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Pre- and post-accident environmental transfer of radionuclides in Japan: lessons learned in the IAEA MODARIA II programme

Keiko Tagami[1,*], Shoji Hashimoto[2], Masashi Kusakabe[3], Yuichi Onda[4], Brenda Howard[5,6], Sergey Fesenko[7], Gerhard Pröhl[8], Andra-Rada Harbottle[9] and Alexander Ulanowski[9]

[1] National Institute of Radiological Sciences, National Institutes for Quantum Science and Technology, Chiba, Japan
[2] Department of Forest Soils, Forestry and Forest Products Research Institute, Tsukuba, Japan
[3] Marine Ecology Research Institute, Onjuku, Japan
[4] Center for Research in Isotopes and Environmental Dynamics, University of Tsukuba, Tsukuba, Japan
[5] School of Bioscience, University of Nottingham, Loughborough, United Kingdom
[6] UK Centre for Ecology and Hydrology, Lancaster, United Kingdom
[7] Russian Institute of Radiology and Agroecology, Obninsk, Russia
[8] Consultant, Freising, Germany
[9] International Atomic Energy Agency, Vienna, Austria
[*] Author to whom any correspondence should be addressed.

E-mail: tagami.keiko@qst.go.jp

Keywords: pre and post-accident, Fukushima Daiichi Nuclear Power Plant, transfer parameters, radiocaesium, radioiodine

Abstract

An international review of radioecological data derived after the accident at the Fukushima Daiichi nuclear power plant was an important component of activities in working group 4 of the IAEA Models and data for radiological impact assessment, phase II (MODARIA II) programme. Japanese and international scientists reviewed radioecological data in the terrestrial and aquatic environments in Japan reported both before and after the accident. The environmental transfer processes considered included: (a) interception and retention radionuclides by plants, (b) loss of radionuclides from plant and systemic transport of radionuclides in plants (translocation), (c) behaviour of radiocaesium in soil, (d) uptake of radionuclides from soil by agricultural crops and wild plants, (e) transfer of radionuclides from feedstuffs to domestic and wild animals, (f) behaviour of radiocaesium in forest trees and forest systems, (g) behaviour of radiocaesium in freshwater systems, coastal areas and in the ocean, (h) transport of radiocaesium from catchments through rivers, streams and lakes to the ocean, (i) uptake of radiocaesium by aquatic organisms, and (j) modification of radionuclide concentrations in food products during food processing and culinary preparation. These data were compared with relevant global data within IAEA TECDOC-1927 'Environmental transfer of radionuclides in Japan following the accident at the Fukushima Daiichi Nuclear Power Plant'. This paper summarises the outcomes of the data collation and analysis within MODARIA II work group 4 and compares the Japan-specific data with existing radioecological knowledge acquired from past and contemporary radioecological studies. The key radioecological lessons learned are outlined and discussed.

1. Introduction

The accident in the Fukushima-Daiichi Nuclear Power Plant (FDNPP) occurred on 11 March 2011 led to a release of radionuclides that were dispersed over both terrestrial and aquatic environments. Due to the prevailing wind directions during the accident, a large fraction of the released radionuclides deposited on the Pacific Ocean whereas a smaller fraction deposited on terrestrial and freshwater environments. The highest depositions on land occurred in a north-westerly direction from the FDNPP (International Atomic Energy Agency (IAEA) 2015).

The deposition of radionuclides on terrestrial and aquatic systems immediately triggered the setup of monitoring campaigns and research activities to study the transfer of radionuclides in the environment.

