Evaluating radiocesium transfer for white-spotted char (*Salvelinus leucomaenis*) in forested headwater streams

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by

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ABSTRACT

Radioceium (^{137}Cs) released from the Fukushima Dai-ichi Nuclear Power Plant (FDNPP) accident severely contaminated forested areas in Fukushima and adjacent prefectures. ^{137}Cs could move across terrestrial and aquatic ecosystems in forested headwater streams because of tight linkages between riparian areas and stream channels. White-spotted char (*Salvelinus leucomaenis*) is considered as a top predator in food web of headwater streams and important fishery resources. Although white-spotted char consumes various food sources from forest and stream, complex interactions for ^{137}Cs movement from their multiple food sources have not been fully examined. Therefore, the primal objective of this thesis was to understand ^{137}Cs movement and contamination levels of organisms in headwater streams. I developed a new method for investigating ^{137}Cs transfer to white-spotted char by considering their multiple dietary sources with respective contamination levels. Then, I evaluated changes in ^{137}Cs contaminations from 1st to 5th years after the accident for examining the temporal patterns of ^{137}Cs activity concentrations in biota. Finally, I discussed the fate of ^{137}Cs contamination levels in future and management applications for reducing contamination levels in complex headwater ecosystems.

Firstly, I reviewed approximately 100 previous studies related to ^{137}Cs contamination in freshwater fish around the world since 1974. About 40 fish species in rivers and lakes were summarized. ^{137}Cs activity concentrations in fish species such as brown trout (*Salmo trutta*) and perch (*Perca fluviatilis*) was associated with fallout volume of contaminants in rivers and lakes. Hence, the contamination levels of freshwater fish in similar fallout volume varied substantially with 1 to 2 orders of magnitude. Variability of ^{137}Cs contaminations tended to be high in rivers species compared to the species of lake habitats in Fukushima. Differences of contamination levels of fish species in similar fallout were associated with food consumptions and physiological

conditions. Among them, dietary conditions are one of the important factors for controlling the contamination levels.

For examining dietary ^{137}Cs contributions with multiple prey items, I developed food web-based transfer factor (TF_{web}) for white-spotted char in headwater streams draining Japanese cedar (*Cryptomeria japonica*) forest in Fukushima and Gunma. I estimated dietary contributions based on stable carbon (δ^{13}C) and nitrogen isotope ratios (δ^{15}N) using samples collected in 2012 and 2013. TF_{web} was calculated as the activity concentration in char divided by the sum of activity concentrations in all prey items, using the lower and upper estimates of dietary contributions. Both terrestrial and aquatic species such as mayflies (*Ephemera japonica*), spider crickets (Raphidosphoridae gen. spp.), and freshwater crab (*Geothelphusa dehaani*) contributed 3 to 12% of the fish diet. Despite the differences of ^{137}Cs activity concentrations of char in Fukushima (704 to 6082 Bq kg^{-1}-dry) and Gunma (193 to 618 Bq kg^{-1}-dry), TF_{web} had similar ranges from 1.1 to 3.8 in Fukushima, and from 1.3 to 4.3 in Gunma. TF_{web} values suggested that ^{137}Cs tended to be accumulated from prey to white-spotted char. TF_{web} using multiple prey items also provided consistent values compared to the other transfer factors using single prey-predator.

Seasonal variations of TF_{web} for ^{137}Cs activity concentrations in white-spotted char was examined because dominant food items of white-spotted char changes from terrestrial ones in summer to aquatic ones in winter. In Fukushima, TF_{web} tended to be consistent throughout seasons in summer (mean ± SD: 3.9 ± 1.3) and in winter (6.5 ± 1.5). In Gunma, the greatest TF_{web} occurred in winter (7.5 ± 1.6) and the lowest values was estimated in summer (2.6 ± 0.9). Because the metabolic rate of char in summer was four times greater than that in winter, high excretion rate of ^{137}Cs in summer promoted the low summer TF_{web} in Gunma. Hence, the contamination of food resources in Fukushima were 5-folds greater than those in Gunma, seasonal differences of TF_{web} was not apparent by overwhelmed ^{137}Cs intake relative to excretion rates.

Because changes in ^{137}Cs activity concentrations over time is important for projecting contamination levels in future, I examined ^{137}Cs activity concentrations of char in the 1st and 5th years after the accident in Fukushima. Ecological half-life (T_{eco}) of ^{137}Cs in char differed between samples collected in summer (1.4 y) and autumn (6.6 y). T_{eco} in summer samples was within the ranges of the previous studies (1.2 to 2.7 y), while autumn samples exhibited longer T_{eco}. Rate for decreases in ^{137}Cs activity concentrations in aquatic food items (29%) tended to be slower compared to those of terrestrial food items (82%). Therefore, high seasonal dependencies for terrestrial food items in summer induced greater reduction of ^{137}Cs activity concentrations together with high metabolic rates. Similarly, short T_{eco} in large-bodied char in summer samples possibly associated with high terrestrial food dependencies compared to small-sized char. The findings of this study suggested that differences of sampling season as well as body size need to be considered for the long-term projection using T_{eco}.

In conclusion, dietary contributions and their seasonal changes are important for explaining the variability of ^{137}Cs contamination of char. Such seasonal conditions and body sizes were not incorporated into the sampling campaign in most of the previous studies. Therefore, the sources of variability for ^{137}Cs contamination were not fully included in ^{137}Cs transfer modeling. Findings of this study will help for improving a model of ^{137}Cs transfer in ecosystems which had suggested by international organization of radiation projections and rehabilitation.

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CHAPTER 1

INTRODUCTION

1.1. Radionuclide contamination as global issue

Radionuclide contamination occurred all over the world due to nuclear weapon test, nuclear power generation and nuclear power plant accident (Beresford, 2005; Leieveld et al., 2012). The nuclear weapon test left a legacy of the test sites, while enormous amounts of radioactivity released following the major nuclear power plant accidents in the world such as the Khystym, the Chernobyl and the Fukushima accidents (Beresford, 2006). Moreover, mining and milling works of radioactive ores originated large amount of radioactive materials as a result of uranium decay (Batista et al., 2005; Carvalho et al., 2007). Once radionuclide released into the environments, contaminated the surrounding ecosystems which ultimately pose potential risk to the human health (Barnett et al., 2006; Prăvălie, 2014). Therefore, understanding the behavior of radionuclide in the ecosystems is essential which are linked to the development of radionuclide transfer models and/or management practices.

1.2. Radionuclide transfer in ecosystems

Radionuclide can be mobilized in the ecosystems through biological and ecological processes (Tikhomirov and Shcheglov, 1994; Avery, 1996; Monte et al., 2009; Murakami et al., 2014). Biological processes consist of the various activities of an animal's body such as intake of food, digestion, absorption and excretion; while the ecological processes consider energy flow from lower to higher trophic level animals (Figure 1.1). Between the biological and ecological processes individual is the key for linking of these two processes. For evaluating the radionuclide contamination of individuals' species, International Commission on Radiological Protection [ICRP] (2005) had introduced the 'Reference Species Approach' as an indicator of ecosystems contamination. Both the terrestrial and aquatic species were used as references species for a given ecosystem. For instance, deer, rat, bee, earthworm, grass, pine tree and brown seaweed were considered as terrestrial and frog, trout, flatfish, crab and duck were aquatic (ICRP, 2005).

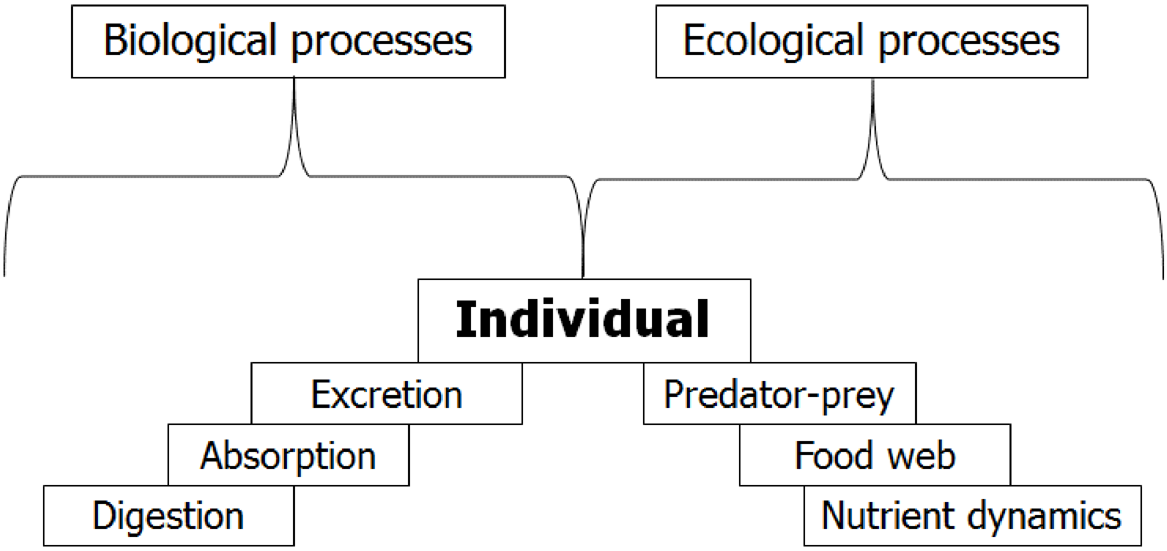


Figure 1.1. Conceptual diagram of radionuclide transfer via biological and ecological processes in ecosystems.

1.3. The Fukushima Dai-ichi Nuclear Power Plant accident

The Fukushima Dai-ichi Nuclear Power Plant (hereafter noted as FDNPP) was one of the promising source of electricity in Japan with 6 reactors of which first three reactors were in operation, and the rest three were undergoing routine maintenance on the date of accident (Miyashita, 2012). The Great East Earthquake of Japan which was M 9.0 with a hypocentral region of 500 km long and 200 km width attacked the northeast Honshu Island, Japan on March 11, 2011 (Hirose, 2012). Just after the earthquake, gigantic tsunami severely damaged the water circulation systems of reactors 1, 2 and 3; and resulted enormous amount of radioactive materials released to the atmosphere which caused serious environmental problems in Fukushima and north Kanto regions (Tagami et al., 2011; Hashimoto et al., 2012; Hirose, 2012; TEPCO, 2012; Teramage et al., 2014).

1.3.1. Radioactive materials released from the FDNPP accident

The contamination levels of the radioactive materials were not clearly known at the time of early release (Tagami et al., 2011); therefore radioactive plume detections were made at several monitoring stations in Japan by the Ministry of Education, Culture, Sports, Science and Technology (MEXT) and others (Chino et al., 2011; Yasunari et al., 2011; Wetherbee et al., 2012) following the accident to determine the radioactivity. About 900 PBq radioactive materials emitted to the atmosphere and ocean because of this accident, and increased levels of environmental radioactivity was found in Fukushima prefecture compare to the other parts of Japan (TEPCO, 2012). A number of different radionuclides like gamma emitting radionuclides (^{131}I, ^{132}Te, ^{134}Cs, ^{136}Cs and ^{137}Cs), alpha emitting volatile radionuclides (^{89}Sr, ^{90}Sr, ^{103}Ru and ^{106}Ru), pure beta-emitting radionuclides (^{3}H, ^{14}C and ^{35}S), gaseous radionuclides (^{85}Kr, ^{133}Xe and ^{135}Xe), radionuclides with very long physical half-lives (^{36}Cl, ^{99}Tc and ^{129}I) and some actinides (^{236}U) released from the FDNPP accident to the environment (Steinhauser, 2014). Among them, it was estimated that 3.3×10^{16} Bq of radiocesium (hereafter noted as ^{137}Cs) was emitted to the atmosphere (NISA, 2011). ^{137}Cs are considered as most important radioactive materials because they decay very slowly, accumulate in biota due to their high assimilation and slow clearance capacity from the tissue (Kolehmainen, 1972; Ugedal et al., 1992). ^{137}Cs also expected to cause long-term contamination in the environment because of their long physical half-lives (i.e., 30.2 years) (Sakai et al., 2014). Therefore, monitoring the ^{137}Cs dispersion across the ecosystems is essential to determine the transfer processes, factors and resulted contamination levels in biota, and to mitigate the radiological risks to the environment (Ashraf et al., 2014; Sakai et al., 2016b).

1.3.2. ^{137}Cs contamination in forest ecosystem

The ^{137}Cs dispersed in the environment can be deposited onto the forest ecosystems (IAEA, 2009). Forest ecosystems consist of tree biomass (e.g., boles, branches, leaves and roots), small dead organic matter on the soil surface (e.g., litter), dead trees on the soil surface (e.g., coarse woody litter) and soil (Hashimoto et al., 2012). Forested areas in Japan were the primal sources of radioactive materials because about 70% of contaminated land is covered by forest (Teramage et al., 2014). However, the affected areas in Fukushima are mostly covered by montane forest i.e., coniferous cedar (*Cryptomeria japonica*; Japanese name: Sugi) plantation forest (Kuroda et al., 2013). ^{137}Cs captured by the Japanese cedar (*C. japonica*) canopy (Kato et al., 2012; Takahashi et al., 2015), and then transferred to the forest floor through litterfall (Teramage et al., 2014). The litter layer on the forest floor is considered as one of the most contaminated components in forest ecosystems (Hashimoto et al., 2012; Murakami et al., 2014; Teramage et al., 2014; Sakai et al., 2016b) because litter layer continuously receive ^{137}Cs via litterfall and cause secondary contamination in forest floor (Teramage et al., 2014; Sakai et al., 2016b).

In such montane forests, well-developed stream networks characterized the terrestrial and aquatic ecosystems (Gomi et al., 2002). Despite the facts of high contamination in ecosystems, the movement of ^{137}Cs in ecosystems, and the consisted biota can be complex in hydrological, geomorphological and biological processes (Evrard et al., 2015). Radionuclide contaminants also affect both physical and biological processes of stream networks in forested catchment (Monte et al., 2009; Murakami et al., 2014; Sakai et al., 2016b). Therefore, ^{137}Cs in forest ecosystem are considered as primal source of contamination for forested headwater streams in riparian ecosystems (Sakai et al., 2016b).

1.3.3. ^{137}Cs dynamics in forested headwater streams

Headwater streams are the headmost area within channel networks, and are characterized by tight linkages among hydrologic, geomorphic and biological processes from hillslope to stream channels, and from terrestrial to aquatic environments (Gomi et al., 2002). Headwater streams are considered as abundant and unique components of a river network (Meyer et al., 2007). These streams are critical for nutrient dynamics, and provide habitats for macroinvertebrates, fish and amphibians within the watersheds (Meyer and Wallace, 2001). The processes in headwater streams are the key to comprehend the ^{137}Cs dynamics in riparian and riverine ecosystems because ^{137}Cs would move from the terrestrial to aquatic ecosystems via nutrient transfer because of tight linkage between forests and stream channels (Figure 1.2) (Gomi et al., 2002).

Understanding the hydrogeomorphic (Sidle et al., 2000) and biological (Richardson, 1992; Wallace et al., 1999; Sakai et al., 2016b) processes from forest to stream channels is important to elucidate the functional linkage among the ecosystem components in headwater streams for addressing the relevant ecological processes (Gomi et al., 2002; Sakai et al., 2016). For instance, the released ^{137}Cs can be transported in headwater catchment though fluvial (Onda et al., 2012) and biological processes (Figure 1.3) (Sakai et al., 2016b). The movement, depositions and accumulations of ^{137}Cs in forested headwater streams may induce contamination of biota within the habitats (Sakai et al., 2016b). The aquatic ecosystems strongly depends on the terrestrial energy sources such as leaf litter and terrestrial insects. For instance, leaf litter enter into the stream channels as a primal source of contaminations (Sakai et al., 2016b), decomposed and/or leached into the water column; and transferred to other substances such as clay materials (Sakai et al., 2015). ^{137}Cs attached to soil, organic matter and clay particles transported via channel networks from up-to down-streams, and finally reached to the ocean (Tanaka et al., 2014; Evrard et al., 2015). These contaminants tend to be accumulated in sediments on stream bed and coastal areas (Lepage et al., 2016). Moreover, wash off of ^{137}Cs from the catchment become a long-term source of ^{137}Cs in aquatic ecosystems (IAEA, 2009). Local agricultural irrigation ponds and check dams also potentially accumulate ^{137}Cs that are transported from the upstream regions (Yamada et al., 2015). Therefore, understanding the ^{137}Cs exposure pathways across ecosystems is essential to elucidate the impact of ^{137}Cs contamination on landscape-level processes (Sakai et al., 2016b).

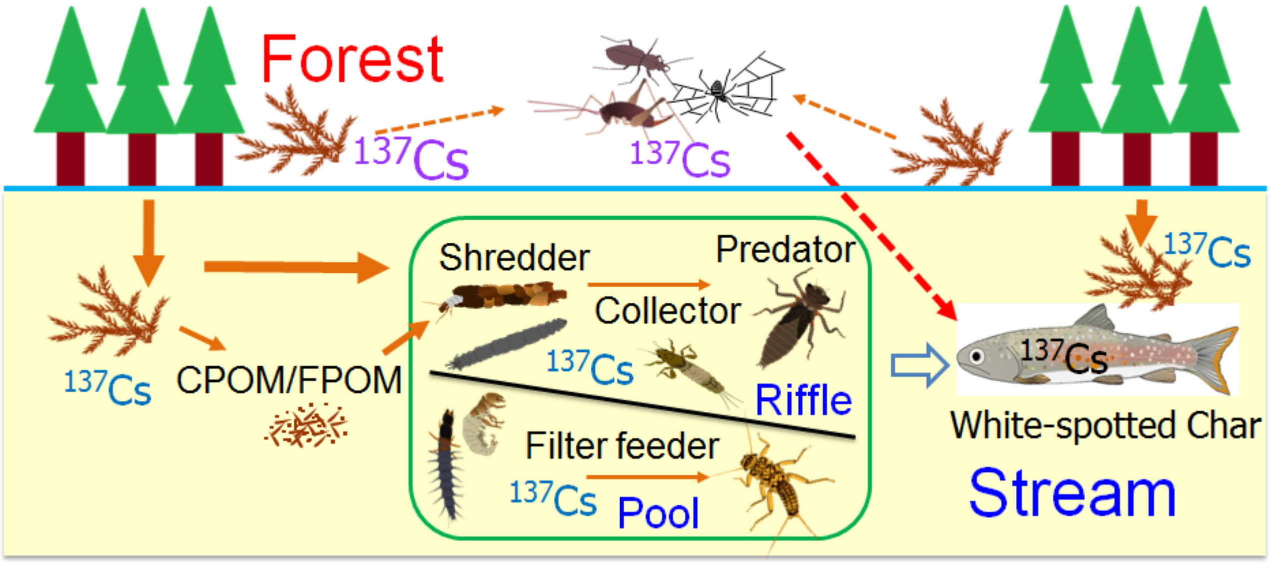


Figure 1.2. Schematic illustration of ^{137}Cs transfer from forest to stream.

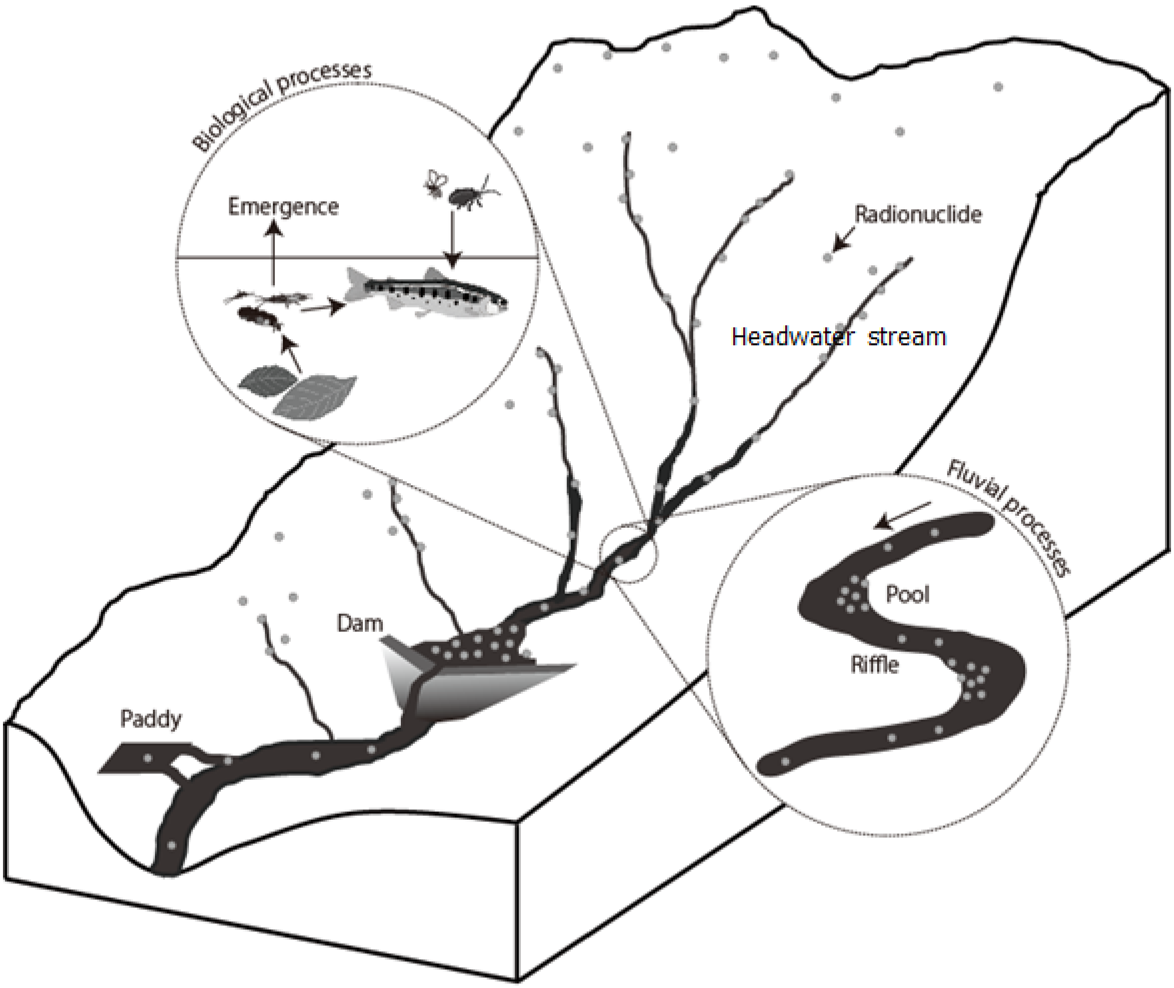


Figure 1.3. Schematic illustration of ^{137}Cs transfer in watershed.

1.3.4. ^{137}Cs exposure pathways in biota of forested headwater streams

External exposure may occur in organisms which inhabited in the contaminated areas within and/or around the streams and rivers along with internal exposure by consumption of contaminated food sources. Direct uptake from the water column may lead to both direct irradiation and internal exposure if the ^{137}Cs becomes assimilated and distributed within the organisms (ICRP, 2009). This internal exposure may continue via food webs in ecosystems from the producers to the animals of higher trophic levels in aquatic and/or stream ecosystems (Murakami et al., 2014; Sakai et al., 2016b). The conceptual view of ^{137}Cs exposure pathways in biota of forested headwater streams were illustrated in Figure 1.4. Moreover, ^{137}Cs has a long physical half-life (Sakai et al., 2014), and can be potentially accumulated in the organisms (Sakai et al., 2016b; Wada et al., 2016). For instance, Teramage et al., (2014) showed that about 68% of ^{137}Cs were deposited from the forest canopy to the forest floor via litterfall in coniferous forest in Fukushima. Onda et al. (2012) showed that ^{137}Cs would deposited into the water bodies through geomorphic processes. Murakami et al. (2014) showed that detrital channels are the primary route for ^{137}Cs transfer via trophic levels in both terrestrial and aquatic ecosystems in Fukushima. Similarly, Sakai et al. (2016b) demonstrated that ^{137}Cs transferred via detrital based food web from primary producer (i.e., litter) to organisms of higher trophic levels in both forest and headwater streams in Fukushima.

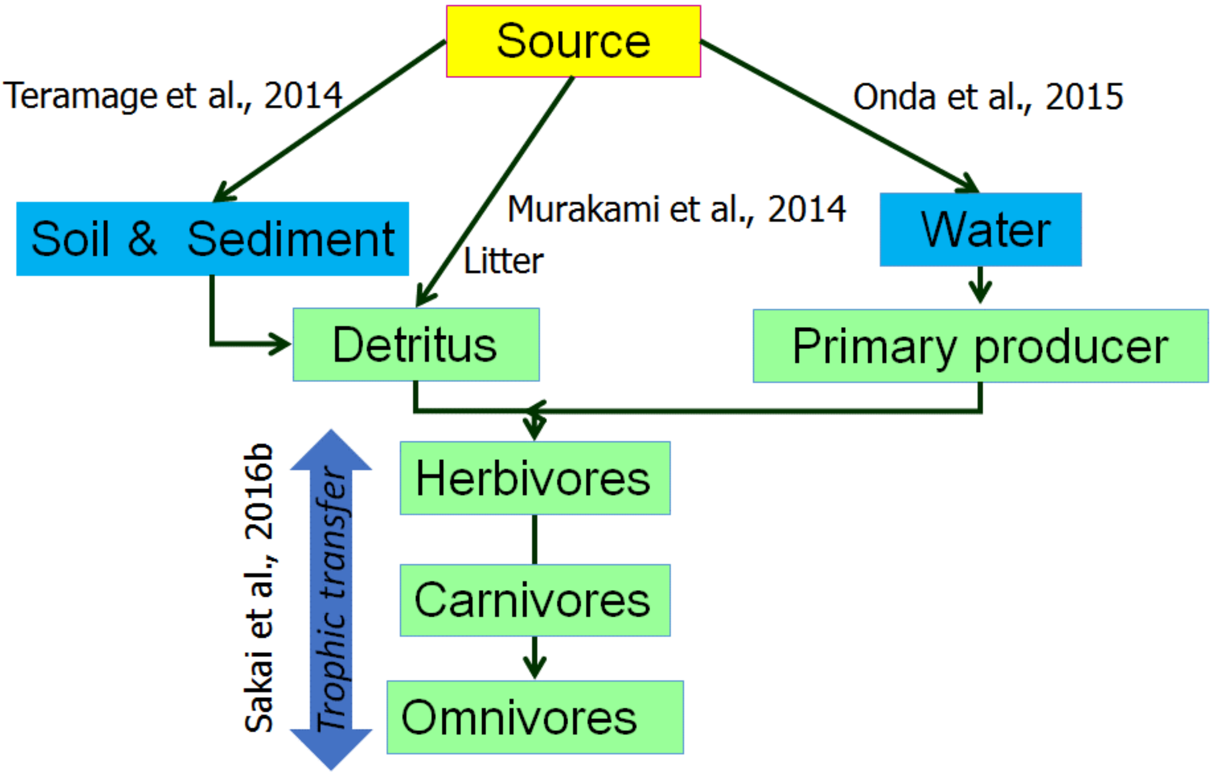


Figure 1.4. Conceptual view of ^{137}Cs exposure pathways in biota of headwater streams.

1.4. Importance of fish as bio-indicator

Fish are considered as bio-indicator because of long life span, easy sampling, and can accumulated ^{137}Cs easily (Anim-Gyampo et al., 2013). Fish are one of the important food sources for our lives; investigation of transfer and biological accumulation in fish are essential for resources management within watersheds. Predation in aquatic food web leads to transfer of ^{137}Cs to higher trophic animals, including fish (ICRP, 2009). Fish is also considered as an important indicator of given environment for transfer of ^{137}Cs from terrestrial to aquatic ecosystems via geomorphic and biological processes (Onda et al., 2012; Murakami et al., 2014; Sakai et al., 2016b). Moreover, areas with contaminated watersheds are important for freshwater fisheries which are one of the primal local industries. Since ^{137}Cs had long physical half-life, long-term prediction based on scientific investigations is also necessary to understand the long-term decay in biota, essential for planning and establishing proper strategies to manage the ecosystems. The conceptual illustration regarding the ^{137}Cs accumulation in fish via food subsidies in aquatic ecosystems were presented in Figure 1.5. Ugedal et al. (1995) described ^{137}Cs accumulation in fish by a simple model: accumulation = intake – excretion. Thus, any factors affecting either intake or excretion may contribute to the contamination levels of the given fish species. ^{137}Cs in freshwater fish species are mainly accumulated through consumption of contaminated food stuffs, while intake through gill and body surface due to concentration gradient is very negligible (King, 1964; Hewett and Jefferies, 1978). Because of chemical similarity of ^{137}Cs with K^+ it has known to influence the ^{137}Cs accumulation in fish (Blaylock, 1982). Previous studies had shown strong inverse relationship between ^{137}Cs activity concentrations and potassium concentration in water (Blaylock, 1982; Smith et al., 2000a). White-spotted char (*Salvelinus leucomaenis*; Pallas, 1814) (Japanese name: Iwana) used in this study is a unique headwater salmonid fish species.

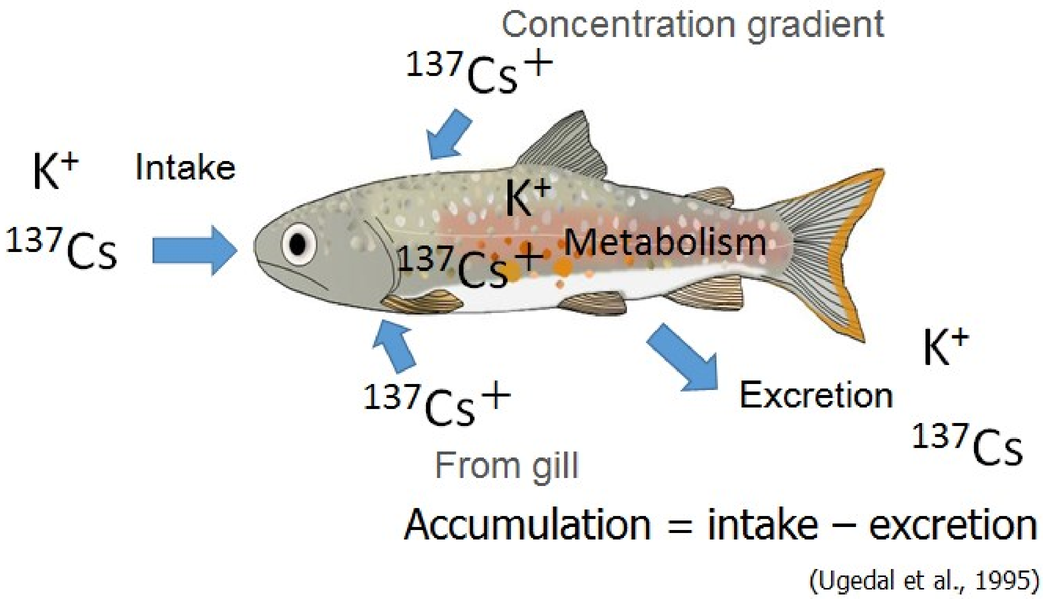


Figure 1.5. Schematic illustration of ^{137}Cs accumulation in fish.

1.4.1. White-spotted char (*Salvelinus leucomaenis*; Pallas, 1814)

White-spotted char (*S. leucomaenis*) (Japanese name: Iwana) is cold-water adapted freshwater salmonid fish species in far-eastern Asia (Yamamoto et al., 2004). Like other salmonids, its distribution ranges from higher altitudes to lower latitude regions (Nakano et al., 1996). At higher latitudes, char uses different habitats within a river from headwaters to the mouth (Fausch et al., 1994). They are common in Honshu Island, Japan (Dunham et al., 2008), key-top predator in headwater streams, and also considered as an important source of fishery resources in Japan (Nakano and Murakami, 2001; Miyasaka et al., 2003). Through the size and weight of char varies according to the age of char; however, Esin and Sorokin (2012) reported the maximum size, weight and age of char are 285 mm, 245 g and 7 years, respectively. They are insectivorous, aggressive drift forager and relying on both terrestrial and aquatic food sources (Nakano and Furukawa-Tanaka, 1994; Nakano and Murakami, 2001; Miyasaka et al., 2003). Dipteran larvae, raphidosphorids, tetragnathids among the terrestrial insects, and stream macroinvertebrates such as mayfly, stonefly, midges and amphipods are the dominant prey items of char (Nakano and Furukawa-Tanaka, 1994; Miyasaka et al., 2003; Sakai et al., 2016b). The mean wet weight of dietary compositions of char was 7.57 ± 6.78 mg, while mean diet ratio was 1.91 ± 0.80 mg/5 min per gram of body weight (Nakano and Furukawa-Tanaka, 1994). Char also have seasonal dietary preferences depends on terrestrial prey items in summer and aquatic prey items in winter (Kawaguchi and Nakano, 2001; Nakano and Murakami, 2001). Recently, Sakai et al. (2016b) showed that ^{137}Cs activity concentrations in terrestrial insects were 4 times greater than those of aquatic in forested headwater stream. Therefore, multiple dietary sources with their respective ^{137}Cs contamination levels along with shifting food source habitats of char across season need to be considered in studying ^{137}Cs transfer for char in forested headwater streams.

1.5. Necessity and justification of this studies

The environmental problems caused by the radioactive materials couldn't be minimized without increased awareness and complex regulations (Stark et al., 2006). To reduce the problems caused by local and global environmental pollutions, both biota and ecosystems are need to be protected from the impact of contaminants (UNCED, 1992). Recently, ICRP has been expanded their radiation framework to protect the environment by preventing or reducing the deleterious effects of radioactive materials to a minimal levels for the conservation and maintenance of the species, habitats, communities and ecosystems (Valentin, 2007). However, scientific studies are necessary to reach a better understanding of the circumstances of the nuclear accident, the behaviour of radioactive materials in different environmental media, and their associated countermeasure practices.

However, the dynamics of ^{137}Cs emitted from the FDNPP accident is different than those of previous major accident, such as the Chernobyl accident because of the differences in landscape structures, environmental conditions and data collection date between the contaminated areas affected by these two major accidents (Hilton et al., 2008; Shinomiya et al., 2014). Different landscape structures and climatic conditions possibly affect hydrological and biological processes for ^{137}Cs dynamics. The ^{137}Cs dynamics in forested headwater stream of riparian ecosystem after the FDNPP accident aren't studied well, though the headwater streams are considered as source of contaminations for downstream regions. Therefore, considering the current situation of ^{137}Cs threats following the FDNPP accident, it is crucial to carry out research for the investigation of the effects of ^{137}Cs in the stream-riparian ecosystem and their biota, particularly fish because they harbour both terrestrial and aquatic organisms including fish to meet the following general and scientific needs:

General

1. To assess the current and predict the future levels of contamination in biota predominantly fish of headwater streams to justify remedial actions and long-term counter-measurements.
2. To inform the general people in contaminated areas about the persistence, seasonal and annual variability of ^{137}Cs contamination levels in fish, and to give dietary advice about fish and other aquatic resources accordingly to reduce ^{137}Cs intake in human being.
3. To inform the general people in contaminated areas about changing radiological conditions in order to relieve public concerns.

Scientific

1. To determine ^{137}Cs transfer processes to fish in headwater streams to specify the predictive models for contaminated areas, and for potential future radioactive releases.
2. To determine the long-term behavior of ^{137}Cs and factors affecting ^{137}Cs contamination levels in fish of forested headwater streams to clarify persistence of ^{137}Cs, and explore remediation possibilities with special attention to processes of importance for contribution to biota doses.

There are many identified research directions based on the ^{137}Cs dynamics in forested headwater streams such as (i) how does the ^{137}Cs movement, transfer and deposit in headwater streams?; (ii) how does the ^{137}Cs accumulate in biota of forest and stream ecosystems?; (iii) how does terrestrial energy subsidize to the biota including fish of stream networks?; (iv) how does the ^{137}Cs accumulate in fish via food webs of headwater streams?; (v) how does the ^{137}Cs activity concentration in fish vary according to the season in Japanese channel networks?; (vi) how does the factor affecting ^{137}Cs activity concentrations in fish in stream-riparian ecosystem? Aiming to solve these questions, I conducted comprehensive field observations, collected the biotic samples including fish for measuring ^{137}Cs activity concentrations, and stable isotope ratios to estimate the trophic position in food webs by following proper scientific methods. In addition, I investigated the food sources of fish in my study sites, and analysed the gut contents to determine their food and feeding habits.

The research presented in this thesis was conducted for better understanding of ^{137}Cs accumulation in fish of forested headwater streams with complex energy subsidies between forest and stream in riparian ecosystems. Parallelly, this study considered possible recommendation for improving ^{137}Cs transfer and/or accumulation models that would help the policy makers to take necessary steps for successful decontamination practices and associated efforts for implementations.

This study was mostly focused on two headwater streams with similar surrounding environmental conditions but different fallout volume affected by the FDNPP accident. Osawaga-stream at Nihonmatsu city in Fukushima prefecture is located about 45 km away from the FDNPP. Oya-san stream at Midori city in Gunma prefecture is about 190 km away from the FDNPP. Based on governmental airborne investigation on June 2012, the mean air dose rate and ^{137}Cs fallout inventory in Fukushima site were 1.0–1.9 μSv h^{-1} and 100–300 kBq m^{-2}, respectively, while those of Gunma site were 0.2–0.5 μSv h^{-1} and 30–60 kBq m^{-2}, respectively (MEXT, 2012). The drainage area of Fukushima site was 170 ha and that of Gunma site was 93 ha. Both of the study sites were typical Japanese headwater channels consisting of sequences of steps and pools and draining by Japanese cedar (*C. japonicus*) plantation forest. Freshwater salmonid, white-spotted char (*S. leucomaenis*) is the top predator of these headwater streams, mostly insectivorous, and depends on the energy fluxes of forest and stream ecosystems.

1.6. Objectives and structure of this thesis

The objectives of this study were (1) *to develop a method of transfer factor for examining ^{137}Cs transfer via food web structures for white-spotted char in forested headwater streams*; (2) *to clarify the seasonal variability of newly developed ^{137}Cs transfer factor in white-spotted char from headwater streams*; and (3) *to examine the time-series change of ^{137}Cs activity concentrations in white-spotted char of headwater stream after 5 years of the FDNPP accident*. I also discussed about the management practices for decontamination of ^{137}Cs within channel networks for the sustainable management of streams and/or aquatic ecosystems.

The findings of this study were organized into six chapters (Figure 1.6). Each chapter comprehensively represented the key processes, mechanisms and factors of ^{137}Cs transfer and/or accumulation in white-spotted char in forested headwater streams. Though the approaches that were taken in each chapter differ a little from each other, but the findings of each chapter were interlinked to the previous chapter(s) and were discussed in logical sequences. In chapter 1, I highlighted the background, importance, necessity and justification of this study followed by the objectives and structure of this research. In chapter 2, I demonstrated the variability of ^{137}Cs contamination levels in freshwater fish in association with different transfer processes, accumulation patterns, and spatio-temporal changes of ^{137}Cs activity concentrations in fish based on the review of previous studies. The purpose of chapter 3 was to develop an indicator for examining the ^{137}Cs transfer based on food web structures by integrating the dietary ^{137}Cs contributions of multiple prey items to a target predator (i.e., white-spotted char, *S. leucomaenis*; Japanese name: Iwana). I denominated the indicator as 'food web-based transfer factor' and symbolized by 'TF_{web}' for use in ^{137}Cs dynamics. Chapter 4 evaluated the seasonal variability in TF_{web} of ^{137}Cs to address the impacts of prey items contributions, specific metabolic rate of white-spotted char, and fallout volume in two different headwater streams with similar surrounding environmental conditions. The aims of chapter 5 was to examine the time-series change of ^{137}Cs contamination levels in white-spotted char using ecological half-life (T_{eco}) by making a comparison between the data quantified after 1 and 5 years of the FDNPP accident. Finally, summary and conclusions derived from each chapter of this study, management applications, and importance and possible applications of the findings of this study in Bangladesh were discussed in Chapter 6.

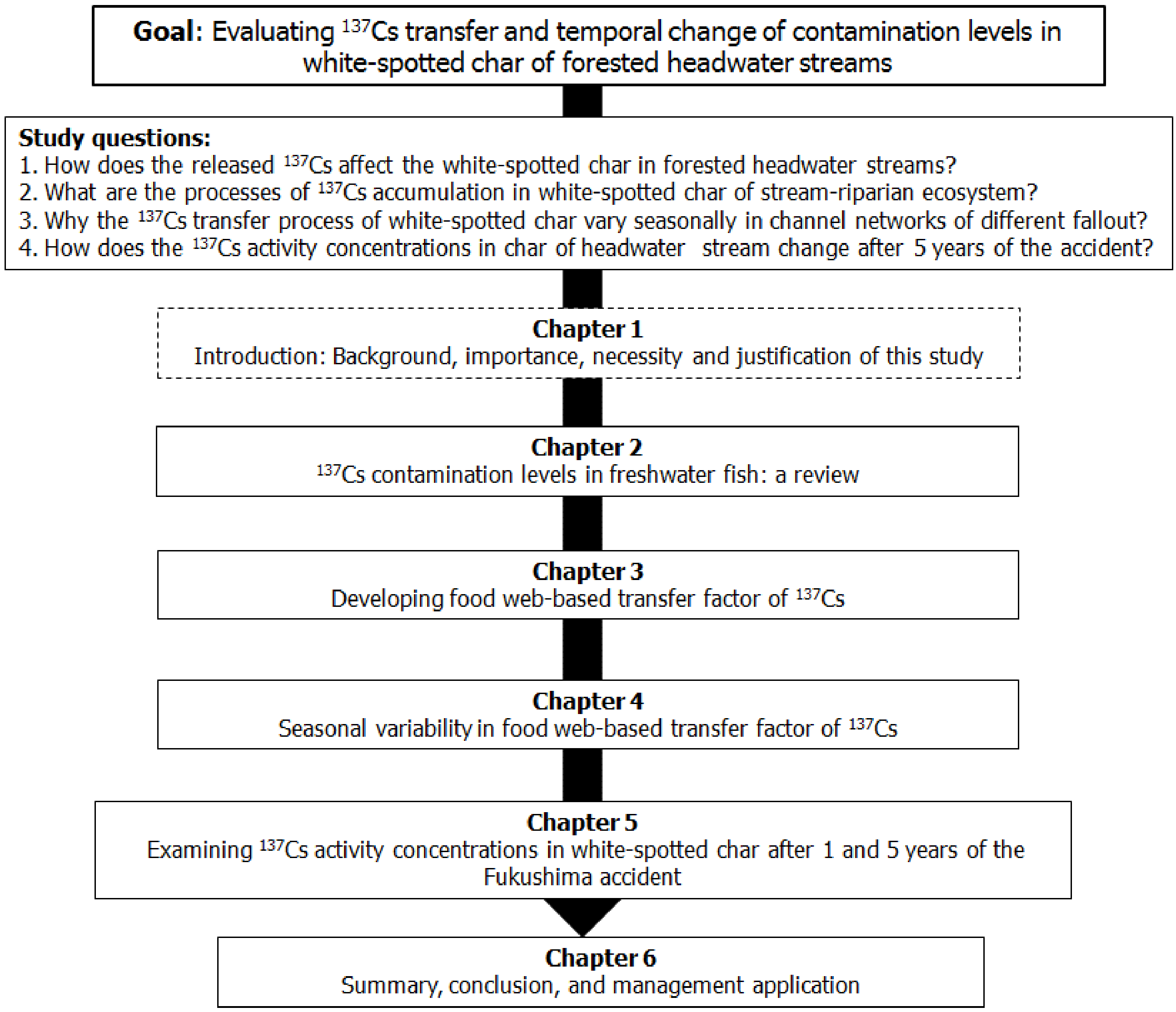


Figure 1.6. Flow chart illustrating the structure of this study.

CHAPTER 2

^{137}Cs CONTAMINATION IN FRESHWATER FISH: A REVIEW

2.1. Introduction

Radioecology is a science that deals about the interactions between radionuclides and environments (Aarkrog, 2000). This study was started after the discovery of *X*-rays in 1895, and the term ‘radioecology’ was first introduced into the scientific vocabulary in 1956 (Alexakhin, 2006). The most extensive creative work related to radioactivity was on radium as a new and enormous source of energy (Kautzleben and Müller, 2014). Later, N.V. Timofeev-Resovsky in 1940 calculated the radiation doses in a living body based on radioactivity works in the vicinity of atomic industry facility in the USSR (Alexakhin, 2006). In 1942, after the nuclear chain reactions at the University of Chicago, the Manhattan Project expanded their research activities on environmental radioactivity across the USA, including Hanford in Washington State (Gray and Becker, 1993; Shaw, 2005). Welander (1946) and Foster (1948) highlighted radiological impact of the Hanford reactors on chinook salmon (*Oncorhynchus tshawytscha*) and rainbow trout (*Salmo gairdneri*) in the Columbia River. During 1950 and early 1960, radionuclides to the environment was increased due to atmospheric weapons testing, then examining the radionuclides contamination levels in various ecosystem components, and the associated effect of radiation on human and other biota become the major concern (Aleksakhin, 2006). In 1951, the Savannah River Ecology Laboratory investigated extensively the effects of radiation on organisms and in ecosystems (Odum, 1956). These researches were extended not only for investigating the contamination level of a given species, but also the processes of contamination transfer in ecosystems (Shaw, 2005).

Forest ecosystems were mostly contaminated by the major nuclear power plant accidents, such as the Kyshtym, Chernobyl and Fukushima accidents (Tikhomirov et al., 1993; Kuroda et al., 2013). In Fukushima, 70-80% of total radioactive materials deposited on the forested areas (Hashimoto et al., 2012), while half of the 30 km zone around the Chernobyl reactor is covered by forest (Kashparov et al., 2000). In forested landscapes, contaminated materials could be secondarily mobilized by their ecosystem processes (Tikhomirov et al., 1993; Sakai et al., 2016b). For instance, litterfall from forest canopy contributed 45% for total ^{137}Cs movement to ground surface, while throughfall and stemflow contributed 53% and 2.3%, respectively (Teramage et al., 2014). Therefore, contamination of litter as primary producer in both forest and stream ecosystems alter the contamination levels of biota because both habitats and dietary sources were contaminated (Sakai et al., 2016b). Hence, Sakai et al. (2016b) showed that ^{137}Cs contamination of aquatic litter was 75% lower than ones in terrestrial associated with leaching of ^{137}Cs. Such differences had potential effects for different contamination levels in food web components between terrestrial and aquatic ecosystems (Sakai et al., 2016b). In these consequences, understanding the complex processes of radionuclide transfer become one of the challenge for radioecological studies (Hinton et al., 2014).

Freshwater fish is one of the bio-indicators for radionuclide transfer and resultant contamination (Anim-Gyampo et al., 2013). Fish is also an important indicator of given environments (i.e., lakes, ponds, wetlands, streams and rivers) for transfer of contaminants from terrestrial to aquatic ecosystem via hydrological and geomorphic processes (Avery, 1996; Onda et al., 2012 Evrard et al., 2015). They are also important local fishery resources and local food supply; thereby contamination of them also critical for human health (Tjahaja et al., 2012; Tsuboi et al., 2015). For instance, perch (*Perca fluviatilis*) located in lake about 232 km away from the Chernobyl contaminated area had 5006 Bq kg^{-1} of ^{137}Cs, which was 50 times greater than the maximum concentration of health standard (Smith et al., 2000a). Similarly, ayu (*Plecoglossus altivelis*), which is one of the important fish resources in Japan had 1770 to 3300 Bq kg^{-1} in river of Fukushima prefecture located 20–40 km away from the FDNPP (Mizuno and Kubo, 2013).

These previous studies also revealed the tempo-spatial variability of radionuclide contamination for a given species, although they were sampled from similar contaminated areas.

For instance, Sundbom et al. (2003) showed that pike (*Esox lucius*) had 153 to 1798 Bq kg^{-1} of ^{137}Cs activity concentrations with contamination levels of 20–30 kBq m^{-2}. Mizuno and Kubo (2013) showed that ^{137}Cs activity concentrations differed two times between salmonids (Salmonidae) and sweetfish (Plecoglossidae) in Fukushima prefecture. Findings of these previous studies showed that understanding the variability of contamination of given fish species are essential for elucidating the factors and processes of contamination in aquatic ecosystems.

Therefore, the objectives of this study were to (1) investigate the characteristics of freshwater fish inhabiting around the contaminated areas; (2) summarize the contamination levels of given fish species; (3) examine the factors for controlling the contamination levels; (4) elucidate tempo-spatial variation of contamination levels in freshwater fish; and (5) develop countermeasures for controlling the contamination level in aquatic ecosystem. In this study, I focused on ^{137}Cs exposure in freshwater fish based on previous studies because it has long physical half-live (i.e., 30.2 years), and common radionuclide in the contaminated areas.

2.2. Characteristics of freshwater fish around the contaminated area

The characteristics of freshwater fish are important for evaluating their ^{137}Cs contamination levels (Braune et al., 1999). ^{137}Cs contamination level in a given fish depends on fish species, feeding behaviour, prey items, and prey's prey in given contaminated area (Håkanson, 1999) as ^{137}Cs accumulated in fish mainly by consumption of contaminated food items (Avery, 1996). Trophic position of freshwater fish is considered as a key parameter for understanding ^{137}Cs accumulation (Zhoa et al., 2001; Sundbom et al., 2003) which usually comprised four well defined trophic levels with phytoplankton as primary producer (planktivores), herbivores that include zooplankton feeding on these, detrivores such as benthic insect larvae and crustaceans, and carnivores feeding on benthic organisms (Braune et al., 1999). For instance, roach (*Rutilus rutilus*), bream (*Abramis brama*), silver carp (*Hypophthalmichthys molitrix*) are planktivores mainly feed on phytoplankton, while ayu (*P. altivelis*), goldfish (*Carrasius auratus*), ruud (*Cardinus erytrophtalmus*) are mainly herbivores feed on algae and plant materials (Table 2.1). Moreover, crussian carp (*Carassius* spp.), tench (*Tinca tinca*) are detrivores feed on detritus and organic matter, whereas white-spotted char (*Salvelinus leucomaenis*), brown trout (*Salmo trutta*), pike (*E. lucius*), perch (*P. fluviatilis*) are carnivores feed on invertebrates, insects and small fish.

Differences in diets with respective contamination levels associated with the functional feeding group of freshwater fish species may influence the variability in their ^{137}Cs contamination levels. For example, Iguchi et al. (2013) showed that the ^{137}Cs activity concentrations in ayu (*P. altivelis*) differed at various locations in accordance to the contamination labels of algae because ayu are hervivours and mostly contaminated by consuming contaminated algae. Rowan et al. (1998) showed that ^{137}Cs activity concentrations in benthivorous fish such as white sucker, johnny darter were about 2 times greater than planktivorous fish (i.e., yellow perch, emerald shinner) in Ottawa river because of large ^{137}Cs inventories. Sundbom et al. (2003) reported that fish of intermediate trophic levels had the highest ^{137}Cs contamination with relatively small effect of trophic structure. Mizuno and Kubo (2013) reported that ^{137}Cs activity concentrations were higher in carnivorous fish species such as masu salmon (*O. masou*) and white-spotted char (*S. leucomaenis*) compare to the herbivorous (*P. altivelis*) and omnivorous (*T. hakonensis*, *C. carpio*, *Carrasius* sp) fish by studying in the Aga river basin in Fukushima. Therefore, characteristics of fish vs. ^{137}Cs relationship of any fish possibly depends on foodstuffs, feeding patterns, assimilation, consumption, ingestion, elimination, growth and differentiation, and indeed for making accurate predictions, detail study is needed.

Table 2.1. List of ^{137}Cs contaminated freshwater fish, their dominant habitats, food items, and location of study.

Name of Freswater Fish		Dominant Habitat	Dominant Consumtion of Food	Location of Study	References
English Name	Latin Name				
Bullhead catfish	*Ameiurus natalus*	Pond	Invertebrates, fish	South Carolina, USA	Peles et al., 2000a
Redfin pickerel	*Esox americanus*	Stream	Invertebrates, benthic organisms	South Carolina, USA	Peles et al., 2000b
Pirate perch	*Aphredoderus sayanus*	Stream	Invertebrates	South Carolina, USA	Peles et al., 2000b
Spotted sunfish	*Lepomis auritus punctatus*	Stream	Invertebrates, detritus	South Carolina, USA	Peles et al., 2000b
Warmouth	*Lepomis auritus gulusus*	Stream	Invertebrates, detritus	South Carolina, USA	Peles et al., 2000b
Bowfin	*Amia calva*	Stream	Invertebrates, fish	South Carolina, USA	Burger et al., 2001
Dusky shiner	*Notropis cummingsae*	Stream	Invertebrates, zooplankton	South Carolina, USA	Burger et al., 2001
Yellow perch	*Perca flavescens*	Stream	Invertebrates, fish	South Carolina, USA	Burger et al., 2001
Red-breast sunfish	*Lepomis auritus*	Stream	Invertebrates	South Carolina, USA	Burger et al., 2001
Black crappie	*Pomoxis nigromaculatus*	Stream	Invertebrates, fish	South Carolina, USA	Burger et al., 2001
Channel catfish	*Ictalurus punctatus*	Stream	Insects, algae, snail, fish	South Carolina, USA	Burger et al., 2001
Silver carp	*Hypophthalmichthys molitrix*	Pond	Phytoplankton, algae, plant materials	Ukraine	Kryshev, 1995
Pike-perch	*Lucioperca luciopera*	Pond	Fish, invertebrates	Ukraine	Kryshev, 1995
Silver bream	*Blicca bjoerkna*	Pond, Reservoir	Benthic organism, invertebrates	Ukraine	Kryshev, 1995
Channel catfish	*Ictalurus punctatus*	River, Pond	Insects, algae, snail, fish	Ukraine	Oleksyk et al., 2002
White fish	*Coregonus lavaretus*	Lake	Benthic organisms, zooplankton	Finland	Rask et al., 2012
Vendace	*Coregonus albula*	Lake	Zooplankton	Finland	Rask et al., 2012
Eel	*Anguilla anguilla*	Lake	Insects, bivalves	Finland	Rask et al., 2012
Roach	*Rutiulus rutilus*	Lake	Plankton, benthic organisms	Finland, Belarus	Smith et al., 2000a
Bream	*Abramis brama*	Lake, Reservoir	Invertebrates, zooplankton, plant materials	Finland, Belarus, Ukraine, Sweden	Sundbom et al., 2003
Perch	*Perca fluviatelis*	Lake, Pond, Reservoir	Invertebrates, fish	Finland, England, Sweden, Ukraine, Belarus	Elliot et al., 1992; Smith et al., 2003; Saxén et al., 2005
Pike	*Esox lucius*	Lake, Pond, Reservoir	Invertebrates, fish	Finland, England, Sweden, Ukraine, Belarus	Elliot et al., 1992; Smith et al., 2003; Saxén et al., 2005
Gudgeon	*Gobio gobio*	Lake	Benthic organisms	Belarus, Sweden	Smith et al., 2003
Ruffe	*Acerina cernua*	Lake	Benthic organisms, fish egg	Belarus, Sweden	Smith et al., 2003
Rudd	*Cardinus erytrophtalmus*	Lake, Reservoir	Algae, phytoplankton, plant materials	Belarus, Ukraine, Sweden	Smith et al., 2003
Tench	*Tinca tinca*	Lake, Reservoir	Invertebrates, plankton, plant material	Belarus, Ukraine, Sweden	Smith et al., 2003
Goldfish	*Carrasius auratus*	Lake, Reservoir	Algae, plant materials	Belarus, Ukraine, Sweden	Smith et al., 2003
Arctic charr	*Salvelinus alpinus*	Lake	Benthos, plankton, fish	Norway, England, Sweden	Forseth et al., 1991; Elliot et al., 1993; Hammar, 1998
Brown trout	*Salmo trutta*	Lake	Drifting insects, zooplankton	Norway, England, Sweden, Japan	Forseth et al., 1991; Elliot et al., 1993; Yoshimura and Yokoduka, 2014
Ayu	*Plecoglossus altivelis*	River	Algae, plant materials	Japan	Wada el al., 2016
Sockeye salmon	*Oncorhynchus nerka*	River	Plankton, bottom dwelling organisms	Japan	Arai, 2014a
Rainbow trout	*Oncorhynchus mykiss*	Lake	Invertebrates, plankton, fish, fish egg	Japan	Yoshimura and Yokoduka, 2014
Pond smelt	*Hypomesus nipponensis*	Lake	Invertebrates, zooplankton	Japan	Wada et al., 2016
Kokanee	*Oncorhynchus nerka*	Lake	Invertebrates, plankton, fish	Japan	Wada et al., 2016
Japanese dace	*Tribolodon hakonensis*	Lake, River	Benthic organisms	Japan	Wada et al., 2016
Masu salmon	*Oncorhynchus masou*	Stream, Lake, River	Insects and fish	Japan	Waada et al., 2016
White-spotted char	*Salvelinus leucomaenis*	Stream, River	Insects	Japan, USA	Wada et al., 2016
Largemouth bass	*Micrpterus salmoides*	Stream, River	Fish, insects	Japan, USA	Burger et al., 2001; Matsuda et al., 2015
Blue gill	*Lepomis macrochirus*	River	Invertebrates, plankton	Japan, USA	Burger et al., 2001; Matsuda et al., 2015
Japanese eel	*Anguilla japonica*	River	Benthic organisms, invertebrates, fish	Japan, Finland	Rask et al., 2012; Wada et al., 2016
Crussian carp	*Carassius* spp.	River, Pond	Benthic organisms, plankton, detritus	Japan, Ukraine	Jagoe et al., 1997; Wada et al., 2016
Common carp	*Cyprinus carpio*	River, Pond	Benthic organisms, plant materials	Japan, Ukraine	Franić and Marović, 2007; Wada et al., 2016

Streams are small, steep gradient chanels including hillslopes, zero-order basins, and first and second order channels.
River is the large water body where many streams meet, including six to twelve order channels.

2.3. ^{137}Cs contamination in freshwater fish species

The ^{137}Cs activity concentrations in freshwater fish varied widely depends on the habitat condition, fallout amount, sampling time, type, size, and feeding habit of fish species. After the Chernobyl accident, the mean ^{137}Cs activity concentrations in brown trout (*S. trutta*) were varied from 404 to 540 Bq kg^{-1} by studying in two different lakes with similar fallout amount (i.e., 4 kBq m^{-2}) in England (Elliot et al., 1992), while that of arctic charr (*S. alpinus*) in another two different lakes with 2–20 kBq m^{-2} fallout of same country ranged from 164 to 204 Bq kg^{-1} during 1986-1988 (Elliot et al., 1993) (Table 2.2). Saxén et al. (2005) showed that ^{137}Cs activity concentrations in perch (*P. fluviatilis*) were ranged from 34 to 59 Bq kg^{-1} by studying in Finnish lakes having 0–4 kBq m^{-2} fallout volume. Forseth et al. (1991) reported that the maximum ^{137}Cs activity concentrations in brown trout (*S. trutta*) and arctic charr (*S. alpinus*) were 16340 and 5460 Bq kg^{-1}, respectively by studying at Høysjøen lake of 50 kBq m^{-2} fallout in Norway in 1986–1989.

Similarly, ^{137}Cs activity concentrations in silver carp (*H. molitrix*) and pike-perch (*Lucioperca lucioperca*) varied from 18000 to 192000 Bq kg^{-1}, and from 4000 to 355000 Bq kg^{-1}, respectively in the Chernobyl Cooling Pond during 1986–1989 (Koulikov and Ryabov, 1992). Broberg et al. (1995) reported that the mean ^{137}Cs activity concentrations in perch (*P. fluviatilis*), roach (*R. rutilus*) and pike (*E. lucius*) were ranged from 850 to 10500 Bq kg^{-1}, from 200 to 2100 Bq kg^{-1}, and from 700 to 9400 Bq kg^{-1}, respectively during 1989–1992 by studying in various lakes in Sweden. The mean ^{137}Cs activity concentrations in carp (*Cyprinus carpio*) and silver bream (*Blicca bjoerkna*) ranged from 15000 to 100000 Bq kg^{-1}, and from 8000 to 110000 Bq kg^{-1} in the Chernobyl Cooling Pond during 1986–1990 (Kryshev, 1995). Koulikov (1996) reported that the minimum and maximum values of ^{137}Cs activity concentrations in goldfish (*Carassius* sp.) and tench (*T. tinca*) of Keiv Reservoir between 1987–1992 ranged from 197 to 1291 Bq kg^{-1}, and from 255 to 1650 Bq kg^{-1}, respectively.

After the FDNPP accident, ^{137}Cs activity concentrations in white-spotted char (*S. leucomaenis*) in headwater stream of 100–300 kBq m^{-2} fallout volume in Fukushima prefecture varied from 704 to 6080 Bq kg^{-1} during 2012–2013 (Sakai et al., 2016b). Yoshimura and Yokoduka (2014) reported that ^{137}Cs activity concentrations in trout (*S. trutta*) ranged from 90 to 190 Bq kg^{-1} in lakes with 0.08–0.12 μSv h^{-1} air dose rate in 2012 in Japan. Mizuno and Kubo (2013) reported that ^{137}Cs activity concentrations in ayu (*P. altivelis*) ranged from 650 to 3300 Bq kg^{-1} in 2011 at Mano and Abukuma river in Japan. Similarly, Iguchi et al. (2013) found that ^{137}Cs activity concentrations of ayu (*P. altivelis*) between 2010–2014 ranged from 1 to 900 Bq kg^{-1} by studying in rivers of Fukushima. ^{137}Cs activity concentrations in white-spotted char (*S. leucomaenis*) ranged from 6.50 to 490 Bq kg^{-1} in rivers of Fukushima prefecture during 2012–2013 (Arai, 2014a). Similarly, Wada et al. (2016) reported that the mean ^{137}Cs activity concentrations in white-spotted char (*S. leucomaenis*), masu salmon (*Oncorhynchus masou*), Japanese dace (*Tribolodon kakonensis*), ayu (*P. altivelis*), common carp (*C. carpio*), crucian carp (*Carassius* sp.) were varied from 345 to 383 Bq kg^{-1}, from 76 to 387 Bq kg^{-1}, from 81 to 396 Bq kg^{-1}, from 57 to 133 Bq kg^{-1}, from 365 to 414 Bq kg^{-1} and from 244 to 782 Bq kg^{-1}, respectively in different rivers with different air dose rates in Fukushima prefecture during 2011-2014. These previous studies confirmed that contamination levels varied in accordance with the fallout amount and/or air dose rate.

However, most of studies after the Chernobyl accident were based on the lake habitats (e.g., Forseth et al., 1991; Elliot et al., 1992, Smith et al., 2003; Sundbom et al., 2003; Saxén et al., 2005; Rask et al., 2012 etc.). Variability of ^{137}Cs activity concentrations in freshwater fish species are tended to be higher in rivers than lakes (Figure 2.1) possibly because of lake morphometry (Särkkä et al., 1995; Matsuda et al., 2015). For instance, ^{137}Cs activity concentrations in white-spotted char (*S. leucomaenis*) of Aga river system varied from 99 to 400 Bq kg^{-1}, while those of lake Hibara varied from 140 to 350 Bq kg^{-1} in Fukushima (Wada et al., 2016). Matsuda et al. (2015)

showed that ^{137}Cs activity concentrations in masu salmon (*O. masou*) had inverse relation with the lake area and water retention time by studying in lake Hayama, Akimoto and Tagokura in Fukushima. In contrast, Särkkä et al. (1995) showed that ^{137}Cs activity concentrations in perch (*P. fluviatilis*), pike (*E. lucius*) and roach (*R. rutilus*) were showed positive relation with water retention time while negative relation with catchment size of the lake. Therefore, habitat specific characteristics need to be examined for understanding the variability of ^{137}Cs contaminations levels in freshwater fish.

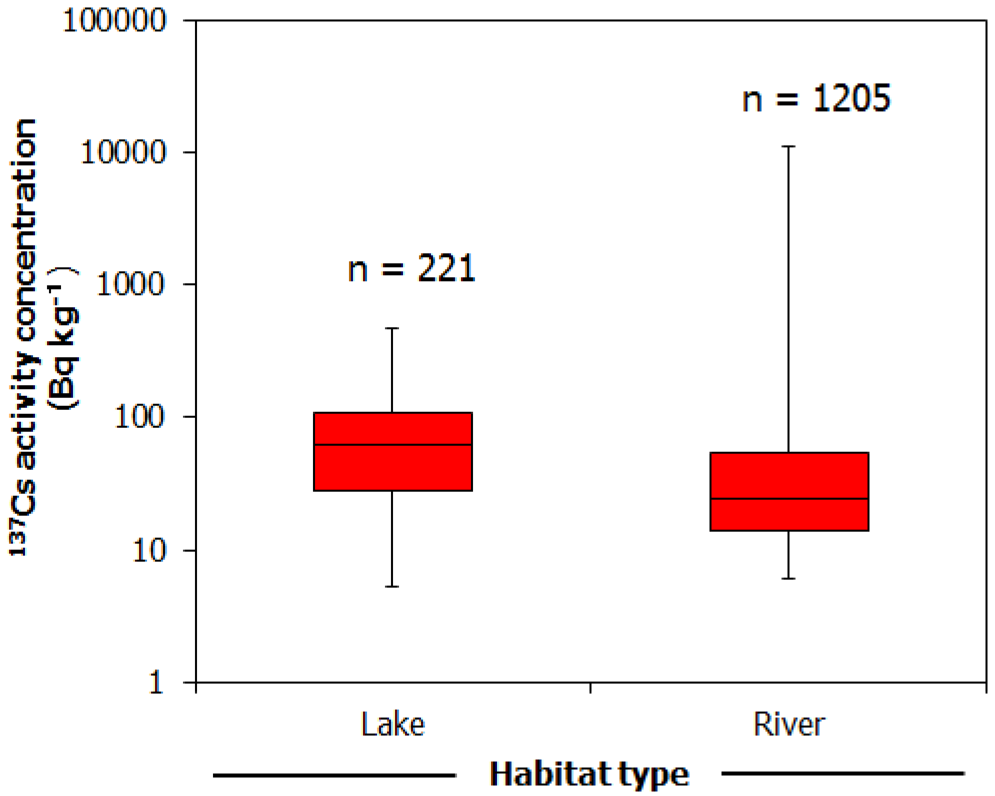


Figure 2.1. Variability of ^{137}Cs activity concentrations of freshwater fish species in lakes and rivers of Fukushima prefecture based on Wada et al. (2016). The lower, middle and upper hinges correspond to the first, second and third quartiles [the 25th, 50th (median), and 75th percentiles]. The whiskers extend from the hinges indicated the highest and lowest values. n indicated the number of samples.

Moreover, ^{137}Cs contamination levels also differed in fish species those were sampled from the same contaminated area. Contamination levels in fish varied from 1 to 2 orders of magnitude within the similar contaminated habitats. For instance, Mizuno and Kubo (2013) showed that ^{137}Cs activity concentrations in Salmonidae fish species in the Aga river systems with air dose rate of 0.30 μSv h^{-1} varied from 17 to 200 Bq kg^{-1} (Table 2.2). Hessen et al. (2000) reported that ^{137}Cs activity concentrations in brown trout (*S. trutta*) in lake of 150 kBq m^{-2} fallout ranged from 443 to 13370 Bq kg^{-1} in Norway (Table 2.2). Rask et al. (2012) showed that ^{137}Cs activity concentrations in perch (*P. fluviatilis*) and pike (*E. lucius*) varied largely from 2200 to 138000 Bq kg^{-1}, and from 8600 to 27000 Bq kg^{-1}, respectively by studying in Finnish lake of 4.07 Bq l^{-1} fallout volumes (Table 2.2). For the assessment of environmental impact of ^{137}Cs on freshwater fish, it is necessary to establish the relationship between exposure (dose rate, accumulated dose), and effects that may be induced in freshwater fish (Real et al., 2004). Therefore, understanding the factors and processes for the variability of contamination levels is crucial for ^{137}Cs dynamics in aquatic ecosystems.

Table 2.2. Summary of ^{137}Cs activity concentrations in freshwater fish.

^{137}Cs Contamination Level		Location of Study		Name of Freswater Fish		^{137}Cs (Bq/kg)	Reference
Fallout volume (kBq/m^2)	Air dose rate (µSv/h)	Habitat	Country	English Name	Latin Name		
10-67	NA	Pond	Ukraine	Channel catfish	*Ictalurus punctatus*	52970	Sugg et al., 1996
0-4	NA	Lake	Finland	Perch	*Perca fluviatilis*	34-59	Saxén et al., 2005
4	NA	Lake	England	Brown Trout	*Salmo trutta*	404-540	Elliot et al., 1992
4	NA	Lake	England	Perch	*Perca fluviatilis*	1192	Elliot et al., 1992
4	NA	Lake	England	Pike	*Esox lucius*	568	Elliot et al., 1992
4-8	NA	Lake	Finland	Perch	*Perca fluviatilis*	21-244	Saxén et al., 2005
8-16	NA	Lake	Finland	Perch	*Perca fluviatilis*	21-324	Saxén et al., 2005
2-20	NA	Lake	England	Brown Trout	*Salmo trutta*	391-462	Elliot et al., 1993
2-20	NA	Lake	England	Arctic charr	*Salvelinus alpinus*	164-204	Elliot et al., 1993
20-30	NA	Lake	Sweden	Perch	*Perca fluviatilis*	2110	Sundbom et al., 2003
20-30	NA	Lake	Sweden	Pike	*Esox lucius*	153	Sundbom et al., 2003
20-30	NA	Lake	Sweden	Roach	*Rutilus rutilus*	118	Sundbom et al., 2003
20-30	NA	Lake	Sweden	Crucian carp	*Carassius carrasius*	789	Sundbom et al., 2003
20-30	NA	Lake	Sweden	Bream	*Abramis brama*	450	Sundbom et al., 2003
16-32	NA	Lake	Finland	Perch	*Perca fluviatilis*	42-1011	Saxén et al., 2005
30-40	NA	Lake	Sweden	Perch	*Perca fluviatilis*	1489	Sundbom et al., 2003
30-40	NA	Lake	Sweden	Pike	*Esox lucius*	1040	Sundbom et al., 2003
30-40	NA	Lake	Sweden	Roach	*Rutilus rutilus*	1292	Sundbom et al., 2003
30-40	NA	Lake	Sweden	Bream	*Abramis brama*	424	Sundbom et al., 2003
32-55	NA	Lake	Finland	Perch	*Perca fluviatilis*	107-7836	Saxén et al., 2005
50	NA	Lake	Norway	Arctic charr	*Salvelinus alpinus*	5460	Forseth et al., 1991
50	NA	Lake	Norway	Broun Trout	*Salmo trutta*	16340	Forseth et al., 1991
60-80	NA	Lake	Sweden	Perch	*Perca fluviatilis*	1626	Sundbom et al., 2003
60-80	NA	Lake	Sweden	Pike	*Esox lucius*	2811	Sundbom et al., 2003
60-80	NA	Lake	Sweden	Roach	*Rutilus rutilus*	1684	Sundbom et al., 2003
60-80	NA	Lake	Sweden	Crucian carp	*Carassius carrasius*	789	Sundbom et al., 2003
60-80	NA	Lake	Sweden	Bream	*Abramis brama*	2127	Sundbom et al., 2003
130	NA	Lake	Norway	Brown trout	*Salmo trutta*	1070-8400	Brittain and Gjerseth, 2010
150	NA	Lake	Norway	Brown Trout	*Salmo trutta*	443-13370	Hessen et al., 2000
482	NA	Lake	Russia	Perch	*Perca fluviatilis*	5006	Smith et al., 2003
1368	NA	Lake	Russia	Perch	*Perca fluviatilis*	60531	Smith et al., 2003
769	NA	Lake	Russia	Perch	*Perca fluviatilis*	4463	Smith et al., 2003
257	NA	Lake	Russia	Perch	*Perca fluviatilis*	527	Smith et al., 2003
515	NA	Lake	Russia	Perch	*Perca fluviatilis*	169	Smith et al., 2003
225	NA	Lake	Russia	Perch	*Perca fluviatilis*	242	Smith et al., 2003
675	NA	Lake	Russia	Perch	*Perca fluviatilis*	1226	Smith et al., 2003
620	NA	Lake	Russia	Perch	*Perca fluviatilis*	24151	Smith et al., 2003
1.43*	NA	Lake	Finalnd	Perch	*Perca fluviatilis*	570-7800	Rask et al., 2012
1.43*	NA	Lake	Finland	Pike	*Esox lucius*	1700-7700	Rask et al., 2012
1.73*	NA	Lake	Finland	Perch	*Perca fluviatilis*	960-20000	Rask et al., 2012
1.73*	NA	Lake	Finland	Pike	*Esox lucius*	3000-12000	Rask et al., 2012
2.12*	NA	Lake	Finalnd	Perch	*Perca fluviatilis*	2800-136000	Rask et al., 2012
2.12*	NA	Lake	Finland	Pike	*Esox lucius*	4700-19000	Rask et al., 2012
4.07*	NA	Lake	Finland	Perch	*Perca fluviatilis*	2200-138000	Rask et al., 2012
4.07*	NA	Lake	Finland	Pike	*Esox lucius*	8600-27000	Rask et al., 2012
NA	0.08-0.12	Lake	Japan	Brown trout	*Salmo trutta*	90-190	Yoshimura and Yokoduka, 2014
NA	0.08-0.12	Lake	Japan	Kokanee	*Oncorhynchus nerka*	50-100	Yoshimura and Yokoduka, 2014
NA	0.08-0.12	Lake	Japan	Rainbow trout	*Oncorhynchus mykiss*	5-80	Yoshimura and Yokoduka, 2014
NA	0.08	Lake	Japan	Crussian carp	*Carassius spp.*	244	Wada et al., 2016
NA	0.08	Lake	Japan	Japanese dace	*Tribolodon hakonensis*	396	Wada et al., 2016
NA	0.18	Lake	Japan	Kokanee	*Oncorhynchus nerka*	400	Wada et al., 2016
NA	0.21	Lake	Japan	White-spotted char	*Salvelinus leucomaenis*	345	Wada et al., 2016
NA	0.21	Lake	Japan	Masu salmon	*Oncorhynchus masou*	217	Wada et al., 2016
NA	0.21	Lake	Japan	Japanese dace	*Tribolodon hakonensis*	377	Wada et al., 2016
NA	0.21	Lake	Jpana	Pond smelt	*Hypomesus nipponensis*	59	Wada et al., 2016
100-300	NA	Stream	Japan	White-spotted char	*Salvelinus leucomaenis*	704-6080	Sakai et al., 2016b
30-60	NA	Stream	Japan	White-spotted char	*Salvelinus leucomaenis*	193-618	Haque et al., 2017a
NA	0.08-0.12	Stream	Jpana	White-spotted char	*Salvelinus leucomaenis*	20-30	Yoshimura and Yokoduka, 2014
NA	0.21-0.26	Stream	Japan	White-spotted char	*Salvelinus leucomaenis*	5-85	Yoshimura and Yokoduka, 2014
6.2	NA	River	Ukraine	Common carp	*Cyprinus carpio*	19.5	Franić and Marović, 2007
NA	0.3	River	Japan	Salmonidae	Salmonidae	17-200	Mizuno and Kubo, 2013
NA	0.26	River	Japan	Masu salmon	*Oncorhynchus masou*	365	Wada et al., 2016
NA	0.26	River	Japan	Japanese dace	*Tribolodon hakonensis*	351	Wada et al., 2016
NA	0.26	River	Japan	Ayu	*Plecoglossidae altivelis*	57	Wada et al., 2016
NA	0.30	River	Japan	White-spotted char	*Salvelinus leucomaenis*	377	Wada et al., 2016
NA	0.30	River	Japan	Masu salmon	*Oncorhynchus masou*	387	Wada et al., 2016
NA	0.30	River	Japan	Japanese dace	*Tribolodon hakonensis*	396	Wada et al., 2016
NA	0.30	River	Japan	Ayu	*Plecoglossidae altivelis*	133	Wada et al., 2016
NA	0.30	River	Japan	Common carp	*Cyprinus carpio*	365	Wada et al., 2016
NA	0.30	River	Japan	Crussian carp	*Carassius* spp.	244	Wada et al., 2016
NA	0.76	River	Japan	White-spotted char	*Salvelinus leucomaenis*	377	Wada et al., 2016
NA	0.76	River	Jpana	Masu salmon	*Oncorhynchus masou*	76	Wada et al., 2016
NA	0.76	River	Japan	Japanese dace	*Tribolodon hakonensis*	77	Wada et al., 2016
NA	1.50	River	Japan	White-spotted char	*Salvelinus leucomaenis*	383	Wada et al., 2016
NA	1.50	River	Japan	Masu salmon	*Oncorhynchus masou*	387	Wada et al., 2016
NA	1.50	River	Japan	Japanese dace	*Tribolodon hakonensis*	81	Wada et al., 2016
NA	1.50	River	Japan	Ayu	*Plecoglossidae altivelis*	105	Wada et al., 2016
NA	1.50	River	Japan	Common carp	*Cyprinus carpio*	414	Wada et al., 2016
NA	1.50	River	Japan	Crussian carp	*Carassius spp.*	782	Wada et al., 2016

* (asterisk mark) values indicated in Bq/L; NA indicated not available

2.4. Factors affecting ^{137}Cs accumulation in freshwater fish

2.4.1. Fallout amount

A number of physical, chemical and biological factors were considered for ^{137}Cs contamination in freshwater fish (Table 2.3). Fallout amount, ecosystem structure, and continual input of radioactive materials from the catchment to the ecosystem have significant roles for contamination of freshwater fish (Poon and Au, 1999). ^{137}Cs enter freshwater systems by both atmospheric fallout, and hydrological processes from the adjacent catchment areas (Renard et al., 1991; Onda et al., 2012). The highest ^{137}Cs activity concentrations in pike (*E. lucius*), perch (*P. fluviatilis*) and vendace (*Coregomus albula*) were found in the lakes, Pielinen and Oulujärvi with the highest ^{137}Cs deposition in Finland after the Chernobyl accident in 1986 (Saxén and Rantavaara, 1987). Särkkä et al. (1995) reported that ^{137}Cs activity concentrations in perch (*P. fluviatilis*), pike (*E. lucius*) and roach (*R. rutilus*) showed the positive correlation with the fallout amount by studying in lakes of different sizes and water qualities with deposition amount of 10-67 kBq m^{-2} in Finland after the Chernobyl accident. Similarly, Hessen et al. (2000) found that ^{137}Cs activity concentrations in brown trout (*S. trutta*) was related to the fallout amount based on the studies on about 1800 samples of trout collected from about 100 localities in Oppland country in south central Norway during 1986-1995. Tsuboi et al. (2015) found increased amount of ^{137}Cs activity concentrations of ayu (*P. altivelis*) in three rivers (Niida, Kido, and Abukuma) with higher fallout volume (> 100kBq m^{-2}) among the five rivers in Fukushima. Similarly, Wada et al. (2016) reported that higher ^{137}Cs activity concentrations within freshwater fish like rainbow trout (*O. mykiss*), ayu (*P. altivelis*), white-spotted char (*S. leucomaenis*), Japanese dace (*T. kokonensis*), common carp (*C. carpio*) were found in areas with higher deposition based on the studies on freshwater fish collected from rivers, lakes and cultural ponds in Fukushima prefecture during 2011-2014. Recently, Sakai et al. (2016b) showed that ^{137}Cs activity concentrations in primary producer (i.e., litter) in riparian zones was associated with ^{137}Cs fallout volume in Fukushima which is an important basal food resource for freshwater fish of streams, and fallout volume possibly reflects ^{137}Cs accumulation in fish via trophic transfer.

In forest ecosystems, a large amount of atmospherically supplied ^{137}Cs was attached to the canopy in coniferous forest, while ^{137}Cs fallout was deposited mostly on the forest floor in deciduous forest because of the absence of leaves (Kato et al., 2012). ^{137}Cs contamination deposited on forest ecosystems has further circulated within the ecosystems (Steiner et al., 2002) and transferred to aquatic ecosystems via litterfall and throughfall (Teramage et al., 2014). Once litter enter into the aquatic ecosystems, ^{137}Cs leaching from the litter (Sakai et al., 2015) cause the ^{137}Cs availability for the freshwater fish (Murakami et al., 2014; Haque et al., 2017a). Hence, ^{137}Cs activity concentrations in litter differ depending on the fallout volume (Sakai et al., 2016a) which possibly responsible for the ^{137}Cs contamination levels in freshwater fish as litter is considered as basal sources of ^{137}Cs in both terrestrial and aquatic ecosystems (Sakai et al., 2016b).

2.4.2. Habitat

Habitat exposure pathways depend on the ecological characteristics and habitat conditions of the organisms. For example, a benthic feeder mostly contaminated from ^{137}Cs present in water column, benthos, organic matter, and deposited sediments whereas bottom feeder may only be exposed to ^{137}Cs in water column (Ashraf et al., 2014). The positive relation between ^{137}Cs activity concentrations in sediments, and fish in several lakes had also been reported from several studies conducted after the Chernobyl and Fukushima accidents (Jagoe et al., 1998; Håkanson, 1999; Fukushima and Arai, 2014; Matsuda et al., 2015). Fukushima and Arai (2014) showed significant positive correlations between ^{137}Cs activity concentrations in sediments and fish by studying on several lakes in northern Japan after the fallout. Matsuda et al. (2015) reported that

^{137}Cs activity concentrations in freshwater fish had significant correlation with their habitat components. The higher ^{137}Cs activity concentrations in sediments is expected to be related to higher ^{137}Cs contamination in food items, resulting in the higher ^{137}Cs activity concentrations in fish of areas with dense deposits (Wada et al., 2016). Moreover, ^{137}Cs activity concentrations in fish habitats decreased with distance from the FDNPP (Mizuno and Kubo, 2013; Matsuda et al., 2015) though contamination levels in freshwater fish varied in accordance to the retention time (Fukushima and Arai, 2014), depth (Broberg et al., 1995), water hardness and conductivity (Hakånson et al., 1992; Särkkä et al., 1995), suspended sediment concentration and temperature (Rowan and Rasmussen, 1994). Arai (2014a) suggested that ^{137}Cs accumulation in freshwater fish depends on ^{137}Cs deposition in the habitat based on the studies of freshwater fish in different regions after the FDNPP accident.

2.4.3. Food bioaccumulation

Freshwater fishes are generally less active in terms of accumulation of ^{137}Cs from ambient water (Man and Kwok, 2000); food intake is the most important pathway of ^{137}Cs accumulation (Elliot et al., 1992; Smith et al., 2000a). For example, ayu (*P. altivelis*) in the fluvial life stage mainly live on algae attached to stone surfaces, and ^{137}Cs accumulated by algae are the main source of ^{137}Cs for ayu through their foraging (Iguchi et al., 2013; Tsuboi et al., 2015). Similarly, white-spotted char (*S. leucomaenis*) in headwater stream are mainly insectivorous depends on both terrestrial and aquatic insects in stream-riparian zones, and through which ^{137}Cs accumulated in char body (Sakai et al., 2016b). Primary producers such as litter, organic matter and detritus in freshwater systems are decomposed by shredders (Merritt and Cummins, 1996; Allan and Castillo, 2007) which are further consumed by filter feeders and gatherers (Vannote et al., 1980; Graca, 2001), and subsequently ^{137}Cs transfer via food web to primary consumers. These primary consumers become one of the important food sources for many freshwater fish, and by which ^{137}Cs transferred to higher trophic levels (Sakai et al., 2016b; Wada et al., 2016). Moreover, Rowan and Rasmussen (1994) reported that trophic level and length of food chain influenced ^{137}Cs accumulation in fish. For example, piscivores fish (e.g., perch, pike) bioaccumulate more than planktivores and benthivores (e.g., brown trout, bream). A higher annual mean ^{137}Cs activity concentrations had been detected in fish of higher trophic levels in some lakes of Finland after the Chernobyl accident (Rask et al., 2012) although species-specific food intake and food availability can cause differences in ^{137}Cs activity concentrations among fish species (Rowan and Rasmussen, 1994). Similarly, Wada et al. (2016) found higher ^{137}Cs activity concentrations in carnivorous and omnivorous fish (e.g., masu salmon, white-spotted char) than herbivorous and planktivorous fish (e.g., ayu, pond smelt) which revealed that fish bioaccumulate ^{137}Cs through the food web. ^{137}Cs bioaccumulation in predatory mangrove snapper (*Lutjanus argentimaculatus*) is resulted via food web transfer from the assimilation of extremely high ^{137}Cs contaminated ingested prey items (Zhoa et al., 2001).

Table 2.3. Summary of factors affecting ^{137}Cs accumulation in freshwater fish.

Factors		
Physical factor	Chemical factor	Biological factor
Fallout amount	Amount of precipitation	Organic matter and detritus
Habitat	Water retension time	Prey items
Landscape structure	Potassium content of water	Dietary intake
Size of catchment area	Phosphorus content of water	Trophic level
Suspended sediment	pH of water	Food web
Size, weight and age of fish	Water hardness	Storage and absorbtion
Physical half-life	Electrical conductivity	Excretion and metabolism
Data collection date	Water temperature	Biological half-life

2.4.4. Metabolic rate

Metabolic rate, which is dependent on water temperature, could affect ^{137}Cs intake, retention and excretion rates (Elliott et al., 1992; Ugedal et al., 1992). Differences in metabolic rates among seasons could also partially explain the differences of ^{137}Cs accumulation between fish species. For instance, Peles et al. (2000a) reported that the ^{137}Cs activity concentration in freshwater largemouth bass (*M. salmoides*) in winter was about two times greater than that in summer because of lower uptake and excretion rate in winter. Furthermore, the metabolic rate of rainbow trout (*O. mykiss*) was three-fold greater in summer than that in winter (Facey and Grossman, 1990). Hammar (1998) reported that arctic char (*S. alpinus*) accumulate more ^{137}Cs in winter than summer which indicating their lower metabolic rate in winter. ^{137}Cs activity concentrations in white-spotted char (*S. leucomaenis*) of headwater stream in Gunma are about two times greater in winter than summer because of lower metabolic rate in winter (Haque et al., 2017b).

2.4.5. Potassium

The potassium (K^+) concentration of water influences ^{137}Cs accumulation and excretion in fish (Elliot et al., 1992; Rowan and Rasmussen, 1994; Furukawa et al., 2012; Rask et al., 2012). By using log-liner regression analysis, it was found that ^{137}Cs bioaccumulation in fish had negative relation with dissolved K^+ (Rowan and Rasmussen, 1994). Due to its chemical similarity of K^+ with ^{137}Cs, K^+ in water had known to influence ^{137}Cs accumulation in fish (Blaylock, 1982). Strong inverse relationships were observed between K^+ in lake water and ^{137}Cs activity concentrations in fish following the nuclear weapons testing (Blaylock, 1982), and the Chernobyl accident (Smith et al., 2000a). Dietary intake of ^{137}Cs was inversely related to K^+ concentration in ambient water (Blaylock, 1982; Rowan and Rasmussen, 1994). In addition, Smith et al. (2000a) reported that ^{137}Cs

concentration ratio in fish were inversely proportional to K^+ concentration of lake water by studying on the lakes of the Chernobyl-contaminated regions. Ugedal et al. (1992) reported longer biological half-lives of ^{137}Cs in fish as fish actively excrete Cs^+ during osmoregulation (Furukawa et al., 2012). These physiological phenomenon along with ^{137}Cs recycling or remobilization within a freshwater system (Comans et al., 1989) triggered long-term contamination in freshwater fish (Bulgakov et al., 2002).

2.4.6. Size, weight, age of fish, and other factors

Size, weight and age of fish are another factor for ^{137}Cs accumulation in fish. For example, ^{137}Cs accumulation in fish increased with accordance to their sizes (Elliott et al., 1992). A 'positive size effect' revealed that older fish were more ^{137}Cs contaminated than juveniles of same species because ^{137}Cs excretion rate were higher in juvenile than in older age classes, and the decrease in ^{137}Cs activity concentrations in water over time also resulted in lower ^{137}Cs levels in fish (Kryshev and Ryabov, 2000). Such type of phenomenon was reported for silver carp (Koulikov and Ryabov, 1992), brown trout (Elliott et al., 1992), bream, roach, pike-perch (Hadderingh et al., 1996) etc. after the Chernobyl accident. Moreover, ^{137}Cs fallout volume in habitat and water retention time showed positive correlation, while the size of catchment area, colour, phosphorus content, pH, and electrical conductivity of water showed inverse correlation with ^{137}Cs activity concentrations in fish (Särkkä et al., 1995).

2.4.7. Multiple interactions

The overall variability of ^{137}Cs contamination in freshwater fish over time was almost related to fallout amount (Forseth et al., 1991; Hessen et al., 2000). Differences in prey items contamination level would reflect the ^{137}Cs activity concentration in predator (Hessen et al., 2000) and shifting food consumption habit of freshwater fish from terrestrial to aquatic sources between summer and winter (Nakano et al., 1999; Sato et al., 2011) might also result in differences in their total ^{137}Cs activity concentrations between seasons. Sakai et al. (2016b) showed that the ^{137}Cs activity concentrations of terrestrial sources were about four times greater than those of aquatic sources. Therefore, multiple interactions by considering various processes within the ecosystem might have potential contributions for controlling the variability in ^{137}Cs contamination levels of freswater fish in aquatic ecosystems.

2.5. Tempo-spatial variations of ^{137}Cs contamination in freshwater fish

2.5.1. Temporal variability of ^{137}Cs contamination in freshwater fish

Both the temporal and spatial fluctuations of ^{137}Cs activity concentrations in freshwater fish are important to understand more insights about variability of ^{137}Cs contamination levels in fish to predict the future. Time is the most important factor for ^{137}Cs variability in freshwater fish as ^{137}Cs activity concentrations in fish of European lakes was decreased with time (Håkanson, 1999). Holloman et al. (1997) reported that temporal patterns of ^{137}Cs in mosquitofish (*Gambusia holbrooki*) indicated seasonal fluctuations in ^{137}Cs contamination by studying in the Savannah River Site (SRS), South Carolina after the Chernobyl accident. By comparing with the results of Newman and Brisbin (1990), it was predicted that these decreasing trends possibly be outflow of ^{137}Cs from the reservoir and/or long-term sequestration deeper in the sediments (Holloman et al., 1997). Peles et al. (2000a) showed that ^{137}Cs activity concentrations in largemouth bass (*M. salmoides*) collected monthly over a one-year period from abandoned reactor cooling reservoir located on the SRS, South Carolina, USA were also decreased with time. Suspended sediment is one of source of ^{137}Cs in freshwater fish, ^{137}Cs activity concentrations decreased exponentially in the suspended sediment in irrigated paddy fields (Yang et al., 2016), and in river (Tanaka et al., 2014) of Fukushima prefecture with ultimately altering ^{137}Cs activity concentrations in fish.

The gradual decreasing trends of temporal variations of ^{137}Cs in freshwater fish were also found in Fukushima prefecture after the FDNPP accident during May 2011 to March 2014 (Arai, 2014a). These results suggested that ^{137}Cs accumulation in terrestrial habitats in Fukushima decreased gradually after the accident, and ^{137}Cs accumulation in freshwater fish were also decreased accordingly because terrestrial contaminations are the main source of contamination for freshwater fisheries resources (Sakai et al., 2016b). However, ^{137}Cs contamination in brown trout (*S. trutta)* in the Norwegian lake still remains after 20 years of the Chernobyl accident because concentration levels were no longer decreased (Brittain and Gjerseth, 2010). It was possibly being continual input of ^{137}Cs from the adjacent catchments, and remobilization in local terrestrial environment (Arai, 2014a; Sakai et al., 2016b). In another study, Arai (2014b) showed that ^{137}Cs activity concentrations in salmonids had lower temporal variations after the FDNPP accident during May 2011 to February 2014 which suggested that ^{137}Cs were widely distributed and remained in natural environment, and the accumulation patterns significantly varied among the salmonids species; freshwater salmonids were highly contaminated than marine salmonids. These results also revealed the widespread contamination in terrestrial environment which in turns become the major source of contamination for freshwater environment.

2.5.2. Spatial variability of ^{137}Cs contamination in freshwater fish

Arai (2014a) reported gradual spatial decreasing trends of ^{137}Cs activity concentrations in freshwater fish based on the studies in five different regions (i.e., Fukushima, Aizu, Sousou, Koriyama and Iwaki) in Fukushima prefecture during 2011 to 2014. The highest ^{137}Cs accumulation was found in the nearest region (i.e., Sousou region) from the FDNPP, and the accumulation trends among the regions were tended to be decreased as a function of distance from the point of accident. This study revealed that the spatial accumulation patterns of ^{137}Cs were related to the natural and/or fallout amounts. ^{137}Cs deposition patterns correspond to the atmospheric circulation; therefore ^{137}Cs levels in Fukushima region were higher which caused higher accumulation in freshwater fish in Fukushima compare to other regions. In freshwater ecosystems, the initial dynamic phase of ^{137}Cs contamination and equilibration lasts up to five years after fallout and those can be determined by various biological processes (Bergan, 2000). After these five years, ^{137}Cs in fish become in steady state condition with slow decreasing rate indeed that is controlled by continuous secondary inputs of ^{137}Cs to the freshwater habitats, and transfer through their food web (Bergan, 2000). The bioavailability of ^{137}Cs for freshwater ecosystems continuously decreased in accordance to the decreased of contamination levels in abiotic components within the systems (Bergan, 2000).

The ^{137}Cs activity concentrations in freshwater fish, ayu (*P. altivelis*) exhibited spatial variations in their accumulation patterns based on the studies of five different rivers in Fukushima during 2011–2013 (Tsuboi et al., 2015). The decrease of ^{137}Cs in riverbed caused by flushing out of contaminated soil from the mountains would be the possible factor of decreasing trends of contamination level in ayu (Tsuboi et al., 2015). ^{137}Cs activity concentrations of fish muscle in European lakes peaked a few years after the Chernobyl disaster (Jonsson et al., 1999; Smith et al., 2000a); conversely those in ayu peaked immediately after the FDNPPs accident (Tsuboi et al., 2015). Therefore, tempo-spatial monitoring of ^{137}Cs contamination levels in freshwater ecosystems are essential for understanding long-term dynamics of ^{137}Cs, and to reveal the radiological effects on each ecosystem.

2.6. Management applications

Freshwater ecosystems such as rivers, streams, lakes, ponds, wetlands, and groundwater have specific characteristics, and site-specific behaviours governed by their own morphological, hydrological, biological and ecological parameters, and their drainage areas. ^{137}Cs remediation of any reservoir depends on its site-specific characteristics (Ashraf et al., 2014). The remediation plans and practices must be based on a cost-benefit basis (Brenner et al., 2003). Though the radiological risks associated with ^{137}Cs in soils, water and sediments, those can't be eliminated completely by using on-going remediation technique, but the contamination levels can be reduced by using an appropriate technique. Scientists believed that majority of ^{137}Cs contaminated areas may not take proper remedial actions by considering the ecological and public health risks associated with the contaminations. However, it becomes a significant challenge to convince the public, regulators, and decision makers that action is not warranted (Ashraf et al., 2014). Remediation practices can be categorized into biological, chemical, and physical remediation. Many techniques are carried out for remediation of chemical, organic compounds and heavy metals, but all are not effective for ^{137}Cs.

2.6.1. Biological remediation

The ^{137}Cs activity concentrations in fish can be decreased by biological dilution based on the theories of buffering (Ashraf et al., 2014) through using various fertilizers (Butler, 2011). Trophic transfer of ^{137}Cs in freshwater system can be reduced by lowering the fish and/or predator stocks, and also by influencing the predation pressure (Morino et al., 2011). By increasing water biomass, ^{137}Cs could be dispersed ultimately causing lower contamination in fish (Lee et al., 2008). Controlling microbial primary producers had potential role in lowering ^{137}Cs accumulation in fish as ^{137}Cs mobilization and cycling in aquatic ecosystems largely depends on these producers (Avery, 1995). Salinity is another factor that influenced freshwater fish to accumulate ^{137}Cs potentially than marine fish (Håkanson, 1991).

2.6.2. Chemical remediation

Chemical remediation techniques can't decontaminate freshwater systems entirely, but aid ^{137}Cs extraction from streambed and sediments. Fertilization and liming were useful, and ecologically relevant methods to remediate ^{137}Cs contaminated freshwater systems (Kinoshita et al., 2011). Because of chemical similarity of K^+ with ^{137}Cs (Blaylock, 1982), use of potassium fertilizer caused substantial reduction in ^{137}Cs uptake (Hirose, 2012) by blocking and/or substituting ^{137}Cs taken up by fish (Moller et al., 2013; Ashraf et al., 2014). Addition of ammonium fertilizer has a potential capacity to displace the sediment bound ^{137}Cs (Evans et al., 1983). Liming is used to increase the proportion of ^{137}Cs in sediment of freshwater bodies that prevents or delays uptake of ^{137}Cs. It was performed based on the destabilized tendency of ^{137}Cs carrying particles is increased by increasing pH, alkalinity and hardness of water (Håkanson et al., 1996; Tsukada et al., 2003). Liming model is considered as an important model for stimulating ^{137}Cs in fish, and would be used to remediate ^{137}Cs from the freshwater systems after the fallout like the Chernobyl and FDNPP (Ottosson and Håkanson, 1997).

2.6.3. Physical remediation

Physical remediation for ^{137}Cs can't refine the contamination level entirely without removal of contaminated sources (soil, sediments and/or water), but it can be able to reduce the potentiality of exposure through the sources (Ashraf et al., 2014). Capping and in situ grouting are generally less cost effective remediation methods (Davoine and Bocquet, 2007). Electrokinetic techniques require dissolution of the contaminant in soil water to facilitate its movement for collecting electrode (Devell et al., 1995). The addition of illite to ^{137}Cs contaminated freshwater system to reduce contamination levels in biota (Galmarini et al., 2011) will be effective to reduce the biological availability of ^{137}Cs. The relative distribution of ^{137}Cs between ecosystem components is highly variable and dynamic, while the behaviour of ^{137}Cs differs considerably in terrestrial, freshwater and marine ecosystems. It is clear that ^{137}Cs undergoes intensive recycling in the environment which owing to the relatively long physical half-life indicates that ^{137}Cs can remain a serious problem for many years following a pulse of contamination (Avery, 1996).

2.7. Knowledge gaps

It was evident from of the previous studies in this chapter is that ^{137}Cs activity concentrations in freshwater fish differed based on the food and feeding habits, habitat characteristics, physiological conditions of the given fish species. Among the factors, contaminations via consumptions of food sources were the dominant for controlling the contamination levels. Contamination of fishes within the same habitat also varied. Fish of rivers are tended to have higher contamination than lakes because of higher catchment size, volume, water retention time, and organic matter. Therefore, it is essential to examine all of the possible factors for understanding the variability of contamination levels of a given fish species in a specific ecosystem.

However, yet there is little information available on processes and factors towards variability in ^{137}Cs contamination levels in freshwater fish in aquatic ecosystems. Most of the previous studies related with simple measurement of ^{137}Cs contamination levels, but studies regarding processes and factors for the variability in contamination levels in fish have not received much more attention still now. ^{137}Cs in aquatic ecosystem bioaccumulate in higher trophic animals including fish via trophic transfer; therefore become threaten for human beings. Major questions that need to be answered concern the relative contributions of ^{137}Cs from natural, terrestrial, aquatic and other sources; identify transfer processes, accumulation patterns, factors affecting contamination, and the biological implications of those different sources.

Moreover, temporal trend information is very limited for ^{137}Cs in freshwater fish because they are based on only two or three sampling intervals. A limited long-term temporal trend data are available for ^{137}Cs activity concentrations in fish after the Chernobyl accident, and comparisons with FDNPP accident are limited because it about six years have passed. This information of freshwater fish is particularly important because contamination levels are decreasing in aquatic ecosystems. Whether this decrease is entirely due to limited anthropogenic inputs and/or transfer to the biota which have been shown to be decreased slowly or whether it is due to some environmental changes which is mobilizing ^{137}Cs is not clearly known yet. Furthermore, the geographical coverage of ^{137}Cs measurements in fish is less detailed; most measurements have been carried out in fish muscles from the adjacent areas of accidents. The freshwater systems – freshwater systems variability within the same or different fish species has not yet been adequately explained. It is difficult to assess whether these systems differences in ^{137}Cs activity concentrations reflect true differences between the systems or are more a reflection of the inherent variability in such measurements. This resolution is important because ^{137}Cs activity concentrations in fish appear to be a function of trophic level, and other aspects of freshwater ecosystem. Very limited data is available based on the geographic coverage of major freshwater fish species. Several studies also shown that localized ^{137}Cs contamination in fish, when considered on a broad regional scale, there is a need to determine whether freshwater fish frequenting the waters within the general area of their living sites. No specific studies carried out based on this idea to examine this question.

2.8. Future research

Our knowledge of ^{137}Cs contamination is mostly based on single species experiments, while in natural condition species exits as a part of multispecies assemblage. In the natural environment, species are exposed differently to ^{137}Cs contamination because of their specific habitats, behaviours, food and feeding habits, ecotones, niches etc. The interactions among fish and the other biota alter the ^{137}Cs contamination levels in fish because fish mainly contaminated by consumption of contaminated flora and fauna. The various biological and ecological responses due to interactions among the ecosystem components alter the ^{137}Cs contamination levels in fish. A conceptual flow chart of possible research by considering interactions among the ecosystem components to elucidate ^{137}Cs contamination levels in freshwater fish of aquatic ecosystems is illustrated in Figure 2.2. In general, fish consume a variety of food items with different biomasses (Bendell and McNicol, 1987), and contamination of such food stuffs may differ due to contamination levels of habitats (Murakami et al., 2014). This may lead to secondary effects which alter the contamination levels. Moreover, fish of higher trophic levels consume multiple food sources with different biomasses (Rowan and Rasmussen, 1994) which possibly determine the ^{137}Cs contamination levels in fish in natural environmental conditions by a complex way due to the differences of dietary contributions and their respective contamination levels. The fate and exposure pathways of ^{137}Cs in freshwater ecosystems have not yet been studied in details, and this information is critical for the study of ^{137}Cs dynamics by considering the complex process-based exposure pathways. Indeed, this review chapter provides the basic information about ^{137}Cs contamination levels and their associated factors and processes in freshwater fish of aquatic ecosystems.

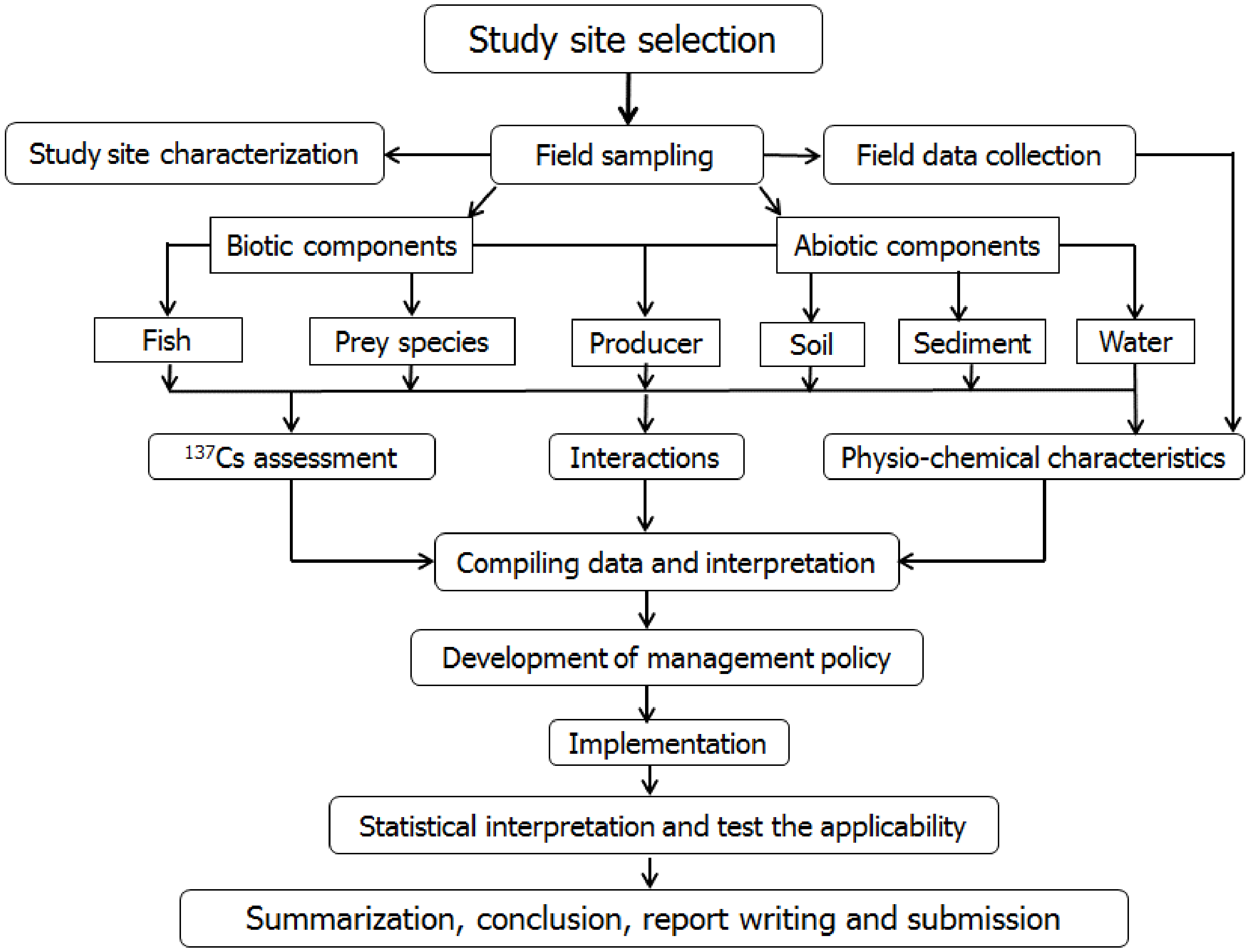


Figure 2.2. Conceptual flow chart of possible research in freshwater systems.

2.9. Summary and conclusions

The FDNPP accident in Japan was the worst nuclear disaster after the Chernobyl accident, and the initial dynamic phase of contamination and equilibration may be still continuing and unstable. ^{137}Cs dynamics in freshwater ecosystems have not yet been revealed, and this information is critical for the removal of ^{137}Cs from the freshwater ecosystems. Understanding the mechanisms and processes affecting variability and dynamics allows the prediction of the fate of ^{137}Cs in freshwater environments. The present study has suggested that ^{137}Cs accumulation in freshwater fish has gradually decreased in different regions followed by the studies on freshwater fish after the Chernobyl and FDNPP accidents. Monitoring the long-term behaviour of ^{137}Cs in the environment by considering different processes and affecting factors is an important issue for estimating possible radiological consequences and associated risks. This monitoring is also important for evaluating the standard limit of ^{137}Cs activity concentration in contaminated food stuffs for human consumption (i.e., 100 Bq kg^{-1}-wet in Japanese health standard), potential use of contaminated areas, and possible effectiveness of remediation activities. Further continuous monitoring is indispensable for understanding ^{137}Cs behaviour in freshwater ecosystems and human health after the accident. This researches and findings of this study can also apply various types of contaminants including heavy metals which are transfer and/or accumulated in fish of freshwater ecosystems via their ecological, hydrological and biological processes. Therefore, the present study might be a meaningful and worthwhile study in radioecological arena in the context of ^{137}Cs released from the nuclear power plant accidents, and its impact on freshwater environments.

CHAPTER 3

DEVELOPING FOOD WEB-BASED TRANSFER FACTOR OF ^{137}Cs

3.1. Introduction

Contaminants from radionuclide fallout can be mobilized in ecosystems through physical and biological processes (Monte et al., 2009; Murakami et al., 2014; Sakai et al., 2016b). Because of tight linkages between riparian areas and stream channels in headwater systems (Vannote et al., 1980; Gomi et al., 2002; Baxter et al., 2005), radionuclides would move across terrestrial and aquatic environments in forested streams. ^{137}Cs emitted due to the FDNPP accident has led to serious contamination of forested areas in Fukushima and the surrounding regions (Hashimoto et al., 2012). ^{137}Cs captured by tree canopies in coniferous forests (Kato et al., 2012) is transferred to the forest floor via litterfall and throughfall (Teramage et al., 2014). Moreover, ^{137}Cs attached to litter can also be mobilized from forest floor to streams (Takahashi et al., 2015). Part of the litter from forest canopy can directly fall into stream channels and ^{137}Cs can be leached into the water column and/or transferred to other substances such as clay minerals (Sakai et al., 2015).

The ^{137}Cs contamination can also enter food webs and transfer across trophic levels (Ashraf et al., 2014). In stream ecosystems, leaf litter in the stream channel is decomposed by bacterial colonization and leaching (Allan and Castillo, 2007). Macroinvertebrates classified as shredders are detritivores that feed on coarse particulate organic matter (CPOM > 1 mm) and break it down into fine particulate organic matter (FPOM < 1 mm) (Merritt and Cummins, 1996). Drifting FPOMs are consumed by filterers whereas gatherers consume depositional FPOMs (Vannote et al., 1980). Grazers and predatory macroinvertebrates consume algae on stream substrate and other macroinvertebtrates. Such macroinvertebrates are important food sources for many fish species (Wallace and Webster, 1996). In addition to aquatic food sources, terrestrial food sources are also important for salmonid fishes in forested headwater streams (Nakano et al., 1999; Allan and Castillo, 2007) because of high availability of terrestrial food sources in summer season (Kawaguchi and Nakano, 2001; Nakano and Murakami, 2001). In such stream-riparian ecosystems in Fukushima, Sakai et al. (2015, 2016b) showed that the contamination levels of biota in terrestrial environments were about 4-16 times greater than those in streams because terrestrial litter had higher contamination level than did aquatic litter. These findings suggested that ^{137}Cs transfer to fishes in forested stream are required to be examined through food webs across forest and stream ecosystems (Bréchignac et al., 2016).

A number of approaches have been developed for estimating radionuclide transfer to biota with various environmental conditions (ICRP, 2009). Transfer factors (*TF*s) are one method for evaluating contamination levels of biota in a given environment (IAEA, 2009). *TF*s are also known as concentration ratios (Tuovinen et al., 2013), concentration factors (Smith et al., 2000a), bioconcentration factors (Marzano et al., 2000), aggregated concentration factors (Garnier-Laplace et al., 2000), and bioaccumulation factors (Vanderploeg et al., 1975). All of these *TF*s were estimated with simple calculations using radionuclide contamination values in biota divided by those in media (i.e., water, sediment and/or litter). I referred these *TF*s as the contemporary transfer factor indicated as TF_c. Various previous researches showed the ranges of TF_c. For instance, Čepanko et al. (2007) showed that TF_c from water to brown trout (*Salmo trutta trutta*) ranged from 2.7 to 3.4 in Lithuanian rivers 18–19 years after the Chernobyl accident. Fukushima and Arai (2014) showed that TF_c based on ^{137}Cs activity concentrations in lake sediment and different fish species including char (*Salvelinus leucomaenis*) and cherry salmon (*Oncorhynchus masou*) ranged from 0.10 to 6.12 in northeastern Japan after the FDNPPs accident in 2011.

Another method for estimating *TF*s is based on the concentration of available food items for a particular species (Brown et al., 2004). Such *TF*s between prey and predator are also called as biotransference factor (Dallinger et al., 1987) or trophic transfer factor (*TTF*) (Zhao et al., 2001), which is estimated by ^{137}Cs activity concentration of a predator divided by ^{137}Cs activity concentration of a single prey item. Prey-predator *TTF* considered that trophic uptake of ^{137}Cs by ingestion of contaminated food sources is much more important than direct uptake from the water column (King, 1964; Dallinger et al., 1987; Sundbom et al., 2003; Johansen et al., 2014). Doi et al. (2012) also emphasized on ^{137}Cs accumulation in fish associated with their diet is much more important in freshwater systems. Although *TTF* is a useful indicator for understanding contamination levels in biota based on their food sources, estimated *TTF*s can vary depending on the selected prey items because a given predator may consume a variety of prey with different biomasses (Bendell and McNicol, 1987). Furthermore, the contamination level of such food items may also differ depending on the contamination level of habitat (Murakami et al., 2014; Sakai et al., 2016b).

To overcome such problems and for understanding ^{137}Cs transfer in food webs, the ^{137}Cs activity concentrations of prey items to a given predator need to be studied in conjunction with their relative dietary contributions to predators, especially for species at higher trophic levels with multiple food sources (Rowan and Rasmussen, 1994; Clements and Newman, 2002). Consumption of multiple prey items may be more pronounced for the food webs between forests and headwater streams because of the reciprocally subsidized food sources (Nakano and Murakami, 2001; Miyasaka et al., 2003). However, there have been no attempts to develop a metric to assess concentration magnitudes by considering the dietary ^{137}Cs contributions of multiple prey items to a given predator. This is also critical for understanding the variability of contamination level within a particular organism (Rowan et al., 1998). Therefore, the primary aim of this study was to develop a food web-based transfer factor (symbolized and hereafter noted as 'TF_{web}') that integrate dietary contributions of multiple prey items with their respective ^{137}Cs activity concentrations to a predator.

I chose white-spotted char (*Salvelinus leucomaenis*; Japanese name: Iwana) inhabiting headwater channels draining forested areas as a target predator because they consume multiple food sources from both terrestrial and aquatic ecosystems. White-spotted char is common in Honshu

Island, Japan (Dunham et al., 2008) and is also commercially valuable in freshwater fisheries. Kawaguchi and Nakano (2001) and Nakano and Murakami (2001) showed that white-spotted char consume more terrestrial than aquatic prey items during the summer due to their higher availability. The dominant prey items of white-spotted char are terrestrial arthropods such as terrestrial insects, dipteran larvae, tetragnathids and raphidosphorids, and stream macroinvertebrates such as mayflies, stoneflies, midges and amphipods (Nakano and Furukawa-Tanaka, 1994; Miyasaka et al., 2003; Sakai et al., 2016b). Therefore, a wide range of prey items with different habitats and contamination levels can be included for examining the applicability of this method.

3.2. Material and methods

3.2.1. Study sites and field samplings

This study was conducted in two locations with similar landscapes but different levels of ^{137}Cs fallout inventory. Osawa-gawa stream is 45 km away from the FDNPP in Nihonmatsu city, Fukushima prefecture (37º36´N, 140º37´E) (Figure 3.1a and b). Oya-san stream is 190 km away from the FDNPP in Midori city, Gunma prefecture (36º33´N, 139º21´E) (Figure 3.1a and c). Based on airborne observations from June 28, 2012, the fallout inventory of ^{137}Cs was 100–300 kBq m^{-2} in the Fukushima site, whereas it was 30–60 kBq m^{-2} in the Gunma site (MEXT, 2012). The mean annual precipitation and air temperature measured at nearby Funehiki AmeDAS automated weather stations in Fukushima were 1248 mm and 11°C. The underlying geology of Fukushima is Lower Cretaceous granites. In the Gunma site, the mean annual precipitation was 1323 mm and mean annual air temperature was 15°C based on the data measured at nearby Kiryu AmeDAS automated weather stations in Gunma. The area is covered by a mélange matrix of middle to late Jurassic accretionary complex. The dominant overstory vegetation in both study sites was 30- to 40-year-old

Japanese cedar (*Cryptomeria japonica*). The dominant riparian shrubs and understory vegetation were Japanese snowball (*Viburnum plicatum*) and silver vine (*Actinidia polygama*) in the Fukushima site and hornbeam maple (*Acer carpinifolium*) and wild beans (*Dumasia truncate*) in the Gunma site.

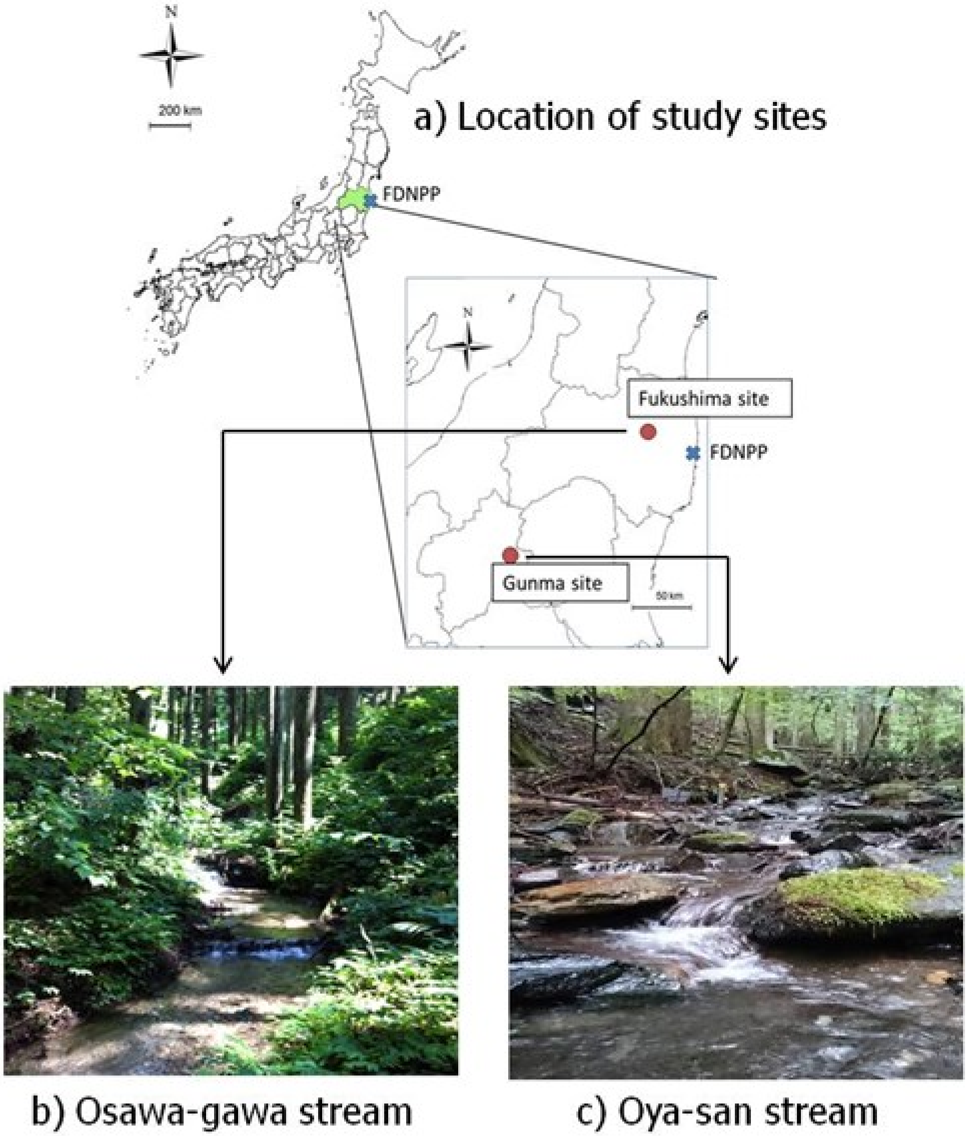


Figure 3.1. Map of the locations of study sites, a) Fukushima and Gunma and overview of b) Osawa-gawa stream in Fukushima; and c) Oya-san stream in Gunma. FDNPP indicates Fukushima Dai-ichi Nuclear Power Plant.

I selected 50-m channel segments for collecting samples in each site. The channel gradient ranged from 5 to 7° with sequences of steps formed by large boulders and pools (Montgomery and Buffington, 1997). The channel width ranged from 1.0 to 5.0 m in both sites, while the channel depth ranged from 5 to 50 cm in Fukushima and from 10 to 100 cm in Gunma. In each site, samples of white-spotted char, prey items and stream litters were collected at four times from August 2012 to May 2013. Samples of stream sediments were collected in August 2012.

White-spotted char were captured using an LR-24 backpack electrofisher (Smith-Root Inc., Vancouver, WA, USA) (Figure 3.2). I examined every pool and riffle in the 50-m channel segments. Twenty-three individual char from Fukushima and 16 individual char from Gunma were selected and brought to the laboratory for further processing (Table 3.1). Standard length (mm) and body weight (g) of char were measured in the field using scales and weight measuring meter (AND HL-300 WP, Japan) (Figure 3.3). Based on the analysis of stomach contents, white-spotted char in Fukushima and Gunma consumed similar prey items (Figure 3.4).

Figure 3.2. White-spotted char sampling using LR-24 backpack electrofisher in headwater stream.

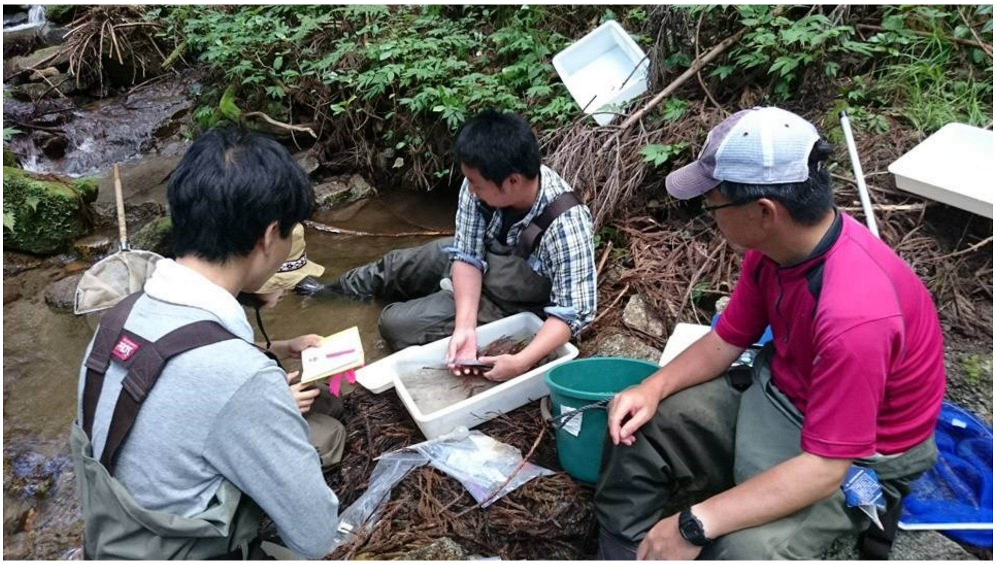


Figure 3.3. Length and weight measurement of white-spotted char in field.

Figure. 3.4. Gut content analysis of white-spotted char in study site.

I collected samples of possible prey items in the stream and riparian zones. I placed 14 plastic cups (diameter: 8 cm, depth: 11 cm) with an attractant (a mixture of beer and a lactic acid beverage) as bait traps and left them on the forest floor for 36 h during each season (Sakai et al., 2016). The collected samples were gently washed using distilled water and stored in plastic bags with respect to each taxon before ^{137}Cs activity concentration analysis. I also set five bucket bait-traps, centrally equipped with attractant-soaked cotton, on an aluminum plate that was wholly covered by a tea strainer to avoid changes in the isotopic ratios (δ^{13}C and δ^{15}N) (Sakai et al., 2016b). These samples were stored in the same manner as for the other bait trap samples and used for stable isotopic analyses.

Aquatic prey items were collected repeatedly from pools and riffles which are the basic channel units with different current velocity of headwater streams. I placed a D-frame net (mesh size: 250 µm) on the stream bed and then disturbed the immediate upstream area with hands and/or feet. The collected samples were placed on a white tray and aquatic prey items were selected using tweezers (Figure 3.5). The samples were identified to the lowest possible taxonomic levels, rinsed gently to remove attached sediments and stored in glass vials with respect to each taxon. I defined each of the prey items of char individually using taxon number from taxon 1 to taxon 12 (Tables 3.3 and 3.4). I also sampled submerged needle litter of Japanese cedar (*Cryptomeria japonica*; Japanese name: Sugi), which was the most dominant basal food resource in my study sites (Sakai et al., 2016b), and stream sediment to estimate TF_c from litter to char and from sediment to char, respectively (Figure 3.9a and b). The stream litter were collected those were attached to the twigs, not dark brown or black in color which indicate decomposition, and buried in the stream channels. Three sediment samples were collected using shovels in each study segment following the methods of Grost et al. (1991). All of the samples were stored in freezers at -20°C prior to sample processing.

Figure 3.5. Sorting and identification of prey items of white-spotted char in laboratory.

3.2.2. Sample processing

Standard length and wet weight were measured for char samples (Figure 3.3). I then calculated condition factor (K), expressed as the following formula based on Fulton (1904):

$$K = \frac{W}{L^3} \times 100, \quad (1)$$

where W is body weight in g and L is standard length in cm. K indicates the relative fatness of fishes, which is related to their feeding behavior (Froese, 2006) and metabolic rate (Seppänen et al., 2009). I hypothesized that K may affect the TFs of ^{137}Cs from prey to chars.

The ages of sampled fish were determined with otoliths using the surface reading method (Brothers, 1987). Muscles were dissected from individual fish and used for measuring δ^{13}C, δ^{15}N and ^{137}Cs activity concentrations. These tissues were dried at 60°C for more than 2 days and ground to a fine powder using an agate mortar and pestle. However, muscle tissues were used for ^{137}Cs measurement other than any parts of char body such as bone, liver, kidney, gastrointestinal tract tissues because most of the ^{137}Cs accumulated and concentrated in muscle tissues of fish body (Forseth et al., 1991; Whicker et al., 1993); while bone exhibited the lowest amount of ^{137}Cs, and the ^{137}Cs in other organs were also very small (Potter et al., 1989). ^{137}Cs in gut contents was not representative without considering whole-body concentration of fish as gut had different ratios of consummated prey items with different contamination levels (Potter et al., 1989; Elliot et al., 1992).

Estimating activity concentrations of ^{137}Cs requires samples with more than about 1 g, so I selected only dominant aquatic and terrestrial prey items that had sufficient mass. I also assumed that these sampled prey items were highly available (i.e., high density) in both terrestrial and aquatic habitats and contributed to the dietary sources of chars. The samples of aquatic and terrestrial prey items and litter were dried at 60°C for 4–10 days. Prey items and char muscle were ground and litter were pulverized using a FM-1 electric mill (Osaka Chemical Co., Ltd., Osaka, Japan). Three replicated samples were used for stable isotopic analysis, while only one to two samples for each prey item were used for ^{137}Cs radioactivity measurements. The stream sediment were dried overnight at 105°C and separated using a 2-mm sieve. Sediment samples with particles < 2 mm were used for ^{137}Cs activity concentration measurements because ^{137}Cs attached with clay minerals (Sakai et al., 2015). Because litter and sediment were abundant, sufficient material was collected to allow for three ^{137}Cs radioactivity measurement replicates.

3.2.3. ^{137}Cs activity concentration measurement

The ^{137}Cs activity concentrations in all samples were measured based on dry weight basis at 661.6 keV using a high purity n-type germanium coaxial gamma-ray detector system coupled with a multi-channel analyzer (GCW2022 coupled with DSA1000 [CANBERRA, Meriden, CT, USA]; and Ortec GEM20-70 coupled with DSPEC jr [ver. 2.0; Ametek-AMT, Beijing, PRC]) (Figure 3.6). The energy and efficiency calibrations for this detector were performed using standard provided by the suppliers and blank (background) samples. Each sample was measured for < 5% error counts per net area count. Natural decay of ^{137}Cs concentration corrected based on the sampling date. Detector systems were tested for proficiency following the test using reference water (n = 2), soil (n = 1) and organic matter (n = 1) provided by the International Atomic Energy Agency.

Figure 3.6. Gamma spectrometer for the measurement of ^{137}Cs activity concentrations in samples.

3.2.4. Analysis of stable isotope ratio and estimating diet contribution of prey

The $\delta^{13}C$ and $\delta^{15}N$ concentrations in the samples were determined using an isotope ratio mass spectrometer coupled with an elemental analyzer (Finningan-MAT 252; Thermo Fisher Scientific, Florence, KY, USA). The results were expressed in delta notation (Table 3.3) and calculated as:

$$\delta X = [(R_{sample}/R_{standard}) - 1] \times 1000\ (‰),$$

where X is ^{13}C or ^{15}N and R is the $^{13}C/^{12}C$ or $^{15}N/^{14}N$ ratio for $\delta^{13}C$ or $\delta^{15}N$, respectively (Peterson and Fry, 1987). Pee Dee Belemnite (derived from the Cretaceous marine fossil, *Belemnitella americana*) carbon and atmospheric nitrogen were used as respective standard reference materials. The working standards of known delta values (e.g., tyrosine and proline) were analyzed to confirm reproducibility and accuracy of isotope analyses. All analytical errors were within 0.1‰ for $\delta^{13}C$ and 0.3‰ for $\delta^{15}N$. This technique identifies both trophic level and food web paradigms in ecosystem ecology (Post, 2002). Typically, the $\delta^{15}N$ of a consumer is enriched by 3–4‰ relative to its diet, which could be used for estimating trophic positions in the predator–prey interactions of a food web (Figure 3.7) (Peterson and Fry, 1987).

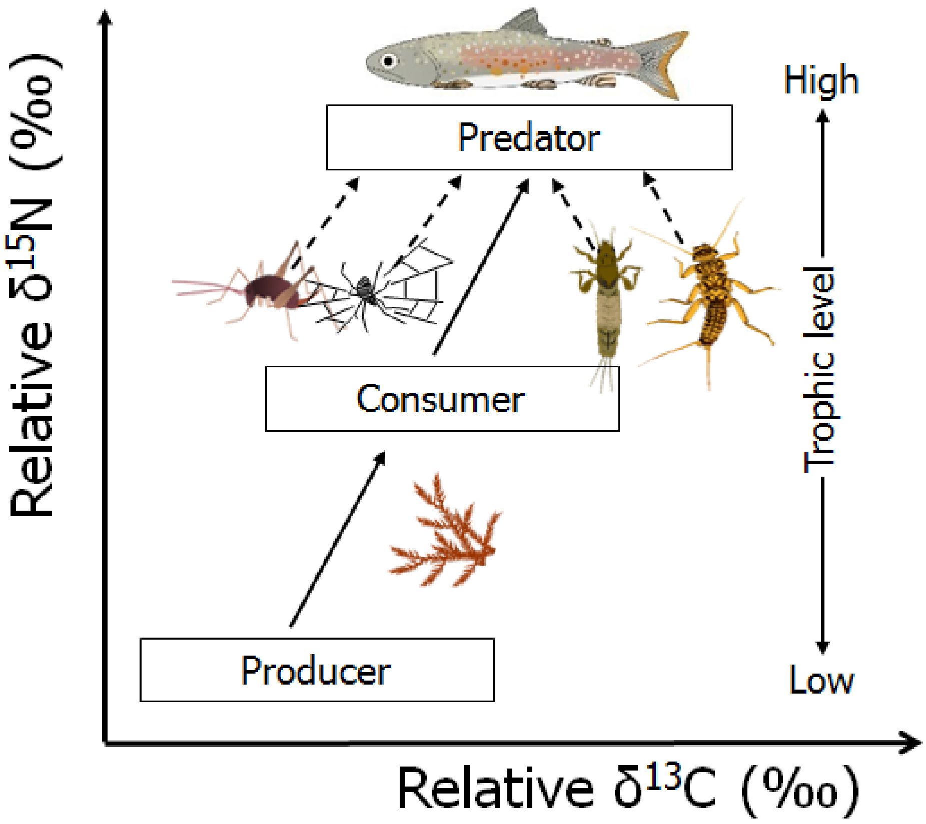


Figure 3.7. Schematic illustration of food web structures in headwater streams.

A Bayesian analysis of a stable isotope mixing model was performed to examine the prey–char dietary contributions (Figure 3.8) (Phillips, 2001). Bayesian analysis of stable isotope mixing models has previously been used to determine the natural variation and partitioning of food sources (Parnell et al., 2010). This model incorporated the $\delta^{13}C$ and $\delta^{15}N$ isotope signatures of predator and prey items to produce lower, upper, mean and mode estimates of dietary contribution of each prey item for a given predator (Parnell et al., 2010). Discrimination factors, or the amount of change in isotopic ratios (means ± standard deviation), in the models were set to 0.47 ± 1.23‰ for $\delta^{13}C$ and 3.41 ± 0.20‰ for $\delta^{15}N$, based on a meta-analysis of trophic fractionation (Vander Zanden and Rasmussen, 2001). I ran 500,000 iterations to ensure proper model function. I then calculated the lower and upper bounds of the 50% credibility intervals of the contributions of each prey item to chars. All models were run using R software (ver. 3.1.2; R Development Core Team, 2014) with the package, SIAR (ver. 4.0; Inger et al., 2006).

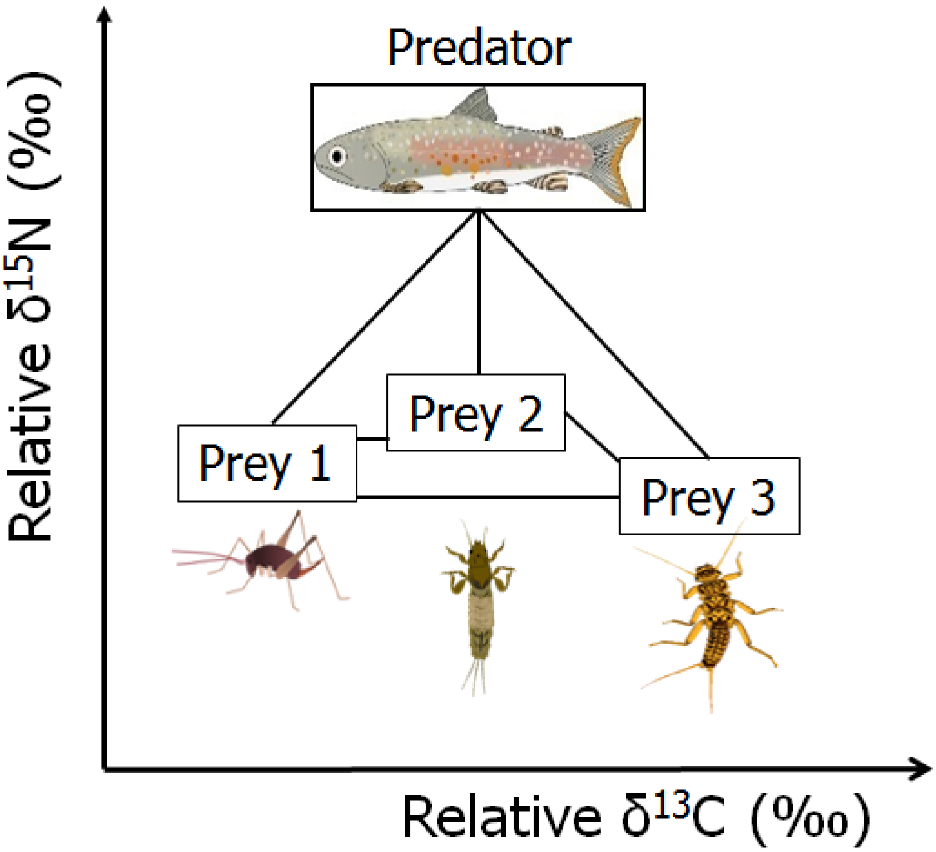


Figure 3.8. Conceptual illustration of mixing model based on stable isotopes for estimating diet contributions of prey items.

3.2.5. Calculation of food web-based transfer factor

TF_{web} is calculated based on predator–prey relationships and described as following equation:

$$TF_{\mathrm{web}} = C_{fish}/C_{total\ preys}, \quad (2)$$

where C_{fish} is the ^{137}Cs activity concentration in char (Bq kg^{-1}-dry). Denominator of TF_{web} is expressed as the following equation (3), based on the dietary contributions of multiple prey items (Figure 3.9d):

$$C_{total\ preys} = \sum_n (C_{prey\ n} \cdot f_{prey\ n}), \quad (3)$$

where $C_{prey\ n}$ is the ^{137}Cs activity concentration in n^{th} prey items (Bq kg^{-1}-dry) and $f_{prey\ n}$ is the dietary contributions of the n^{th} prey items. Dominant 12 prey contributions (n = 1,2, ……,12) were used for this calculation. I used the mean value of ^{137}Cs activity concentration of a given prey as $C_{prey\ n}$. The estimated lower and upper contributions of potential prey items to char based on the Bayesian analysis (see section 3.2.4) were used for $f_{prey\ n}$ for each prey item. Therefore, TF_{web} enabled us to evaluate ^{137}Cs transfers from multiple prey items to a predator. $TF_{\mathrm{web}} > 1$ indicated bioaccumulation, while $TF_{\mathrm{web}} < 1$ indicated biodilution.

In addition to TF_{web}, I estimated TF_{c} and TTF as comparisons. The TF_{c} from contaminated media (e.g., litter and/or sediment) to biota was calculated using the ^{137}Cs activity concentrations of char divided by the ^{137}Cs activity concentrations of either stream litter (litter–char) or sediment (sediment–char), separately (Figure 3.9a and b). To calculate the TTF from one prey item to a char, I used the ^{137}Cs activity concentrations of each prey item as the denominator for the ^{137}Cs activity concentration in each char (Figure 3.9c).

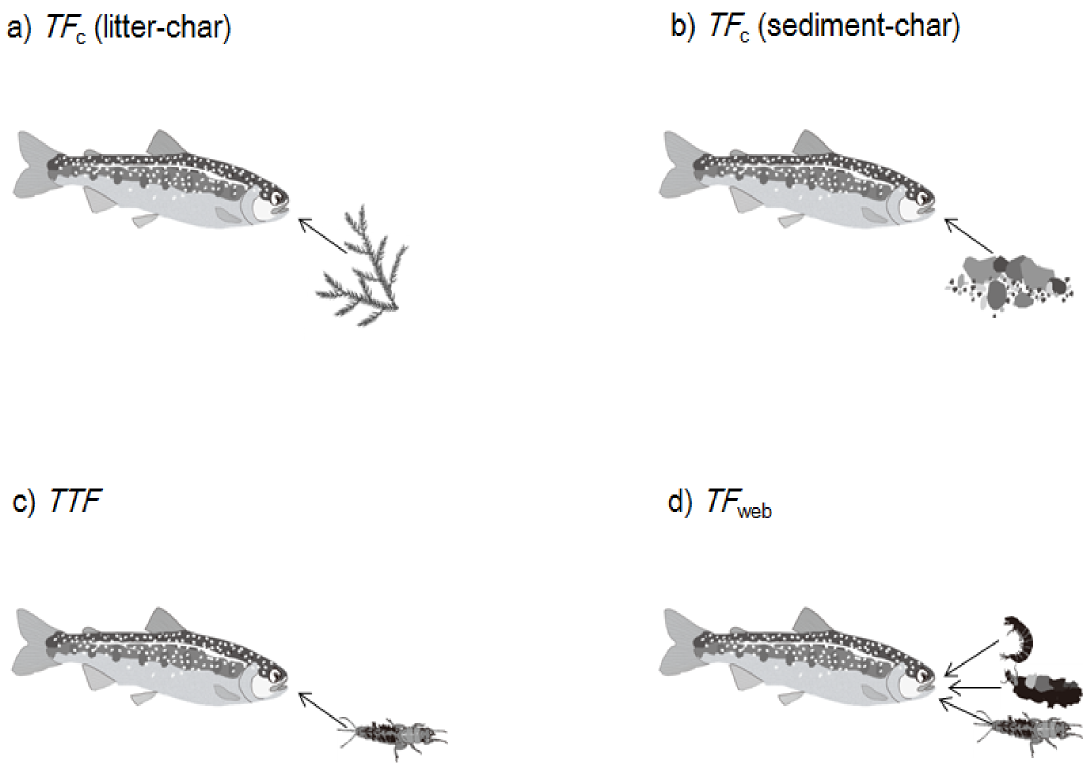


Figure 3.9. Conceptual characteristics of each transfer factor (*TF*; contemporary transfer factor [TF_c], trophic transfer factor [*TTF*], and food web-based transfer factor [TF_{web}]).

3.2.6. Statistical analysis

I calculated mean values and coefficients of variation (CV) to evaluate each *TF*. CV can be used to examine general patterns of *TF* variability. To identify the possible factors controlling TF_{web}, I used generalized linear models (GLMs). GLMs were used separately for all *TF*s (TF_{web}, *TTF*, and TF_{c}). Explanatory variables for the GLMs were study site, standard length and condition factor (*K*). To avoid multicollinearity, I excluded body weight because of the significant positive correlation between standard length and weight (Fukushima: $\rho = 0.98$, $p < 0.001$; Gunma: $\rho = 0.91$, $p < 0.001$ based on Spearman's rank correlation test). A Poisson error distribution was used for the response variables together with a log link function. All the statistical analyses were performed using R (ver. 3.1.2; R Development Core Team, 2014).

3.3. Results

3.3.1. ^{137}Cs activity concentrations in white-spotted char, prey items and the environment

The ^{137}Cs activity concentrations of white-spotted char in Fukushima and in Gunma ranged from 704 to 6082 Bq kg^{-1}-dry (mean ± SD: 2006 ± 1068 Bq kg^{-1}-dry), and from 193 to 618 Bq kg^{-1}-dry (397 ± 128 Bq kg^{-1}-dry), respectively (Table 3.1). The standard lengths of the char ranged from 65 to 200 mm (107 ± 38 mm) in Fukushima, and from 82 to 187 mm (124 ± 26 mm) in Gunma. The body weights ranged from 3.9 to 88.7 g (23.6 ± 24 g), and from 7.2 to 68.1 g (26.9 ± 16.9 g) in Fukushima and Gunma, respectively. *K* varied from 1.1 to 1.9 (1.5 ± 0.2) in Fukushima, and from 0.8 to 2.0 (1.3 ± 0.3) in Gunma. The ages of the sampled fish ranged from 0 to 3 years at both study sites.

I obtained ranges of prey items of char based on stomach contents in both Fukushima and Gunma (Tables 3.3 and 3.4). Freshwater crabs (taxon 1, *Geothelphusa dehaani*) inhabit both terrestrial and aquatic environments. Three taxa such as web-building spiders (taxon 2, Tetragnathidae gen. spp.), spider crickets (taxon 3, Rhaphidosphoridae gen. spp.), and ground beetles (taxon 4, *Carabus* sp.) were found as terrestrial prey items. Dragonflies (taxon 5, *Anotogaster sieboldii*; taxon 6, Gomphidae gen. spp.) were dominant aquatic prey items. Similarly, damselflies (taxon 7, *Mnais costalis*), mayflies (taxon 8, *Ephemera japonica*) and stoneflies (taxon 9, *Kamimuria* sp.; taxon 10, Perlinae gen. spp.; and taxon 11, *Paragnetina* sp.) and craneflies (taxon 12, *Tipula* sp.) were also found as aquatic prey items (Tables 3.3 and 3.4). ^{137}Cs activity concentration in terrestrial prey items in Fukushima ranged from 957 to 3546 Bq kg^{-1}-dry, with the highest concentration being found in spider crickets (Rhaphidosphoridae gen. spp.) and freshwater crabs (*G. dehanni*) (Table 3.4). ^{137}Cs activity concentration in aquatic prey items in Fukushima ranged from 247 to 3592 Bq kg^{-1}-dry. The highest ^{137}Cs activity concentration was found in aquatic prey mayflies (*E. japonica*), followed by craneflies (*Tipula* sp.) as 1702 Bq kg^{-1}-dry (Table 3.4).

Similar ^{137}Cs activity concentration patterns were also observed between the terrestrial and aquatic environments in Gunma (Table 3.4). ^{137}Cs activity concentrations in the terrestrial taxa in Gunma ranged from 158 to 417 Bq kg^{-1}-dry with the highest values occurring in freshwater crabs (*G. dehanni*) and spider crickets (Rhaphidosphoridae gen. spp.). Some aquatic taxa, such as Perlinae gen. spp. and *Paragnetina* sp., were below the detection limit, while mayflies (*E. japonica*) and craneflies (*Tipula* sp.) had values of 658 and 405 Bq kg^{-1}-dry, respectively.

The ^{137}Cs activity concentrations in stream litter tended to be greater than those in biota. ^{137}Cs activity concentrations in litter of stream channels (water and sediment flowing path within the stream banks) in Fukushima ranged from 3959 to 8496 Bq kg^{-1}-dry (mean ± SD: 6439 ± 1510 Bq kg^{-1}-dry), while those in Gunma ranged from 1097 to 7315 Bq kg^{-1}-dry (2839 ± 1675 Bq kg^{-1}-dry) (Table 3.2). ^{137}Cs activity concentrations in stream sediments in Fukushima ranged from 543 to

740 Bq kg^{-1}-dry (624 ± 103 Bq kg^{-1}-dry). In Gunma, ^{137}Cs activity concentrations in stream sediments ranged from 244 to 442 Bq kg^{-1}-dry (325 ± 104 Bq kg^{-1}-dry) (Table 3.2).

Table 3.1. Summary of numbers, standard lengths, weights, condition factors, ages and ^{137}Cs activity concentrations of white-spotted char samples during August 2012-May 2013.

Parameters	Fukushima (n=23)				Gunma (n=16)			
	Min	Max	Mean	±SD	Min	Max	Mean	±SD
Standard length (mm)	65	200	107	38	82	187	124	26
Body weight (g)	3.9	88.7	23.6	24	7.2	68.1	26.9	16.9
Condition factor (*K*)	1.1	1.9	1.5	0.2	0.8	2.0	1.3	0.3
Age (y)	0	3.0	1.5	1.0	0	3.0	1.9	0.9
^{137}Cs activity concentration (Bq kg^{-1}-dry)	704	6082	2006	1068	193	618	397	128

*n indicate the number of chars

Table 3.2. Summary of sample numbers, ^{137}Cs activity concentrations of stream litters and stream sediments during August 2012-May 2013.

Components	Fukushima					Gunma				
	n	^{137}Cs activity concentration (Bq kg^{-1}-dry)				n	^{137}Cs activity concentration (Bq kg^{-1}-dry)			
		Min	Max	Mean	±SD		Min	Max	Mean	±SD
Stream litter	12	3959	8496	6439	1510	12	1097	7315	2839	1675
Stream sediment	3	543	740	624	103	3	244	442	325	104

*n indicate the number of samples.

Table 3.3. Summary of stable isotope ratios of carbon ($\delta^{13}C$) and nitrogen ($\delta^{15}N$) of white-spotted chars and prey items during August 2012-May 2013.

Taxon no	Taxon	English name	Fukushima				Gunma			
			$\delta^{13}C$ (‰)		$\delta^{15}N$ (‰)		$\delta^{13}C$ (‰)		$\delta^{15}N$ (‰)	
			Mean	±SD	Mean	±SD	Mean	±SD	Mean	±SD
Predator	*Salvelinus leucomaenis*	Whitespotted char	-25.5	0.7	6.3	0.6	-24.4	0.4	5.0	0.6
Taxon 1	*Geothelphusa dehaani*	Freshwater crab	-23.9	0.3	1.8	1.0	-24.3	0.5	0.4	0.5
Taxon 2	Tetragnathidae gen. spp.	Web-building spider	-25.8	–	5.0	–	-25.9	–	2.8	–
Taxon 3	Rhaphidosphoridae gen. spp.	Spider cricket	-26.4	0.3	1.7	0.7	-25.4	0.3	0.5	0.4
Taxon 4	*Carabus* sp.	Ground beetle	-26.8	–	2.5	–	-25.9	–	1.0	–
Taxon 5	*Anotogaster sieboldii*	Dragonfly	-26.2	0.1	3.6	0.4	-25.6	–	2.6	–
Taxon 6	Gomphidae gen. spp.	Dragonfly	-25.6	0.2	5.0	0.7	-25.4	0.3	3.8	0.7
Taxon 7	*Mnais costalis*	Damselfly	-26.2	–	4.0	–	-24.9	–	1.9	–
Taxon 8	*Ephemera japonica*	Mayfly	-25.5	0.9	5.5	1.2	-25.1	0.5	2.1	0.9
Taxon 9	*Kamimuria* sp.	Stonefly	-29.4	–	4.6	–	-27.0	–	3.0	–
Taxon 10	Perlinae gen. spp.	Stonefly	-28.1	–	4.2	–	-24.7	–	2.3	–
Taxon 11	*Paragnetina* sp.	Stonefly	-28.7	–	4.0	–	-25.9	–	3.9	–
Taxon 12	*Tipula* sp.	Cranefly	-28.1	1.3	0.8	0.5	-28.5	0.3	-1.1	0.9

3.3.2. Diet contributions and ^{137}Cs activity concentrations

The mean dietary contributions of 12 prey items in both study sites were similar, with lower and upper estimates of 3–4% and 11–12%, respectively (Table 3.4). Freshwater crabs (*G. dehanni*) and spider crickets (Rhaphidosphoridae gen. spp.) tended to have high percentages of contribution. Considering the mean lower and upper estimates, the total dietary contributions based on stable isotope analysis ranged from 84.5 to 94.0% in Fukushima, and from 87.5 to 94.5% in Gunma.

The total diet based on ^{137}Cs uptake (^{137}Cs activity concentration multiplied by the dietary contribution of each taxon) varied substantially (Table 3.4). In Fukushima, the dietary ^{137}Cs contributions of spider crickets (Rhaphidosphoridae gen. spp.) ranged from 141.8 to 425.5 Bq kg^{-1}-dry, based on lower and upper uptake values, respectively (Table 3.4). The dietary ^{137}Cs uptake from mayflies (*E. japonica*) ranged from 107.8 to 395.1 Bq kg^{-1}-dry. Contributions from craneflies (*Tipula* sp.) and freshwater crabs (*G. dehanni*) also tended to be higher compared to other aquatic and terrestrial prey items, respectively. The highest contributors of dietary ^{137}Cs measurements were similar at both sites. In Fukushima, the stoneflies such as *Kamimuria* sp. and *Paragnetina* sp. contributed little, ranging from 8.6 to 31.7 Bq kg^{-1}-dry and from 7.4 to 27.2 Bq kg^{-1}-dry, respectively. In Gunma, the dragonflies (*A. sieboldii*) and stoneflies (*Kamimuria* sp.) had the lowest dietary ^{137}Cs uptake values, at 0.5 to 1.4 Bq kg^{-1}-dry and 1.6 to 4.7 Bq kg^{-1}-dry, respectively.

Table 3.4. Summary of ^{137}Cs activity concentrations and dietary contributions of from each prey item during August 2012-May 2013.

Taxon no.	Taxon	English name	Habitat	Fukushima						Gunma					
				^{137}Cs (Bq kg^{-1}-dry)		Dietary contribution (DC)		^{137}Cs x DC (Bq kg^{-1}-dry)		^{137}Cs (Bq kg^{-1}-dry)		Dietary contribution (DC)		^{137}Cs x DC (Bq kg^{-1}-dry)	
				Mean	±SD	Lower	Upper	Lower	Upper	Mean	±SD	Lower	Upper	Lower	Upper
Taxon 1	*Geothelphusa dehaani*	Freshwater crab	Forest and Stream	1634	746	0.04	0.12	65.4	196.1	417	235	0.04	0.12	16.7	50.0
Taxon 2	Tetragnathidae gen. spp.	Web-building spider	Forest	1167	-	0.03	0.11	35.0	128.4	224	-	0.03	0.12	6.7	26.9
Taxon 3	Rhaphidosphoridae gen. spp.	Spider cricket	Forest	3546	1403	0.04	0.12	141.8	425.5	387	99	0.04	0.12	15.5	46.4
Taxon 4	*Carabus* sp.	Ground beetle	Forest	957	-	0.04	0.12	38.3	114.8	158	-	0.04	0.12	6.3	19.0
Taxon 5	*Anotogaster sieboldii*	Dragonfly	Stream	632	178	0.03	0.12	19.0	75.8	12	-	0.04	0.12	0.5	1.4
Taxon 6	Gomphidae gen. spp.	Dragonfly	Stream	778	443	0.03	0.11	23.3	85.6	250	134	0.04	0.12	10.0	30.0
Taxon 7	*Mnais costalis*	Damselfly	Stream	470	-	0.03	0.11	14.1	51.7	74	-	0.03	0.12	2.2	8.9
Taxon 8	*Ephemera japonica*	Mayfly	Stream	3592	1006	0.03	0.11	107.8	395.1	658	206	0.04	0.12	26.3	79.0
Taxon 9	*Kamimuria* sp.	Stonefly	Stream	288	-	0.03	0.11	8.6	31.7	39	-	0.04	0.12	1.6	4.7
Taxon 10	Perlinae gen. spp.	Stonefly	Stream	476	-	0.03	0.11	14.3	52.4	n.d.	-	0.04	0.12	-	-
Taxon 11	*Paragnetina* sp.	Stonefly	Stream	247	-	0.03	0.11	7.4	27.2	n.d.	-	0.03	0.11	-	-
Taxon 12	*Tipula* sp.	Cranefly	Stream	1702	180	0.04	0.12	68.1	204.2	405	126	0.03	0.11	12.2	44.6

*n.d. indicate under detetion limit

3.3.3. Transfer factors

The TF_{web} based on the lower dietary contribution ranged from 1.41 to 13.53 (mean ± SD: 3.79 ± 2.43) in Fukushima and from 1.92 to 8.56 (4.30 ± 1.66) in Gunma (Table 3.5). TF_{web} based on the upper dietary contribution ranged from 0.38 to 3.45 (1.12 ± 0.61) in Fukushima and from 0.63 to 2.07 (1.30 ± 0.43) in Gunma (Table 3.5). The CV of TF_{web} in Fukushima was 0.64 for the lower dietary contribution and 0.54 for the upper dietary contribution, while these values in Gunma were 0.39 and 0.33 for the lower and upper dietary contributions, respectively.

The litter–char TF_{c} in Fukushima ranged from 0.11 to 0.94 (mean ± SD: 0.31 ± 0.17), while that in Gunma ranged from 0.07 to 0.22 (0.14 ± 0.05). The CV was 0.53 in Fukushima and 0.32 in Gunma (Table 3.5). Similarly, the sediment–char TF_{c} in Fukushima ranged from 1.13 to 9.74 (3.21 ± 1.71) with a CV of 0.53, while those in Gunma ranged from 0.60 to 1.90 (1.22 ± 0.39) with a CV of 0.32 (Table 3.5).

The *TTF* varied considerably depending on the prey selection of a given white-spotted char (Table 3.5). For example, in a white-spotted char with 2,836 Bq kg^{-1}-dry ^{137}Cs activity concentration, the *TTF* was 0.79 for mayflies (*E. japonica*) with 3,592 Bq kg^{-1}-dry ^{137}Cs activity concentration, and 11.48 for *Paragnetina* sp. with 247 Bq kg^{-1}-dry ^{137}Cs activity concentration. The mean *TTF* of 276 combinations between prey items and white-spotted chars in Fukushima ranged from 0.56 to 8.12 (CV: 0.53). The mean *TTF* of 160 combinations between prey items and white-spotted chars in Gunma ranged from 0.60 to 33.07 (CV: 0.32). We excluded two prey taxa (Perlinae gen. spp. and *Paragnetina* sp.) because the ^{137}Cs activity concentrations were below the detection limit.

Table 3.5. Summary statistics of each *TF* at the Fukushima and Gunma sites.

Study site	Transfer factor	Min - Max	Mean	±SD	Coefficient of variation
Fukushima	TF_{web} (lower)	1.41 - 13.53	3.79	2.43	0.64
	TF_{web} (upper)	0.38 - 3.45	1.12	0.61	0.54
	TF_c (litter-char)	0.11 - 0.94	0.31	0.17	0.53
	TF_c (sediment-char)	1.13 - 9.74	3.21	1.71	0.53
	TTF (taxon1-char)	0.43 - 3.72	1.23	0.65	0.53
	TTF (taxon2-char)	0.60 - 5.21	1.72	0.92	0.53
	TTF (taxon3-char)	0.20 - 1.72	0.57	0.30	0.53
	TTF (taxon4-char)	0.74 - 6.35	2.10	1.12	0.53
	TTF (taxon5-char)	1.11 - 9.62	3.17	1.69	0.53
	TTF (taxon6-char)	0.91 - 7.82	2.58	1.37	0.53
	TTF (taxon7-char)	1.50 - 12.94	4.27	2.27	0.53
	TTF (taxon8-char)	0.20 - 1.69	0.56	0.30	0.53
	TTF (taxon9-char)	2.45 - 21.12	6.97	3.71	0.53
	TTF (taxon10-char)	1.48 - 12.78	4.22	2.24	0.53
	TTF (taxon11-char)	2.85 - 24.62	8.12	4.33	0.53
	TTF (taxon12-char)	0.41 - 3.57	1.18	0.63	0.53
Gunma	TF_{web} (lower)	1.92 - 8.56	4.30	1.66	0.39
	TF_{web} (upper)	0.63 - 2.07	1.30	0.43	0.33
	TF_c (litter-char)	0.07 - 0.22	0.14	0.05	0.32
	TF_c (sediment-char)	0.60 - 1.90	1.22	0.39	0.32
	TTF (taxon1-char)	0.46 - 1.48	0.95	0.31	0.32
	TTF (taxon2-char)	0.86 - 2.76	1.77	0.57	0.32
	TTF (taxon3-char)	0.50 - 1.60	1.03	0.33	0.32
	TTF (taxon4-char)	1.22 - 3.91	2.51	0.81	0.32
	TTF (taxon5-char)	16.12 - 51.47	33.07	10.67	0.32
	TTF (taxon6-char)	0.77 - 2.47	1.59	0.51	0.32
	TTF (taxon7-char)	2.61 - 8.35	5.36	1.73	0.32
	TTF (taxon8-char)	0.29 -0.94	0.60	0.19	0.32
	TTF (taxon9-char)	4.96 - 15.84	10.18	3.28	0.32
	TTF (taxon10-char)	–	–	–	–
	TTF (taxon11-char)	–	–	–	–
	TTF (taxon12-char)	0.48 - 1.52	0.98	0.32	0.32

3.3.4. Controlling factors of transfer factors

The *K* value for white-spotted char at Fukushima was not significantly correlated with either standard length (ρ = −0.14, p = 0.52) or body weight (ρ = −0.01, p = 0.93) based on Spearman's rank correlation test. *K* for white-spotted char in Gunma was also not significantly correlated with either standard length (ρ = −0.12, p = 0.64) or body weight (ρ = 0.24, p = 0.36). The GLM analysis, which included site, standard length and *K* as independent variables, showed that the site factor was not significant for most of the *TF*s, except for upper TF_{web}, and *TTF* (taxon3–char, taxon4–char, and taxon7–char) (Table 3.6). Similarly, *K* was not significant for explaining most *TF*s, except *TTF* (taxon3–char) (Table 3.6). Standard length was significant for most *TF*s, except for lower TF_{web}, and *TTF* (taxon1–char, taxon6–char, taxon8–char and taxon12–char) (Table 3.6).

Table 3.6. Summary of the generalized linear models. Each *TF* was analyzed separately. Bold characters indicate statistical significance ($p < 0.05$).

Response variables	Site (Fukushima vs Gunma)			Standard length			Condition factor (*K*)		
	Estimate	*z*	*p*	Estimate	*z*	*p*	Estimate	*z*	*p*
TF_{web} (lower)	-0.902	-1.128	0.259	0.009	1.019	0.308	0.111	0.074	0.941
TF_{web} (upper)	-1.069	-4.040	**<0.001**	0.009	3.289	**0.001**	0.135	0.280	0.780
TF_c (litter-char)	-0.371	-1.106	0.269	0.009	1.992	**0.046**	0.007	0.011	0.991
TF_c (sediment-char)	-0.091	-0.348	0.728	0.008	2.336	**0.020**	-0.045	-0.085	0.933
TTF (taxon1-char)	0.466	1.183	0.237	0.007	1.321	0.186	-0.142	-0.812	0.856
TTF (taxon2-char)	0.056	0.249	0.803	0.008	2.564	**0.010**	-0.074	-0.161	0.872
TTF (taxon3-char)	2.213	17.205	**<0.001**	0.005	3.288	**0.001**	-0.326	-2.035	**0.042**
TTF (taxon4-char)	-0.597	-2.395	**0.017**	0.009	2.914	**0.004**	0.052	0.105	0.917
TTF (taxon5-char)	0.104	0.663	0.508	0.008	3.660	**<0.001**	-0.082	-0.256	0.798
TTF (taxon6-char)	-0.046	-0.102	0.918	0.008	1.327	0.184	-0.056	-0.060	0.952
TTF (taxon7-char)	0.252	2.136	**0.033**	0.008	4.651	**<0.001**	-0.108	-0.453	0.650
TTF (taxon8-char)	-0.301	-0.900	0.368	0.008	1.951	0.051	-0.002	-0.003	0.998
TTF (taxon9-char)	-0.012	-0.070	0.944	0.009	3.941	**<0.001**	-0.079	-0.244	0.823
TTF (taxon12-char)	0.029	0.093	0.926	0.008	1.943	0.052	-0.058	-0.090	0.928

3.4. Discussion

3.4.1. Contributions from prey items to white-spotted char

The results indicated that both terrestrial and aquatic insects were the main prey items of white-spotted char. White-spotted char in headwater stream depend on terrestrial prey items to subsidize their diet of aquatic ones (Nakano and Murakami, 2001). In both Fukushima and Gunma, the dominant aquatic prey taxa were mayflies, *E. japonica* (collectors) and craneflies, *Tipula* sp. (shredders) generally inhabit in depositional environments of stream channels. In such depositional environments, fine particulate matters including sands, clay materials, particulate organic matters and leaf litters generally accumulate, providing substrate as well as food for the aquatic insects (Kato et al., 2004; Iwata, 2007; Yoshimura and Akama, 2014). Dragonflies, *A. sieboldii* and Gomphidae gen. spp. and damselflies, *M. costalis* (insect predators) also occupied depositional environments of headwater streams (Merritt and Cummins, 1996; Abdelsalam, 2012). These aquatic prey items tended to have higher levels of ^{137}Cs activity concentrations because their ^{137}Cs activity concentrations depend on the accumulated ^{137}Cs in the depositional environments of streams (Yoshimura and Akama, 2014). Predatory stoneflies (*Kamimuria* sp., *Paragnetina* sp. and Perlinae gen. spp.) feed on aquatic insects and are consumed by white-spotted char (Miyasaka and Genki-Kato, 2009). They have the chloride cells for actively expel ^{137}Cs and thus result in moderate ^{137}Cs accumulation in their bodies (Yoshimura and Akama, 2014). Small sized freshwater crabs, *G. dehaani* (collectors) generally inhabit both terrestrial and aquatic environments (Okano et al., 2000) were also an important food source of white-spotted char in Fukushima and Gunma.

Because white-spotted char consume drifting invertebrates, this study confirmed that various terrestrial insects such as web-building spiders (Tetragnathidae gen. spp.), spider crickets (Rhaphidosphoridae gen. spp.) and ground beetles (*Carabus* sp.) became important dietary contributions at both sites (Nakano and Furukawa-Tanaka, 1994; Sakai et al., 2016b). In particular, spider crickets (Raphidosphoridae gen. spp.) occasionally approach stream channels although they generally inhabit the forest floor (Taylor et al., 2005). Because behavior-controlling parasites live inside spider crickets may direct them towards streams, spider crickets become an important food source for white-spotted chars (Sato et al., 2011). Web-building spiders (Tetragnathidae gen. spp.) inhabit the riparian forest and construct their webs above streams (Yoshida, 1981). Therefore, such spiders occasionally fall into stream channels and are consumed by white-spotted chars. Ground beetles (*Carabus* sp.), living as predators at the forest edge (ElSayed and Nakamura, 2010), were also considered as important food sources for white-spotted chars in both Fukushima and Gunma.

However, I used the mean values of ^{137}Cs activity concentrations of char and prey items not considering the values of standard deviation (±SD) of ^{137}Cs activity concentrations in char and some prey items in calculation of dietary ^{137}Cs contributions and resulted TF_{web}. I assume that as diet contributions (DC) of all prey items were almost within the similar ranges (Table 3.4), and SD consider both positive and negative values of ^{137}Cs activity concentrations; therefore, possibly the resulted mean dietary ^{137}Cs contributions and TF_{web} using both positive and negative values of SD have not generate large variations in TF_{web} values which indeed need to consider in future experiments. Furthermore, the total consumed ^{137}Cs via dietary sources accumulated fully inside the char body or not still questionable. It is difficult to answer this question as I couldn't measure the amount of ^{137}Cs eliminated from the char body in natural environmental conditions. Further studies need to be done to measure the amount of intake and elimination of ^{137}Cs in char body under laboratory conditions.

3.4.2. Applicability of the food web-based transfer factor

Despite the different levels of fallout in Fukushima and Gunma, TF_{web} of white-spotted char were similar at both sites (Table 3.5). This result showed that prey–predator TF_{web} was at a similar order of magnitude regardless of contamination level. In previous studies using TF_{c} based on ^{137}Cs activity concentration of fish and water, estimated TF_{c} varied substantially depending on the location (Smith et al., 2000a; Čepanko et al., 2007; Tuovinen et al., 2013; Fukushima and Arai, 2014). For example, after the Chernobyl accident, the TF_{c} of perch in the Kiev Reservoir was 1.2, while in the cooling pond it was 3.8 based on the ^{137}Cs activity concentrations of fish and water (Smith et al., 2000a). These TF_{c} values varied due to the variation in habitat contamination levels, which may not directly propagate to the contamination levels of biota. To avoid potential erroneous estimates of TF_{c} associated with different levels of contaminations, TF_{web} would be best used at locations with similar biotic and abiotic characteristics such as riparian vegetation, watershed topography, water temperature and channel morphology but different contamination level. Therefore, understanding the processes of TF_{web} across ecosystems is crucial to extrapolate the variability of *TF*s in fish within the respective food webs.

I found contradictory *TF*s among biodilution and accumulation depending on the calculation methods of *TF*s. By using water–fish ^{137}Cs activity concentrations, the estimated TF_{c} values become substantially high because the activity concentration of ^{137}Cs in water is typically very low (Iwagami et al., 2017). ^{137}Cs activity concentrations were relatively low in organisms compared with their habitats and essentially the high ^{137}Cs activity concentrations were found in litter contaminated by the FDNPP accident (Sakai et al., 2016b; Kato et al., 2017). Even in aquatic environments, where ^{137}Cs activity concentrations were reduced by leaching (Sakai et al., 2015), litter–char TF_{c} indicated biodilution. Such biodilution was also reported by Murakami et al. (2014).

The inconsistency of *TF*s was more pronounced for *TTF*s using a single prey taxon and predator. The estimated *TTF* values indicated both dilution and accumulation from prey to predator, depending on the prey item selected. The variability of *TTF*s in this study was much greater than those in previous studies (Zhao et al., 2001; Tuovinen et al., 2013). For instance, Zhao et al. (2001) indicated that ^{137}Cs *TTF* values ranged from 1.0 to 4.4 (median: 2) for the predatory fish, mangrove snapper (*Lutjanus argentimaculatus*), in Hong Kong. In lakes in Finland, the ^{137}Cs prey–predator *TTF* between fish species in aquatic food chains was 2.9 ± 0.23 (Tuovinen et al., 2013). The greater variability of the estimated *TTF* values (Table 3.5) compared with previous studies may be explained by the complex contamination levels of the prey items (Table 3.4). Because the system included strong linkages between riparian areas and streams, contamination of prey insects varied depending on whether their habitat was between the forest and streams or within streams (pools and riffles) (Sakai et al., 2016b). Indeed, mayflies (*E. japonica*) and craneflies (*Tipula* sp.) generally inhabit pools (Kato et al., 2004; Iwata, 2007; Yoshimura and Akama, 2014) where greater amounts of contaminated terrestrial detritus can be deposited. Mayflies (*E. japonica*) and craneflies (*Tipula* sp.) also directly consume organic matters as collectors and shredders, respectively (Merritt and Cummins, 1996). On the other hand, stoneflies (*Kamimuria* sp., *Paragnetina* sp. and Perlinae gen. spp.) inhabiting riffles with high water velocity tended to have lower ^{137}Cs activity concentrations. Therefore, TF_{web} would provide insight into the ecological processes of ^{137}Cs transfer in predator–prey systems in terms of stability of *TF*s values in complex ecosystems.

Through this new approach has only been tested for two forested headwater streams in Fukushima and Gunma but I assumed that this approach possibly be applicable for aquaculture practice of white-spotted char in streams, rivers, lakes and/or ponds of different locations with various fallout inventories (Wada et al., 2016) because of the presence of possible contaminated prey items within the respective aquaculture habitats which transform and/or modify this approach as a validated model of ^{137}Cs transfer factor from one conditions to another.

3.4.3. Factors affecting food web-based transfer factor

Standard length of chars significantly altered TF_{web} because large-bodied char had a higher consumption rate of contaminated food compared to small-bodied char. In general, total food consumption increases with body size in salmonid fishes (Handeland et al., 2008). Large individuals tend to consume more terrestrial food sources because of greater biomass and dietary efficiency (Nakano and Murakami, 2001). Moreover, both metabolic and elimination rates are lower in older fish with greater body size (Ugedal et al., 1992). Therefore, higher exposure to contaminated terrestrial food sources and lower elimination rates increased the ^{137}Cs in large fish. *K* was not significant, possibly because it is not sensitive enough to respond to the contaminants (Linde-Arias et al., 2008).

The other possible factor for controlling *TF*s is potassium (K^+) concentration as suggested by the various previous studies (Rowan and Rasmussen, 1994; Kryshev and Ryabov, 2000; IAEA, 2010; IAEA, 2014). Although I did not measure K^+ concentration of water, K^+ concentration in headwater streams is generally lower compared to the downstream area with agricultural runoff and soil erosion (Ueda et al., 2013). I think that effects of K^+ concentration on TF_{web} can be applicable when the streams of research area could be extended from headwaters to downstream with various sources of K^+. In addition to the chemical characteristics of water, various other factors including time sequences (or age) of exposures to contaminations need to be examined for more comprehensive understanding of processes using the TF_{web}.

Furthermore, although the present investigation evaluated the effects of fallout volumes, multiple dietary sources, body size and condition factor of char on TF_{web}, but the effects of population density of char and natural disturbance (e.g., typhoon and/or flood) on TF_{web} were not examined. As the area of headwater streams are small, char possibly was not densely populated in such headwater streams which was also reflected in sample sizes although sustainable sampling was done. Therefore, within the small population size, large-bodied char occupy at the inlet of pool to ensure the better chance for consuming highly contaminated terrestrial prey items in headwater streams (Nakano and Furukawa-Tanaka, 1999; Nakano and Murakami, 2001) which possibly had effects on TF_{web} that need to be considered in further research.

Moreover, high water flow due to typhoon, storm, flood or any other natural disturbances could accelerate the transportation of ^{137}Cs from source areas of the streams such as riparian zones of the forested areas (Ohte et al., 2012). Litter was considered as primal source of ^{137}Cs contamination (Sakai et al., 2016b), and subsequently litter and detritus could form the biggest pool of ^{137}Cs in forested ecosystems from which ^{137}Cs could transfer from lower to higher trophic levels via food web (Ohte et al., 2012). Ohte et al. (2012) showed that ^{137}Cs contamination levels in suspended matter decreased very rapidly after 1 day of the storm, and the highest contamination was observed in the downstream areas compare to the headwaters in forested catchment. Therefore, I assume that though I didn’t sampling in or after any natural disturbance, but the early research showed that contamination level within the headwater streams of riparian zones become its previous state immediate after the disturbances (Ohte et al., 2012). Thus, the disturbance possibly have no effect on TF_{web}; of course, to justify these phenomena, an intensive research need to be conducted based on ^{137}Cs transfer due to any natural disturbance in forested headwater streams in coming years.

3.5. Summary and conclusions

To examine ^{137}Cs transfer in white-spotted char, I developed a TF_{web} based on the respective dietary contributions and contaminations of prey items using stable isotope analysis of samples taken from sites in Fukushima and Gunma. Under similar environmental conditions and food web structure but different fallout levels, the mean TF_{web} were similar between two locations. In both sites, white-spotted char tended to accumulate ^{137}Cs via prey consumption at similar orders of magnitude. Therefore, this method could be used to study transfer through food webs. This approach will be beneficial for understanding patterns of concentrations in other organisms, as well as contaminants in other ecosystems (Dallinger et al., 1987; Clements and Newman, 2002). Ecosystem-based approaches, including species interactions and their dynamics and contributions to the ecosystem, are essential for protecting the environment from contamination by radiation (Bréchignac et al., 2016). Thus, TF_{web} method can also be used to develop ecosystem-based modelling of contaminant movements. For long-term projections and possible decontamination practices, process-based models of contaminant movement within complex ecosystems are needed (Clements and Newman, 2002). There is still a knowledge gap between empirical parameters and complex, process-based modelling of ^{137}Cs transfer in ecosystems (Hinton et al., 2014). Therefore, combining food web structure analysis with ^{137}Cs research will help to elucidate the fate and transport of contaminants in food webs.

CHAPTER 4

SEASONAL VARIABILITY IN FOOD WEB-BASED TRANSFER FACTOR OF ^{137}Cs

4.1. Introduction

Radioactive contaminants emitted due to the FDNPP accident were distributed widely and deposited in surrounding forested areas (Hashimoto et al., 2012; Kuroda et al., 2013). These radioactive materials entered both terrestrial and aquatic ecosystems via leaf litter (Teramage et al., 2014; Sakai et al., 2016b; Kato et al., 2017). For instance, 64% of the ^{137}Cs inventory was transported from the canopy to the forest floor as mixed broad-leaved litter in areas 40 km from the FDNPP (Kato et al., 2017). ^{137}Cs transfer from riparian forests to streams is another important process that drives contamination movement from forests to headwater streams (Sakai et al., 2016b) because of close associations between forests and stream channels (Gomi et al., 2002). Once ^{137}Cs enters streams, leaching and decomposition of litter alters ^{137}Cs availability (Sakai et al., 2015).

Because submerged litter in headwater streams is one of the most important basal food resources, ^{137}Cs attached to litter can transfer to biota from lower to higher trophic levels via food webs (Sakai et al., 2016b). In these food webs, freshwater salmonids such as white-spotted char (*Salvelinus leucomaenis*) are considered as top predators (Nakano and Murakami, 2001; Sakai et al., 2016b). Previous researches has shown that ^{137}Cs contamination is highly variable in top predatory fish. For instance, Mizuno and Kubo (2013) showed that the ^{137}Cs activity concentration in white-spotted char (*S. leucomaenis*) and cherry trout (*Oncorhynchus masou*) in the Abukuma river of western Fukushima ranged from 17 to 200 Bq kg^{-1} in the first year after the FDNPP accident. ^{137}Cs activity concentrations in white-spotted char (*S. leucomaenis*) in Fukushima rivers at locations with a mean air dose rate of 0.30 and 1.50 μSv h^{-1} ranged from 8 to 400 and from 10 to 900 Bq kg^{-1}, respectively (Wada et al., 2016).

To assess radionuclide transfer in ecosystems, transfer factors are used to estimate the order of magnitude of change in ^{137}Cs transfer to biota (ICRP, 2009). Transfer factors of freshwater fishes are calculated by dividing the radionuclide concentration in fish by the radionuclide concentration either of sediment or water (IAEA, 2010). In addition, Zhao et al. (2001) developed trophic transfer factor (*TTF*) based on the relationships between the ^{137}Cs activity concentrations of a predator and its prey. In previous chapter, a food web-based transfer factor (TF_{web}) of ^{137}Cs was developed based on possible dietary contributions of multiple prey items and their respective ^{137}Cs activity concentrations. TF_{web} has an advantage over other transfer factors indicating ^{137}Cs transfer from lower to upper trophic levels.

The ^{137}Cs activity concentrations and transfer factors in fishes may vary seasonally because of food consumption patterns and variation in contamination levels. Moreover, such variability is associated with elimination related to metabolic rate (Kolehmainen, 1974; Ugedal et al., 1995). For instance, Peles et al. (2000a) reported that the ^{137}Cs activity concentration in freshwater largemouth bass (*Micropterus salmoides*) in winter was up to 2 times greater than that in summer because uptake and excretion were lower in winter. In addition, the metabolic rate of rainbow trout (*Oncorhynchus mykiss*) in summer tended to be three-fold greater than that in winter (Facey and Grossman, 1990). Shifting food consumption from terrestrial to aquatic sources between summer and winter (Nakano et al., 1999; Sato et al., 2011) might also result in different total ^{137}Cs intakes in summer and winter. For instance, Sakai et al. (2016b) showed that the ^{137}Cs activity concentration of terrestrial sources were about four times greater than those of aquatic sources.

Although possible factors that can affect the seasonal variability of transfer factors have been identified, seasonal patterns and their associated causes have not been fully examined. Understanding the seasonal variability of transfer factors is important to determine the dominant processes that control contamination levels. Therefore, the objective of this study was to evaluate the seasonal variability of transfer factors in white-spotted char (*S. leucomaenis*) based on their seasonal patterns of ^{137}Cs consumption. I examined two areas with different amounts of fallout because differences in contamination levels could alter the balance between ^{137}Cs consumption and elimination rates. TF_{web} was used as this method can help to identify food web-based exposure pathways in ecosystems.

4.2. Materials and methods

4.2.1. Study sites

The study was conducted in two headwater streams with similar landscape structures and surrounding channel environments. Osawa-gawa stream in Nihonmatsu city, Fukushima prefecture (latitude: 37°36′N, longitude: 140°37′E) is located 45 km from the FDNPP, while Oya-san stream in Midori city, Gunma prefecture (latitude: 36°33′N, longitude: 139°21.2′E) is located approximately 190 km away from the FDNPP (Figure 4.1). Based on governmental aircraft monitoring conducted in June 2012, the ^{137}Cs inventories in Fukushima and Gunma were 100–300 and 30–60 kBq m^{-2}, respectively. The Fukushima site has a mean annual precipitation of 1248 mm and a mean air temperature of 11°C, while the Gunma site has a mean annual precipitation of 1323 mm and a mean annual air temperature of 15°C. The underlying geology at the Fukushima site is composed of lower Cretaceous granites and the Gunma site is covered by melange matrix consisting of Jurassic accretionary complex. Both of the headwater catchments are covered mostly by Japanese cedar (*Cryptomeria japonica*) and some cypress (*Chamaecyparis obtusa*). To investigate fish and their prey items, 50 m channel segment were selected. The channel segments consisted of large boulders with sequences of step and riffle pools (Montgomery and Buffington, 1997) with a flow depth of 0.05–1 m and channel width of 1–5 m.

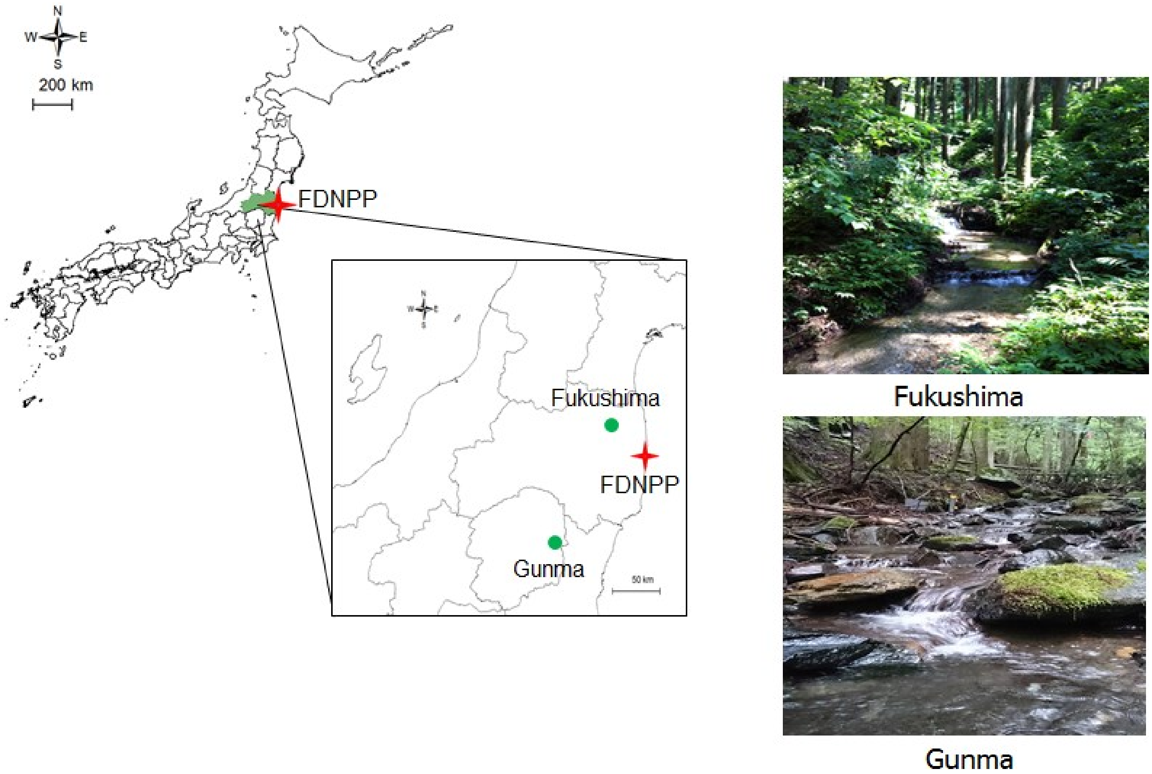


Figure 4.1. Study sites in Fukushima and Gunma with the location of Fukushima Dai-ichi Nuclear Power Plant (FDNPP).

4.2.2. Field sampling

White-spotted char (*S. leucomaenis*) and their possible prey items were collected in four consecutive seasons: summer (August) and autumn (November) in 2012; winter (February) and spring (May) in 2013. Fish were sampled using a backpack electrofisher (LR-24, Smith-Root Inc., Vancouver, Washington, USA) by skimming in 50-m channel segments. Possible prey items of char in riparian zones were collected with 14 bait traps (diameter: 8 cm, depth: 11 cm) and five buckets using beer and lactic acid bacteria beverage as an attractant. I left the traps for 36 h to collect a sufficient number of terrestrial insects for the analysis. Samples of possible prey items were washed to exclude organic and inorganic materials attached to body surfaces using distilled water. Aquatic prey items were sampled using a D-frame net (mesh size: 250 μm) on the streambed after disturbing the streambed with hands and feet. The collected samples were placed in white plastic trays and sorted using tweezers. Prey items were visually identified to the lowest possible taxonomic level and stored in separate glass vials for each taxon in the field; they were immediately transported to the laboratory and stored at -20°C until further processing.

4.2.3. Laboratory analysis

For the char samples, standard length and wet weight were measured. Char were dissected to isolate muscle tissue and wet weight of muscle was measured. Then we dried the muscles at 60°C for 2 days. For small fish, the samples included all tissues without dissection step. After drying the samples, we ground them with an agate mortar and pestle. The ground samples were used for the ^{137}Cs activity concentration and stable isotope ratio analyses.

The terrestrial and aquatic prey samples were dried at 60°C for more than 2 days and milled before the ^{137}Cs analysis. Sampled individuals of a given prey type were pooled into one or two samples to obtain sufficient amounts for measuring ^{137}Cs activity concentration.

The ^{137}Cs activity concentrations of the char and prey item samples were determined using gamma-ray spectroscopy based on dry-weight basis. Gamma-ray emissions at an energy level of 661.6 keV were measured using a high-purity germanium coaxial gamma-ray detector system coupled to a multichannel analyzer (GCW2022 coupled to DSA1000, Can-berra, Meriden, CT, USA; Ortec GEM20–70 coupled to DSPEC jr. 2.0, Ametek-AMT, Beijing, China). Gamma-ray peaks of 661.6 keV were used to determine the ^{137}Cs activity concentrations. The energy and efficiency calibrations for this detector were performed using standard and blank (background) samples. Each sample was measured so that there were less than 10% error counts per net area counts. If sample measurements exceeding 3 days did not detect the gamma-ray emission peaks, the ^{137}Cs activity concentrations in the samples were considered to be below the detection limit. All activities were corrected for decay from the sampling dates. Detector systems were examined using the proficiency test by the International Atomic Energy Agency.

Carbon ($\delta^{13}C$) and nitrogen stable isotope ratios ($\delta^{15}N$) were determined to calculate dietary contributions of prey items to the white-spotted char. The white-spotted char and prey item samples were analyzed using an isotope ratio mass spectrometer coupled to an elemental analyzer (Finnigan-MAT252, Thermo Fisher Scientific, Florence, KY, USA). Approximately 1–3 mg of each sample was used for this analysis. The isotope ratios are expressed as $\delta^{13}C$ and $\delta^{15}N$ corresponding to the relative differences per mill (‰) in the $^{13}C/^{12}C$ and $^{15}N/^{14}N$ between samples and standard reference materials. The analytical precision was lower than 0.1‰ for $\delta^{13}C$ and 0.3‰ for $\delta^{15}N$.

4.2.4. Dietary contribution, transfer factor and metabolic rate

The dietary contributions of prey items for each individual white-spotted char were estimated using the Bayesian framework, Stable Isotope Analysis in R ver. 4 (SIAR, V4) (Inger et al., 2006). This approach is based on a mixing model of dietary contributions for a given predator (Phillips, 2001). The model incorporated the $\delta^{13}C$ and $\delta^{15}N$ isotope signatures of predator and prey items to produce upper, mean, mode and lower estimates of dietary contribution of each prey item for a predator (Parnell et al., 2010). Discrimination factors (mean ± standard deviation) in the models were set to 0.47 ± 1.23‰ for $\delta^{13}C$ and 3.41 ± 0.20‰ for $\delta^{15}N$ based on a meta-analysis of freshwater ecosystems (Vander Zanden and Rasmussen, 2001). For each model, I ran 500,000 iterations to assure proper model function. The $\delta^{13}C$ and $\delta^{15}N$ data were used to calculate the 50% credibility intervals of the contribution of each prey item to white-spotted char (Inger et al., 2006). The upper and lower contributions were obtained based on a quantile-based probability interval of $\delta^{13}C$ and $\delta^{15}N$. I used the upper and lower values of dietary contributions in the following estimates.

The TF_{web} is based on the predator–prey relationship in a given trophic link. I used the TF_{web} to measure the extent of ^{137}Cs transferred from lower (prey) to higher (predator) trophic levels. The ^{137}Cs activity concentration in multiple prey items with their respective dietary contributions was calculated as:

$$C_{total\ prey} = \sum_n (C_{prey\ n} \cdot f_{prey\ n}), \qquad (1)$$

where $C_{prey\ n}$ indicates the ^{137}Cs activity concentration of n^{th} prey items and $f_{prey\ n}$ indicates the dietary contributions of n^{th} prey. The mean ^{137}Cs activity concentration of a given prey was used as $C_{prey\ n}$. $f_{prey\ n}$ was estimated from the stable isotope analysis. For this study, number of samples from 1st to 12th prey items (n = 1 to 12) were used and then calculated the upper and lower values of diet-based ^{137}Cs contributions (Bq kg^{-1}) for each season. TF_{web} was calculated as the ^{137}Cs activity concentration in a white-spotted char divided by the ^{137}Cs activity concentration in multiple prey items using the following equation:

$$TF_{\mathrm{web}} = C_{fish}/C_{total\ preys}\ , \qquad (2)$$

where C_{fish} indicates the ^{137}Cs activity concentration in fish. This calculation was conducted for 23 white-spotted char from the Fukushima site and 16 white-spotted char from the Gunma site. $TF_{\mathrm{web}} > 1$ was indicative of bioaccumulation, while $TF_{\mathrm{web}} < 1$ was indicative of biodilution.

In addition, the specific metabolic rate of char (B-Watt/kg) was analyzed using the following equation based on Brown et al. (2004):

$$B = b_0 M^{-\frac{1}{4}} e^{-\frac{E}{kT}}\ , \qquad (3)$$

where b_o is a normalization constant ($e^{18.47}$), M is the body mass (Kg), E is the activation energy (0.63 eV), k is Boltzmann's constant (8.6173×10^{-5} eV K^{-1}) and T is the absolute temperature (K). The mean water temperature during the sampling dates in each season was used in this calculation. Then, Watt/kg was converted to kJ/hr/kg by multiplying 3.6 as J (Joule) is more informative for metabolic rate.

One-way analyses of variance (ANOVAs) were conducted to test the significance of seasonal differences in TF_{web} values (upper, mean and lower), and dietary-based ^{137}Cs contributions (terrestrial, aquatic and total prey items) at the Fukushima and Gunma sites. When a significant difference was detected, I conducted a Fisher's multiple comparison test to identify the specific season. All of the analyses were conducted using R ver. 3.1.2 (R Development Core Team, 2014).

4.3. Results

4.3.1. ^{137}Cs activity concentrations in white-spotted char and prey

The ^{137}Cs activity concentrations of white-spotted char (*S. leucomaenis*) at the Fukushima site were about four times greater in winter than those of a similar size class at the

Gunma site (Table 4.1). At the Fukushima site, the ^{137}Cs activity concentration in white-spotted char in summer samples ranged from 1365 to 2836 (mean ± SD: 1986 ± 631) Bq kg^{-1}-dry, and from 1674 to 3057 (mean ± SD: 2131 ± 545) Bq kg^{-1}-dry in winter. At the Gunma site, the ^{137}Cs activity concentration in white-spotted char ranged from 193 to 441 (mean ± SD: 318 ± 105) Bq kg^{-1}-dry in summer, and from 367 to 618 (mean ± SD: 516 ± 96) Bq kg^{-1}-dry in winter. The ^{137}Cs activity concentrations of the Fukushima and Gunma site samples differed more in winter than summer. The ^{137}Cs activity concentrations at the Fukushima site did not differ significantly among seasons, while those at the Gunma site differed significantly and the highest values were observed in winter based on the multiple comparisons test ($p < 0.05$).

Table 4.1. Summary of the number of samples by season, standard length, weight, ^{137}Cs activity concentration, specific metabolic rate, TF_{web} (upper, mean and lower) of white-spotted char in Fukushima and Gunma in 2012 and 2013.

Study site	Season	Number of char (n)	Standard length (mm)	Weight (g)	^{137}Cs (Bq kg^{-1}-dry)	Specific metabolic rate (kJ/hr/kg)	Food-web based transfer factor (TF_{web})		
							Upper	Mean	Lower
			Mean ± SD	Mean ± SD	Mean ± SD	Mean ± SD	Mean ± SD	Mean ± SD	Mean ± SD
Fukushima	Summer	4	95 ± 28	17 ± 14	1986 ± 631	0.013 ± 0.003	1.89 ± 0.61	3.93 ± 1.30	5.98 ± 2.00
	Autumn	6	121 ± 33	30 ± 26	1827 ± 822	0.005 ± 0.001	1.75 ± 0.82	3.50 ± 1.68	5.26 ± 2.55
	Winter	5	111 ± 45	26 ± 28	2131 ± 545	0.004 ± 0.001	2.89 ± 0.70	6.51 ± 1.45	10.12 ± 2.27
	Spring	8	101 ± 44	20 ± 28	2073 ± 1649	0.007 ± 0.002	2.55 ± 1.97	5.81 ± 3.86	9.07 ± 5.76
	Year-round		107 ± 38	24 ± 25	2006 ± 1068	0.007 ± 0.004	2.30 ± 1.32	5.03 ± 2.75	7.77 ± 4.21
Gunma	Summer	4	135 ± 35	30 ± 35	318 ± 105	0.013 ± 0.002	1.30 ± 0.44	2.62 ± 0.89	3.94 ± 1.33
	Autumn	3	126 ± 20	26 ± 12	424 ± 121	0.005 ± 0.001	1.65 ± 0.50	3.58 ± 0.61	5.51 ± 0.83
	Winter	5	115 ± 27	21 ± 18	516 ± 96	0.003 ± 0.001	3.45 ± 0.74	7.51 ± 1.64	11.56 ± 2.57
	Spring	4	124 ± 21	32 ± 12	306 ± 74	0.006 ± 0.001	1.46 ± 0.34	3.01 ± 0.62	4.56 ± 0.90
	Year-round		124 ± 26	27 ± 17	397 ± 128	0.007 ± 0.004	2.08 ± 1.08	4.43 ± 2.39	6.77 ± 3.71

Among the collected prey items at both sites, four taxa were from terrestrial habitats and eight taxa were from aquatic ones (Table 4.2). The ^{137}Cs activity concentrations in the terrestrial prey at both study sites (Fukushima site, mean ± SD: 2225 ± 1400 Bq kg^{-1}-dry; Gunma site, mean ± SD: 356 ± 157 Bq kg^{-1}-dry) tended to be higher than those of the aquatic prey (Fukushima site, mean ± SD: 1096 ± 1079 Bq kg^{-1}-dry; Gunma site, mean ± SD: 301 ± 276 Bq kg^{-1}-dry). The lowest ^{137}Cs activity concentration was observed in stonefly (*Paragnetina* sp.), and the highest was observed in spider crickets (Rhaphidosphoridae gen. spp.) in summer samples at the Fukushima site. At the Gunma site, the lowest ^{137}Cs activity concentration was observed in dragonflies (*Anotogaster sieboldii*), and the highest was observed in mayflies (*Ephemera japonica*) in spring. Terrestrial prey items tended to be more abundant in summer at both study sites (Table 4.2). Among these prey items, freshwater crabs (*Geothelphusa dehaani*) were found in all seasons except winter, while ground beetles (*Carabus* sp.) were found only in summer at both study sites. Similarly, among aquatic prey, dragonflies (Gomphidae gen. spp.) were observed in all seasons at both sites, while mayflies (*E. japonica*) were found at Gunma site in all seasons.

Table 4.2. Summary of seasonal ^{137}Cs activity concentrations (Bq kg^{-1}-dry) of prey items in Fukushima and Gunma in 2012 and 2013.

Ecosystem	English Name	Taxon	Fukushima					Gunma				
			Year-round	Season				Year-round	Season			
			Mean ± SD	Summer	Autumn	Winter	Spring	Mean ± SD	Summer	Autumn	Winter	Spring
Terrestrial	Freswater crab	*Geothelphusa dehaani*	1634 ± 746	2843	1511* (2)	n.a.	1152* (2)	417 ± 235	649	179	n.a.	422
	Web-building spider	Tetragnathidae gen. spp.	1167	1167	n.a.	n.a.	n.a.	224	224	n.a.	n.a.	n.a.
	Spider cricket	Rhaphidosphoridae gen. spp.	3546 ± 1403	4246* (3)	1447	n.a.	n.a.	387 ± 99	403* (3)	243	n.a.	481
	Ground beetle	*Carabus* sp.	957	957	n.a.	n.a.	n.a.	158	158	n.a.	n.a.	n.a.
Aquatic	Dragonfly	*Anotogaster sieboldii*	632 ± 178	687	432	n.a.	776	12	n.a.	n.a.	n.a.	12
	Dragonfly	Gomphidae gen. spp.	778 ± 433	1529	514* (2)	546* (2)	1015	250 ± 134	126* (2)	435	331	233
	Damselfly	*Mnais costalis*	470	n.a.	470	n.a.	n.a.	74	n.a.	74	n.a.	n.a.
	Mayfly	*Ephemera japonica*	3592 ± 1006	n.a.	n.a.	2880	4304	658 ± 206	372	750	658	850
	Stonefly	*Kamimuria* sp.	288	n.a.	n.a.	n.a.	288	39	n.a.	n.a.	n.a.	39
	Stonefly	Perlinae gen. spp.	476	n.a.	n.a.	476	n.a.	n.d.	n.d.	n.d.	n.d.	n.d.
	Stonefly	*Paragnetina* sp.	247	247	n.a.	n.a.	n.a.	n.d.	n.d.	n.d.	n.d.	n.d.
	Cranefly	*Tipula* sp.	1702 ± 180	n.a.	1574	1829	n.d.	405 ± 126	n.a.	315	494	n.a.

Note: n.a.: no available sample
n.d.: not detected
multiple samples for ^{137}Cs analysis is indicated as * (asterisk mark)
number in brackets shows replication

4.3.2. Dietary contributions of prey items and food web-based transfer factor

The dietary contributions of prey items to white-spotted char (*S. leucomaenis*) were similar between the Fukushima and Gunma sites in all seasons. Significant seasonal differences were observed in the diet-based contributions of both terrestrial and aquatic prey items at the Fukushima site ($p < 0.01$). The mean and lower total diet-based ^{137}Cs contributions of all prey items per white-spotted char were highest in autumn and lowest in winter, while the highest and lowest contributions were found in spring and winter, respectively in case of upper total diet-based ^{137}Cs contributions (Figure 4.2a). Similarly, significant seasonal differences were observed for both terrestrial and aquatic diet-based ^{137}Cs contributions ($p < 0.01$) but not for the lower dietary-based ^{137}Cs contributions of aquatic prey items at the Gunma site. The contributions of terrestrial items to diet-based ^{137}Cs contributions were up to 62% for the lower case and 57% for upper case of total prey, with a maximum in autumn (mean ± SD: 2,537 ± 18 Bq kg^{-1}-dry) and minimum in winter (mean ± SD: 0 ± 0 Bq kg^{-1}-dry) at the Fukushima site (Figure 4.2a). Total dietary-based ^{137}Cs contributions of all prey items at the Gunma site were high in summer (324–982 Bq kg^{-1}-dry) and low in winter (225–751 Bq kg^{-1}-dry) (Figure 4.2b). Contributions of terrestrial prey items to total diet-based ^{137}Cs contributions were 61–63%, with a maximum in summer at the Gunma site (Figure 4.2b).

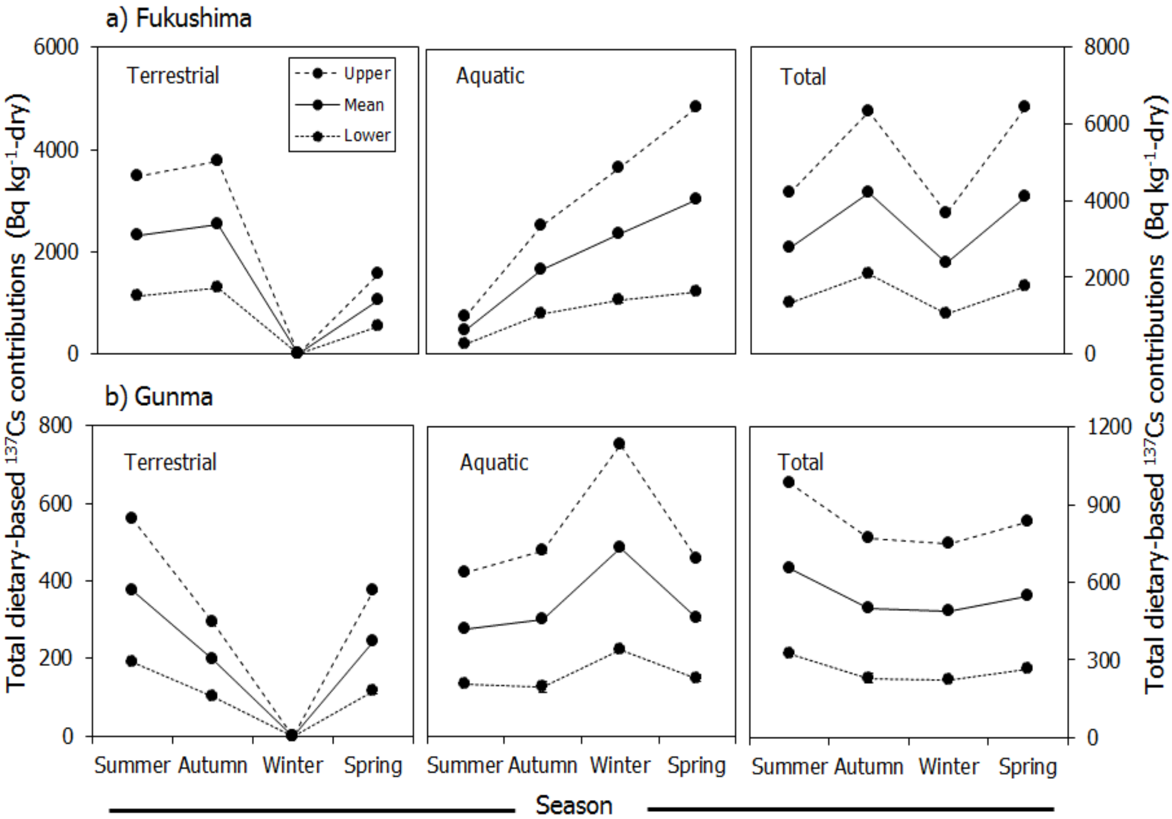


Figure 4.2. Seasonal variations of the total dietary-based ^{137}Cs contributions of terrestrial, aquatic and total prey items in a) Fukushima and b) Gunma. Left side of Y-axis showed the values of terrestrial and aquatic, while right side of Y-axis showed the values of total prey items.

All seasonal TF_{web} values at the Fukushima and Gunma sites were > 1, indicative of bioaccumulation (Table 4.1). At the Fukushima site, the mean upper and lower TF_{web} values varied from 2.30 ± 1.32 to 7.77 ± 4.21, respectively. There were no significant seasonal differences in the upper, mean and lower TF_{web} values at the Fukushima site. Similarly, at the Gunma site, the mean upper and lower TF_{web} values ranged from 2.08 ± 1.08 to 6.77 ± 3.71, respectively. TF_{web} values at the Gunma site revealed significant differences among seasons ($p < 0.01$). The upper, mean and lower TF_{web} values (3.45, 7.51 and 11.56, respectively) were significantly higher in winter at the Gunma site based on the multiple comparison test (Table 4.1 and Figure 4.3a and b).

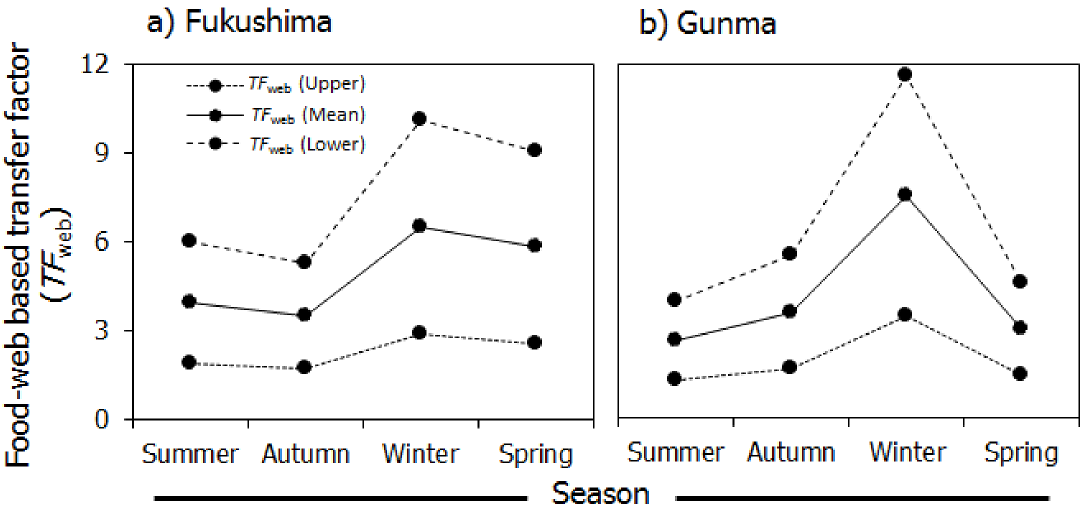


Figure 4.3. Seasonal variation of TF_{web} in a) Fukushima and b) Gunma.

Significant differences in the specific metabolic rate of white-spotted char were found according to sampling season at both sites ($p < 0.01$); however, these values were similar between sites, 0.003–0.016 kJ/hr/kg at the Fukushima site and 0.002–0.014 kJ/hr/kg at the Gunma site (Table 4.1). Significant relationships between the mean TF_{web} and specific metabolic rate of white-spotted char were observed at both the Fukushima ($p = 0.05$) and Gunma ($p < 0.01$) sites. The mean TF_{web} tended to be lower with a high specific metabolic rate for white-spotted char at both study sites (Figure 4.4).

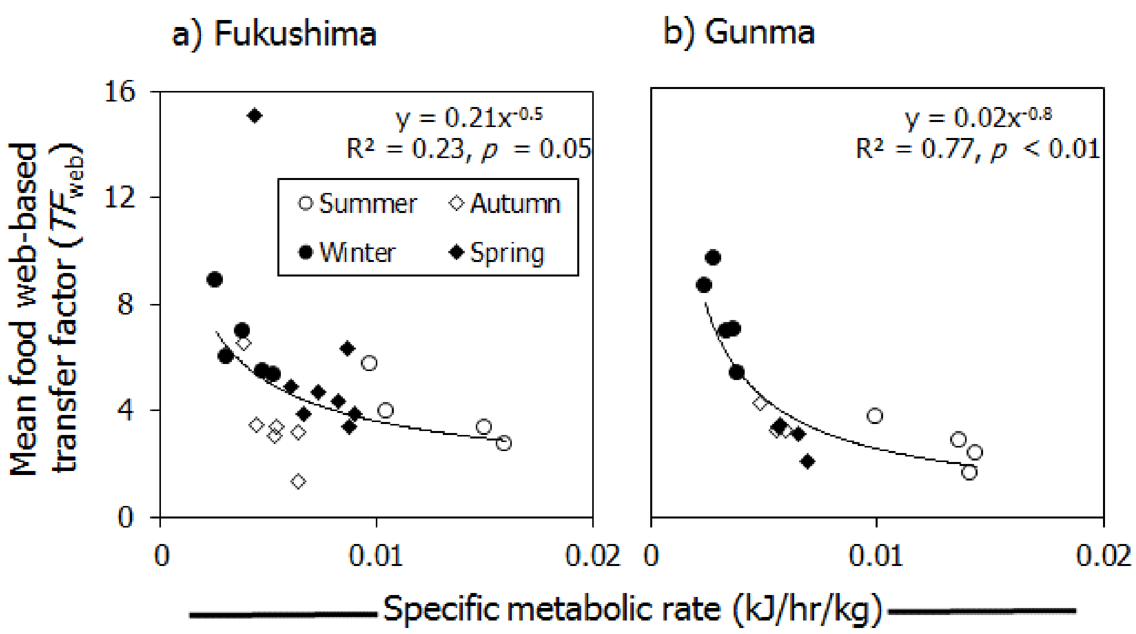


Figure 4.4. Relationship between mean TF_{web} and specific metabolic rate of white-spotted char in a) Fukushima and b) Gunma.

4.4. Discussion

The ^{137}Cs transfer factors from prey to white-spotted char (*S. leucomaenis*) were indicative of bioaccumulation at both the Fukushima and Gunma sites in all seasons. Therefore, the persistence of seasonal differences in TF_{web} with the highest TF_{web} values in winter might be associated with food consumption patterns of intake and excretion of ^{137}Cs (Figure 4.3). Terrestrial prey items were more prevalent in summer than winter, which is in agreement with previous researches, including Nakano et al. (1999) and Kawaguchi and Nakano (2001). Based on gut contents, the white-spotted char mostly consumed terrestrial prey items at the Fukushima site, although their diet included both terrestrial and aquatic prey (Table 4.2). In addition, the ^{137}Cs activity concentrations of terrestrial prey were greater than those of aquatic prey (Sakai et al. 2016b), because forest ecosystems are more contaminated than aquatic ecosystems. For instance, the mean ^{137}Cs activity concentration of spider crickets (Raphidosphoridae gen. spp.) was about six times higher than that of dragonflies (*A. sieboldii*) and damselflies (*Mnais costalis*) at the Fukushima site. Moreover, terrestrial prey ^{137}Cs activity concentrations were two times greater than concentrations in aquatic prey at the Gunma site (Table 4.2). Such differences in ^{137}Cs activity concentrations between terrestrial and aquatic biota are associated with a decline in ^{137}Cs activity concentrations in leaf litter due to leaching (Sakai et al. 2015).

Seasonal differences in macroinvertebrate assemblage structures possibly contributed to the differences in TF_{web} values. For instance, freshwater crabs (*G. dehaani*) were absent in winter at both sites. Freshwater crabs generally shelter by digging burrows on the forest floor (Minei, 1968) or remaining between holes between sediment and boulders near stream channels during winter (Shimotsukasa and Wada, 1995). In contrast, I obtained a high biomass of ground beetles (*Carabus* sp.) in summer: 957 Bq kg^{-1}-dry at the Fukushima site and 158 Bq kg^{-1}-dry at the Gunma site. Spider crickets (Rhaphidosphoridae gen. spp.) tend to be important food sources, particularly in summer (Sato et al., 2011). Regarding aquatic insects, dragonflies (Gomphidae gen. spp.) are much more abundant and have a higher biomass in summer than winter. In contrast, mayflies (*E. japonica*) occur throughout the year at the Gunma site and are an important food source for char. The results of this study are in agreement with a previous report that larvae of mayfly (*E. japonica*) remain in the water column throughout the year (Koyabashi and Kagaya, 2002).

However, I didn't found mayflies (*E. japonica*) in summer and autumn at the Fukushima sites which possibly related to the life cycles of mayflies. At the Fukushima site, the samplings were done little lately (23-25 August [summer 2012], and 20-22 November [autumn 2012]) compare to those of the Gunma site in summer (16-18 August, 2012) and autumn (26-27 November, 2012). These differences of sampling periods possibly associated with the abundance and/or presence of mayflies. For instances, Kuroda et al. (1984) showed that nymphs of mayflies (*E. japonica*) were absent in late summer (late August and September) in the streams because mayflies emerge out as adults in late summer. The absence of mayflies (*E. japonica*) in autumn at the Fukushima site also possibly be associated with the gaps of life cycles among the generations which varied depending on the locations of streams (Watanabe and Kuroda, 1985). In addition, litter retention in the different units of stream channels (e.g., pools and riffles) in association with the hydraulic conditions of headwater streams were corresponded to the macroinvertebrates assemblage including mayflies (Koyabashi and Kagaya, 2002). Moreover, data of the Gunma site in this chapter (Table 4.2) and those of the Fukushima site in next chapter (Table 5.2) of this thesis showed that ^{137}Cs activity concentrations in mayflies (*E. japonica*) were relatively lower in summer seasons; therefore I think that as the seasonal TF_{web} was developed by considering the total dietary ^{137}Cs contributions from all prey items in each season, and mayflies (*E. japonica*) of summer and autumn tended to had lower ^{137}Cs contamination levels which possibly have no remarkable effects on seasonal TF_{web} at the Fukushima site. Indeed, to clarify this phenomena further studies are need to be conducted in future.

The ^{137}Cs consumed from food sources by white-spotted char is generally eliminated, although some ^{137}Cs can be stored in muscle tissue (Yamamoto et al., 2014). The biological half-life of ^{137}Cs in freshwater salmonids varies from 138 to 636 days (Forseth et al., 1998). For instance, the half-lives in brown trout (*Salmo trutta*) and Arctic char (*Salvelinus alpinus*) are 138 and 148 days, respectively (Forseth et al., 1998). Because biological half-life is related to metabolic rate, ^{137}Cs can remain longer in the white-spotted char body during winter than in summer (Forseth et al., 1991). Given that the metabolic rate during winter was about 60% lower than in summer at both sites, this may have caused the higher TF_{web} values during winter. Gillooly et al. (2011) found similar seasonal patterns due to the temperature and body mass of fish under laboratory conditions in the United States.

The different seasonal variation in TF_{web} between the two sites was associated with different concentration gradients between food intake and excretion. The landscape and stream characteristics of the headwater streams at the sites were identical. Based on Sakai et al. (2016b), Japanese cedar litter is the basal energy source of the food web of both of these stream ecosystems. Furthermore, Sakai et al. (2016a) also showed that cedar litter was the vital food sources for stream macroinvertebrates including detritivores by studying in coniferous forest with dominant Japanese cedar plantation. In addition, the estimated metabolic rates of the white-spotted char were similar based on similar stream temperature patterns. Therefore, because the concentration of consumed ^{137}Cs was much higher at the Fukushima site than at the Gunma site, ^{137}Cs remained in the white-spotted char body longer, resulting in higher concentrations at the Fukushima site. ^{137}Cs intake tended to be greater than excreted ^{137}Cs in fish from the Fukushima site (Figure 4.5). Conversely, ^{137}Cs excretion via metabolism at the Gunma site reflected the significant seasonal patterns of ^{137}Cs

in white-spotted char and the resulting TF_{web} patterns. At the site with low contamination levels, equilibrated ^{137}Cs intake and excretion in white-spotted char possibly drove seasonal patterns associated with physiological conditions of the fish, while an imbalance between ^{137}Cs intake and excretion at the highly contaminated site masked these seasonal patterns (Figure 4.6).

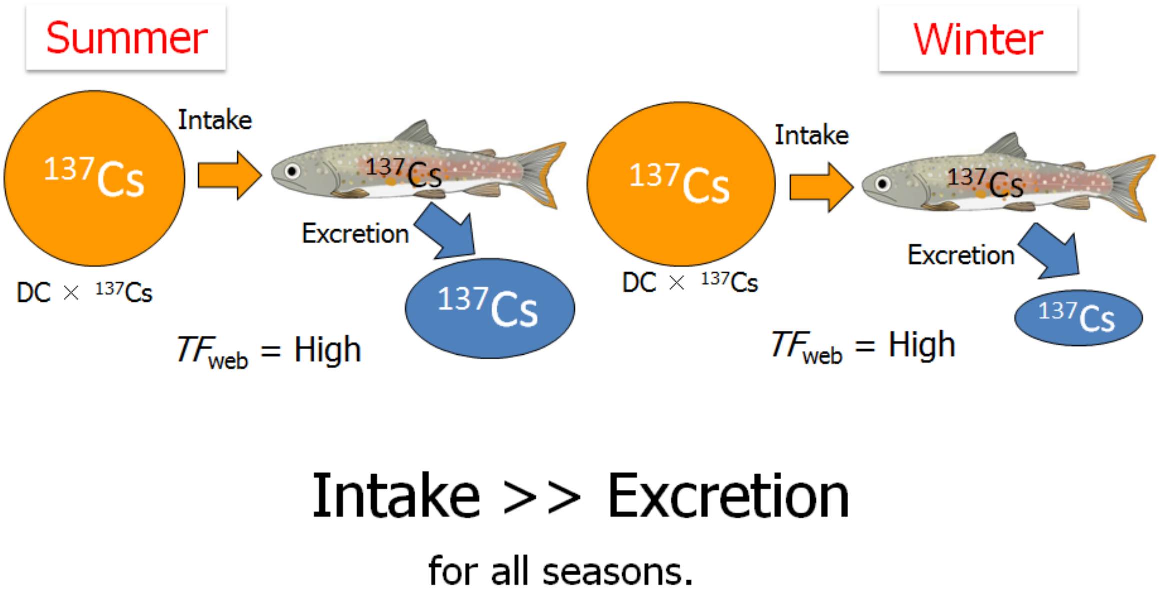


Figure 4.5. Schematic explanation for non-significant seasonal patterns of TF_{web} in Fukushima.

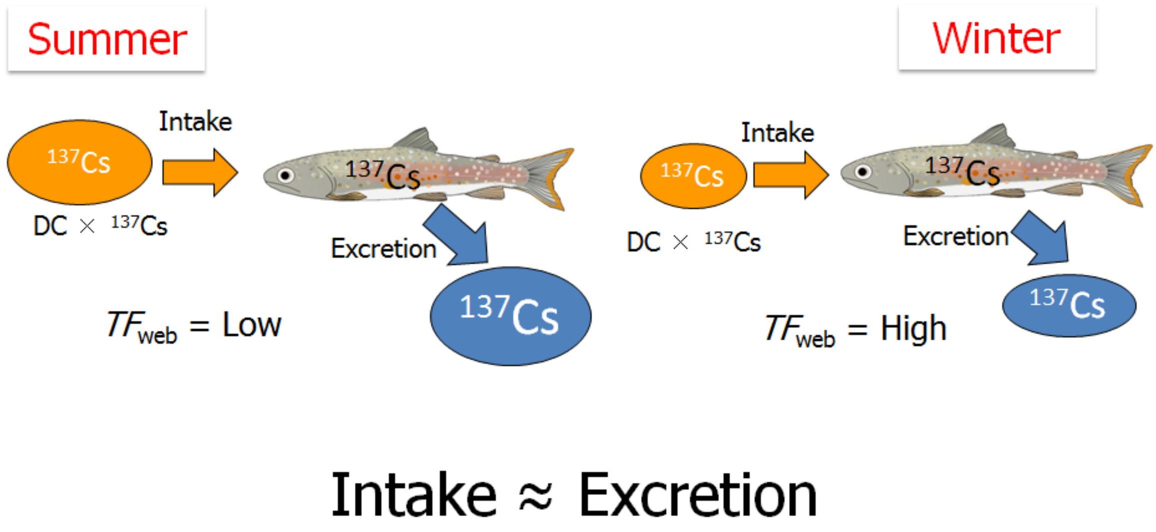


Figure 4.6. Schematic explanation for significant seasonal patterns of TF_{web} in Gunma.

4.5. Summary and conclusions

The TF_{web} in white-spotted char (*S. leucomaenis*) from headwater streams in four seasons exhibited bioaccumulation from prey to char at the Fukushima and Gunma sites even having different amount of contamination levels. High ^{137}Cs levels from prey were provided by terrestrial food sources because of their higher contamination levels (Murakami et al. 2014; Sakai et al. 2016b). These results emphasize the effects of various factors on the TF_{web} in char from the two study sites. Understanding the seasonal variation and factors affecting TF_{web} in freshwater fish is necessary to elucidate the dominant processes that control bioaccumulation and biodilution in food webs. ^{137}Cs can remain continuously in the white-spotted char body throughout the year in highly contaminated areas of Fukushima, because of differences in ^{137}Cs intake and excretion rates. The approaches and results of this study will help to identify factors that affect variation in TF_{web} in process-based modelling for predicting long-term patterns of ^{137}Cs transfer in complex ecosystems.

CHAPTER 5

EXAMINING THE ^{137}Cs ACTIVITY CONCENTRATIONS IN WHITE-SPOTTED CHAR AFTER 1 AND 5 YEARS OF THE FUKUSHIMA ACCIDENT

5.1. Introduction

Forest ecosystems were severely contaminated by the nuclear plant accidents such as the Chernobyl and the FDNPP accidents. Forest occupies 30-40% of the contaminated areas around the Chernobyl reactor, and 60-90% of the total deposition were intercepted by the tree foliage (IAEA, 2001). In Fukushima, forests extend over 70% land areas, and 70-80% of total radioactive materials were deposited in the forested areas (Hashimoto et al., 2012). After the Chernobyl accident, leaf litter is considered as most contaminated part of the forest ecosystems, as 45-90% forest contamination were concentrated in litter. All initially deposited ^{137}Cs reside in upper layer of soil (i.e., 5-8 cm) as bioavailable form, and still continues to a major potential sources for ^{137}Cs transfer into biota (IAEA, 2001). Similarly, in Fukushima, ^{137}Cs remain in the litter and upper soil layers of the forest floor as a dominant source of contamination to the biota (Hashimoto et al., 2013; Sakai et al., 2016b). Understanding temporal changes of ^{137}Cs activity concentrations in forest components (such as litter, soil etc.) is important because contamination of such media alter the contamination levels of biota (Sakai et al., 2016b).

Knowledge regarding the decline of ^{137}Cs activity concentrations within terrestrial components is critical because the contaminated materials could be mobilized by ecosystem processes in forested catchments (Tikhomirov et al., 1993; Sakai et al., 2016b). The decline in ^{137}Cs mobility and bioavailability over the first few years after any contamination events is controlled by slow diffusion of ^{137}Cs into media (soil, water) which determine the contamination levels in terrestrial biota (Smith et al., 2000b). Temporal decrease of ^{137}Cs activity concentrations within natural ecosystems is quantified by means of ecological half-life (T_{eco}) (Hessen et al., 2000; Peles et al., 2002b). T_{eco} is defined as the amount of time needed for the ^{137}Cs in an ecosystem or one of its components to decrease by 50% as a result of physical, chemical and biological processes (Paller et al., 1999; Peles et al., 2002b; Wada et al., 2016). ^{137}Cs decline rates within various ecosystem components has become important for evaluating the length and severity of potential risks to resident species, and for the need of remediation efforts within contaminated ecosystems (Paller et al., 1999; Peles et al., 2002c). Thus, knowledge regarding decay of ^{137}Cs in terrestrial components relative to other ecosystem components is necessary to fully understand the long-term changes of ^{137}Cs activity concentrations within all components of a given contaminated ecosystem (Peles et al., 2002c).

Decay of terrestrial components is differed than those of aquatic ecosystems possibly because of time lag that required for transfer of contamination from forests to aquatic environments for the availability of contamination to the exposure of the biota in aquatic ecosystems. The variability of long-term behaviour of ^{137}Cs in aquatic ecosystems are much more pronounced than forested components because of higher organic matter contents, lower pH, and lower potassium contents (Pröhl et al., 2006). Decay processes in aquatic ecosystems also varied depending on the size, volume, and physiochemical properties of habitats. For instance, Hessen et al. (2000) showed that T_{eco} of brown trout (*Salmo trutta*) was relatively longer in large shallow lakes compare to small deep lakes by studying in the lakes of Norway after the Chernobyl accident. Peller et al. (1999) showed that the ^{137}Cs decline rate was much faster in biota of lotic ecosystems compared to the lentic habitats. T_{eco} of upstream were relatively shorter than those of downstream because of shorter water retention time (Paller et al., 1999). However, T_{eco} is not consistent after any contamination event that emphasize what is monitored in natural environmental conditions in a particular situation because it is strictly depends on the site-specific characteristics, observation periods, and input-output of contamination sources with other ecosystems components (Paller et al., 1999; Wada et al., 2016).

Headwater streams are characterized by strong linkages among hydrologic, geomorphic and biological processes from hillslopes to stream channels, and from terrestrial and aquatic ecosystems (Gomi et al., 2002). They are considered as the headmost area, abundant and unique components within channel networks (Meyer et al., 2007). These streams are also crucial for nutrient dynamics in riparian ecosystems, and provide habitats for macroinvertebrates, fish including salmonid species, and other aquatic fauna within the watersheds (Meyer and Wallace, 2001; Nakano and Murakami, 2001; Sakai et al., 2016b). White-spotted char (*Salvelinus leucomaenis*) are considered as top predator in such headwater streams (Nakano and Murakami, 2001; Sakai et al., 2016b). Char consume multiple food sources from both terrestrial and aquatic ecosystems (Nakano and Murakami, 2001; Haque et al., 2017b). They also changed their food consumption patterns of intake and excretion from terrestrial to aquatic food sources across seasons (Nakano et al., 1999; Sato et al., 2011; Haque et al., 2017b). Char consume more terrestrial food sources in summer than aquatic ones because of higher availability in forested headwater streams (Kawaguchi and Nakano, 2001; Nakano and Murakami, 2001).

Dietary ^{137}Cs contributions from terrestrial and aquatic food sources to char differed by their different contamination levels (Haque et al., 2017b) because ^{137}Cs activity concentrations in terrestrial food sources were 4 times greater than those of aquatic ones in headwater stream (Sakai et al., 2016b). Differences of ^{137}Cs contamination levels in such food sources due to different fallout inventories altered seasonal patterns in the contamination levels of char (Haque et al., 2017b). This finding implies that seasonal pattern in contamination levels of char possibly occur after reduction of ^{137}Cs contamination levels in terrestrial environments. Moreover, terrestrial and aquatic food sources have different decay processes (Pröhl et al., 2006) which may affect the ^{137}Cs contamination levels in char.

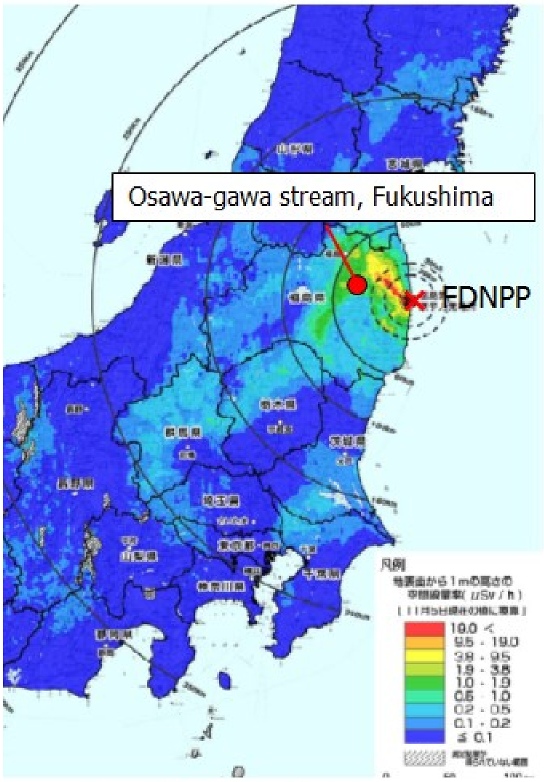
Since 6 years have passed after the FDNPP accident, very little is known how the ^{137}Cs activity concentrations in char of headwater streams change with time, and do the ^{137}Cs activity concentrations in char differ across seasons with dependence of terrestrial food sources.

Understanding the time-series decline of ^{137}Cs activity concentrations in char will help for prediction of ^{137}Cs in char which is also important for fishery resources and management. Therefore, the objectives of this study were to (i) examine the ^{137}Cs activity concentrations in white-spotted char after 1 and 5 years of the FDNPP accident and (ii) compare the ^{137}Cs activity concentrations in char of summer and autumn samples collected during 2012 and 2016.

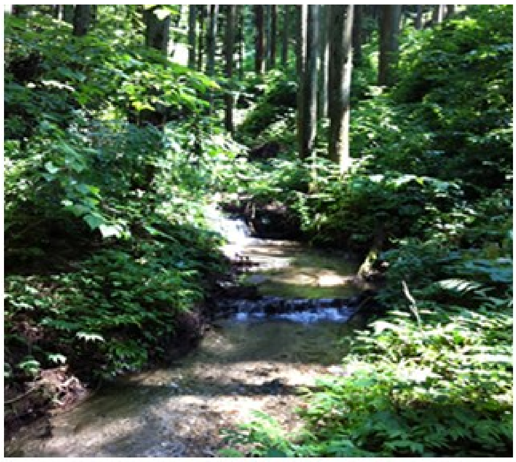
5.2. Methodology

5.2.1. Study site

This study was conducted after 1 and 5 years of the FDNPP accident in the year of 2012 and 2016, respectively in a forested headwater stream draining with Japanese cedar (*Cryptomeria japonica*) plantation forest. The Osawa-gawa stream in Nihonmatsu city, Fukushima prefecture (37°36′N, 140°37′E) is located about 45 km away from the FDNPP (Figure 5.1). The drainage area of the study site is about 170 ha. Based on governmental airborne investigation on June 2012, ^{137}Cs fallout inventory were 100–300 kBq m^{-2} (MEXT, 2012). The mean air dose rates were 1.0–1.9 μSv h^{-1} in 2012, while those in 2016 were 0.2–1.0 μSv h^{-1}. In 2012, the mean annual precipitation were 1248 mm, and mean air temperature was 11°C, while those in 2016 were 1157 mm and 11.6°C, respectively according to the measurement of the nearby (18 km south from the study site) automated weather stations, Funehiki AMeDAS in Fukushima. The underlying geology was lower Cretaceous granite. The dominant overstory vegetation was Japanese cedar (*C. japonica*), while the dominant understory vegetations were Japanese snowball (*Viburnum plicatum*) and silver vine (*Actinidia polygama*). The study site is a typical headwater channel, and 50-m channel segments were selected for collecting char and prey items. The channel segments were consisting of sequences of steps and riffle pools (Montgomery and Buffington, 1997) with 1–5 m channel width, and channel depth of 5–50 cm.



a) Location of study site



b) Overview of Osawa-gawa stream in 2012

c) Overview of Osawa-gawa stream in 2016

Figure 5.1. Map of study site a) Location of study site in Fukushima, and an overview of b) Osawa-gawa stream in 2012 and c) Osawa-gawa stream in 2016

5.2.2. Sample collection

White-spotted char (*S. leucomaenis*) were collected in two consecutive seasons in each sampling year after 1 and 5 years of the FDNPP accident: 23-25 August (summer) and 20-22 November (autumn) in 2012; and 11-13 August (summer) and 26-27 November (autumn) in 2016. Char samples were captured using a backpack electrofisher (LR-24, Smith-Root Inc., Vancouver, WA, USA) by skimming in both pools and riffles within the 50-m channel segment. Six individual white-spotted char were selected for each sampling season (except summer 2012), while 4 individual char were selected only for summer 2012 (23-25 August, 2012). All the selected white-spotted char were brought to the laboratory, and stored at -20°C prior to further processing.

Terrestrial prey items were collected along the stream-riparian zones using bait traps. Fourteen bait traps (diameter: 8 cm, depth: 11 cm), and five buckets (diameter: 29 cm, depth: 24 cm) were installed using beer and lactic acid beverage (1:1 ratio of mixture) as an attractant on the forest floor. Then, I left them for 36 h in each season for collecting enough terrestrial prey items for ^{137}Cs analysis (Sakai et al., 2016b). Samples collected from the bait traps were washed thoroughly using distilled water to remove the attached organic and inorganic matters from their body surfaces.

Aquatic prey items were collected from the pools and riffles using a D-frame net (mesh size: 250 μm) on the stream bed by disturbing the upstream with hands and/or feet. Both the collected terrestrial and aquatic samples were placed into the white plastic trays, and sorted manually using tweezers. All prey items were visually identified to the possible lowest taxonomic levels, and stored in glass vials with respect to each taxonomic group in the field (Sakai et al., 2016b). Both terrestrial and aquatic samples were immediately transported to the laboratory, and stored at -20°C prior to further processing.

Japanese cedar (*C. japonica*) leaf litter from the forest floor were collected which were fully intact, and attached to the twigs, light brown in color, and were not partially buried in the soil matrix (Sakai et al., 2015). Cedar litter were also sampled from the stream those were not black in color which indicated decomposition, submerged in stream channels, and those were considered as basal energy source of the food web in the forested headwater stream (Sakai et al., 2016b).

5.2.3. Sample processing

Standard length and wetted-body weight were measured for the white-spotted char samples. The ages of char were also determined using otoliths following the surface reading method (Brothers, 1987). Muscle tissues were dissected from the individual char sample, and measured wet weight of the muscle tissues. Dissection was avoided for small-sized fish whose standard length were < 100 mm. Muscle tissues and/or fish as a whole (those weren't dissected due to small body size) were dried at 60°C for more than 2 days. After drying the samples, I ground them to a fine powder using agar mortar and pestle; and used these dry powder forms of samples for the measurement of ^{137}Cs activity concentrations.

Terrestrial and aquatic prey items those had sufficient amount were selected for ^{137}Cs analysis because the accurate measurement of ^{137}Cs activity concentrations require sufficient mass of samples (i.e., 1-5 g). The prey items were dried at 60°C for more than 2 days, and ground as the same manner used for muscle tissues of char. Samples that did not have enough mass for ^{137}Cs analysis were pooled into one or two samples to obtain sufficient amount for measuring ^{137}Cs activity concentrations.

Forest and stream litter were also dried at 60ºC for more than 2 days. The litter samples were pulverized using an electrical mill (FM-1; Osaka Chemical Co., Ltd., Osaka, Japan), and then ground samples were used for ^{137}Cs activity measurements.

5.2.4. ^{137}Cs activity concentration measurement

^{137}Cs activity concentrations of all samples were determined using gamma-ray spectroscopy. Gamma-ray emissions at energy level of 661.6 keV were measured using a high-purity germanium coaxial detector system (GCW2022; Canberra, Tokyo, Japan) coupled to a multichannel analyzer (DSA1000, Canberra). The energy and efficiency calibrations for this detector were performed using standard provided by the suppliers and blank (background) samples. For the analysis of ^{137}Cs activity concentrations, each sample was measured for less than 5% error counts/net area counts. ^{137}Cs activity concentration of each sample was corrected for decay according to the sampling dates. ^{137}Cs activity concentrations were calculated based on dry-weight basis (Bq kg^{-1}-dry) for all samples. The detector systems were examined for proficiency following the test established by the International Atomic Energy Agency.

5.2.5. Estimation of ecological half-life for ^{137}Cs in white-spotted char

The decline of ^{137}Cs activity concentrations in white-spotted char from its maximum contamination levels were estimated using the single-component exponential model based on Jonsson et al. (1999) and Wada et al. (2016):

$$A_t = A_0\, e^{-\lambda t} \qquad (1)$$

where A_t and A_o represent the ^{137}Cs activity concentrations of char at time *t* (d) and 0, λ strand for decay rate constant (d^{-1}), and *t* denotes the number of days after the maximum ^{137}Cs activity concentrations of char. This equation allows to calculate ecological half-life (T_{eco}) using the following formula (Jonsson et al., 1999):

$$T_{eco} = \ln 2/\lambda \qquad (2)$$

Due to small sample size, to get a robust estimate of T_{eco}, I used the mean value of ^{137}Cs activity concentrations of white-spotted char in each season from the period 2012 and 2016. Moreover, ^{137}Cs activity concentrations of char of different standard lengths (e.g., 50 mm, 100 mm and 150 mm) in summer and autumn of 2012 and 2016 were calculated using the fitted equations separately those were derived from the relationships between standard lengths and ^{137}Cs activity concentrations in char (Figure 5.3). Afterwards, size-based ^{137}Cs activity concentrations were used to estimate size-based T_{eco}'s for elucidating the relation between body size and T_{eco}'s of char in summer and autumn separately after 5 years of the accident. I referred these size-based T_{eco}'s as $T_{eco\text{-}size}$, and expressed based on the standard lengths of char as $T_{eco\text{-}50}$, $T_{eco\text{-}100}$ and $T_{eco\text{-}150}$ for char having standard length of 50 mm, 100 mm and 150 mm, respectively.

5.3. Results

5.3.1. ^{137}Cs activity concentrations of litter and prey items

^{137}Cs activity concentrations of forest cedar litter in summer 2012 ranged from 30890 to 35996 Bq kg^{-1}-dry (mean ± SD: 33330 ± 2560 Bq kg^{-1}-dry), while those in summer 2016 ranged from 4212 to 8430 Bq kg^{-1}-dry (mean ± SD: 6295 ± 2109 Bq kg^{-1}-dry) (Table 5.1). In autumn 2012, ^{137}Cs activity concentrations of forest cedar litter ranged from 22684 to 31200 Bq kg^{-1}-dry (26655 ±

4287 Bq kg^{-1}-dry), and those in autumn 2016 ranged from 1439 to 5697 Bq kg^{-1}-dry (3969 ± 2240 Bq kg^{-1}-dry). In case of stream litter, ^{137}Cs activity concentrations in summer 2012 ranged from 3959 to 6250 Bq kg^{-1}-dry (4856 ± 1223 Bq kg^{-1}-dry), and those in summer 2016 ranged from 4212 to 8430 Bq kg^{-1}-dry (5149 ± 1247 Bq kg^{-1}-dry). In autumn 2012, ^{137}Cs activity concentrations in litter of stream channels ranged from 6956 to 8496 Bq kg^{-1}-dry (7922 ± 841 Bq kg^{-1}-dry), while those ranged from 884 to 5024 Bq kg^{-1}-dry (2463 ± 2238 Bq kg^{-1}-dry) in autumn 2016 (Table 5.1).

^{137}Cs activity concentrations of terrestrial and aquatic prey items in summer 2012 ranged from 957 to 4246 Bq kg^{-1}-dry (2303 ± 1546 Bq kg^{-1}-dry), and from 247 to 1529 Bq kg^{-1}-dry (821 ± 651 Bq kg^{-1}-dry), respectively while those in summer 2016 ranged from 132 to 695 Bq kg^{-1}-dry (474 ± 301 Bq kg^{-1}-dry), and from 172 to 1010 Bq kg^{-1}-dry (464 ± 448 Bq kg^{-1}-dry), respectively (Table 5.2). In autumn 2012, ^{137}Cs activity concentrations of terrestrial prey items ranged from 1447 to 1511 Bq kg^{-1}-dry (1479 ± 45 Bq kg^{-1}-dry), and those in autumn 2016 ranged from 263 to 990 Bq kg^{-1}-dry (657 ± 367 Bq kg^{-1}-dry). ^{137}Cs activity concentrations of aquatic prey items in autumn 2012 ranged from 230 to 1574 Bq kg^{-1}-dry (644 ± 531 Bq kg^{-1}-dry), and those in autumn 2016 ranged from 71 to 1488 Bq kg^{-1}-dry (406 ± 443 Bq kg^{-1}-dry). ^{137}Cs activity concentrations of terrestrial and aquatic prey items decreased 82% and 29%, respectively in summer, while those in autumn decreased 42% and 47%, respectively after 5 years of the accident (Figure 5.5). Spider cricket (Raphidosphoridae gen. spp.) and freshwater crab (*Geothelphusa dehaani*) were common terrestrial prey items in both study years (Table 5.2). In addition, among the aquatic prey items, dragonflies (*Anotogaster sieboldii*) and cranefly (*Tipula* sp.) were common during the sampling periods in both study years while mayflies (*E. japonica*) were also found in 2016 (Table 5.2). ^{137}Cs activity concentrations of spider crickets decreased about 84% in summer, while those of dragonflies decreased about 51% in autumn after 5 years of the FDNPP fallout (Table 5.2).

Table 5.1. Summary of temporal change of ^{137}Cs activity concentrations (Bq kg^{-1}-dry) in forest and stream litter monitored during 2012 and 2016.

Components	Sampling year							
	2012				2016			
	Summer		Autumn		Summer		Autumn	
	Min - Max	Mean ± SD	Min - Max	Mean ± SD	Min - Max	Mean ± SD	Min - Max	Mean ± SD
Forest litter	30890 - 35996	33330 ± 2560	22684 - 31200	26655 ± 4287	4212 - 8430	6295± 2109	1439 - 5697	3969 ± 2240
Stream litter	3959 - 6250	4856 ± 1223	6956 - 8496	7922 ± 841	3716 - 5986	5149 ± 1247	884 - 5024	2463 ± 2238

Table 5.2. Summary of temporal change of ^{137}Cs activity concentrations (Bq kg^{-1}-dry) in prey items of white-spotted char monitored during 2012 and 2016.

English name	Taxon	Habitat	Sampling year			
			2012		2016	
			Season		Season	
			Summer	Autumn	Summer	Autumn
Freshwater crab	*Geothelphusa dehaani*	Forest	2843	1511[a] (2)	596	263
Ground beetle	*Carabus* sp.	Forest	957	n.a.	132	n.a.
Spider cricket	Rhaphidosphoridae gen. spp.	Forest	4246[a] (3)	1447	695	990
Web-buidling spider	Tetragnathidae gen. spp.	Forest	1167	n.a.	n.a.	n.a.
Wood louse	*Ligidium japonicus*	Forest	n.a.	n.a.	n.a.	718
Caddisfly	*Lepidostoma* sp.	Stream	n.a.	n.a.	1010	n.a.
Caddisfly	Hydrophychidae gen. spp.	Stream	n.a.	n.a.	n.a.	71
Cranefly	*Tipula* sp.	Stream	n.a.	1574	n.a.	556
Dragonfly	*Anotogaster sieboldii*	Stream	687	432	172	210
Dragonfly	*Dabidus nanus*	Stream	n.a.	n.a.	267	218
Damselfly	*Mnais costalis*	Stream	n.a.	470	n.a.	238
Dragonfly	Gomphidae gen. spp.	Stream	1529	514[a] (2)	n.d.	n.d.
Fishing spider	*Dolomedes* sp.	Stream	n.a.	n.a.	n.a	786
Mayfly	*Ephemera japonica*	Stream	n.a.	n.a.	872	1488
Stonefly	*Scopura longa*	Stream	n.a.	n.a.	n.a.	314
Stonefly	*Paragnetina* sp.	Stream	247	n.a.	n.d.	n.a.
Stonefly	*Kamimuria* sp.	Stream	n.a.	n.a.	n.a.	95
Stonefly	Perlodidae gen. spp.	Stream	n.a.	230	n.a.	n.d.
Water bug	*Appasus major*	Stream	n.a.	n.a.	n.a.	80

Number in parenthesis shows replication
n.a. No available sample, n.d. not detected
[a] Multiple samples for ^{137}Cs analysis

5.3.2. ^{137}Cs activity concentrations of white-spotted char

^{137}Cs activity concentrations of white-spotted char (*S. leucomaenis*) in summer and autumn were almost similar after 1 year of the FDNPP accident; while after 5 years of the accident, the ^{137}Cs activity concentrations of char in autumn were about 4 times greater than those of similar size class in summer (Table 5.3). In summer 2012, ^{137}Cs activity concentrations of white-spotted char ranged from 1365 to 2836 Bq kg^{-1}-dry (1986 ± 631 Bq kg^{-1}-dry), and those in autumn 2012 ranged from 704 to 3258 Bq kg^{-1}-dry (1827 ± 822 Bq kg^{-1}-dry) (Table 5.3 and Figure 5.4a). ^{137}Cs activity concentrations of char after 5 years of the accident significantly differed in autumn than summer (p = 0.05). ^{137}Cs activity concentrations of white-spotted char ranged from 66 to 956 Bq kg^{-1}-dry (304 ± 338 Bq kg^{-1}-dry) in summer 2016, and from 215 to 2855 Bq kg^{-1}-dry (1201 ± 949 Bq kg^{-1}-dry) in autumn 2016 (Table 5.3 and Figure 5.4b). ^{137}Cs activity concentrations of char in summer decreased about 85%, while those in autumn decreased 34% after 5 years of the FDNPP (Table 5.3 and Figure 5.2).

The standard lengths of char samples those were sampled after 1 year of the accident ranged from 69 to 124 mm (95 ± 28 mm) in summer, and from 89 to 173 mm (121 ± 33 mm) in autumn (Table 5.3). The body weights ranged from 4.5 to 33.1 g (16.9 ± 14.1 g), and from 10.0 to 77.5 g (30.3 ± 25.9 g) in summer and autumn, respectively. The age of the sampled char ranged from 0 to 3 years in both summer and autumn with mean ± SD of 1.3 ± 1.5 and 2.0 ± 0.9, respectively. After 5 years of the accident in 2016, the standard lengths ranged from 69 to 173 mm (130 ± 36 mm), and from 77 to 186 mm (124 ± 42 mm) in summer and autumn, respectively. The body weights ranged from 6.1 to 67.7 g (37.7 ± 22.8 g) in summer 2016, and from 7.9 to 92.0 g (34.9 ± 31.2 g) in autumn 2016. The age of char ranged from 0 to 3 years in summer and autumn 2016 with mean ± SD of 1.5 ± 1.2 years and 2.0 ± 1.2 years, respectively (Table 5.3).

Table 5.3. Summary of numbers, standard lengths, weights, ages and ^{137}Cs activity concentrations of white-spotted char monitored in summer and autumn during 2012 and 2016.

Season	Sample no	Sampling year							
		2012				2016			
		Standard length (mm)	Weight (g)	Age (y)	^{137}Cs (Bq kg^{-1}-dry)	Standard length (mm)	Weight (g)	Age (y)	^{137}Cs (Bq kg^{-1}-dry)
Summer	1	124	33.1	2	2836	69	6.1	0	80
	2	112	24.5	3	2044	128	29.7	2	144
	3	69	5.7	0	1699	115	23.7	0	66
	4	73	4.5	0	1365	136	40.9	2	375
	5	–	–	–	–	158	58.3	2	203
	6	–	–	–	–	173	67.7	3	956
Autumn	1	93	10.0	1	704	186	92.0	3	2855
	2	89	10.3	1	1566	152	45.7	2	927
	3	109	20.7	2	1852	131	29.9	2	1341
	4	112	21.1	2	1768	116	24.4	2	1463
	5	147	42.0	3	1814	77	7.9	0	407
	6	173	77.5	3	3258	81	9.7	0	215

5.3.3. Estimated T_{eco}

The estimated T_{eco} for ^{137}Cs of white-spotted char in autumn was more than 4 times greater than that in summer. T_{eco} for ^{137}Cs of char in summer was 1.4 years, while that in autumn was 6.6 years (Table 5.4). $T_{eco\text{-}size}$ of char in summer were lower than those in autumn (Table 5.5). $T_{eco\text{-}50}$, $T_{eco\text{-}100}$ and $T_{eco\text{-}150}$ of char in autumn were 1.9, 2.8 and 5.1 years, respectively; while those in summer were 3-5 times lower than autumn (Table 5.5). The differences of $T_{eco\text{-}size}$ in summer and autumn tended to increase with the increase of body size of char. ^{137}Cs activity concentrations of char in summer after 1 and 5 years of the accident had tended to increase with increase the standard lengths of char ($p > 0.10$) (Figure 5.3a). In contrast, significant relationships were found between ^{137}Cs activity concentrations and standard lengths of white-spotted char in autumn of similar study years ($p < 0.05$) (Figure 5.3b).

Table 5.4. Summary of T_{eco} of white-spotted char in summer and autumn after 5 years of the FDNPP accident.

Season	D	A_0 (Bq kg^{-1}-dry)	A_t (Bq kg^{-1}-dry)	λ (d^{-1})	T_{eco} (y)
Summer	1249	1986	304	1.2×10^{-3}	1.4
Autumn	1267	1827	1201	2.8×10^{-4}	6.6

D, number of days between seasonal sampling period after 1 and 5 years of the accident

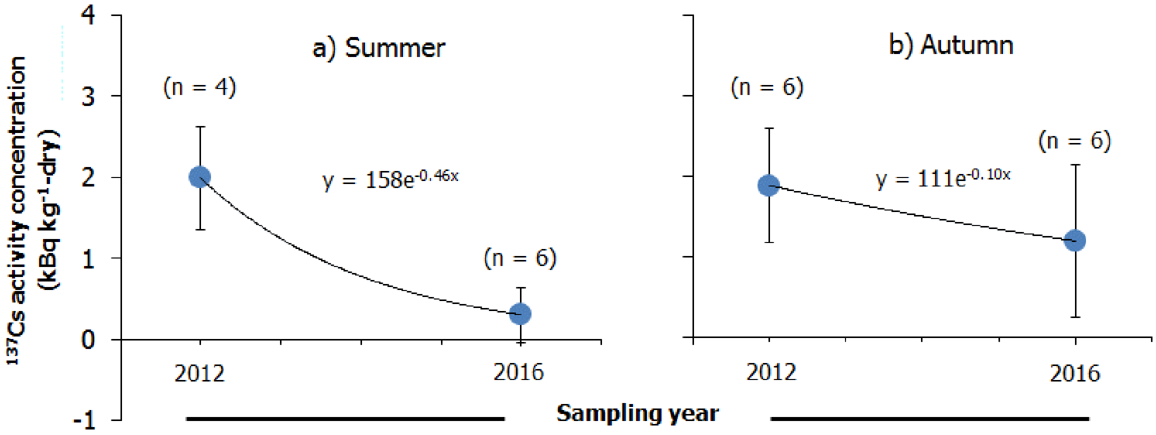


Figure 5.2. Decreasing patterns of ^{137}Cs activity concentrations of white-spotted char in a) summer and b) autumn during 2012 and 2016. Error bars indicate standard deviation (±SD), and n indicated the sample size in each season.

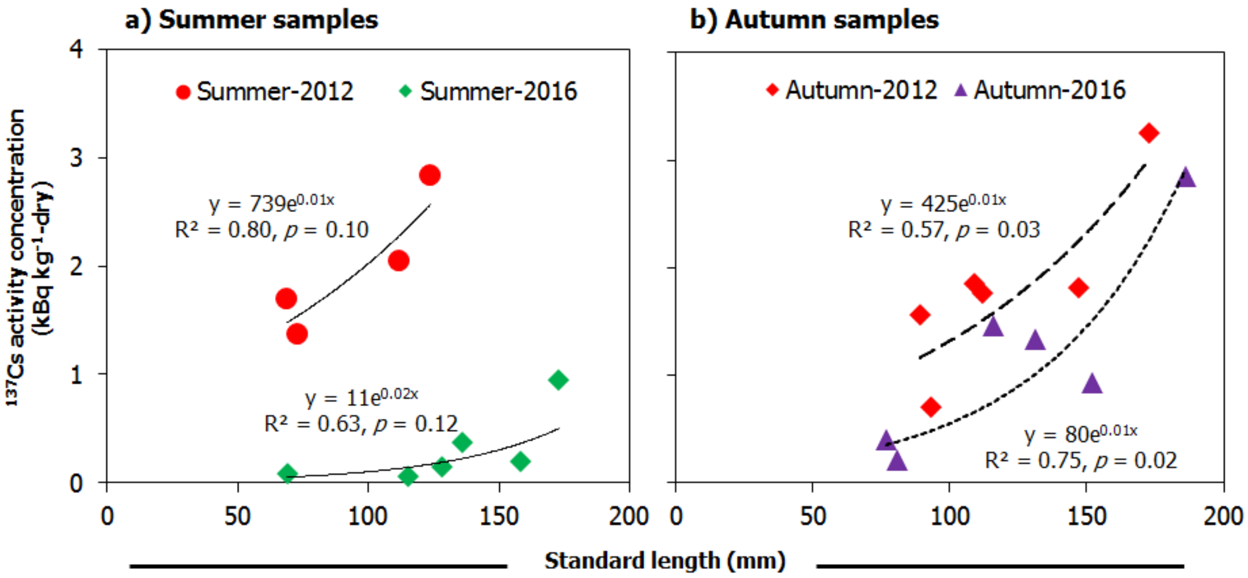


Figure 5.3. Relationship between standard lengths and ^{137}Cs activity concentrations of white-spotted char in a) summer and b) autumn collected in 2012 and 2016

Table 5.5. Summary of $T_{eco\text{-}size}$ of white-spotted char after 1 and 5 years of the FDNPP accident.

Season	D	Standard length of char		
		50 mm	100 mm	150 mm
		$T_{eco\text{-}50}$ (y)	$T_{eco\text{-}100}$ (y)	$T_{eco\text{-}150}$ (y)
Summer	1249	0.6	0.8	1.0
Autumn	1267	1.9	2.8	5.1

D, number of days between seasonal sampling period

5.4. Discussion

5.4.1. Ecological decay of ^{137}Cs in white-spotted char

Ecological decay of ^{137}Cs in white-spotted char was quantified by estimated T_{eco} after 5 years of the FDNPP accident. The result showed that T_{eco} of char ranged from 1.4 to 6.6 years which were considerably shorter than the physical half-life of ^{137}Cs (i.e., 30.2 years). T_{eco} of ^{137}Cs in char were the combined results of interactions among various physical, hydrological, biological and ecological processes that lowered ^{137}Cs contamination levels in given ecosystems (Paller et al., 1999; Wada et al., 2016). However, due to the unique characteristics of decay processes, initial decay of a given species are relatively higher, while slower decay rates are observed in the later phase.

Contamination levels, site-specific characteristics, time after the fallout, and observation periods are possibly alter the T_{eco}. For instance, Jonsson et al. (1999) showed that T_{eco} of brown trout (*Salmo trutta*) was 0.6 years in Norwegian lake with fallout amount of 50 kBq m^{-2} during the first 3 years after the Chernobyl accident because ^{137}Cs initially deposited on the lake surface, and washed out from the catchment before being adsorb or absorb into the soil (Table 5.6). Brittain et al. (1991) showed that T_{eco} of brown trout (*Salmo trutta*) become 3.0 years due to increased input from the catchment at the studied lake Øvre Heimdalsvatn in Norway with 130 kBq m^{-2} fallout inventory during periods from 1986 to 1989 after the Chernobyl accident (Table 5.6).

In contrast, relatively long observation periods prolonged the T_{eco} of a given fish species. For instance, Sundbom et al. (2003) showed that T_{eco} of perch (*Perca fluviatilis*) at lake Ekholmssjön, Sweden with 20-30 kBq m^{-2} fallout inventory after the Chernobyl accident become 14 years during 1996-2000 because of slow decline rate, large individual variations, and long observation periods (Table 5.6). T_{eco} of Arctic char (*Salvelinus alpinus*) in lake of Norway with fallout amount of 50 kBq m^{-2} was 22.4 years during 1989 - 1998 due to slow leaking of ^{137}Cs from the catchment, and recycling within the lake (Jonsson et al., 1999) (Table 5.6). Therefore, longer T_{eco} strongly suggest continuous uptake of ^{137}Cs from contaminated food sources (Wada et al., 2016), resulted higher accumulation within the fish body, and prolonged observation periods (Sundbom et al., 2003).

T_{eco} of char also differed based on the percentage of ^{137}Cs activity concentrations of samples above the detection limit. For instance, T_{eco} in white-spotted char (*Salvelinus leucomaenis*) sampled from the Northern Abukuma River Systems was 1.5 years, while those of the Aga River Systems become nearly 2 times greater (i.e., 2.7 years) during similar observation period from 2011 to 2014 in Fukushima because of underestimation of the decreasing rate constant and/or the overestimation of T_{eco} in char sampled from the Aga River Systems (Table 5.6) (Wada et al., 2016). Moreover, my estimated T_{eco} of char (1.4 to 6.6 years) were longer than the biological half-life of salmonids fish species. For example, the biological half-life of brown trout (*S. trutta*) and Arctic char (*S. alpinus*) were 0.3 and 0.4 years, respectively (Forseth et al., 1998). These results suggest that the main contamination sources of intake of char is contaminated prey items in the headwater stream (Wada et al., 2016), and dietary differences were reflected in the decay rates, and resulted T_{eco} (Hessen et al., 2000).

Table 5.6. Summary table of previous studies related to T_{eco} in fish.

Study site	Source of contaminants	Fallout inventory/air dose rate	Habitat type	Habitat characteristics	Fish species: English name	Fish species: Latin name	T_{eco} (y)	Body length (cm)	Duration	Season	Reference
NABR, Fukushima	FDNPP	1.5 $\mu Sv\ h^{-1}$	River	Attitude - 205 m	White-spotted char	*Salvelinus leucomaenis*	1.5	-	2011-2014		Wada et al., 2016
AGR, Fukushima	FDNNP	0.30 $\mu Sv\ h^{-1}$	River	Altitute - 580 m	White-spotted char	*Salvelinus leucomaenis*	2.7	-	2011-2014		Wada et al., 2016
Lake, Norway	CNPP	>150 $kBq\ m^{-2}$	Lake	-	Brown trout	*Salmo trutta*	1.2-4.2	25-40	1987-1995		Hessen et al., 2000
Devoke water, North-East England	CNPP	10-20 kBq m-2	Lake	Drainage area - 2.4 km^2	Brown trout	*Salmo trutta*	1.6	-	1986-1988		IAEA, 2000
Øvre Heimdalsvatn, Norway	CNPP	130 kBq m-2	Lake	Drainage area - 0.78 km^2	Brown trout	*Salmo trutta*	3.0	-	1986-1990		Brittain, 1991
Steel Creek, SRS	Nuclear reactor, SRS	9.66 × 1013 Bq	Steel Creek	Drainage area - 290 km^2	Largemouth bass	*Micropterus salmoides*	5.4	-	1974-1981		Peles et al., 2000
Lake Ekholmssjon, Sweden	CNPP	20-30 kBq m-2	Lake	Drainage area - 7.8 km^2	Perch	*Perca fluviatilis*	14	6.0-9.6	1996-2000		Sundbom et al., 2003
Lake, Norway	CNPP	50 $kBq\ m^{-2}$	Lake	-	Brown trout	*Salmo trutta*	0.6	-	1986-1889		Jonsson et al., 1999
							7.7	-	1989-1998		
			Lake	-	Arctic char	*Salvelinus alpinus*	1.1	-	1986-1889		
							22.4	-	1989-1998		
This study	FDNPP	100-300 $kBq\ m^{-2}$	Headwater stream	Drainage area - 1.7 km^2	White-spotted char	*Salvelinus leucomaenis*	1.4	6.9-17.3	2012-2016	Summer	
							6.6	7.7-18.6	2012-2016	Autumn	

FDNPP indicates Fukushima Dai-ichi Nuclear Power Plants, CNPP indicates Chernobyl Nuclear Power Plant

Headwater streams are small, steep gradient chanels including hillslopes, zero-order basins, and first and second order channels.

River is the large water body where many streams meet, including six to twelve order channels.

Steel Creek is a small to medium-sized natural stream.

5.4.2. Size depended T_{eco}

Standard length of char altered the ^{137}Cs activity concentrations, and resulted $T_{eco\text{-}size}$ because large-bodied char consume more contaminated terrestrial food sources in summer than the small-bodied char. The relation between the standard lengths and ^{137}Cs activity concentrations of char in summer and autumn samples collected after 1 and 5 years of the accident showed different patterns (Figure 5.3). In general, total food consumption rate increase with the increase of body size in salmonid fishes (Handeland et al., 2008). Large-bodied char are aggressive drift foragers, size-based dominance hierarchies are recognized in the headwater stream, making their own territories, have specific niche, and occupy near the inlet of pool to consume more terrestrial food sources (Nakano and Furukawa-Tanaka, 1999). Large char had the more chance to consume terrestrial prey items in summer with greater biomass because of its good position in food web of headwater streams (Nakano and Furukawa-Tanaka, 1999; Nakano and Murakami et al., 2001). In contrast, small fish tend to consume aquatic prey items even in summer (Haque et al., 2017b) due to interspecific competition; the small char are deprived to consume terrestrial food sources (Nakano and Furukawa-Tanaka, 1999). Moreover, ^{137}Cs activity concentrations in terrestrial prey items decreased about 82% in summer during the period from 2012 to 2016 (Figure 5.5). Therefore, higher decreasing rates of ^{137}Cs activity concentrations of terrestrial food sources, contamination levels of char in summer 2012 and 2016 varied largely in accordance to their body size in headwater stream (Figure 5.3a).

Moreover, char of summer and autumn 2012 showed similar pattern of contamination levels with their body size, while those in 2016 had the differences in their seasonal contamination levels according to the body size (Figure 5.4). These finding is in agreement with previous chapter including Haque et al. (2017b). No difference in contamination levels of char in summer and autumn 2012 because of high fallout volume, while differences in contamination levels with higher values in autumn 2016 possibly due to decrease of habitat contamination levels which was supported by the studies in headwater streams of Fukushima and Gunma prefectures with different fallout inventories (Haque et al., 2017b).

$T_{\text{eco-size}}$ in char tended to increase with the increase of standard lengths of char. Sundbom et al., (2003) showed that the T_{eco} in different fish categories increased with the size effect by studying in Norwegian lakes after the Chernobyl accident. The estimated $T_{\text{eco-size}}$ in char differ in summer and autumn samples possibly because of differences of feeding habits, contamination levels of prey items, life cycles, decline rates, sample sizes across seasons. Long observation periods, and large sample size are needed to consider for understanding more insights about T_{eco}'s, and for prediction of ^{137}Cs in char of forested headwater stream.

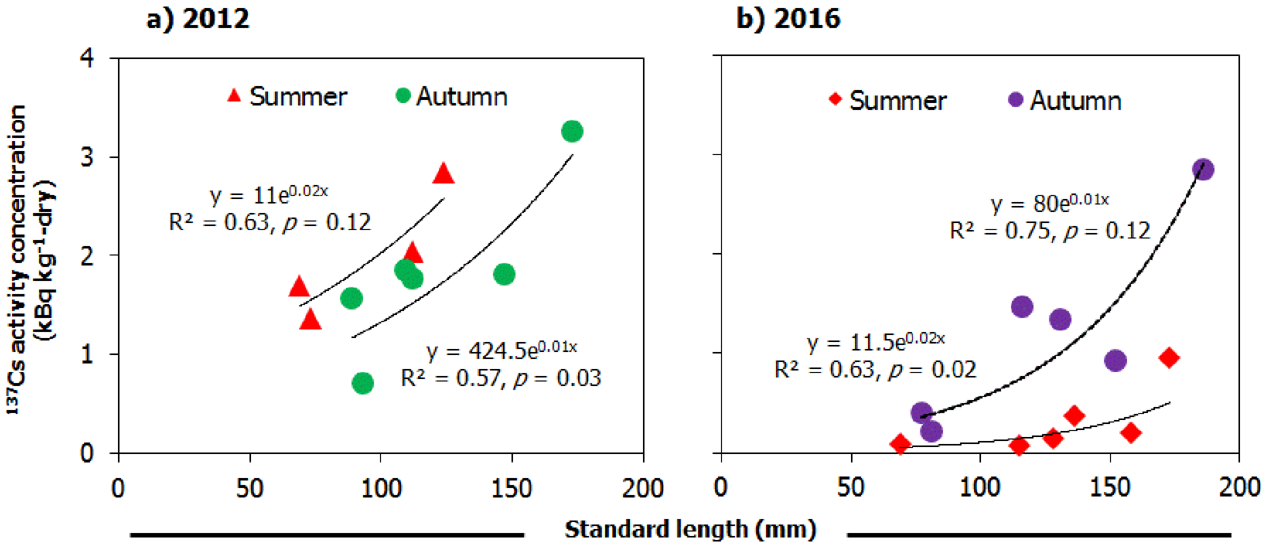


Figure 5.4. Relationship between standard lengths and ^{137}Cs activity concentrations of white-spotted char of Fukushima in summer and autumn in a) 2012 and b) 2016.

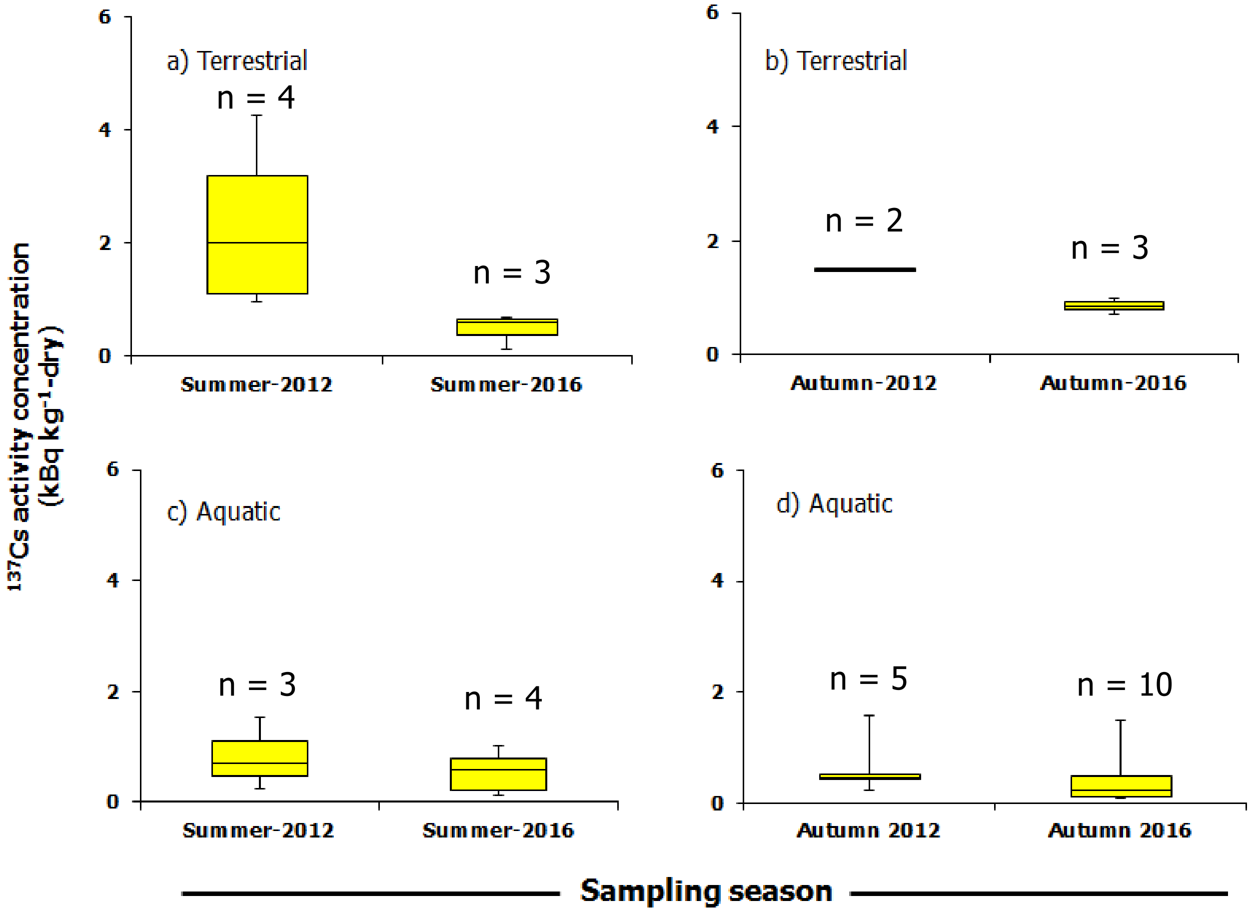


Figure 5.5. ^{137}Cs activity concentrations in prey items a) terrestrial prey items in summer 2012 and 2016, b) terrestrial prey items in autumn 2012 and 2016, c) aquatic prey items in summer 2012 and 2016 and d) aquatic prey items in autumn 2012 and 2016. The lower, middle and upper hinges correspond to the first, second and third quartiles [the 25th, 50th (median), and 75th percentiles]. The whiskers extend from the hinges indicated the highest and lowest values. n indicated the number of samples.

5.4.3. Seasonal T_{eco}

The estimated T_{eco} of white-spotted char showed seasonal variability; lower in summer (1.4 y) and higher in autumn (6.6 y) after 5 years of the FDNPP accident (Table 5.4). The persistence of seasonal variability of T_{eco} in char might be associated with the intake and excretion rate between summer and autumn. Indeed, seasonal variability of T_{eco} depending on the sampling season which is important for char of headwater streams because char is relying on terrestrial and aquatic food sources in riparian ecosystems (Haque et al., 2017b). Furthermore, char have the seasonal dietary preferences in a year. For instance, char mostly consume terrestrial prey items in summer than any other seasons (Nakano et al., 1999; Kawaguchi and Nakano, 2001, Haque et al., 2017b). In addition, terrestrial prey items were highly contaminated because of higher contamination levels in forest ecosystems (Sakai et al., 2016b; Haque et al., 2017b). Terrestrial prey items contributed about 83% in summer and 60% in autumn to the total diet of char based on the dietary ^{137}Cs contributions to char in headwater stream (Haque et al., 2017b). However, ^{137}Cs activity concentrations of terrestrial prey items were decreased to 82% in summer and 42% in autumn, over 4-year study period from 2012 to 2016 (Figure 5.5). For instance, the mean ^{137}Cs activity concentrations of spider crickets (Raphidosphoridae gen. spp.) in summer become 695 Bq kg^{-1}-dry after 5 year of the accident which were 6 times lower than those in summer (4246 Bq kg^{-1}-dry) after 1 year of the accident (Table 5.2). Moreover, metabolic rate of char in summer is higher than autumn (Haque et al., 2017b). Char of autumn samples mostly consume low contaminated aquatic prey items, have lower metabolic rate (Haque et al., 2017b), and the ^{137}Cs activity concentrations of aquatic prey items decreased about 47% during 2012 to 2016 (Figure 5.5). Therefore, differences in exposure to prey items, and metabolic rate of char in summer and autumn reflected on the seasonal variability of T_{eco}.

Higher T_{eco} in autumn might be associated with the consumption patterns of intake and elimination. Life cycle of a given fish species have an effect on T_{eco} (Wada et al., 2016) such as char become sexually mature in autumn indicated their limited food consumption habits (Brittain et al., 1991) related to lower metabolic rate in autumn (Haque et al., 2017b) possibly reflect on longer T_{eco}. In contrast, higher water temperature, and slow stream flow during summer (Nakano and Furukawa-Tanaka, 1999) associated with shorter biological half-life (Udegal et al., 1992) possibly affect T_{eco} (Hessen et al., 2000; Wada et al., 2016). Therefore, understanding the seasonal variability of T_{eco} with the dependence of prey items is crucial to predict the fate of ^{137}Cs in char of headwater stream.

The result showed that ^{137}Cs activity concentrations of char in summer and autumn samples after 1 year of the accident had no remarkable differences (p = 0.75) because of higher concentrations of consumed ^{137}Cs across seasons associated with higher fallout volume in Fukushima (Haque et al., 2017b), while those after 5 years of the accident differed significantly between summer and autumn samples (p = 0.05) with the dependence of terrestrial food sources. Higher ^{137}Cs activity concentrations in char of autumn samples after 5 year of the accident because char had 60% dietary contributions from terrestrial prey items in autumn (Haque et al., 2017b), and the ^{137}Cs activity concentrations of terrestrial prey items decreased only 31% during 2012-2016 (Figure 5.5). In contrast, ^{137}Cs activity concentrations of char in summer were lower than autumn through char mostly depends on terrestrial prey items as dietary sources in summer because ^{137}Cs activity concentrations in terrestrial prey items decreased 82% in the period from 2012 to 2016 (Figure 5.5). Another reason behind the lower contamination levels in summer can be explained due to higher increased growth rates, and higher food availability with higher metabolic rate of char in summer (Brittain and Gjerseth, 2010).

T_{eco} is one of the most important indices to predict the fate of ^{137}Cs which possibly varied depending on the ^{137}Cs activity concentrations of prey items. Most of the previous studies not considering seasonal variability of ^{137}Cs activity concentrations but for the accurate estimation of T_{eco}, seasonal sampling is important, and shifting of diet sources also need to be considered. Though I didn't consider winter and spring samples in this study, but I think that for estimation of T_{eco} using winter sample will be most appropriate for monitoring T_{eco} of white-spotted char in headwater stream because of its unique feeding characteristics with different ^{137}Cs activity concentrations in terrestrial and aquatic prey items across seasons, and the ^{137}Cs activity concentrations of char in winter might be the highest among the seasons which was already evident in previous chapter (chapter 4). This study suggests that T_{eco} of winter in white-spotted char possibly tended to be higher which will help the local people and/or fishermen for fishing and management of fishery resources in terms of human consumptions below the Japanese standard limit of ^{137}Cs contamination levels (i.e., 100 Bq kg^{-1}-wet) to avoid risk for public health. Indeed, further process-based research need to be conducted for examining the variability of T_{eco} in ecosystems.

5.5. Summary and conclusions

T_{eco} of ^{137}Cs in white-spotted char showed seasonal variability that demonstrate the importance of incorporating information in determining the length and severity of potential risks to fisheries resources of headwater streams in different seasons of a specific year. The results of this chapter also emphasized the effects of standard length on T_{eco} of char in summer and autumn after 5 years of the FDNPP accident. Because of unique characteristics of headwater stream, receiving ^{137}Cs continuously through litterfall from the adjacent catchments (Sakai et al., 2016b) which provide excellent opportunities for studying principles related to contaminant decay processes. ^{137}Cs decay processes of media and/or food sources are essential to consider for predicting the temporal change of ^{137}Cs contamination levels in fish (Hessen et al., 2000; Pröhl et al., 2006). Such decay processes of ^{137}Cs across seasons revealed the need for incorporating the concept of seasonal variability of T_{eco} with attention to the size-effect for potential risk assessment, and for predicting the fate of ^{137}Cs in char of riparian ecosystem. Indeed, such predictions will be useful to generate policies for the management and conservation of headwater streams after the fallout, and for preserving public health by avoiding consumption of contaminated fish.

CHAPTER 6

SUMMARY, CONCLUSION, AND MANAGEMENT APPLICATION

6.1. Main findings of the thesis

The ^{137}Cs is considered as an environmental contaminants after the FDNPP accident because of its long physical half-life (Sakai et al., 2014). Once ^{137}Cs enter into the forested headwater streams via litterfall, leaching and decomposition of leaf litter alters ^{137}Cs availability (Sakai et al., 2015). Leaf litter in forested headwater stream is considered as a dominant source of ^{137}Cs for contamination of biota, and ^{137}Cs transfer from lower to higher tropic levels including white-spotted char (*Salvelinus leucomaenis*) through food webs (Sakai et al., 2016b).

A key question of this research is related to the understanding of transfer processes and factors that affect ^{137}Cs accumulation in white-spotted char of forested headwater streams. Through numerous previous studies considered ^{137}Cs transfer factor to determine the environmental conditions of ^{137}Cs to a specific species by considering ^{137}Cs transfer from abiotic (e.g., water, soil and/or sediment) to biotic component (e.g., fish), not considering any other potential processes (Garnier-Laplace et al., 2000; Marzano et al., 2000; Smith et al., 2000a). In addition, Zhao et al. (2001) considered trophic transfer factor using single prey item to fish without considering quantitatively the complex consumption habits of fish in natural environmental conditions for evaluating ^{137}Cs accumulation of the given fish species. Moreover, the values of these transfer factors were extremely large and widely variable indicated both dilution and accumulation which might be questionable in the studies of ^{137}Cs dynamics. On the other hand, few previous studies identified some possible factors that can affect the transfer factor values, but seasonal variability of transfer processes with their affecting factors were not clearly understood. Moreover, attention need to be given on understanding the seasonal variation of transfer factor to elucidate the dominant processes that control ^{137}Cs contamination levels in char. Though it is 6 years have passed after the fallout, ^{137}Cs still transfer from the forest to stream channels via litterfall, and subsequently contaminated the biota including char. In addition, multiple food sources of char may also have challenge for estimating temporal changes of ^{137}Cs activity concentration. Based on the previous studies, ecological half-life of terrestrial and aquatic components differed (Klemt and Zibold, 2003; Kanisch, 2002; Pröhl et al., 2006). Such differences of decay processes possibly affect the temporal pattern of ^{137}Cs in white-spotted char. Therefore, monitoring the long-term behavior, and time-series changes of ^{137}Cs contamination levels in char is essential to predict the fate of ^{137}Cs in char of forested headwater stream which is also important for evaluating the potential use of contaminated areas, and for addressing the risks to human health though consumption of contaminated fish (Peles et al., 2000b; Pröhl et al., 2006). Understanding the temporal decline of ^{137}Cs activity concentrations in char with seasonal variability will helpful for the prediction of ^{137}Cs in char, which is also important for fishery resource management.

In this thesis, I addressed the above knowledge gaps through six chapters. Chapter 1 introduced the background, importance, necessity, and justification for studying ^{137}Cs transfer in fish of forested headwater streams by highlighting the main objectives and structure of this thesis. Chapter 2 represented the variability of ^{137}Cs contamination levels in freshwater fish based on the review of some previous studies. I also evaluated the characteristics of freshwater fish species with special emphasis on their food and feeding habits around the contaminated areas all over the world, different transfer processes and factors that induce variability in association with spatio-temporal variations in their contamination levels. The objective of chapter 3 was to develop an indicator for examining ^{137}Cs transfer by food web. In particular, food web based transfer factor (TF_{web}) was developed in this chapter by integrating the dietary ^{137}Cs contributions of multiple prey items to white-spotted char which suggested bioaccumulation from prey items to predator. This approach possibly be applicable for studying transfer process of any contaminant via food web structures in complex ecosystems. Chapter 4 addressed the seasonal patterns in the TF_{web} of ^{137}Cs in white-spotted char for understanding the mechanism of ^{137}Cs accumulation in biota across season by studying in forested headwater streams of similar surrounding environmental conditions with different fallout volumes. Such information is very effective to elucidate the dominant processes and associated factors that affecting ^{137}Cs contamination levels in different habitat contamination levels. Chapter 5 examined the ^{137}Cs activity concentrations in white-spotted char in association with their seasonal accumulation patterns with comparison temporal changes in their contamination levels by estimated ecological half-life (T_{eco}) using the data after 1 and 5 years of the accident. Chapter 6 was summary of important findings, the processes and factors that control contamination levels in white-spotted char of forested headwater streams was also discussed along with conclusions and management applications. The importance and possible applications of the findings of this thesis in Bangladesh were also discussed in this chapter. The key findings of these six chapters are summarized in the following section.

6.1.1. The importance of ^{137}Cs dynamics in headwater streams (chapter 1)

Headwater streams are small (bankfull < 2 m), steep gradient (> 0.10) channels, including hillslopes, zero-order basins (Tsukamoto et al., 1982), and first and second order channels (Strahler, 1957). Processes from hillslopes to streams, and from terrestrial to aquatic ecosystems are tightly linked in forested headwater streams (Gomi et al., 2002). The maximum drainage area of headwater systems is 1 km^2 based on the continuum of hydrogeomorphic processes (Woods et al., 1988; Swanson et al., 1998). Such small headwater streams are considered as habitats for macroinvertebrates (Richardson, 1992), and these macroinvertebrates become important food sources for the salmonid fish species including white-spotted char (Nakano and Furukawa-Tanaka, 1994; Miyasaka et al., 2003; Sakai et al., 2016b).

The ^{137}Cs dynamics in forested headwater streams are largely depend on the riparian conditions, watershed topography, water temperature, and channel morphology (Sakai et al., 2016b).

Understanding how the biological and ecological processes of the riparian ecosystems control the ^{137}Cs movement is crucial because headwater streams continuously received ^{137}Cs via the litterfall from the adjacent catchments as a primal source of contamination for biota (Sakai et al., 2016b).

Litterfall from the adjacent forest canopy is the main transport route to the ground surface (Teramage et al., 2014), along with stream channels draining within such forests (Sakai et al., 2015). Once litter enters into the streams, ^{137}Cs leaching and decomposition from the contaminated litter induce the ^{137}Cs availability in the stream channels (Sakai et al., 2015). ^{137}Cs attached to the litter can transfer from lower to higher trophic levels via food weds in forested stream ecosystems (Sakai et al., 2016b). Therefore, understanding the processes controlling the ^{137}Cs contamination levels in biota is critical for predicting the effects of ^{137}Cs on forested headwater streams. There is also a need to better understanding how these processes control the ^{137}Cs contaminations levels in biota of headwater streams to reduce the ^{137}Cs loads in headwater streams (Ranalli and Macalady, 2010).

This study combined the processes and factors that control the ^{137}Cs accumulation in biota of headwater streams. The objective of this study were to understand the effects of ^{137}Cs on fish habitats, transfer processes and time-series decline with their seasonal variability, and the factors governing the accumulation pattern associated with the temporal change of contamination levels in white-spotted char in forested headwater streams.

6.1.2. Variability of ^{137}Cs contamination levels in fish (chapter 2)

The ^{137}Cs released as a fission product from the nuclear power plants accidents to the environment, and contaminated both terrestrial and aquatic ecosystems. The movement, deposition and accumulation patterns of ^{137}Cs in the freshwater systems may induce variability in contamination levels of freshwater fish. Various physical, chemical, biological factors and their associated processes have potential importance in regulating the variability of ^{137}Cs contamination levels of freshwater fish with the interactions of both biotic and abiotic components of an ecosystem.

Recently, ^{137}Cs dynamics has become the subject of multidisciplinary studies in the radioecology arena. In this review chapter, for addressing the variability of contamination levels in freshwater fish with integrative dimensions, I emphasized the characteristics of previous studied major freshwater fish in the contaminated areas, their major food items, differences in their contamination levels, and the associated factors and processes which are linked to induce the variability. Understanding the different processes and associated factors for ^{137}Cs contamination in freshwater fish is essential to elucidate the potential mechanisms that determine the contamination levels of fish with indication of both bioaccumulation and/or biodilution.

This chapter highlighted how ^{137}Cs effects combine in a broader ecological context at higher levels of biological organizations. It also focused on the current knowledge of tempo-spatial variations of ^{137}Cs contamination levels in freshwater fish. The knowledge gaps due to the relatively few studies on ^{137}Cs dynamics of freshwater fish, and the possible remediation practices are also discussed for contaminated freshwater ecosystems. However, this review chapter will serve as a guideline for the researchers to identify the key factors and processes for variability in contamination levels, and help to make answers of necessary questions that have potential impacts on ^{137}Cs transfer and accumulation in fish of freshwater environments.

6.1.3. Food web-based transfer factor (chapter 3)

TF_{web} was developed as a novel approach to study contaminant movements from multiple prey items to a predator based on the dietary contributions of multiple prey items with their respective contamination levels. TF_{web} was applied for evaluating ^{137}Cs transfer to white-spotted char based on the trophic structures of stream–riparian ecosystem in forested headwater streams draining Japanese cedar plantation forest.

The applicability of this method for ^{137}Cs accumulation in white-spotted char was examined by comparing sites with different contamination levels but similar surrounding environments in Fukushima and Gunma prefectures. The dietary contributions from both aquatic and terrestrial prey items to white-spotted char were analyzed using stable carbon and nitrogen isotope ratios. TF_{web} was calculated using ^{137}Cs activity concentration of white-spotted char divided by the sum of ^{137}Cs activity concentrations of all prey items calculated with their lower and upper values of diet contributions.

The ^{137}Cs activity concentrations in white-spotted char ranged from 704 to 6082 Bq kg^{-1}-dry in Fukushima, and from 193 to 618 Bq kg^{-1}-dry in Gunma. Dominant prey taxa were mayflies (*Ephemera japonica*), spider crickets (Rhaphidosphoridae gen. spp.), and freshwater crabs (*Geothelphusa dehaani*); and each of them accounted for 3 to 12% of the fish diet, based on lower and upper contributions, respectively. Moreover, in both Fukushima and Gunma, white-spotted char tended to be accumulated ^{137}Cs by multiple prey items consumption with similar order of magnitude.

The TF_{web} ranged from 1.12 to 3.79 in Fukushima, and from 1.30 to 4.30 in Gunma which suggested bioaccumulation from prey items to predator. Large size white-spotted char tended to have greater TF_{web} than small bodied fish possibly associated with metabolic characteristics. On the other hand, transfer factor based on media–char and/or single prey–char had wide ranges of values with indication of both dilution and accumulation. This method using TF_{web} can be appropriate not only for examining ^{137}Cs transfer in predator-prey systems with complex food web structures of stream-riparian ecosystems, but also can be applicable for other contaminants for modelling the transfer of contaminants in complex ecosystems.

6.1.4. Seasonal variations of food web-based transfer factor (chapter 4)

The seasonal variability TF_{web} of ^{137}Cs in white-spotted char was examined by selecting two headwater streams as study sites with similar landscape characteristics, but different amounts of fallout in Fukushima (100–300 kBq m^{-2}) and Gunma (30–60 kBq m^{-2}) prefectures, Japan. TF_{web} of predator–prey systems was based on the dietary contributions of multiple prey items estimated from stable isotope ratios compared to their respective ^{137}Cs activity concentrations.

The ^{137}Cs activity concentrations in white-spotted char in Fukushima were 6 times greater than those in Gunma. White-spotted char consumed more terrestrial food sources in summer with higher contamination levels than in winter based on their dietary-based ^{137}Cs contributions. TF_{web} values in Fukushima and Gunma in all seasons indicated ^{137}Cs bioaccumulation from multiple prey items to predator. TF_{web} in Gunma significantly varied among seasons with the greatest values in winter from 2.37 to 14.93 (mean ± SD: 7.51 ± 1.64), and the lowest ones in summer (mean ± SD: 2.62 ± 0.89), while TF_{web} in Fukushima did not vary seasonally.

Despite similar consumption patterns, and the specific metabolic rates of white-spotted char; the different seasonal patterns of TF_{web} at the two sites can be explained by the relative excretion rate with respect to the concentration of ^{137}Cs intake. ^{137}Cs levels can be remained relatively constant in the white-spotted char body throughout the seasons in the areas with high contamination, such as Fukushima, possibly because ^{137}Cs intake overwhelmed to the excretion rate. These findings suggested that seasonal patterns in the transfer processes of ^{137}Cs in white-spotted char can be important for understanding dominant processes of ^{137}Cs movement that controls contamination levels in an ecosystem.

6.1.5. ^{137}Cs activity concentrations in char after 5 years of the accident (chapter 5)

The ^{137}Cs activity concentrations in char were measured to present the current contamination status, and to evaluate the time-series changes in contamination levels of summer and autumn samples after 5 years of the accident. Six individual char were sampled in each sampling season except summer 2012. The standard lengths of char ranged from 69 to 124 mm (mean ± SD: 95 ± 28 mm) in summer, and from 89 to 173 mm (121 ± 33 mm) in autumn after 1 year of the accident, while those after 5 year of the accident ranged from 69 to 173 mm (130 ± 36 mm), and from 77 to 186 mm (124 ± 42 mm) in summer and autumn, respectively.

The mean ^{137}Cs activity concentrations of char in summer decreased from 1986 ± 631 Bq kg^{-1}-dry in 2012 to 304 ± 338 Bq kg^{-1}-dry in 2016, while those in autumn decreased from 1827 ± 822 Bq kg^{-1}-dry in 2012 to 1201 ± 949 Bq kg^{-1}-dry in 2016. The decline of ^{137}Cs activity concentrations of char were quantified by means of ecological half-life (T_{eco}). Estimated T_{eco} was 1.4 years in summer and 6.6 years in autumn. The persistence of seasonal variability in T_{eco} possibly due to the differences in dietary sources with their contamination levels, and associated with the metabolic rates of char across seasons.

^{137}Cs activity concentrations in char were significantly related to the fish body size. Hence, greater increase in ^{137}Cs with respect to standard length was greatest in summer 2012 than summer 2016. $T_{\text{eco-size}}$ of char in summer were relatively lower than those in autumn can be explained by the differences of available dominant food sources with their respective contamination levels and decreasing rates of contamination. Examining the seasonal ^{137}Cs activity concentrations, and resulted T_{eco} in char after 5 years of FDNPP accident are essential for understanding the length and severity of potential risk of ^{137}Cs to fisheries resources, and for the prediction of the fate of ^{137}Cs in char of forested headwater stream.

6.2. Summary for interactions between biological and ecological processes

I summarized the findings of this study by considering the interactions between biological and ecological processes (Figure 6.1). TF_{web} is process-based approach for evaluating ^{137}Cs transfer in predator-prey systems. First of all, consumption of multiple prey items and their respective ^{137}Cs activity concentrations in predator-prey systems is important processes for ^{137}Cs transfer to white-spotted char in forested headwater streams. Biological processes such as metabolic rate is important to understand ^{137}Cs elimination and/or excretion. The variability of site and season is also related to the balancing of intake and excretion of ^{137}Cs from char body. Left side of the figure I developed in this study, while the right side of the figure is already known ecological processes. Such knowledge of relationship between biological and ecological processes can be helpful for developing the ^{137}Cs transfer and accumulation model of char (e.g., ERICA Tool) and their variability.

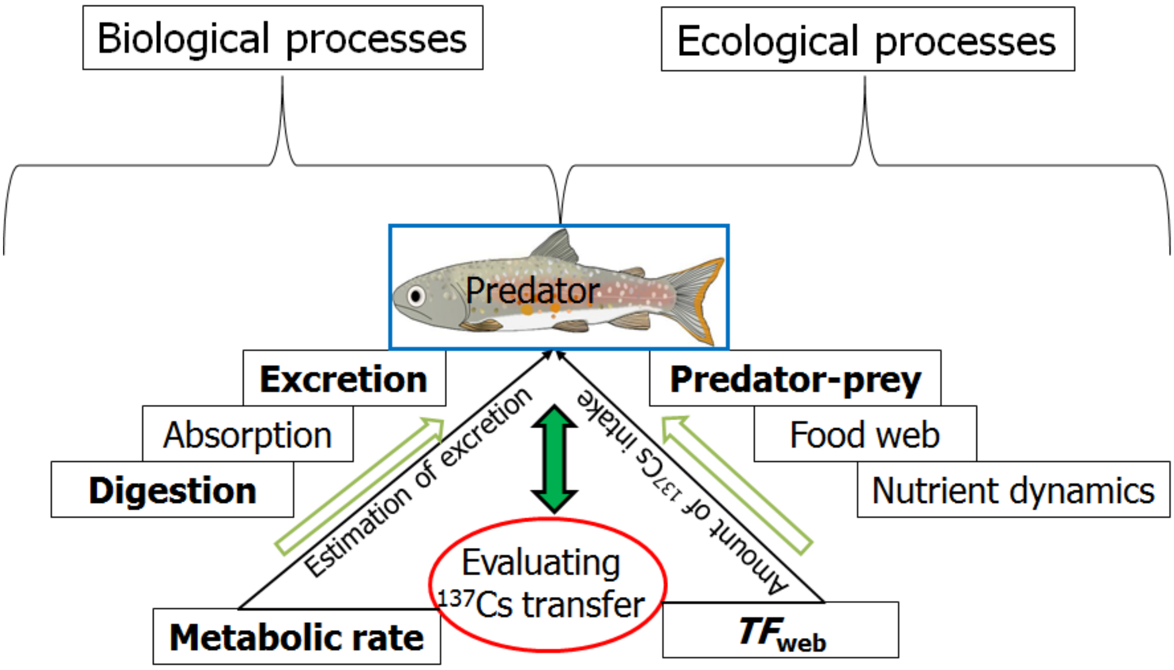


Figure 6.1. Schematic illustration of summary for interactions between biological and ecological processes in ecosystems.

6.3. Summary for interactions between terrestrial and aquatic ecosystems

Findings of this study also showed the importance of interactions between terrestrial and aquatic ecosystems (Figure 6.2). It was evident from this study that seasonal changes of terrestrial and aquatic food sources affected the seasonal pattern of ^{137}Cs transfer and accumulation in white-spotted char. ^{137}Cs contamination levels in terrestrial prey items decay very quickly, while those of aquatic prey items showed slower decay process. Moreover, different decay processes of ^{137}Cs activity concnetrations in terrestrial and aquatic systems affected the ^{137}Cs decay pattern of char. T_{eco} possibly varied with season depending on the changes of ^{137}Cs activity concentrations in terrestrial and aquatic prey items. For accurate estimation of T_{eco}, seasonal sampling is important to elucidate the factors affecting T_{eco}. Further process-based research need to be conducted for examining the variability of ^{137}Cs and resulted T_{eco} in ecosystems.

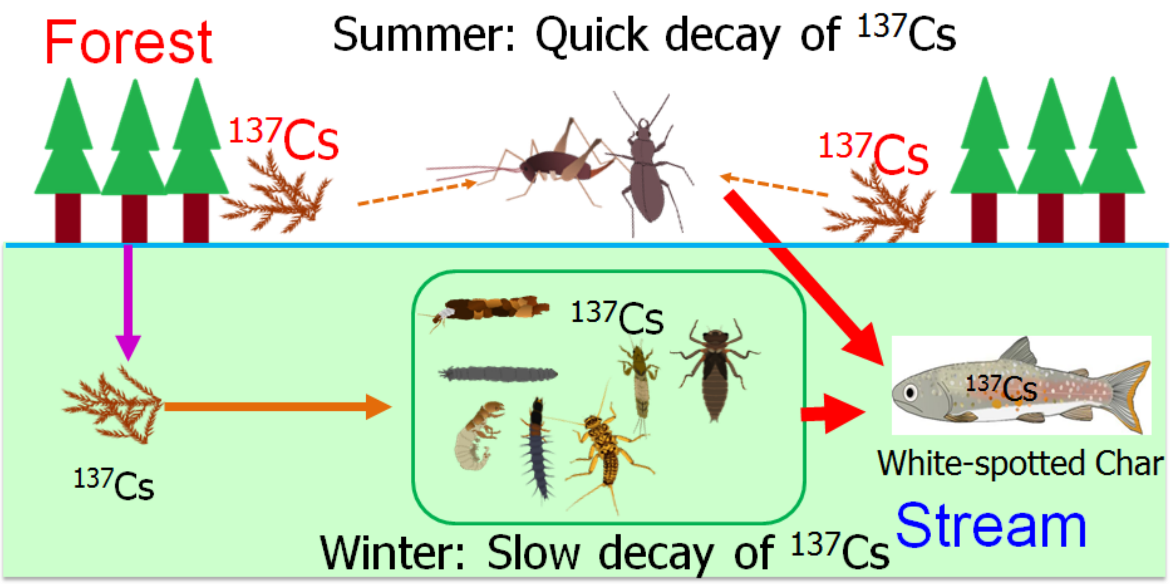


Figure 6.2. Schematic illustration of summary for interactions between terrestrial and aquatic ecosystems.

6.4. Conclusions

Understanding the processes and factors is the fundamental approach to comprehend the ^{137}Cs dynamics in an ecosystem related to hydrology, geomorphology and biology. Based on the specific hypotheses and/or aims and objectives, scientists and/or researchers carried out different studies to elucidate the processes and factors that control contamination levels in biota at various spatial and temporal scales; thus explanations may vary among the results and observations. For instance, after 1 – 2 years of the Fukushima accident, chapter 3 in this study showed that under similar environmental conditions and food web structures with different fallout volumes, the mean values of TF_{web} were similar in both Fukushima and Gunma. By considering the seasonal values of TF_{web}, and associated factors such as seasonal amount of contamination through consumption of multiple prey items, and specific metabolic rate of white-spotted char in chapter 4 showed that TF_{web} were varied among the seasons, and was the highest in winter in Gunma with low fallout volume without having any seasonal variations in area of high fallout amount, such as Fukushima. However, chapter 5 of this thesis showed seasonal variability in ^{137}Cs accumulation patterns, and resulted T_{eco} in white-spotted char in forested headwater stream of Fukushima after 5 years of the accident possibly due to differences in their dietary prey items, dietary ^{137}Cs contributions, ^{137}Cs decreasing rate of prey items with time, and metabolic rate of char across seasons. Thus, potential relationships between processes and factors should be estimated to improve our understandings of the ^{137}Cs dynamics in forested headwater streams (Figure 6.3).

For the sustainable protection of the environment from the contamination by radiation, attention need to be focused on the ecosystem based approaches including the species interactions, their dynamics and contributions to the ecosystems (Bréchignac et al., 2016). Processes based modelling of contaminant transfer with complex ecosystems are also essential for predicting the fate of contaminants, and associated decontamination practices (Clements and Newman, 2002). Indeed, we still have knowledge gaps between empirical parameters, and processes based modelling of ^{137}Cs transfer in complex ecosystems (Hinton et al., 2014). Hence, combination of researches for understanding processes and factors in association with food web structure those govern ^{137}Cs contamination levels in biota to understand the fate and transport of ^{137}Cs in a given ecosystem.

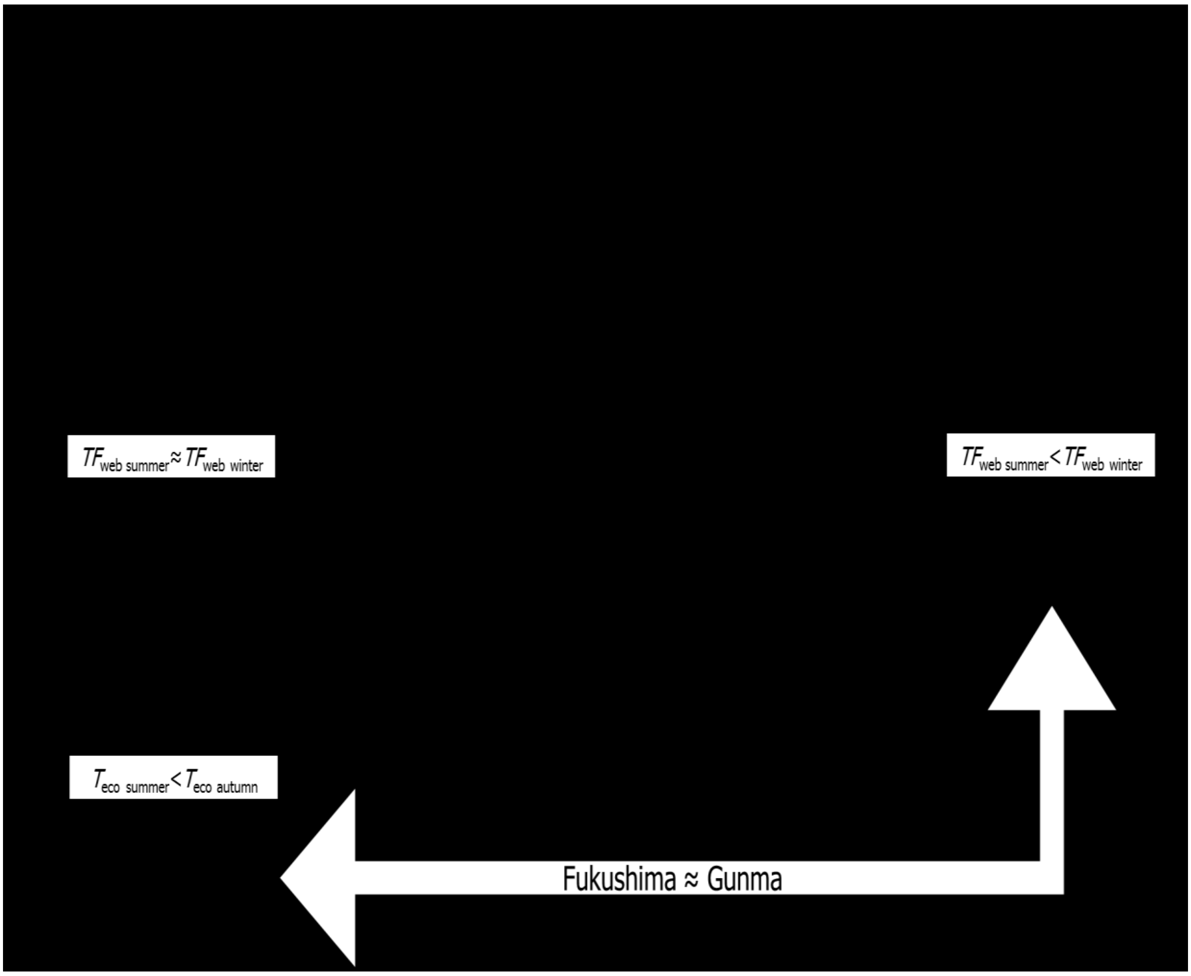


Figure 6.3. Summary of findings of this thesis.

6.5. Management applications

Radioactive materials released from the FDNPP accident exerted a massive influence on various aspects of Japanese life. Through any acute death or health damage due to released radioactive contaminants were not reported, but people are concerns about long-term health effects of living in the affected areas because of long physical half-life of ^{137}Cs. ^{137}Cs activity concentrations exceeding national guideline levels (i.e., 100 Bq kg^{-1}-wet) has been found in ecosystems, particularly in forested area due to persistent recycling of ^{137}Cs. These high levels of ^{137}Cs in forested landscapes are considered to persist for a couple of decades (Miyashita, 2012). Thus, it is important to monitor the long-term behavior of ^{137}Cs in the contaminated areas, and need a lot of efforts to develop the mitigation, remediation and management practices.

Understanding the movement of ^{137}Cs through ecosystems is essential for the management of radiation contamination, and risk assessment in both forest and stream environments. The major sources of knowledge regarding the behavior of ^{137}Cs in forest ecosystems were derived from the previous researches carried out in the Europe after the Chernobyl accident in 1986. But, the forests in Europe differ than those in Japan due to differences in landscape and geological structures, climatic conditions and data collection date etc.; therefore the findings and/or developed management practices of those studies are not directly applicable for the management of contaminated areas affected by the FDNPP accident. Understanding the long-term recirculation of ^{137}Cs in ecosystems is fundamental to management and possible remediation (Shaw, 2013). Thus, due to the environmental differences between the countries, care must be exercised when referencing information that is obtained from Russia, Ukraine, and Scandinavia. For comprehensive comparison between Japan and these countries, intensive and long-term monitoring of various forested ecosystems is required in Japan specially the northeastern parts of Japan.

To reduce the ^{137}Cs contamination from the environment previous studies have suggested management application with respect to minimizing the sources of contamination. For example, Tikhomorov et al. (1993) reported about engineering-based countermeasures for reducing the contamination levels in forested areas after the Chernobyl accident. Shaw et al. (2001) reported different countermeasures for the application in contaminated forests after the Chernobyl accident such as cutting of tress, soil improvement, use of fertilizer, liming, salt licks, etc. to reduce contamination levels in trees, soil, reduction in uptake of ^{137}Cs to tress and grazing animals, respectively. ICRP (2009) suggested reducing dose rate from the contaminated sources via (i) ingestion doses from the consumption of contaminated foodstuffs; (ii) external doses from surfaces contaminated by deposited ^{137}Cs; and (iii) inhalation doses from resuspended materials. IAEA (2012) suggested the followings for reducing the contamination in aquatic systems for ingestion of aquatic biota: (i) reducing direct contamination input into water body; (ii) altering the chemical properties of water to reduce direct ^{137}Cs uptake (such as via fish gills), and transfer to the animals of higher trophic levels via food web; and (iii) reducing contaminated feed consumption by farmed fish. In general, population doses from aquatic pathways are often lower than from terrestrial pathways, depending on food habits. In the context of atmospheric fallout of ^{137}Cs to both terrestrial and aquatic systems, it has been shown that doses from terrestrial sources are more significant than those of aquatic sources (Voitsekhovitch et al., 1996; Sakai et al., 2016b).

The countermeasures in aquatic ecosystems have considered both direct (restrictions) and indirect intervention measures to reduce doses such as restrictions on water use or changing to alternative supplies, restrictions on fish consumption, water flow control measures (e.g., dykes and drainage systems), uptake by fish and aquatic foodstuffs from contaminated sources; and preparation of fish prior to consumption (Wauters et al., 1996; Smith et al., 2001). Smith et al. (2003) showed that use of potassium chloride fertilizer to the lake resulted 40% decrease in ^{137}Cs

activity concentrations of different fish species. Biological dilution based on the theories of buffering (Ashraf et al., 2014) through using fertilizer and liming, ^{137}Cs activity concentrations in fish can be decreased (Butler, 2011; Kinoshita et al., 2011). Lowering the fish and/or predator stocks are also useful for reducing trophic transfer of ^{137}Cs in fish (Morino et al., 2011). Controlling microbial primary producers are effective as ^{137}Cs mobilization and cycling in freshwater ecosystems largely depends on these producers (Avery, 1995). Measures to reduce doses via freshwater foodstuffs may be required over longer timescales as a result of bioaccumulation of ^{137}Cs through the aquatic food chain.

However, these practices hardly be implemented in forested headwater streams because of tight linkages among the various processes from hillslopes to stream channels, and from forest to stream environments. Moreover, forested headwater stream received continuously ^{137}Cs from the adjacent catchments via litterfall (Sakai et al., 2016b). Headwater streams have specific characteristics, and site-specific behaviors that were governed by their hydrological, geomorphological, biological, and ecological parameters. ^{137}Cs remediation of any reservoir depends on its site-specific characteristics (Ashraf et al., 2014). Though the radiological risks associated with ^{137}Cs in litter, soils, sediments, water and biota those can't be eliminate completely, but the contamination levels can possibly be reduced by using appropriate technique.

Radiological protection of the environment is developing rapidly at both the national and international levels, but there are still gap of internationally agreed recommendations for how radiological protection of the environment should be carried out (IAEA, 2002). For the radiological protection of the environment, two essential components of the ecosystems need to be considered, such as media (water, soil, sediment) dominated by physiochemical processes, and living biota dominated by biological processes. Both are in complex interactions within the ecosystems via ecological processes. Moreover, ecosystem-based approaches such as trophic and prey-predatory interactions are not addressed to induce potential effect of radiation through interactions with community and ecosystems processes (Brèchignac and Doi, 2009). Thus, equal emphasis is required on both ecosystems components for radiological protection of the environment.

Moreover, it is essential to develop internationally agreed approach for environmental protection from radiation. For addressing the environmental protections from radiation, it is necessary to understand how effects are combined across different levels of biological organizations, and an enhance collaboration is needed to understand insights about the mechanisms of radiological damage and associated relevance to the populations (Hinton et al., 2004). Consideration will be given to the linkages between radiation dose to biota, identification of the contaminated biota, modeling and risk assessments, limitations on the accuracy and precision of dose measurements, allowance for accommodation of new scientific findings; and the application approaches within an overall protection system (IAEA, 2002).

It is clear that ^{137}Cs undergoes intensive recycling in the environment which owing to the relatively long physical half-life indicates that ^{137}Cs can remain in the ecosystems for many years following contamination event (Avery, 1996). Currently, most of the ^{137}Cs still remained in the forested environment (Yoschenko et al., 2017) after the FDNPP accident, and possibly continouisly moving from forest to stream. Therefore, it is very difficult to attain a holistic understanding of ^{137}Cs transfer as well as the management, and disturbance regimes on stream ecosystems. This study examined and emphasized the dietary contributions, their seasonal changes and temporal decline to explain the variability of ^{137}Cs contamination levels of char in forested headwater streams. These findings are also useful for predicting the fate of ^{137}Cs in complex ecosystems. The approach of TF_{web} of ^{137}Cs in char of headwater streams will help for developing a

^{137}Cs transfer model by combining biological and ecological processes which had suggested by national and international organizations related to the different disciplines of radioecology. Long-time scale based observations with combinations of various processes need to be incorporated not only for char but also for the other species for developing a more conceptual model of ^{137}Cs dynamics, and/or developing management practices for reducing the contamination levels in ecosystems. The approaches of this study will helpful for improving reference species approach (ICRP, 2005), and for modeling the ^{137}Cs transfer in ecosystems such as ERICA Tool.

6.6. Importance and possible application of this study in Bangladesh

Recently, the necessity of Nuclear Power Plant is well recognized in Bangladesh because nuclear power is environment friendly and cost effective option for producing electricity. Presently, there are 437 nuclear power plants in operation in 31 countries around the globe, 68 nuclear power plants are under construction in 14 countries. Establishment of 168 nuclear power plants in 27 countries are in progress which is expected to be completed by 2022. Among them 30 nuclear power plants will be constructed in the new comer countries, Bangladesh is one of them.

The project site is located in Rooppur of Pabna district in Bangladesh with smack dab on top of a potent earthquake fault line (Figure 6.4). It's not totally a population free area, so any accidents at the NPP can endanger nearest people. This is also a coastal area. So, cyclone, tornado and/or any other natural disasters are not uncommon in this place. Radiation will be exposed to environment, and people on low dose which will not be so alarming, but will be able to create havoc if disasters occur. Moreover, once the project started, a major problem will be the disposal, and storage of the radioactive waste material produced by the plant. Around 20–30 tons of high-level wastes are produced per month per nuclear reactor. Though several methods have been suggested for final disposal of high-level waste, including deep burial in stable geological structures, transmutation, and removal to space but none of these methods have been implemented. The main problem of nuclear plants is release of radioactivity. This is a serious concern for environment. It includes deterioration of radioactive waste containers due to radioactive, and thermal effects, and the consequential leakage and contamination of groundwater. Moreover, the behavior of radioactive wastes in ecosystems needs to be concern for successful management now, and even after possible contamination events. Effective and proper waste management system must be launched and implemented. Nuclear disaster is not expected, but not an uncommon incident. So, everyone need to concern about their duties after any natural disasters from the learning of past nuclear accident like the Chernobyl and the Fukushima accidents in the world.

Figure 6.4. Location of the Rooppur Nuclear Power Plant (RNPP) site at Pabna district of Bangladesh. RNPP indicate Rooppur Nuclear Power Plant.

In my Ph.D. study at Tokyo University of Agriculture and Technology (TUAT), Japan, I am focusing on the transfer of ^{137}Cs in fish of forested headwater stream, and how the ^{137}Cs accumulated in fish via food web by considering diet contributions of their prey items. The information, knowledge and skills that I gained through my study in TUAT will allow me to forecast possible effects of the contaminants, and their biological effects on aquatic ecosystems. In Bangladesh, we are struggling to establish foundation of research activities, and to reach to an advanced level as it requires skills, deep knowledge, and philosophy in performing research activities. Through experiencing wide range of research activities at TUAT in Japan, and UC Davis in the USA during internship, I have been expanding my knowledge and skills, which will significantly contribute to enhance research levels in my home country, and will allow me to solve environmental problems including the radioactive wastes from RNPP in aquatic ecosystems. Moreover, due to performing research on radioactive contaminants will allow me to improve my problem solving skills in the research field of eco-toxicology. The findings of my doctoral study will be helpful in maintaining the environmental safety of any contaminants for sustainable ecosystem management. The information and findings that I have gained from my Ph.D. will be useful to establish an integrated knowledge based framework that will further help to measure and mitigate the harmful effects of contaminants such as industrial effluents, heavy metals, pesticides, insecticide, toxins of algal bloom etc. on aquatic ecosystems in Bangladesh. The knowledge, experiences and skills can be applied together for developing process-based transfer modelling of such contaminants which are transfer and/or accumulated in aquatic ecosystems via their hydrological and biological processes. Therefore, this study will be a meaningful in the context of aquatic contaminants including the possible hazards from any contamination sources, and its impact on biological environments.

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