



# Salmon Migration Patterns Revealed the Temporal and Spatial Fluctuations of the Radiocesium Levels in Terrestrial and Ocean Environments

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## Abstract

The disabling of the Fukushima Daiichi Nuclear Power Plant (F1NPP) resulted in the release of radionuclides, including  $^{134}\text{Cs}$  and  $^{137}\text{Cs}$ , into the air and the ocean. The unpredicted nuclear accident is of global concern for human health and the ecosystem. Although investigations of radionuclides in environments were performed shortly after the accident started, the temporal and spatial impacts and fluctuations on the releasing radionuclides to natural environment remain unclear. I focused on salmon, which migrate from inland to the open ocean globally, to reveal the three-year (May 2011 to February 2014) fluctuations and accumulations of  $^{134}\text{Cs}$  and  $^{137}\text{Cs}$  from terrestrial to open ocean environments after the F1NPP accident. The  $^{134}\text{Cs}$  and  $^{137}\text{Cs}$  concentrations in six salmonids exhibited lower temporal variations for three years after the F1NPP accident, suggesting that these radionuclides are widely distributed and these radionuclides remain in the natural environment globally with less convergence. The accumulation patterns were significantly different among the different salmon species. Fluvial (freshwater residence) type salmonids exhibited significantly higher accumulation in  $^{134}\text{Cs}$  (25.3–40.2 Bq kg<sup>-1</sup> in mean) and  $^{137}\text{Cs}$  (41.4–51.7 Bq kg<sup>-1</sup> in mean) than did the anadromous (sea-run) type salmonids (0.64–8.03 Bq kg<sup>-1</sup> in mean  $^{134}\text{Cs}$  and 0.42–10.2 Bq kg<sup>-1</sup> in mean  $^{137}\text{Cs}$ ) suggesting widespread contamination in terrestrial environments versus the coastal and open ocean environments. Salmonids are the most highly migratory animals and are characterised by their strong tendency to return home to their natal site for reproduction. Salmonids have a potential to be a good indicator as an effective monitoring animal.

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**Data Availability:** The author confirm that all data underlying the findings are fully available without restriction. Japanese Ministry of Agriculture, Forestry, and Fisheries. Results of the inspection on radioactivity materials in fisheries products. <http://www.jfa.maff.go.jp/e/inspection/index.html>.

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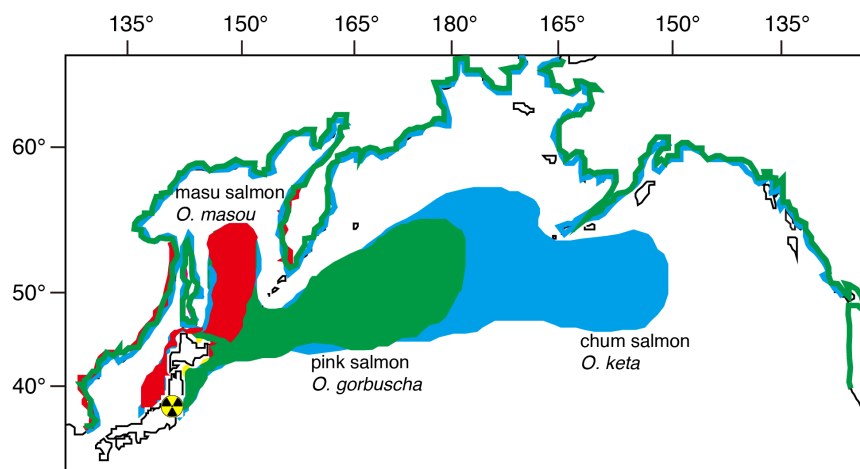
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## Introduction

The Great East Japan Earthquake of magnitude 9.0 on Friday 11 March 2011 caused considerable damage in the region, and the large tsunami it created caused much more. Following a major earthquake, a 15-m tsunami disabled the power supply and the cooling of three Fukushima Daiichi reactors, causing a nuclear accident at that date. Radioactive contamination following the Fukushima Daiichi Nuclear Power Plant (F1NPP) accident was the most significant artificial radioactive liquid release into the environment ever known, on a short time and space scale basis [1–5]. The amount of radionuclides discharged from the F1NPP immediately after the accident was estimated to be 5–10 PBq to the atmosphere and 3–6 PBq to the sea, the latter being caused by direct leakage of the contaminated cooling water [5–8]. Afterwards, trace quantities of radioactive particles from the incident have since been detected around the world, including  $^{131}\text{I}$ , iodine and  $^{134}\text{Cs}$  and  $^{137}\text{Cs}$  [1,3]. Fukushima-derived  $^{134}\text{Cs}$  and  $^{137}\text{Cs}$  were found throughout a 150,000 km<sup>2</sup> area of the Pacific Ocean of Japan [4]. In Europe, the first signs of the release were detected seven days later, while the first peak of the activity level was observed between 28 March and 30 March 2011 [3].

Among the radionuclides discharged by the accident, the effects of  $^{134}\text{Cs}$  and  $^{137}\text{Cs}$  are the most serious because of their high levels and long decay periods. These radionuclides are considered to most substantially contribute to the contamination of various organisms due to the contamination of their environments. Prior to the accident, the concentrations of  $^{137}\text{Cs}$  in the surface water of the Pacific Ocean were in the range of 1–4 Bq m<sup>-3</sup> [9–11], which mainly resulted from global fallout due to atmospheric nuclear weapon tests, reprocessing plants, fallout from the accident of the Chernobyl nuclear power plant, and solid waste dumping at sea [12]. Recently, Bailly du Bois et al. [12] proposed that 12–41 PBq of  $^{137}\text{Cs}$  was discharged to the Pacific Ocean between March 25 and July 28, 2011 and predicted an increase of approximately 6 Bq m<sup>-3</sup> after dilution in the surface ocean. In addition to this direct release into the ocean, the accident also dispersed approximately 15 PBq of  $^{137}\text{Cs}$  into the atmosphere, dominated by a leak from 12 March to 6 April 2011 [2]. These nuclides, once discharged into the atmosphere, can have a global effect through fast atmospheric circulation. These phenomena seriously suggest that the contamination by the F1NPP is a global issue and has spread widely throughout the water, soil, air and biota.



**Figure 1. Migratory route and scale from Japanese habitats and the distribution of anadromous salmonids in the Pacific region used in the present study.** Chum salmon (blue) have the largest natural range of any Pacific salmon, and undergo the longest migrations within the genus *Oncorhynchus*. Chum are found around the north Pacific, in the waters of Korea, Japan, and the Okhotsk and Bering seas, British Columbia in Canada, and from Alaska to California in the United States. The native range of pink salmon (green) is from the Arctic and Pacific drainages from Mackenzie River delta, the Northwest Territories, to the Sacramento River drainage, California (occasionally as far south as La Jolla, southern California) and in the west from the Lena River in Siberia to Korea. Populations in Asia occur as far south as Honshu in Japan. Masu (cherry) salmon (red) found in the Western Pacific Ocean along East Asia, range from the Kamchatka, Kuril Islands, Sakhalin and Primorsky Krai south through Korea, Taiwan and Japan.

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Several agencies have conducted investigations to monitor the radioactive quantities of water, soil, air and biota after the F1NPP accident, and considerable data have been reported to understand the present status of radionuclide pollution levels [1–4]. Although it has been three years since the disaster by the F1NPP accident, the radionuclide pollution issues have not converged yet, and there is little information about radionuclide behaviour in the natural environment and ecosystem globally. A recent study found that the Pacific blue tuna transported Fukushima-derived radionuclides from Japan to California, and such highly migratory animals transport radionuclides globally as a vector [13], leading to the cause of the distribution of the radionuclides globally. In such a

situation, systematic and comprehensive investigations for radionuclides in natural environments are required. However, a number of animals are living in a certain restricted area, e.g., aquatic animals generally can survive in either freshwater or seawater throughout their lives, and thus, it was quite difficult to understand the radionuclide behaviour systematically and comprehensively in various environments from inland water to open ocean environments.