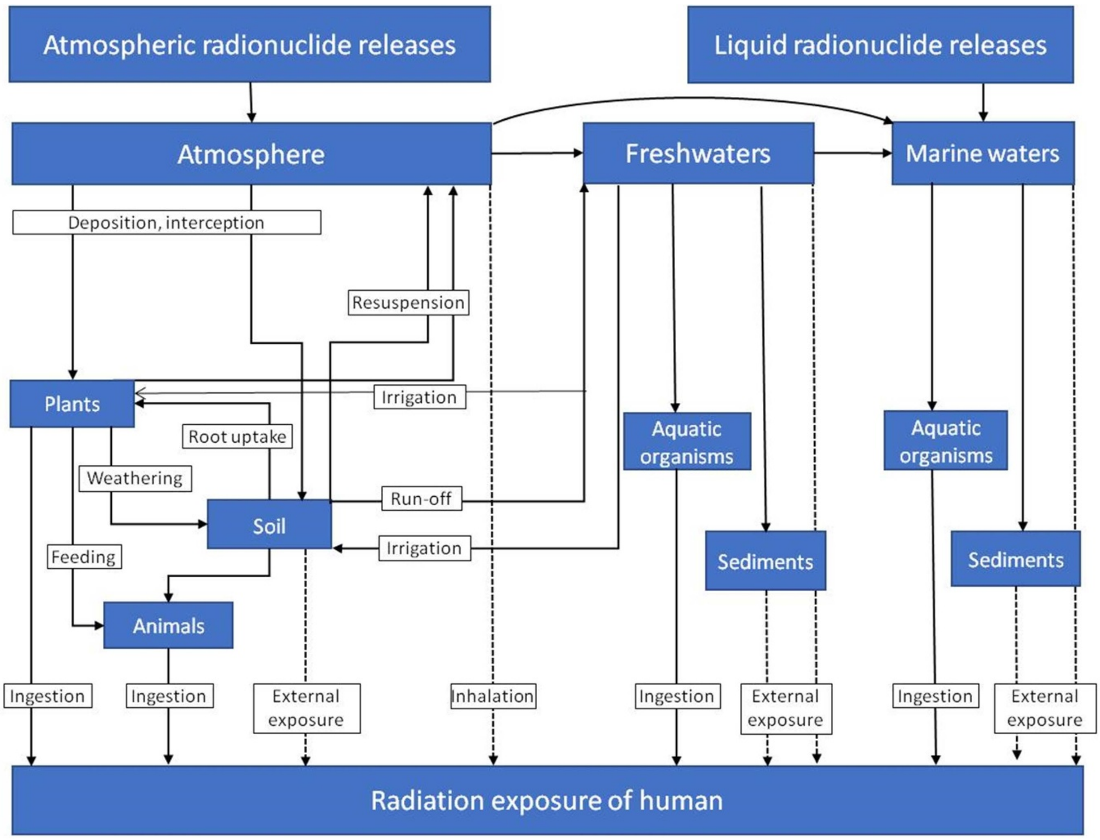

Figure 1. Example of major exposure pathways to humans. The dotted lines show pathways that are not considered in IAEA TECDOC-1927 (as published by Fesenko *et al* 2020a). Reproduced with permission from Fesenko *et al* (2020a). © IAEA, Vienna.

Within the activities of the IAEA MODARIA II programme (Brown *et al* 2022, this volume), Working Group 4 (Transfer Processes and Data for Radiological Impact Assessment) focused on the radioecological understanding gained in Japan following the nuclear accident at the FDNPP. The group—in cooperation with scientists from leading Japanese institutes in the field of radioecology—compiled, analysed, and evaluated results of monitoring and research activities that documented the transfer of radionuclides in the environment in areas affected by the accident. The outcomes of the Working Group have been collated in the IAEA TECDOC-1927 (International Atomic Energy Agency (IAEA) 2020) which brings together the results of Japanese studies that identified and quantified key radioecological processes and related transfer parameters in terrestrial and aquatic environments as shown in figure 1. Site- or region-specific factors influencing the transfer of radionuclides through the environment such as climate, landscape, agriculture and food processing practices, lifestyle and national dietary habits were identified. The experience gained was put into context with existing global radioecological information (International Atomic Energy Agency (IAEA) 2009, 2010).

This paper extracts the key radioecological findings elaborated in the IAEA TECDOC 1927 (2020) and focuses on specific aspects of the environmental transfer of radionuclides under the prevailing conditions of the areas affected by the deposition of radionuclides.

2. Environmental conditions in the areas affected by the deposition of radionuclides

Fukushima Prefecture is predominantly mountainous with about 70% of the area covered by forests. The deposition of radionuclides affected inhabited areas, agricultural land (including paddy fields) and both deciduous and evergreen coniferous forests. Many lakes, ponds, rivers, and creeks are also present in the area (Hashimoto *et al* 2012, 2020a). Usually, hills are steep, and the annual precipitation is high leading to occasional instances of substantial run-off and flooding. Freshwater bodies constitute important resources for drinking and irrigation water, and for industrial purposes.

A wide spectrum of radionuclides was released after the FDNPP accident that were mostly short-lived (IAEA 2015) of which, I-131 (half-life: 8.02 d) was the most important. For long-term considerations, only radiocaesium (comprising both Cs-134 [half-life: 2.06 y] and Cs-137 [half-life: 30.1 y]) is relevant when considering possible consequences for the human foodchain and for other organisms (IAEA 2015).

The accident happened at the transition period from winter to spring in north-east Japan. The growing period had only just started for a few types of crops such as early green vegetables. Radionuclides were deposited on above ground parts of perennial plants, such as fruit trees and tea, but many of these crop species were still in their dormant period. Consequently, most crops were not affected by foliar deposition, because they are usually planted or seeded somewhat later in the year. Therefore, only the uptake of radiocaesium from soil was relevant for the long-term contamination for most crops. Farm animals kept for producing milk, meat, and eggs, were kept indoors, and fed on stored feed. As for trees in forests, deciduous trees did not have leaves while coniferous trees had. Therefore, evergreen trees intercepted a larger fraction of radionuclides deposited to the ground (see below).

3. Sources of information on the environmental transfer of radionuclides

Immediately after the accident, intensive monitoring and research programmes were initiated to determine radionuclide activity concentrations and their time-dependence in environmental media such as soils, plants, foodstuffs, freshwater in streams, rivers, lakes and reservoirs, bottom sediments of freshwater bodies, and aquatic species. The fate of radionuclides deposited on or released directly to the Pacific Ocean was addressed by measuring activity concentrations in seawater, suspended and bottom sediments and marine species in the ocean around Japan.

The results of monitoring programmes were analysed to study the behaviour of radiocaesium in the environment. Many research projects were initiated to identify and quantify different radionuclide transfer processes (see figure 1). Table 1 shows an overview on the main topics addressed in the IAEA TECDOC-1927 (IAEA 2020), which covers a wide spectrum of investigations of the behaviour and dynamic of radionuclides in agricultural systems, forests, freshwater, and marine environments. The vast majority of studies are related to radiocaesium (Cs-134 and Cs-137) although studies carried out on the initial retention of radionuclides by, and loss from, vegetation also include the short-lived I-131.

4. Key findings

For all the topics compiled in table 1, TECDOC-1927 provides detailed information on sampling, environmental and agricultural characteristics, data analysis, and quantification of transfer processes. The key findings identify features that affect radionuclide transport under the specific conditions of the affected areas of Fukushima Prefecture including local climate, landscape and terrain, agriculture practices, traditional diet, and living habits. Systematic comparisons were made between the Japanese data and that of studies carried out before the FDNPP accident in Japan and other countries. Many, but not all, of the transfer processes that were identified and quantified were similar to those reported from other parts of the world before the FDNPP accident in 2011.

4.1. Key finding 1: uptake of radiocaesium by dormant vegetation

At the time of the deposition, leaves of fruit trees had not emerged in Eastern Japan, so radiocaesium was only deposited onto bark and branches. Radiocaesium measurements in fruit in 2011 indicated that the deposition on bark was more important in determining the radiocaesium activity concentrations in fruit at harvest than the uptake via soil (Sato *et al* 2015). This process was most pronounced in 2011 but was also observed in the following years.

The same phenomenon was found in tea plants. In 2012, radiocaesium activity concentrations in tea leaves were higher than that which would arise via the uptake of radiocaesium from soil. Intercepted radiocaesium was stored overwinter in the tea plants and redistributed to the new shoots and leaves as the new growing period started (Tagami *et al* 2020c).

The uptake of radiocaesium via foliage, bark and soil depended on the stage of development of the plants at the time of deposition, the growth characteristics, the plant structure, and fruit type. After the FDNPP accident, the uptake of radiocaesium via the bark was identified as being potentially important for the radiocaesium transfer to tree leaves (needles), branches and wood. This process was identified because, in contrast to previous accidents, the deposition occurred outside of the growing season. If the deposition had taken place during the vegetation growth period, the uptake of radiocaesium via the bark and branches may not have been identified, as translocation via the foliage of fruit trees and tea plants would probably have been more important.

Table 1. Main radioecological processes and parameters addressed in the IAEA TECDOC-1927 (IAEA 2020).

Topic (reference)	Processes and parameters
Radionuclides in agricultural systems (Tagami *et al* 2020a)	Radionuclides deposited on **vegetation** • Mass interception factor f_B and interception factor f • Radiocaesium in green tea after foliar deposition • Effective half-lives for Cs-137 and I-131 on leafy vegetables and wild herbs Behaviour of radiocaesium in **agricultural soil** • Radiocaesium interception potential • Radiocaesium in rice paddies • Relationship of distribution coefficient K_d and Cs-137 uptake by rice from soil • Uptake of radiocaesium from soil by rice and other agricultural crops • Relationship of the potassium status in soil and root uptake of radiocaesium • Time trends of the uptake of radiocaesium from soil Radiocaesium in **fruit trees** • Uptake from soil • Uptake via foliar deposition • Uptake via bark during dormancy • Time trends of radiocaesium in fruit following deposition Radiocaesium in **farm animals** • Transfer to beef, pork, and chicken • Transfer to cow and goat milk
Radiocaesium in forest (Hashimoto *et al* 2020a)	• Interception and weathering of radiocaesium • Transfer of radiocaesium from tree crowns to the forest floor • Uptake of radiocaesium from soil by trees • Time trends of radiocaesium in tree needles, barks, and branches • Effect of potassium on the uptake of radiocaesium from soil • Radiocaesium in pollen • Radiocaesium migration forest soils • Uptake of radiocaesium by mushrooms and wild edible plants • Transfer of radiocaesium to game and time trends in game
Radiocaesium in catchments and rivers (Onda *et al* 2020a)	• Radiocaesium run-off in catchments • Radiocaesium in river water • Distribution coefficients K_d for radiocaesium in rivers • Uptake of radiocaesium by freshwater fish • Effective half-lives of radiocaesium in lake organisms
Radiocaesium in marine systems (Takata *et al* 2020)	• Uptake of radiocaesium by marine organisms • Time trends for radiocaesium in marine organisms • Time trends of radiocaesium in seawater and marine sediments • Distribution coefficients K_d for radiocaesium in marine sediments • Long-term observations of radiocaesium in fish and sediments covering pre- and post-accidents periods
Food and culinary processing (Tagami *et al* 2020b)	• Processing of cereals • Processing of soybeans, vegetables, fruit, and mushrooms • Preparation of tea • Preparation of meat and fish

4.2. Key finding 2: availability of radiocaesium in soil

The radiocaesium uptake by plants from soil depends strongly on soil properties such as clay content and type, exchangeable K^+ concentrations and organic matter content (Cremers *et al* 1988, Cornell 1993, Vidal *et al* 1995, Nakamaru *et al* 2007). The evaluation of the radiocaesium availability in soils and relationships between soil properties and its transfer to crops is based on the K_d at equilibrium and the concentration of exchangeable potassium. Another important linked parameter reflecting the sorption capacity and selectivity of soil for trace amounts of radiocaesium, independent of chemical composition of soil solution is radiocaesium interception potential (RIP). After the Chernobyl accident elevated radiocaesium transfer to plants were identified in some radioecologically sensitive semi-natural environments, mainly associated with soils characterised by insufficient potassium supply, low clay, and high organic matter contents and low pH, leading to a low RIP (IAEA 2010). There are no comparable environmental conditions in the areas affected by the Fukushima accident where the soils are generally well-fertilised and the soil RIPs in most contaminated areas of Fukushima Prefecture are relatively high, resulting in relatively low uptake rates of radiocaesium from soil (Nakao *et al* 2015).

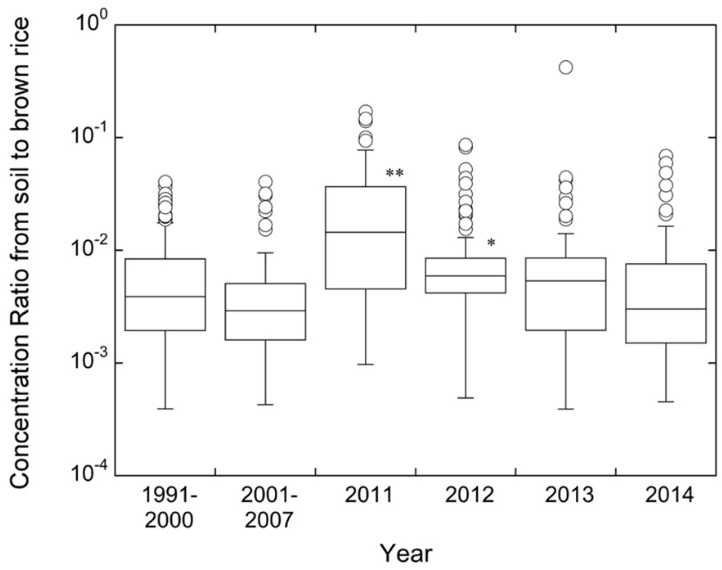

Figure 2. Comparison of the soil-to-brown rice concentration ratio before and after the FDNPP accident, for field data with normal fertiliser conditions. The upper and lower quartiles are given in the box, and whiskers show 10% and 90% values. Box with * shows ANOVA test results ($^{**}p < 0.01$ and *<0.05) (as published by Tagami *et al* 2020a). Reproduced with permission from Tagami *et al* (2020a). © IAEA, Vienna.

4.3. Key finding 3: uptake of radiocaesium by rice from soil

Japanese scientists had previously published data on radiocaesium transfer to rice that was included in recent IAEA data compilations (IAEA 2009, 2010). However, radioecological data for rice before the Fukushima Daiichi NPP accident was relatively sparse compared with other agricultural crops of similar importance as an internationally important staple crop.

As with other cereals, rice had not been planted at the time of the Fukushima deposition. The soil-to-rice geometric mean CR[10] values for radiocaesium in 2011 ranged 0.012–0.04 (DM), depending on soil and potassium supply, i.e. these were 3–10 times higher than the average CR observed in Japan prior the accident (figure 2). The uptake of radiocaesium from soil was high during the first years but tended to return to concentration ratios observed before the FDNPP accident from the third year after the accident (IAEA 2020).

A decade after the accident there is now much more information on the transfer of radiocaesium to rice, including how this varies with soil type to a limited extent. In Japan it was quickly realised that the transfer of radiocaesium to rice was highly correlated with a key factor, namely the exchangeable K^+ in the soil. The CR values for rice grown under flooded conditions in Japan can be predicted from the distribution coefficient K_d and largely depended on the concentration of exchangeable potassium in the soil solution; other soil constituents did not have a major impact on CR. Therefore, remediation for paddy fields focused on ensuring that soils had an adequate supply of potassium to ensure that radiocaesium activity concentration in rice were below the national standard limits.

Irrigation water was not a major source of radiocaesium to rice even though the irrigation ponds are often fed by run-off water from contaminated forested catchments. In 2011, during normal cultivation including flooding, puddling, irrigation, and drainage on experimental rice paddies, not more than 1% of the Cs-137 inventory was lost to the receiving surface water downstream. In the following years, when radiocaesium was mixed with deeper soil layers, and Cs-137 loss rates were below 0.05% per year (Wakahara *et al* 2014, Miyazu *et al* 2016). Most of the deposited Cs-137 will be kept in the soil for a long time.

4.4. Key finding 4: uptake of radiocaesium by other agricultural crops

From the results of monitoring programmes, mean CR values for soybeans and buckwheat derived for 2012 were 0.0034 and 0.021 with geometric standard deviations of 2.6 and 2.4, respectively. The CR values declined by up to a factor of 10 from 2012 to 2015, beyond 2015 the decline of CR was less pronounced (Li *et al* 2019).

The CR values for leafy vegetables (geometric mean [GM] = 6.9×10^{-4} kg soil DM kg^{-1} crop FM[11])and fodder grass (GM = 4.8×10^{-2} kg soil DM kg^{-1} crop FM) derived from the measurements in affected areas

[10] Concentration ratio, CR: ratio of activity concentrations in crops (DM) and soil (DM); DM: dry mass.

[11] For practical use, CR value for leafy vegetables and fodder grass was defined as ratio of activity concentrations in crops (FM) and soil (DM); FM: fresh mass.

are consistent with pre-2011 worldwide experience. The variation in CR values within soil type was high, varying by a factor of 5–50, depending on the crop. For most of the crops, the highest radiocaesium transfer was observed on brown forest soils, followed by brown lowland and andosols. Variations in CR values calculated for each type of vegetables were high and additional data on soil properties, such as potassium status, organic matter, pH, and clay content would be required to explain some of the variation.

4.5. Key finding 5: effective half-life of Cs-137 in crops

The decline in Cs-137 transfer from soil to crops could be described by exponential functions with one (fast loss) or two (fast and slow loss) components. For the first six years after the FDNPP accident, the Cs-137 uptake declined according to effective half-lives ranging from 0.4 years for pasture grass to 3.2 years for spinach. The second component followed half-lives in the order of ten years. For rice no decrease was observed beyond 2015 (Fesenko *et al* 2020b).

For soybeans and buckwheat, the first half-lives of Cs-137 CR values were estimated to be 0.76 years and 1.1, whilst the second half-lives were 5.9 and 4 years, respectively (IAEA 2020).

4.6. Key finding 6: use of potassium to deduce uptake of radiocaesium from soil

The experience gained after the accident in FDNPP in planting crops in the fields with different potassium status allowed a robust quantification of the required potassium application for reduction of radiocaesium transfer to plants (Yamamura *et al* 2018). A level of 25 mg kg^{-1} of exchangeable potassium was used after the accident at the FDNPP in Japan to identify agricultural fields which needed additional potassium fertiliser to reduce radiocaesium in rice. For buckwheat and soybeans, exchangeable potassium values of 30 mg kg^{-1} and 25 mg kg^{-1} of soil have been applied respectively.

The application of elevated amounts of potassium fertiliser to reduce radiocaesium uptake as a countermeasure was well known before the Fukushima accident and was used also in areas affected by the Chernobyl accident (International Atomic Energy Agency (IAEA) 2012). However, in contrast to Japan, often further measures were needed in addition to K enhancement such as application of phosphorus in the optimal rate to K and in some cases liming (Fesenko *et al* 2006). Similar countermeasures were implemented after the Kyshtym accident where deep ploughing was normally accompanied with addition of potassium and phosphorus (Alexakhin 2009).

4.7. Key finding 7: transfer of radiocaesium to animal products

In contrast to the Chernobyl accident, animal products were less important as a source of internal exposure after the FDNPP accident (UNSCEAR 2013). This was because animal management practices in Japan led to low radionuclide intake by animals and a relatively low importance of farm animal products in the Japanese diet. Additionally, radioiodine transfer to milk was nearly negligible as dairy animals were housed indoors and fed stored feed at the time of accident.

Live monitoring of animals which was widely applied in the Chernobyl affected areas was also implemented in Japan to a limited extent due to the relatively low radiocaesium activity concentrations in agricultural animals. Application of this techniques allowed the estimation of biological half-lives T_b of Cs-137 in cattle. A recent review of T_b data suggested that there are two components of loss for Cs with half-lives of 1.4 d (80% of total activity) and 15 d (20% of total activity) for milk and 9 d (56% of total activity) and 53 d (44% of total activity) for beef (Brown *et al* 2022).

The results of studies on the transfer of radiocaesium to milk are in general agreement with those published in the international literature before the accident at the FDNPP.

4.8. Key finding 8: radiocaesium in forest ecosystems

Shortly after the accident, intensive monitoring programmes were initiated to determine interception of radionuclides by the forest canopies and their further redistribution in the forest ecosystems. Existing experimental arrangements that could be immediately used after the accident for radioecological studies provided valuable information allowing the significant broadening of the pre-2011 experience as presented in IAEA (2009, 2010) and Calmon *et al* (2009).

Coniferous forests tended to have higher interception fraction[12] values ($f = 0.2$–0.9) than deciduous broadleaf (e.g. beech, oak) forests ($f = 0.06$–0.34) because the deposition occurred in early spring when there were no leaves on deciduous trees (Kato *et al* 2012).

Radionuclides intercepted by the canopy are lost to the forest floor via throughfall, stemflow and litterfall. The decline of the radiocaesium inventory in a tree canopy during the early phase after the accident can be

[12] Interception fraction (f): Ratio of the activity initially retained on the plant per unit area (A_i in Bq m^{-2}) and the total activity deposited on the terrestrial surface (soil plus vegetation) (A_t in Bq m^{-2}).

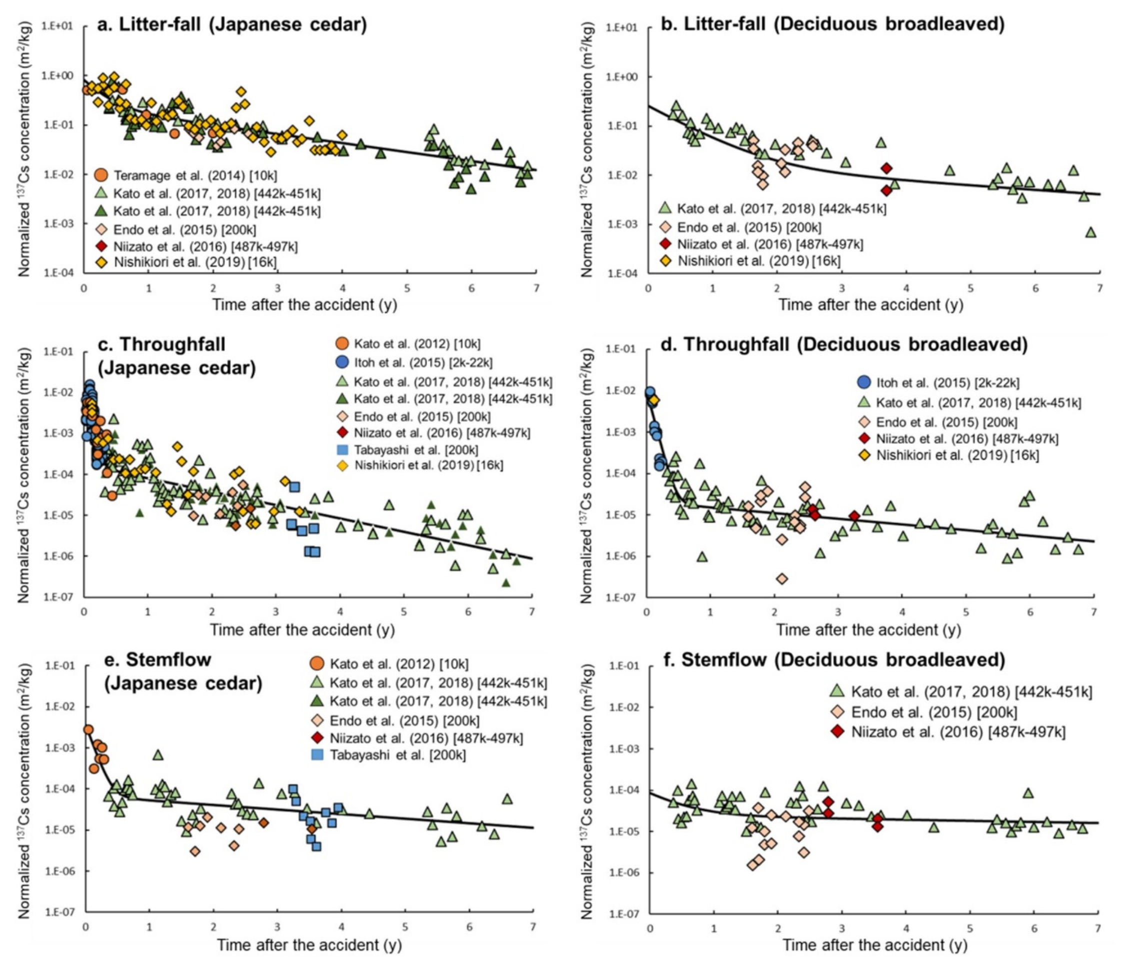

Figure 3. Time trends of the normalised Cs-137 activity concentrations in litterfall (DM), throughfall, and stemflow (as published by Hashimoto *et al* 2020a). