ^{137}Cs activity. The activity of dissolved ^{137}Cs increased when the bottom DO concentration decreased, and a nonlinear causality test revealed significant causal forcing of dissolved ^{137}Cs activity by bottom DO. The fact that NH_4-N concentrations in bottom waters were higher in the summer suggested that remobilization of ^{137}Cs from sediments could result from highly selective ion-exchange with NH_4-N. Despite the shallow depth of Lake Kasumigaura, winds had little influence bottom DO concentrations or dissolved ^{137}Cs activities. The fact that seasonal means of ^{137}Cs activities in pond smelt and crucian carp were positively correlated with the seasonal means of dissolved ^{137}Cs activities suggested that remobilized ^{137}Cs may have influenced the seasonal dynamics of radiocesium in fish through food-chain transfer, but higher feeding rates in warm water could may have also contributed to the seasonal dynamics of ^{137}Cs activity in fish.

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Our findings suggest that in shallow lakes, intermittent but repeated hypoxic events may enhance remobilization of radiocesium from sediments, and remobilized radiocesium may contributed to long-term retention of radiocesium in aquatic organisms.

1. Introduction

The release of large amounts of radionuclides as a result of accidents at nuclear power plants can cause large-scale contamination of aquatic ecosystems (Nakata and Sugisaki, 2015; Okamura et al., 2016; Rowan et al., 1998; Sundbom et al., 2003). Long-term studies of the Chernobyl and Fukushima nuclear power plant accidents have clearly shown that contamination of freshwater fish by radiocesium can persist in lakes for many years (Jonsson et al., 1999; Smith et al., 2000; Wada et al., 2016). The rapid decline in the levels of radiocesium in fish during 3–4 years after a nuclear power plant accident is accounted for by hydraulic dilution, accumulation in bottom sediments, reduced runoff from the lake catchment, and loss of radiocesium through outflow (Jonsson et al., 1999; Smith et al., 1999). After the first few years, however, the net removal rate of radiocesium declines, mainly because of radiocesium recycling within the lake ecosystem (Brittain and Gjerseth, 2010; Jonsson et al., 1999; Smith and Comans, 1996; Sundbom et al., 2003). When the radiocesium activities in water and fish vary seasonally, lack of understanding of the processes and drivers of those seasonal dynamics may delay cancellation of restrictions on fish consumption (Matsuzaki et al., 2016). Although it has been nine years since the Fukushima Daiichi Nuclear Power Plant (FDNPP) accident on 11 March 2011, radiocesium activities of some species of freshwater fish in Fukushima, Ibaraki, and other prefectures still exceed the Japanese regulatory limit of 100 Bq/kg (Ishii et al., 2020; Wada et al., 2016). Understanding the factors and mechanisms that determine the seasonal dynamics of radiocesium in lakes can enable more informed predictions of long-term trends of fish radiocesium concentrations and allow earlier decision-making with confidence (Buesseler, 2012).

Although radiocesium binds strongly to clay minerals in sediments, earlier work has suggested that radiocesium can be remobilized from anoxic sediments to overlying water by diffusion. This remobilization may be one of the most important mechanisms by which radiocesium recycling is enhanced in lakes (Comans et al., 1989; Smith and Comans, 1996). One of the major pathways of radiocesium remobilization is highly selective ion exchange in anoxic sediments with ammonium ions (NH_4^+) that have a similar ionic radius (Comans et al., 1989; Evans et al., 1983). The transfer of sediment-bound radiocesium to benthic animals through benthic food chains could be another pathway (Wang et al., 2016). In deep lakes, the occurrence of intense summer thermal stratification creates persistent anoxic conditions in the hypolimnion, and NH_4^+ reaches high concentrations in anoxic pore waters. These high NH_4^+ porewater concentrations facilitate ion exchange with radiocesium, and increase the remobilization of radiocesium from anoxic sediments. This remobilization of radiocesium can lead to a seasonal cycle of radiocesium activities in the water column (Davison et al., 1993; Dominik and Span, 1992; Evans et al., 1983). However, these previous studies, albeit extensive, were conducted in deep lakes. Although Comans et al. (1989) have suggested that analogous remobilization may occur in shallow lakes, it remains unclear how large a role such remobilization plays in the seasonal dynamics of radiocesium in the water and fish. Although shallow lakes are mixed frequently (i.e., they are polymictic), they can be temporarily stratified by meteorological conditions such as summer heat waves and calm weather (Bartosiewicz et al., 2016; Deng et al., 2018; Jalil et al., 2018; Martinsen et al., 2019; Taguchi et al., in press; Welch and Cooke, 2005). In the case of a shallow, hypereutrophic lake, even temporary stratification may lead to oxygen depletion in bottom waters and increase the mobility of radiocesium. Because thermally stratified shallow lakes are easily destratified by wind stress and surface heat fluxes (Imberger, 1985; Masunaga and Komuro, 2020), remobilized radiocesium may be more quickly transported to the epilimnion and taken up by aquatic biota in shallow lakes versus deep lakes.

Remobilized radiocesium can be taken up by fish through the lake food web, but few empirical studies have assessed the relationship between seasonal variations in the concentrations of dissolved radiocesium and that in fish. Previous studies have reported a seasonal dynamics of radiocesium levels in fish, but those studies have focused only on the effects of ecological and physiological factors, including fish size, diet, and temperature (Brittain and Gjerseth, 2010; Lonvik and Koksvik, 1990; Peles et al., 2000; Ugedal et al., 1997; Ugedal et al., 1992). For example, because physiological considerations suggest that the rate of elimination of radiocesium by fish should increase with increasing water temperature, fish would be expected to contain less radiocesium in summer. However, if bottom waters are depleted of oxygen in the summer, radiocesium levels in fish may temporarily increase as a result of increased remobilization of radiocesium. Focusing on the seasonal dynamics of both dissolved radiocesium and that in fish is thus essential for understanding the mechanisms that control long-term retention of radiocesium in the fish of shallow lake ecosystems.

In this study, we examined the long-term declines and seasonal dynamics of the activities of ^{137}Cs dissolved in the water and ^{137}Cs in fish of Lake Kasumigaura, a shallow, hyper-eutrophic lake in Japan. Our goal was to explore the role of ^{137}Cs remobilization from the sediments of that lake. We measured dissolved ^{137}Cs activities every three months at three stations over a period of five years (2011—2016) after the FDNPP accident and analyzed the relationships between those ^{137}Cs activities and environmental factors (surface-water temperature and bottom water DO concentrations). We used convergent cross-mapping (CCM), a nonlinear causality test (Sugihara et al., 2012), to determine the presence and direction of causal relationships between environmental variables and dissolved ^{137}Cs activities. To determine whether NH_4^+ concentrations in bottom-waters (bottom NH_4^+) increased during the summer, we also sampled bottom waters just above the sediment during the study period and examined the relationship between concentrations of bottom NH_4^+ and dissolved ^{137}Cs activity. In addition, we used the monitoring databases gathered and published by the Ministry of Health, Labor, and Welfare of Japan after the Fukushima Nuclear Power Plant accident, determine whether there were seasonal changes of the ^{137}Cs activities in planktivorous pond smelt (*Hypomesus nipponensis*, Fig. 1) and omnivorous crucian carp (*Carassius*. spp., Fig. 1) in Lake Kasumigaura during 2011–2016. We compared the seasonal patterns of the activities of the ^{137}Cs dissolved in the water and the ^{137}Cs in fish. Because oxygen depletion occurs intermittently in the bottom waters of Lake Kasumigaura, especially during the summer (Hosomi and Sudo, 1987; Kamiya et al., 2017; Masunaga and Komuro, 2020), we hypothesized that even temporary oxygen depletion during the summer would enhance the production of NH_4^+ in the sediments and subsequently the release of ^{137}Cs from the sediments. The result would be seasonal variations of the activities of dissolved ^{137}Cs and the ^{137}Cs in fish.

2. Materials and methods

2.1. Study lake

Lake Kasumigaura, located approximately 60 km northeast of Tokyo, is the second-largest lake in Japan, with a surface area of 167.7 km^2, a

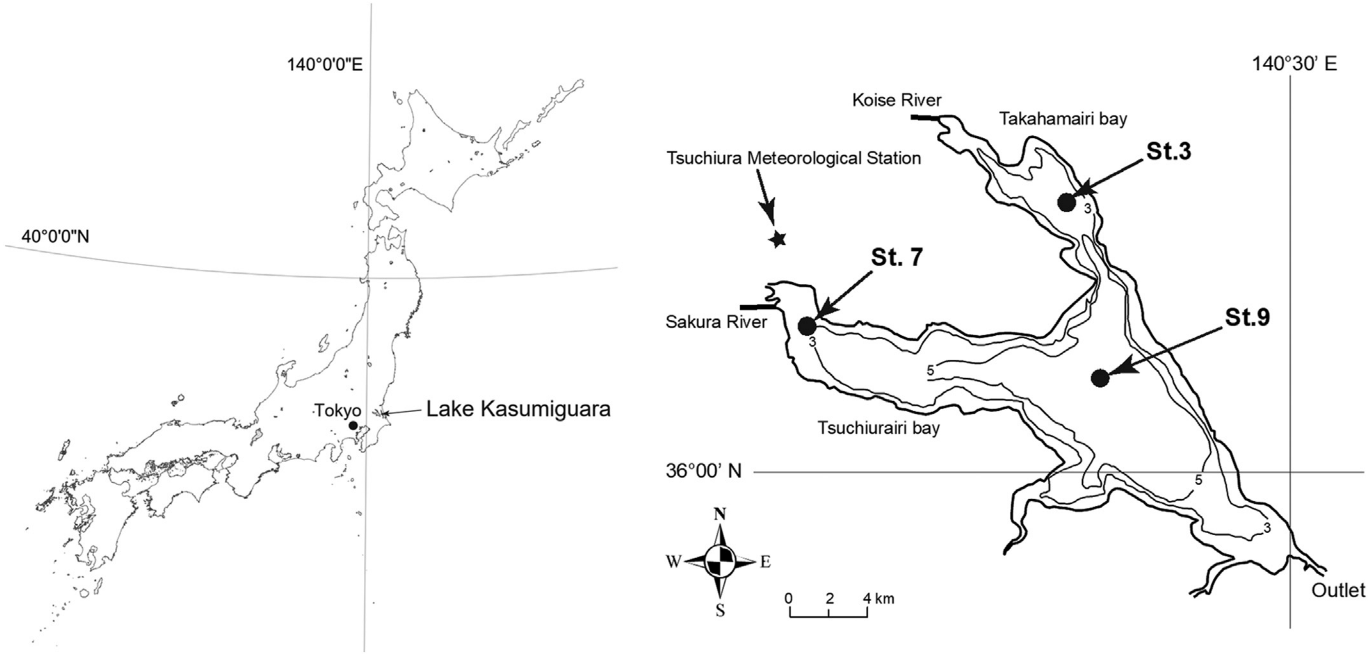

Fig. 1. Maps of Lake Kasumigaura within Japan and the sampling stations in Lake Kasumigaura. The numbers of the stations correspond to the numbers of the Lake Kasumigaura Long-term Monitoring Project. Station 9 is at the center of the lake. Depth contours are in meters. The Sakura River and Koise Rivers are the two major tributary rivers. The star indicates the position of the Tsuchiura Meteorological Station.

total volume of 622 million m^3, a mean depth of approximately 4 m, and a maximum depth of 7.4 m. The average water residence time is approximately 200 days. Because it receives extremely high loads of organic matter and nutrients, Lake Kasumigaura is a well-known hypereutrophic lake. The National Institute for Environmental Studies (NIES) has been conducting monthly monitoring of Lake Kasumigaura since 1976 as part of the Lake Kasumigaura Long-term Monitoring Project (Matsuzaki et al., 2018; Takamura and Nakagawa, 2012). The lake is also registered as a core site of the Japan Long-term Ecological Research Network (JaLTER). The monitoring datasets are available from the Lake Kasumigaura Database (http://db.cger.nies.go.jp/gem/moni-e/inter/GEMS/database/kasumi/index.html).

2.2. Water sampling

We collected water samples of ~20 L from depths of 0–2 m for ^{137}Cs analysis at three stations in Lake Kasumigaura (Stations 3, 7, and 9, Fig. 1) with a 2 m-long column sampler (diameter, 50 mm). The water depths at stations 3, 7, and 9 were approximately 4.3 m, 3.4 m, and 6.3 m, respectively. After the FDNPP accident, we collected water samples at approximately three-month intervals between the summer of 2011 and the spring of 2016 (20 times in total at each station). Most of the samples were collected on the routine monthly monitoring dates, although several surveys were conducted at different times, but within one week of the monitoring dates. For surface water temperature and DO data, we used monthly monitoring data from the Lake Kasumigaura Long-term Monitoring Project. During this routine monitoring, water temperature and DO were measured every 1 m from the surface to near the bottom (ca. 10 cm from the sediment) at each station. We used water temperatures at a depth of 1 m and DO concentrations near the bottom in the subsequent statistical analyses.

To examine the seasonal changes of ammonium (NH_4-N) concentrations in bottom waters, bottom waters were collected from just above the sediment at the three stations during the routine monthly monitoring throughout the study period. For logistical reasons, bottom water sampling was not conducted during May 2013 and May 2014. Sediment cores were collected using a gravity core sampler with an acrylic pipe and acrylic tube (10-cm diameter, 50-cm length) and subsequently we collected bottom water from just above the sediment (approximately 5 cm from the sediment) with a syringe. The collected bottom water samples were immediately filtered on the boat through a precombusted GF/F glass fiber filter (pore size: 0.7 μm). The water samples were transported back to the laboratory in an ice chest. NH_4-N concentrations were determined using an auto-analyzer (Quattro–2HR, BLTEC, Tokyo, Japan). All the analyses were performed in accord with the directions in the manufacturer's manual.

2.3. Wind data

Windy conditions can increase sediment resuspension by mixing the water column. To examine the effect of the wind on bottom DO concentrations and activities of dissolved ^{137}Cs, we obtained wind velocity data on the water sampling dates from a meteorological station in Tsuchiura, which is near Lake Kasumigaura. We used daily mean and maximum wind velocity.

2.4. Radiocesium measurement of water samples

In the laboratory, we sequentially filtered batches of ~200 mL of the 20 L lake water samples onto Whatman GF/C glass fiber filters (1.2-μm pore size; GE Healthcare Whatman). The filtrates were further concentrated with an Empore Caesium Rad Disk (3 M Japan, Ltd., Tokyo, Japan) to trap dissolved radiocesium (Tsuji et al., 2019). Each of the Rad Disks was placed in a U8 container (90-mL volume). The ^{137}Cs activities in the Rad disks were measured with a pair of coaxial, high-purity germanium detectors (GC2518 and GCW7023, Mirion Technologies Canberra Japan Co. Ltd., Tokyo, Japan) using Spectrum Explorer analysis software (Canberra Japan Co. Ltd.) and a set of gamma ray detectors (GWL-450-15-S, LOAX-70550/30, GEM65P4-83, and GMX45P4-76; Seiko EG&G Co. Ltd.) using the Gamma Studio software (Seiko EG&G Co. Ltd., Tokyo, Japan). The counting time for each sample was at least 50,000 s. The detection limit (below 0.01 Bq/L) was defined as the activity equal to three standard deviations (i.e., 3σ) of the background counts. The ^{137}Cs radioactivity of each sample was decay-corrected to the sampling day. Efficiency of each detector was calibrated with standard volume radioactivity sources and filter-shaped standard source (MX033U8PP, The Japan Radioisotope Association).

2.5. Fish radiocesium monitoring data

The Ministry of Health, Labor, and Welfare (MHLW) of Japan has been intensively monitoring the radioisotope contamination in various foods, including freshwater fish, since the FDNPP accident in March 2011 to prevent highly contaminated foods from being distributed in the market. From the MHLW database (https://www.mhlw.go.jp/english/topics/2011eq/index_food.html), we collected radiocesium measurement data for freshwater fish in Lake Kasumigaura from July 2011 to March 2016. We focused on the ^{137}Cs activities in pond smelt and crucian carp for the following three reasons. First, these fish are among the most important species in the Lake Kasumigaura Fishery, and there are radiocesium monitoring data available for these species. Second, pond smelt and crucian carp are the dominant planktivorous and omnivorous fish in Lake Kasumigaura, respectively (Matsuzaki et al., 2018). Third, these two fish species have different diets, habitats, and life histories. Pond smelt is a small fish (~10 cm in length as an adult) and planktivorous, and its lifespan is one year (Sakamoto et al., 2014). In contrast, crucian carp is a relatively large fish (25 cm in length as an adult) and omnivorous, and its lifespan is ~10 years. Whereas pond smelt prefer pelagic habitats, crucian carp prefer benthic and littoral habitats. Ibaraki Prefecture has collected pond smelt and crucian carp samples from Lake Kasumigaura in accord with the MHLW radiocesium monitoring scheme. However, those collections include few crucian carp sampled during 2011 and 2013. The adult fishes have been caught randomly from Lake Kasumigaura (the collection sites have not been unpublished). Whole bodies of pond smelt and whole bodies or muscle tissue of crucian carp were pooled for radiocesium analysis (some individuals are treated as one sample) and radiocesium activities were measured with a germanium gamma-ray spectrometer.

2.6. Statistical analyses

We fitted the time series of dissolved ^{137}Cs and fish ^{137}Cs activities using a single-component exponential decay model and a two-component exponential decay model with a fast (first) and slow (second) component (Jonsson et al., 1999; Smith et al., 2000; Suzuki et al., 2018).

$$\text{Single-component model}: Q_t = Qe^{-kt}$$

$$\text{Two-component model}: Q_t = Q_1 e^{-k1} t + Q_2 e^{-k2} t$$

where t is the number of elapsed days since 11 March 2011; Q is the ^{137}Cs activity at time $t = 0$ (initial value) of the single-component model, and Q_1 and Q_2 are the ^{137}Cs activities at time $t = 0$ of the fast and slow components in the two-component model, respectively; Q_t is the ^{137}Cs activity after t days; k, k_1, and k_2 are the decay rate constant (day^{-1}) of the single-component model, and the decay rate constants of the fast and slow components of the two-component model, respectively. All the parameters of the decay models were estimated using Sigma Plot 14.0 (Systat Software, Inc. U.S.A). We selected the best model based on r^2 values. When r^2 was higher for the two-component model, the rate of decline of dissolved ^{137}Cs or fish ^{137}Cs activity decreases over time. Using the k, k_1, or k_2 values, we also estimated the environmental half-life (T_{env}) of ^{137}Cs in the water and the ecological half-life (T_{eco}) of ^{137}Cs in fish with the following equation:

$$T_{env} \text{ or } T_{eco} = \ln(2)/(k, k_1, \text{or } k_2)$$

We used multiple regression analysis to identify factors that explained seasonal variations of the activities of dissolved ^{137}Cs and ^{137}Cs in fish. To consider temporal autocorrelation, we used a generalized least-squares regression (GLS) model that included a term to correct for first-order autoregressive processes, AR(1). For dissolved ^{137}Cs activities, we included sampling year, monitoring station, surface water temperature, and bottom DO concentrations as explanatory variables. Because there was high collinearity between surface water temperatures and bottom DO concentrations ($r^2 = 0.88$, Fig. S1), we used only bottom DO concentrations in the GLS model. For the activity of ^{137}Cs in fish, we considered the effects of sampling year and season (spring/summer/autumn/winter) in the model. The activities of dissolved and ^{137}Cs in fish were log-transformed to better meet the assumption of normality. Maximum likelihood was used for estimating the model parameters. GLS models were analyzed using the *gls* function from the *nlme* package (Pinheiro et al., 2014) in R (version 3.2.3).

Finally, we applied CCM to determine the presence and direction of causal relationships between dissolved ^{137}Cs activities and environmental variables. Details of the CCM algorithm can be found in Sugihara et al. (2012). Briefly, CCM is based on an algorithm that compares the ability of lagged components of one process to estimate the dynamics of another. Effectively, if variable X is influencing a paired observed variable Y, then based on the generalized Takens' theorem, we can expect that variable X can be reliably predicted from the time-series history of variable Y (Sugihara et al., 2012). Thus, CCM measured the extent to which the recent historical record of the affected variable Y reliably estimates states of a causal variable X. This prediction skill (ρ) is called the cross-map skill and is quantified by calculating the Pearson correlation coefficient between the predicted and observed values of X. The value of ρ is expected to improve with the length of the time series L until it converges to a maximum level if two variables are causally coupled (i.e., convergence).

CCM can be applied to time series of roughly 30 or more sequential observations (Sugihara et al., 2012). Ecological time series, however, are often shorter, and the application of CCM to such data is challenging. However, Clark et al. (2015) have recently developed a method (multispatial CCM) that expands the application of CCM to short time series that are spatially replicated (e.g., data from multiple plots). We applied the multispatial CCM algorithm to our data, because our time-series data at each station were short ($N = 20$). We assumed that data from the three stations came from the same dynamical system ($N = 60$) because dissolved ^{137}Cs activities and the values of environmental variables did not differ among the sampling stations (Fig. 2). We confirmed that the ρ values were improved with increasing sample length (i.e., $L = 20$, $L = 40$, and $L = 60$). Before the CCM analysis, all time-series data were first-differenced, meaning the first value of the series was subtracted from the second value, the second value from the third value, and so on, to remove trends.

We used a simplex-projection to determine the optimal embedding dimension (E) (Sugihara et al., 2012; Tsonis et al., 2015). The selection of E was based on leave-one out cross-validation by removing one observation from the time series and using the rest of the time series to predict its state. The best E for each time series was determined from 2 to 8 dimensions according to the prediction skill (here the dimension with the highest Pearson correlation coefficient, ρ). To investigate nonlinearity of a system, we analyzed the relationships between predictor power and prediction interval (the amount of time in advance for which the prediction is made), because if the system is nonlinear, then predictive power should significantly decrease as the prediction interval increases (i.e., negative correlation) (Clark et al., 2015). We confirmed that all the variables displayed nonlinear dynamics.

We then calculated ρ to determine the presence and direction of the causal interactions among the first-differenced variables. The multiple CCM applies a non-parametric bootstrapping method to test the statistical significance of causal relationships (Clark et al., 2015). This bootstrap method tests whether the value of ρ is greater at the longest available L than at the shortest available L and whether the ρ at the longest L (ρ_{Lmax}) is greater than zero. Note that the longest L is determined by data availability, whereas the shortest L is determined by E. The

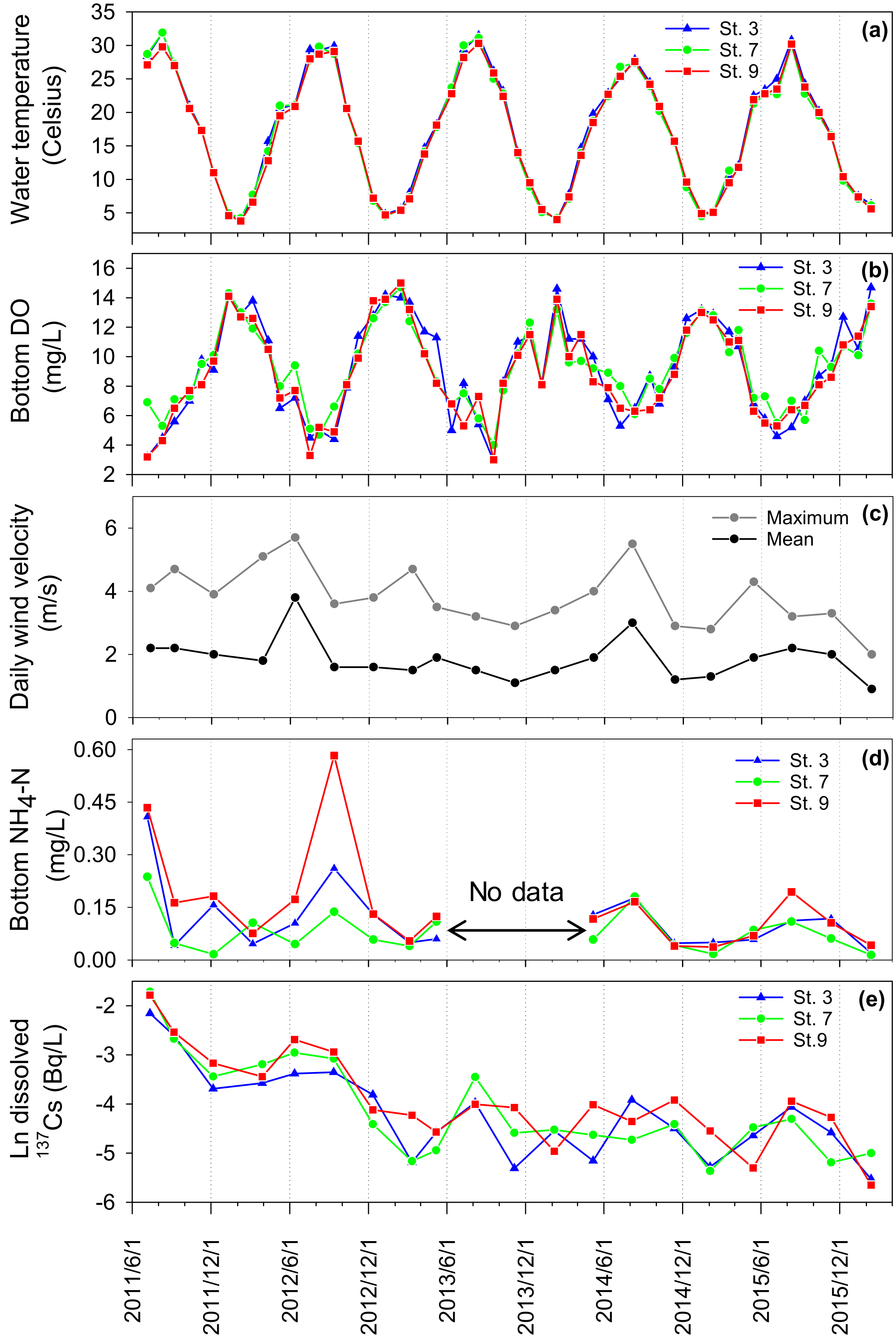

Fig. 2. Temporal changes of (a) surface-water temperature, (b) bottom DO concentration, (c) daily wind velocity, (d) NH_4-N concentration in bottom waters just above the sediments, and (e) dissolved ^{137}Cs activity from July 2011 to February 2016 at the three stations in Lake Kasumigaura. Note that there were no NH_4-N data from May 2013 to May 2014.

number of bootstrapped iterations was 10,000 for all CCM estimates. The multispatial CCM was performed in the R programming language using the R package *multispatialCCM* (Clark et al., 2015).

Seasonality can lead to misidentification of causality. In addition to CCM, we therefore performed partial cross mapping (PCM) (Leng et al., 2020) to distinguish causal relationships from seasonality-driven synchronization. The PCM integrates three basic data analysis methods from nonlinear dynamics and statistics: phase-space reconstruction, cross mapping, and partial correlation. The integration of these methods enables the PCM to identify direct causations from indirect ones in cases where the variables of the underlying dynamical system are non-separable and weakly or moderately interacting. In our case, water temperature changed seasonally and could influence other variables. We used the PCM to determine whether there was the direct causation between bottom DO concentrations and dissolved ^{137}Cs activities by eliminating the indirect causal influences of water temperature on those variables. The PCM analysis was conducted with Mathematica (version 10.2, Wolfram Research, Inc., Champaign, Illinois, USA).

3. Results and discussion

3.1. Dissolved ^{137}Cs dynamics

Dissolved ^{137}Cs activities at all three stations declined rapidly during the first two years after the FDNPP accident, but the rates of decline subsequently slowed (Fig. 2e). The two-component model explained the variance of dissolved ^{137}Cs better than the single-component model (Table S1, Fig. S2a). The T_{env} based on the short-term decay rate was 27 days, but the T_{env} based on the long-term decay rate was 533 days (Table S1). Dissolved ^{137}Cs activities declined over the study period, but also changed seasonally. The GLS models showed that the seasonal changes of the dissolved ^{137}Cs activities were related to bottom DO concentrations (Table 1). Bottom DO concentrations were low every summer (Fig. 2b), and dissolved ^{137}Cs activities (i.e., the residuals after controlling for the year) increased when bottom DO concentrations decreased (Fig. 3a). Furthermore, the CCM identified a significant causal forcing of dissolved ^{137}Cs activity by bottom DO concentrations (Table 2, Fig. S3). Bottom DO concentrations were in turn forced by surface water temperature, but dissolved ^{137}Cs activities were not directly forced by surface-water temperatures. The PCM identified a significant causal relationship between bottom DO concentrations and dissolved ^{137}Cs activities (partial correlation coefficient = 0.495, $P = 0.004$) even when the indirect effects of water temperature on those variables were removed (Table S2). Our results clearly suggested that bottom DO concentrations influenced the seasonal dynamics of dissolved ^{137}Cs activities.

Table 1
Summary of the GLS models explaining temporal variations in dissolved ^{137}Cs ($N = 60$), pond smelt ^{137}Cs ($N = 77$), and curucian carp ^{137}Cs ($N = 78$) in Lake Kasumigaura. Significant effects ($P < 0.05$) are shown in bold.

Predictors	Estimate	SE	P
Dissolved ^{137}Cs			
Year	**−0.43**	**0.05**	**<0.001**
Station (St. 3 vs. St. 7)	0.10	0.17	0.548
Station (St. 3 vs. St. 9)	0.24	0.17	0.161
Bottom dissolved oxygen	**−0.11**	**0.02**	**<0.001**
Pond smelt ^{137}Cs			
Year	**−3.61**	**0.69**	**<0.001**
Season (autumn vs. spring)	2.53	2.58	0.331
Season (autumn vs. summer)	**4.82**	**1.90**	**0.013**
Season (autumn vs. winter)	1.85	2.16	0.393
Crucian carp ^{137}Cs			
Year	**−16.51**	**1.33**	**<0.001**
Season (autumn vs. spring)	**13.38**	**2.43**	**<0.001**
Season (autumn vs. summer)	**12.82**	**2.67**	**<0.001**
Season (autumn vs. winter)	0.27	4.09	0.948

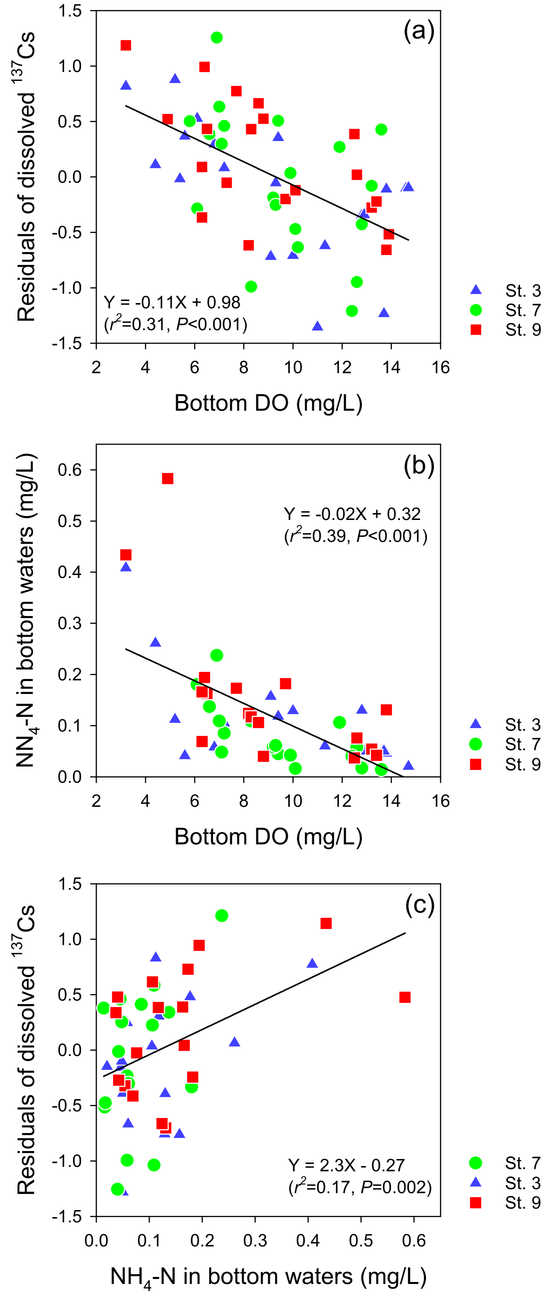

Fig. 3. (a) The relationship between bottom DO concentrations and the residuals from the GLS model of dissolved ^{137}Cs activities against the sampling year. (b) Relationships between bottom DO concentrations and bottom NH_4-N concentrations. (c) Relationships between bottom water NH_4-N concentrations and the residuals from the GLS model of dissolved ^{137}Cs activities against the sampling year.

Earlier studies of deep lakes have demonstrated that radiocesium can be remobilized from sediments to the overlying waters through highly selective ion exchange with NH_4^+ in anoxic sediments, and that remobilization can lead to seasonal changes of dissolved radiocesium activities (Alberts et al., 1979; Davison et al., 1993; Dominik and Span, 1992; Evans et al., 1983; Pinder et al., 2010). Evans et al. (1983) found a positive relationship between NH_4-N concentrations in bottom waters and the percent of ^{137}Cs released from sediments. We observed that the NH_4-N concentrations in bottom water just above the sediment (bottom NH_4-N) increased in summer, although some data were missing (Fig. 2d). The fact that bottom NH_4-N concentrations increased significantly with decreasing bottom DO concentrations (Fig. 3b, Table S3) indicated that low bottom DO concentrations enhanced production and

Table 2
Summary of results of the convergent cross-mapping (CCM) for the relationships between variables. *E* is the best embedding dimension. ρ_{Lmax} is the cross-map skill (ρ, Pearson correlation coefficient between observed and predicted values) obtained at the maximal library length (*Lmax*). The statistical significance of the CCM was determined via a bootstrap test with 10,000 iteration. Bold indicates significance at P < 0.05.

X	Y	Cross-map skill (X → Y)			Cross-map skill (Y → X)		
		E	ρ_{Lmax}	P	*E*	ρ_{Lmax}	P
Surface water temperature	Bottom dissolved oxygen	2	**0.96**	**<0.001**	7	0.91	0.091
Surface water temperature	Dissolved ^{137}Cs	2	0.50	0.131	7	0.41	0.349
Bottom dissolved oxygen	Dissolved ^{137}Cs	7	**0.80**	**0.016**	7	0.45	0.319

accumulation of NH_4^+ in pore waters. Hosomi and Sudo (1987) have reported that NH_4^+ concentrations in pore waters of Lake Kasumigaura increased dramatically in summer. A significant positive relationship was found between bottom water NH_4-N concentrations and dissolved ^{137}Cs residuals after controlling for year (Fig. 3c, Table S3). Our results indicate that remobilization of ^{137}Cs occurs through highly selective ion exchange with NH_4^+. However, we note that the significant correlation between NH_4^+ and bottom DO concentrations is insufficient to show that there is a cause-and-effect relationship between the increase in NH_4^+ concentrations under hypoxic conditions and the dynamics of dissolved ^{137}Cs.

Pinder et al. (2010) have showed that remobilization of radiocesium did not occur in a shallow reservoir (maximum and mean depths: 4 m and 1.6 m, respectively), because the reservoir was too shallow to maintain a hypoxic hypolimnion for more than a short time. However, some studies of shallow lakes have reported that transient oxygen depletion of bottom waters may occur during the summer when thermal stratification is strong (Bartosiewicz et al., 2016; Deng et al., 2018; Martinsen et al., 2019). Furthermore, Taguchi et al. (in press) recently reported that even shallow eutrophic ponds (mean depths <2 m) are strongly stratified with persistent hypolimnetic anoxia during the summer when warmer temperatures promote stratified conditions. Although bottom DO concentrations below 2 mg/L were not observed in this study (Fig. 2b), the bottom waters of Lake Kasumigaura become temporarily hypoxic in the summer when stratification is intense and strong winds do not blow for several days (Hosomi and Sudo, 1987; Kamiya et al., 2017; Masunaga and Komuro, 2020). More serious oxygen depletion, including short periods of anoxia, is likely to occur within a thin layer at the top of the sediment (i.e., at the sediment-water interface) in hypereutrophic bodies of water similar to Lake Kasumigaura. Hypoxic conditions in Lake Kasumigaura have been reported to disappear when the water column is mixed by strong winds or weak heat flux (Masunaga and Komuro, 2020). In shallow lakes, alternating periods of stratification and mixing can occur during the day and night in spring and summer (Andersen et al., 2017; Imberger, 1985). We found that surface-water temperatures were a more influential driver of bottom DO concentrations than wind velocity (Table S4). Results indicate that temperature-induced, transient but repeated hypoxic events during summer can increase the mobilization of ^{137}Cs in shallow lakes.

In fact, there was no significant relationship between wind velocity and dissolved ^{137}Cs activities (Fig. S4). Conditions were calm during all sampling dates (and during the five days before each sampling date) and the daily mean and maximum wind velocities were less than 3.8 m/s and 5.7 m/s, respectively (Fig. 2c). In accord with Shinohara et al. (2016), we calculated the significant wavelength and bottom shear stress at each sampling station (Text S1). Our calculation showed that wind-driven sediment resuspension was unlikely to occur over the range of observed wind velocities (Fig. S5). Therefore, the effects of wind waves on sediment resuspension would be small, at least on our sampling dates. However, the effects of strong winds and typhoon events on the resuspension of sediment-bound ^{137}Cs (Robbins and Jasinski, 1995) and the bioavailability of sediment-bound and particulate ^{137}Cs (Wang et al., 2016) should be examined in detail.

3.2. Fish ^{137}Cs dynamics

The ^{137}Cs activities of both pond smelt and crucian carp gradually declined between 2011 and 2012, but the rate of decline was much slower after 2013 (Fig. 4a, c). For pond smelt, the two-component model was better than the single-component model at predicting the decline of ^{137}Cs activity (Table S1, Fig. S2). Although the two-component model was also slightly better for ^{137}Cs activity in crucian carp, the difference in r^2 between the single- and two-component models was small because the sample sizes were low during 2011 and 2013. The T_{eco} values for the short-term and long-term decay rates of the ^{137}Cs activity in fish were 94 days and 3466 days, respectively, for pond smelt, and 365 days and 990 days, respectively, for crucian carp (Table S1). These slow rates of elimination were consistent with previous studies that have reported the ecological half-lives of ^{137}Cs in pond smelt in Japanese lakes after the FDNPP accident (Suzuki et al., 2018; Wada et al., 2016).

However, despite the ecological differences in pond smelt and crucian carp, the ^{137}Cs activities in both fish species showed a clear seasonal pattern (Table 1, Fig. 4b, d). The ^{137}Cs activities in pond smelt were significantly higher in summer than in other seasons, and even after the year effect was removed, and the ^{137}Cs activities in crucian carp also had significantly higher in spring and summer than other seasons. This seasonality was consistent with the patterns of dissolved ^{137}Cs activity (Fig. 2e). The GLS models showed significant positive correlations between seasonal mean dissolved ^{137}Cs activities and seasonal mean fish ^{137}Cs activities (Fig. 5, pond smelt $P < 0.001$, crucian carp $P < 0.001$); indicating that remobilization of ^{137}Cs from the sediments affected the seasonal dynamics of ^{137}Cs activities in fish, regardless of fish species. Given that alternating periods of hypoxia (due to stratification) and mixing occur repeatedly during the summer in Lake Kasumigaura, the simultaneous increases of dissolved ^{137}Cs activities and ^{137}Cs activities in fish suggest that in shallow lakes, remobilized ^{137}Cs is quickly transported to the epilimnion and enters the food web. In shallow, eutrophic lakes such as Lake Kasumigaura, the high abundance and short turnover time of plankton might also contribute to the rapid transfer of ^{137}Cs from plankton to fish (Marzano et al., 1996; Robbins and Jasinski, 1995). Bottom-up linkages between nutrient, phytoplankton, zooplankton (especially rotifers and cyclopoids), and pond smelt has been reported in Lake Kasumigaura (Matsuzaki et al., 2018). The results of this study highlight the differences in the behavior and dynamics of radiocesium in shallow and deep lakes.

Because the rate at which fish eliminate ^{137}Cs increases with water temperature (Doi et al., 2012; Ugedal et al., 1992), the activity of ^{137}Cs in fish is predicted to decrease during the summer (Peles et al., 2000). However, the feeding rates of fish increase with temperature, and fish feeding rates can increase more rapidly than elimination rates in warm lakes (Forseth et al., 1991; Ugedal et al., 1997). The fact that the ^{137}Cs activities in pond smelt and crucian carp were higher during the summer indicated the increase of feeding rates associated with the higher water temperatures in summer increased the intake of prey that had assimilated remobilized ^{137}Cs. Furthermore, differences in the feeding and elimination rates between different species of fish as a function of temperature might explain why only crucian carp had higher

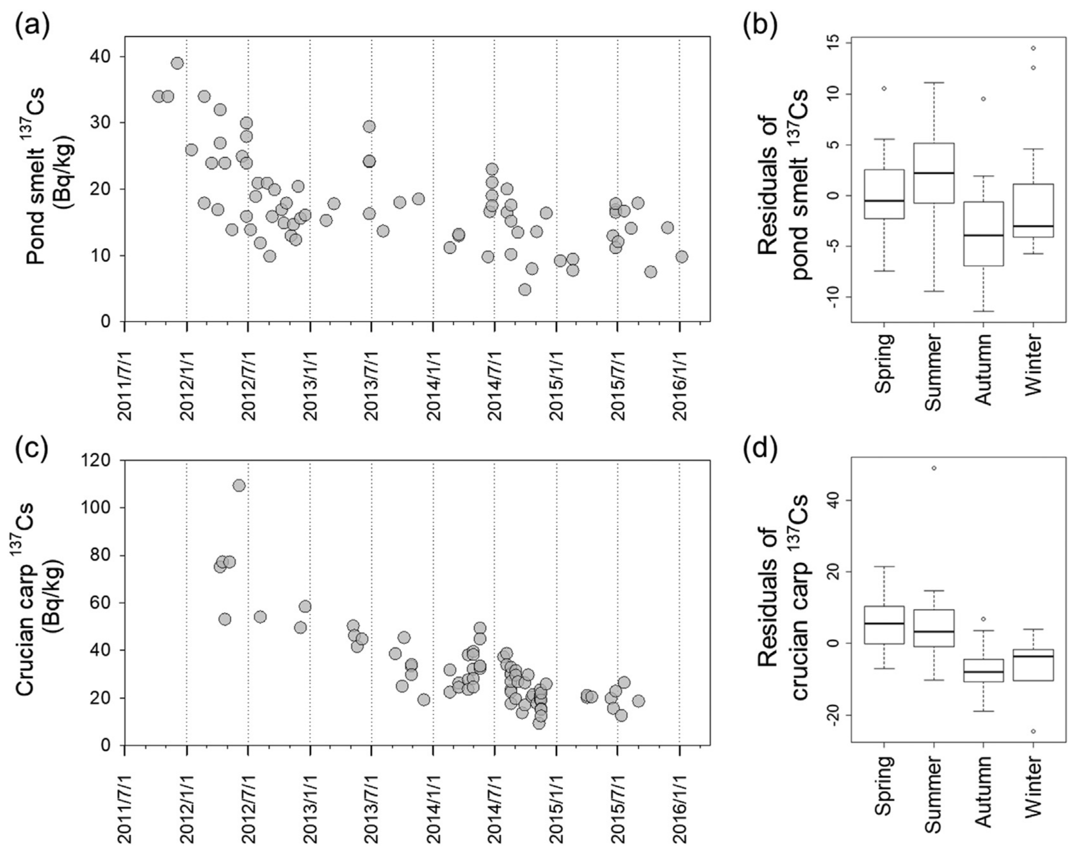

Fig. 4. Temporal changes (a) and seasonal variations (b) in pond smelt ^{137}Cs activity, and temporal changes (c) and seasonal variations (d) in crucial carp ^{137}Cs activity in Lake Kasumigaura. Note that the *y*-axis in (b) and (d) shows residuals after removing the effect of year in the GLS model. In (b) and (d), the horizontal lines represent the median, the box encloses the 25th and 75th percentiles, and the whiskers represent the 5th and 95th percentiles.

^{137}Cs activities in both spring and summer (Forseth et al., 1998). Remobilization of ^{137}Cs from sediments and the high feeding rates of fish during late spring and summer explain the seasonal dynamics of the ^{137}Cs activities of pond smelt and crucian carp in Lake Kasumigaura.

Other factors, including the body size of fish and their prey, can affect the seasonal dynamics of radiocesium in fish (Forseth et al., 1998; Rowan and Rasmussen, 1995; Ugedal et al., 1992).The rate of elimination of ^{137}Cs can decrease with increasing body size (Sundbom et al., 2003; Ugedal et al., 1992). Whereas the lifespan of crucian carp is relatively long (~10 years) and their body size changes slowly, the standard body length of pond smelt changes throughout the year; from <10 mm in March–April to about 6 cm in July, and reaching 10 cm in December (Sakamoto et al., 2014). The ^{137}Cs activity in pond smelt is therefore expected to increase from spring to winter. This prediction based on body size, however, does not explain the seasonal variations of ^{137}Cs activities that we observed. Fish radiocesium activities can also change with ontogenetic shifts in diet (Rowan et al., 1998). Pond smelt are planktivores, but they often consume chironomid pupae when the pupae emerge from late summer to winter (Yoshioka et al., 1994). In Lake Kasumigaura, two main chironomid species are dominant. *Chironomus plumosus* emerges three times between May and October, whereas *Propsilocerus akamusi* emerges once between November and December (Iwakuma, 1992). Because the radiocesium activity is generally higher in benthic macroinvertebrates than in zooplankton (Rowan et al., 1998), the ^{137}Cs activity of pond smelt is expected to increase as their diet shifts from zooplankton to chironomid pupae. However, the times when we observed an increase of ^{137}Cs in pond smelt were not consistently associated with the times of emergences of chironomid pupae. Because crucian carp, which are omnivorous, consume benthic invertebrates such as chironomid larvae as well as other prey, the dominant prey consumed by adult individuals is unlikely to shift seasonally. These other factors are therefore unlikely to influence the seasonal changes of the ^{137}Cs activities in pond smelt and crucian carp.

Seasonal dynamics of dissolved ^{137}Cs in a lake can be influenced by the runoff of ^{137}Cs from the catchment. Although we did not measure dissolved or particulate ^{137}Cs activity in the rivers that discharge into Lake Kasumigaura, the influx of ^{137}Cs from rivers would be expected to increase with discharge. We obtained monthly discharge data for the two major tributaries of Lake Kasumigaura (the Sakura River and Koise River) from the Kasumigaura River Office of the Ministry of Land, Infrastructure, Transport and Tourism, Japan, and examined the seasonal discharge patterns (Text S2, Fig. S6). In both rivers, early summer and autumn tended to be times of high discharge, due to rainy and typhoon seasons, respectively. The month of peak discharge differed only slightly among the study years. Tsuji et al. (2019) found that the ^{137}Cs inventory in the sediments of Lake Kasumigaura increased near the mouths of urban rivers shortly after the FDNNP accident, but the distribution of the sedimentary ^{137}Cs inventory did not change in most areas of the lake. Previous studies have also reported that secondary inputs from the catchment have little influence on the dynamics of dissolved ^{137}Cs activity in lakes (Smith et al., 2000; Smith et al., 1999; Sundbom et al., 2003). The seasonality of discharge from the catchment is therefore an unlikely explanation for the seasonal dynamics of the activities of dissolved ^{137}Cs and the ^{137}Cs in fish during the period of our study.

3.3. Caveats and future directions

Overall, our analyses suggested that the remobilization of radiocesium from sediments may contribute to the seasonal dynamics and long persistence of radiocesium in the water and fish of shallow, eutrophic lakes. Understanding of these seasonal dynamics will help

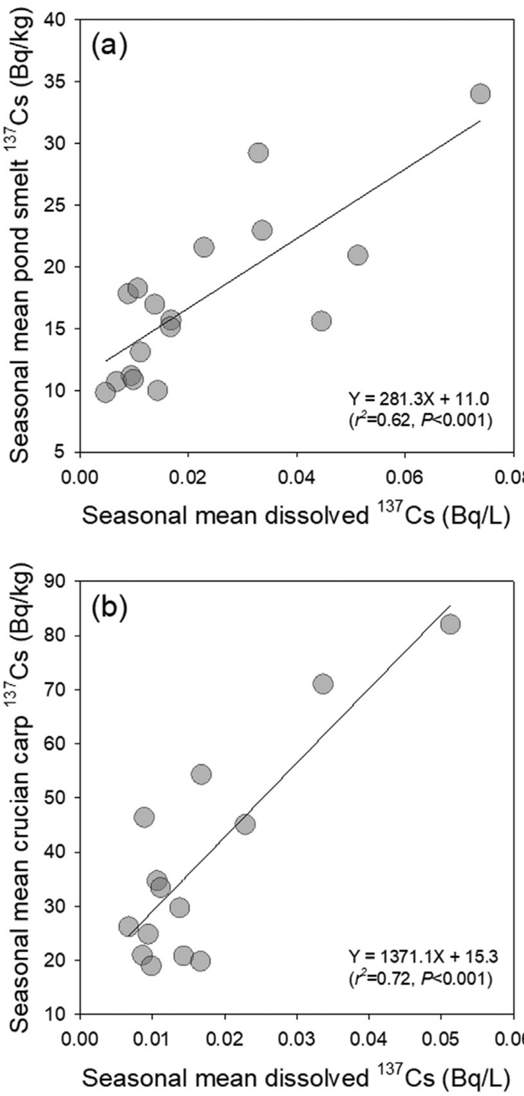

Fig. 5. The linear relationships between seasonal mean dissolved ^{137}Cs activities and seasonal mean (a) pond smelt ^{137}Cs activities and (b) crucial carp ^{137}Cs activities.

inform decisions about whether to cancel restrictions on the distribution of fish. There are at least three caveats to the extension of our results to other shallow lakes. First, we assessed the possibility of radiocesium remobilization based on radiocesium activities and environmental factors in only the water column. The seasonal dynamics and vertical distribution of radiocesium and NH_4^+ in pore waters and sediments should also be studied. Second, we did not measure radiocesium activities in phytoplankton and zooplankton. Measurements of the radiocesium activities in zooplankton could enable a more detailed assessment of the transfer of remobilized radiocesium through the food webs of shallow lakes. Because much of the organic matter in plankton is in forms that can be rapidly decomposed and recycled (Mori et al., 2017), the contribution of such decomposition to fish radiocesium activities should also be considered. Third, our results are based on radiocesium data collected every three months over a five-year period. The DO concentrations in the bottom waters of shallow lakes can vary hourly, daily, and weekly as a function of the intensity of thermal stratification, and the vertical distribution of oxygen at the sediment-water interface can also vary between time intervals much shorter than three months (Gerling et al., 2014; Jabbari et al., 2019). Higher frequency measurements of dissolved ^{137}Cs activities and ^{137}Cs activities in fish, environmental and climatic variables, and sediment oxygen profiles are critically needed to quantify the fluxes of remobilized ^{137}Cs. More frequent and extensive sampling could thus help to predict or prevent releases of not only radiocesium but also nutrients, heavy metals, and methane from the sediments of shallow, hypereutrophic lakes.

CRediT authorship contribution statement

Shin-ichiro S. Matsuzaki: Conceptualization, Data Analysis, Writing, Visualization, Project administration, **Atsushi Tanaka**: Methodology, Investigation, **Ayato Kohzu**: Investigation, Resources, **Kenta Suzuki**: Data Analysis, Visualization, **Kazuhiro Komatsu**: Investigation, Resources, **Ryuichiro Shinohara**: Investigation, Data Analysis, Writing, Resources, **Megumi Nakagawa**: Investigation, **Seiichi Nohara**: Investigation, **Ryuhei Ueno**: Investigation, **Kiyoshi Satake**: Investigation, **Seiji Hayashi**: Writing, Supervision, Funding acquisition.

Declaration of competing interest

The authors declare that they have no known competing financial interests or personal relationships that could have appeared to influence the work reported in this paper.

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Appendix A. Supplementary data

Supplementary data to this article can be found online at https://doi.org/10.1016/j.scitotenv.2020.143257.

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