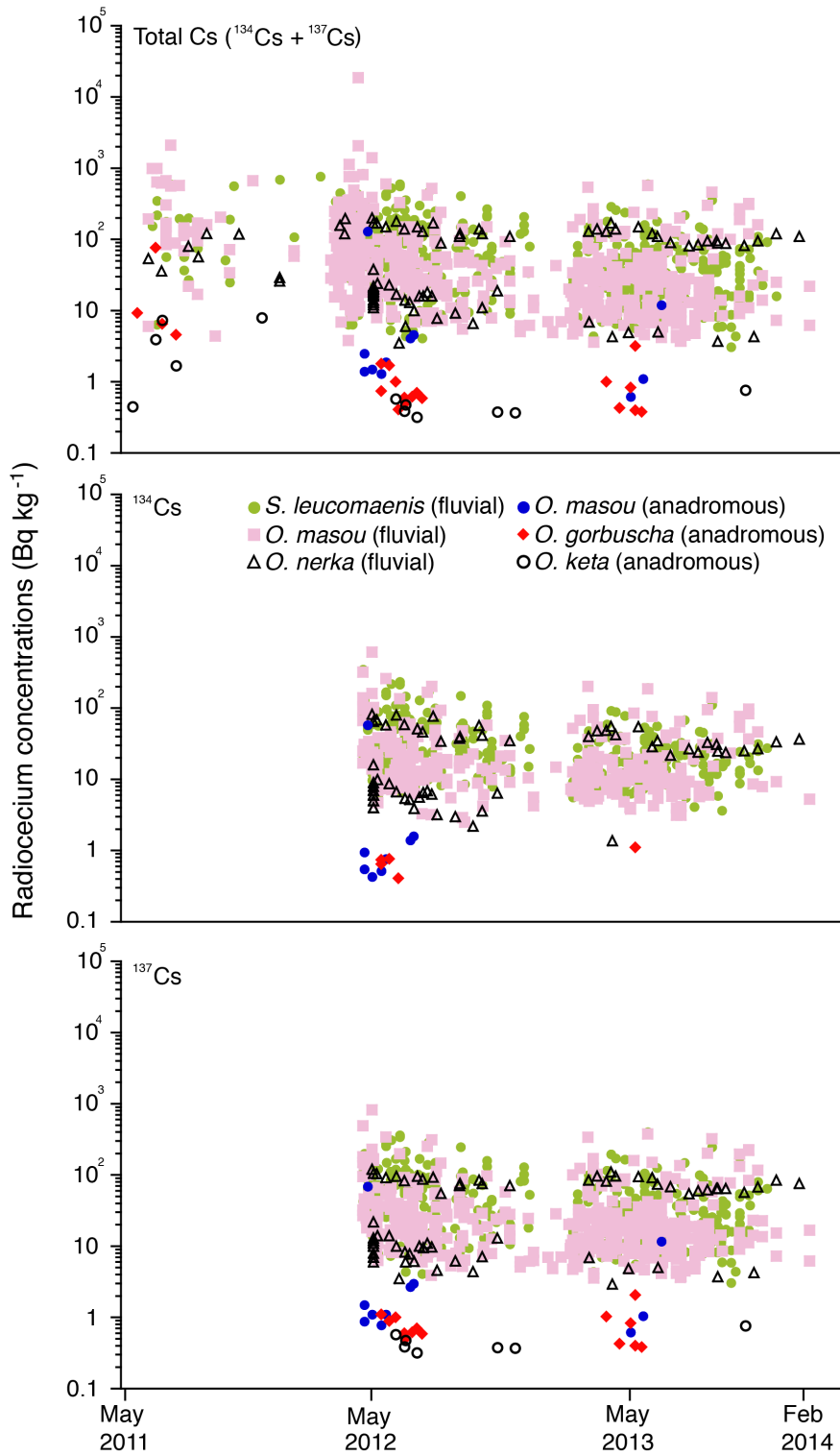
Salmonids are widely distributed in cool and cold waters of all northern continents occurring from the Arctic drainages of Europe, Asia and North America, south as far as the Mediterranean and northern Africa [14]. All of salmonids spawn in

**Table 1. Information of life history in the salmonids examined between May 2011 and February 2014 in the present study.**

Species	Migration scale (migration type)	Freshwater life period before descending to sea	Marine life period before spawning
<i>S. leucomaenis</i> (whitespotted charr)	freshwater (fluvial)	whole life	n/a
<i>O. masou</i> (masu salmon)	freshwater (fluvial)	whole life	n/a
<i>O. nerka</i> (sockeye salmon)	freshwater (fluvial)	whole life	n/a
<i>O. masou</i> (masu salmon)	freshwater-coastal (anadromous)	1–2 year(s)	1–2 year(s)
<i>O. gorbuscha</i> (pink salmon)	freshwater-coastal-open ocean (anadromous)	immediately after hatching	1–2 year(s)
<i>O. keta</i> (chum salmon)	freshwater-coastal-open ocean (anadromous)	immediately after hatching	2–8 years

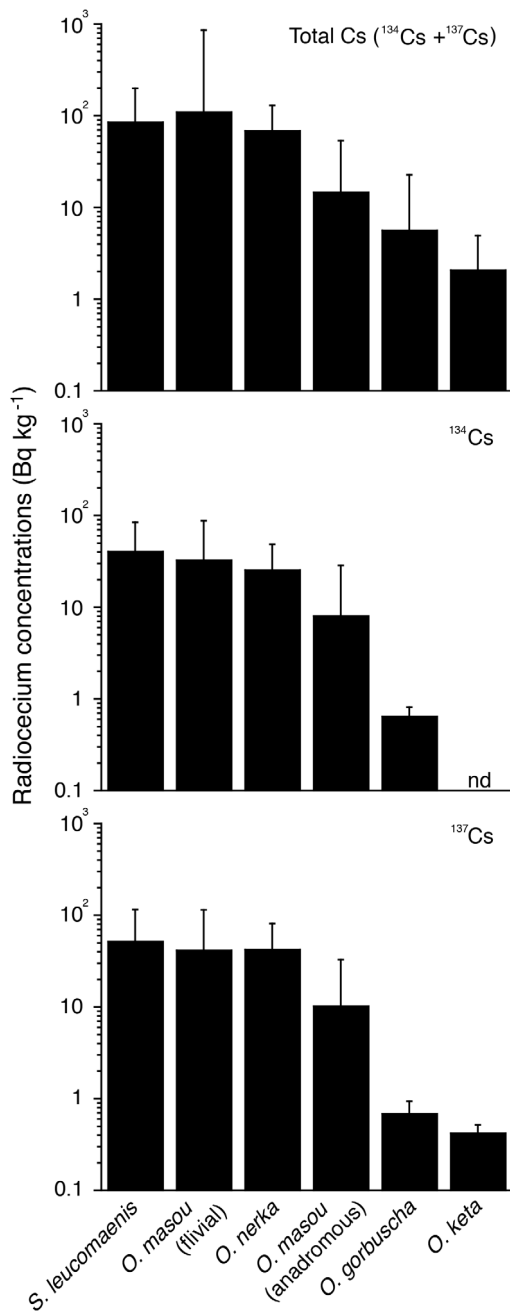
n/a: not applicable.

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**Figure 2. Fluctuations of the radiocesium levels in the total Cs (top) between May 2011 and February 2014, <sup>134</sup>Cs (middle) and <sup>137</sup>Cs (bottom) between April 2012 and February 2014 after the F1NPP accident.** No significant temporal trends were found for all types of salmon suggesting the continued widespread distribution of these radionuclides with less convergence and indicating that these radionuclides remain in the natural environment for both terrestrial and marine ecosystems. All radiocesium data in the salmonids was from the information published by the Fisheries Agency of the Japanese Government [16] between May 2011 and February 2014. nd: not detectable due to levels below the detection limits.

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**Figure 3. Differences in the radiocesium accumulations among the salmon species between May 2011 and February 2014 after the F1NPP accident.** Fluvial salmon exhibited significantly higher concentrations in each radiocesium than those of the anadromous salmon. The detectable rates were also higher in fluvial salmon than in anadromous salmon as presented in Table 2. doi:10.1371/journal.pone.0100779.g003

freshwater [15] but anadromy (sea-run) is strongly and widely represented. The animal is one of the most studied due to their great importance for food and angling. Thus, the biology and ecology are well understood. Some salmonids highly migrate to far offshore locations, such as the Pacific and Atlantic oceans (Fig. 1); however, all salmon come back to freshwater environments for spawning. Thus, this animal can be useful indicator to monitor for the spread of radionuclides from inland water to open ocean environments without allocation of a significant catching effort

and time because a large number of salmon return to their natal rivers.

A number of salmonid species exist in Japan, and the Fisheries Agency of the Japanese Government has examined radionuclides in the animals (Table 1) [16]. In this study, I compared radiocesium (<sup>134</sup>Cs, <sup>137</sup>Cs and these total) levels among salmonid species using a total of 3,024 samples between May 2011 and February 2014. The levels of accumulation were significantly different in accordance with their life patterns; fluvial (freshwater residence) salmon exhibited the highest level, influenced by the higher radionuclide levels in terrestrial environments, a coastal migrant exhibited a moderate level because the salmon migrate between freshwater and coastal water, and the long-distance migrants to the open ocean exhibited the lowest level, being less impacted by the radiocesium because they live in the open ocean for almost all of their lives after hatching. The present study indicates that migration patterns of the different types of salmon can reveal temporal and spatial fluctuations of radiocesium globally, ranging from terrestrial to open ocean environments.

## Materials and Methods

All radiocesium data in the salmonids was from the information published by the Fisheries Agency of the Japanese Government [16] between May 2011 and February 2014 in the eastern and northern parts of Japan from 3 to 15 prefectures. The radiocesium concentrations were determined as a total between May 2011 and March 2012, while thereafter each <sup>134</sup>Cs and <sup>137</sup>Cs concentration was determined in each sample (Fig. 2). All data were shown in the wet weight basis. Each radiocesium concentration among the salmon species was compared using the Kruskal-Wallis test. Significance of the correlation coefficient and regression slope were tested by Fisher's  $\chi$ -transformation.

## Results

The radiocesium concentrations in six salmonids (five different species and a different migratory type in a same species) (Table 1) exhibited less temporal variations for three (total Cs) or 2 (<sup>134</sup>Cs and <sup>137</sup>Cs) years after the F1NPP accident (Fig. 2). No significant temporal trends were found for all of the types of salmon (Fisher's  $\chi$ -transformation,  $p > 0.05$ ), suggesting a continued widespread distribution of these radionuclides with less convergence and that these radionuclides remain in natural environment for both terrestrial and marine ecosystems. Fluvial salmon, such as *Salvelinus leucomaenis*, *Oncorhynchus masou* and *O. nerka*, only reside in a freshwater environment, and thus they accumulate these radionuclides from the terrestrial environment. In contrast, anadromous (sea-run) salmon *O. masou*, *O. gorbuscha* and *O. keta* accumulate radionuclides from terrestrial, coastal and open ocean environments depending on their migration scale (Table 1). The highest concentration of 18,700 Bq/kg in total cesium was found in fluvial *O. masou* in March 2012, one year after the F1NPP accident, and there were still detectable amounts in salmon of over 100 Bq kg<sup>-1</sup> (Japanese safety limit) of <sup>134</sup>Cs and <sup>137</sup>Cs, even three years after the F1NPP incident (Fig. 2).

However, the accumulation patterns were significantly different among the salmon species. In general, fluvial-type salmon, such as *S. leucomaenis*, *O. masou* and *O. nerka*, exhibited significantly higher accumulation of <sup>134</sup>Cs (25.3–40.2 Bq kg<sup>-1</sup> in mean) and <sup>137</sup>Cs (41.4–51.7 in mean) than the anadromous-type salmon of *O. masou*, *O. gorbuscha* and *O. nerka* (0.64–8.03 Bq kg<sup>-1</sup> in mean <sup>134</sup>Cs and 0.42–10.2 Bq kg<sup>-1</sup> in mean <sup>137</sup>Cs) (Table 2, Fig. 3) ( $p < 0.005$ –0.0001). Among the anadromous salmon, the highest concentration was found in *O. masou*, the second highest was in *O. gorbuscha*

**Table 2.** Radiocesium data information and radiocesium concentrations (above the detection limit) in the salmonids examined between May 2011 and February 2014 in the present study.

Species	Number of the data above detection limit (Ratio of detectable (%))			Mean $\pm$ SD (Bq/kg) (minimum-maximum (Bq/kg))		
	Total Cs*	$^{134}\text{Cs}$	$^{137}\text{Cs}$	Total Cs*	$^{134}\text{Cs}$	$^{137}\text{Cs}$
<i>S. leucomaenis</i> (whitespotted charr)	524 (70.6)	317 (45.6)	429 (61.3)	84.7 $\pm$ 112 (3.10–840)	40.2 $\pm$ 43.5 (5.63–350)	51.7 $\pm$ 62.8 (3.07–490)
<i>O. masou</i> (masu salmon)	670 (58.8)	327 (31.6)	493 (48.2)	109 $\pm$ 740 (3.60–18700)	32.4 $\pm$ 54.3 (2.50–610)	41.4 $\pm$ 72.2 (3.57–820)
<i>O. nerka</i> (sockeye salmon)	85 (88.0)	68 (73.0)	74 (82.0)	68.4 $\pm$ 60.1 (3.50–200)	25.3 $\pm$ 22.7 (1.37–82)	42.2 $\pm$ 38.4 (2.94–120)
<i>O. masou</i> (masu salmon)	11 (11.7)	8 (10.5)	11 (18.0)	14.6 $\pm$ 38.4 (0.62–130)	8.03 $\pm$ 20.2 (0.43–58)	10.2 $\pm$ 22.3 (0.62–69)
<i>O. gorbusha</i> (pink salmon)	20 (71.6)	5 (20.2)	14 (48.4)	5.60 $\pm$ 16.9 (0.38–76.7)	0.64 $\pm$ 0.16 (0.41–1.11)	0.69 $\pm$ 0.25 (0.38–2.07)
<i>O. keta</i> (chum salmon)	12 (1.6)	0 (0)	7 (1.3)	2.06 $\pm$ 2.82 (0.32–8.00)	nd	0.42 $\pm$ 0.09 (0.32–0.77)

\*Total Cs =  $^{134}\text{Cs} + ^{137}\text{Cs}$ .

Total Cs was available between May 2011 and February 2014.

$^{134}\text{Cs} + ^{137}\text{Cs}$  were available between April 2012 and February 2014.

nd: all samples were below the detection limit.

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and the lowest was in *O. keta* (Table 2, Fig. 3). Furthermore, the detectable numbers were also different among either fluvial- or anadromous- type salmon. There were 529–772 samples of *O. keta* investigated for three years; however, all of the samples were below the detection limit in  $^{134}\text{Cs}$ , and only seven samples were found to be above the limit. In contrast, radionuclide levels in fluvial salmon could be determined, being generally over 50% (Table 2).

## Discussion

The present study clearly demonstrated that accumulations of radiocesium were different depending on the migratory patterns and migration scale of the different types of salmon. The highest accumulation was found in the fluvial species (type); their concentrations were similar to those of the other freshwater fish, such as Ayu (*Plecoglossus altivelis*) [17], used to investigate the radiocesium contamination levels in the freshwater ecosystem after the F1NPP accident. Thus, these salmon can also be used to document the  $^{134}\text{Cs}$  and  $^{137}\text{Cs}$  contamination levels in the terrestrial environments in East Japan. In contrast to the fluvial salmon,  $^{134}\text{Cs}$  and  $^{137}\text{Cs}$  concentrations were quite low in the anadromous salmon, such as *O. masou*, *O. gorbusha* and *O. nerka*. The detectable ratios of the cesium isotopes were the lowest in *O. keta*, which is the species that has the longest distance of migration, spending almost all of their lives in the North Pacific Ocean (Fig. 1). Among the anadromous salmon, *O. masou* accumulated higher  $^{134}\text{Cs}$  and  $^{137}\text{Cs}$  concentrations than those of the other types of salmon. The species remain in freshwater for one to two year(s) after hatching and thereafter go downstream to the sea to growth. In addition, the salmon tends to migrate to a coastal area along the northern Japan for growth (Fig. 1). Thus, anadromous *O. masou* probably have a higher risk for accumulation of radionuclides compared to *O. gorbusha* and *O. keta*. *O. gorbusha* and *O. keta* migrate to sea immediately after hatching, and further migrate along the coastal to the open ocean for growth (Fig. 1); as a result,

these salmon probably exhibit a lower risk to accumulate these radionuclides in their lives.

Although the higher levels of radiocesium in fluvial salmon are caused by the serious contamination in the terrestrial environments of East Japan after the F1NPP accident, marine fish are usually approximately 100 times lower in  $^{137}\text{Cs}$  than are freshwater fish because potassium, which is more abundant in seawater, blocks the uptake of cesium by marine organisms [18]. Thus, the differences in physiological mechanism, especially ionic uptake and elimination of the related osmoregulation, might also accelerate higher  $^{137}\text{Cs}$  accumulation in the fluvial salmon. The F1NPP accident released large amounts of radioactive substances into the environment and contaminated the soil of Tohoku and the Kanto (East) districts in Japan, and the radiocesium levels in water, soil and biota in terrestrial environment are much more higher than those of the coastal and marine environments [4,19]. The natural habitats of fluvial salmon, such as *Salvelinus leucomaenis* and *O. masou* are widespread in East Asian countries, as they reside streams, ponds and lakes; as a result, these salmon can be useful indicators for radiocesium pollutions in terrestrial environments.

After the F1NPP accident, the  $^{137}\text{Cs}$  concentrations in a 30-km perimeter around the plant exceeded 10 Bq L<sup>-1</sup> [12]. Chum salmon *O. keta* used in the present study might have migrated in the North Pacific at the time of the F1NPP accident because their oceanic migration in North Pacific takes two to eight years. In fact, the *O. keta*  $^{134}\text{Cs}$  and  $^{137}\text{Cs}$  concentrations were below the detection limit and 0.42 $\pm$ 0.09 Bq kg<sup>-1</sup> (mean  $\pm$  SD), respectively, for the 529 samples. These levels were much lower than the highly migratory marine animals of the Pacific blue fin tuna in the North Pacific Ocean (4.0 $\pm$ 1.4 Bq kg<sup>-1</sup> in  $^{134}\text{Cs}$  and 6.3 $\pm$ 1.5 Bq kg<sup>-1</sup> in  $^{137}\text{Cs}$ ) [13]. The tissue concentrations of radiocesium in the tuna might vary depending on the time spent near the Japanese coast, foraging strategies and the timing of the migration [13]. During open-ocean migration by chum salmon for growth, the salmon has never returned to the Japanese coast and are migrating in the

North Pacific Ocean (Fig. 1); and thus the salmon might not accumulate radiocesium of F1NPP accident origin. Nevertheless, trace amounts of radiocesium might have a potential to accumulate during their open ocean migration in the North Pacific Ocean, it could metabolite and eliminate during spawning migration to Japanese coasts. These suggest that the present data of radiocesium levels in chum salmon might be used as a background level, and continuous investigations might reveal the fluctuation of radionuclide pollution levels in North Pacific Ocean. The current distribution of chum salmon extends from approximately 45–70°N to 140°E–125°W longitude (Fig. 1). The species has been recorded globally from Russia, United States, Canada, Japan and Korea. Chum salmon has higher homing rate to the natal river, and thus each country could use the salmon to monitor radionuclide levels during their open ocean migration.

The life history, ecology and evolution of salmonid fishes (salmon, trout and char) are dominated by their strong tendency to return home to their natal site for reproduction. Homing characterises the family but is best studied in the anadromous (i.e., ‘sea run’) forms of salmon. Typically, salmon spawn in streams or lakes, and after a variable period of freshwater residence (zero to three years), depending on the species and population, the offspring migrate to the ocean. Salmon remain in the ocean until they begin to mature, and then they return to their natal site to spawn. Thus, salmon can have a great potential to

monitor the fluctuation of radionuclides in inland, coastal and open ocean environments as we do not need to catch salmon in coastal and off shore regions because anadromous salmon return to their natal river mouth in spite of their migration scale. Anadromous salmon as well as fluvial salmon are also useful to monitor the radionuclide levels considering the migration scale. In contrast to anadromous salmon, fluvial salmon are suitable indicators for investigating terrestrial environments to reveal the fluctuations of local pollution patterns from the F1NPP event. The present study indicates that the levels of  $^{134}\text{Cs}$  and  $^{137}\text{Cs}$  pollution after the F1NPP accident did not converge during the three subsequent years (Fig. 2). Furthermore, Pacific Bluefin tuna, which are highly migratory marine animals, transported Fukushima-derived radionuclides from Japan to California [13], suggesting the widespread distribution of radionuclide pollution globally. Thus, continuous systematic and comprehensive monitoring, taking advantage of the characteristics of salmon migration patterns, might be needed to reveal the temporal and spatial fluctuations of the radionuclides in various environments in the future.

### Author Contributions

Conceived and designed the experiments: TA. Performed the experiments: TA. Analyzed the data: TA. Contributed reagents/materials/analysis tools: TA. Contributed to the writing of the manuscript: TA.

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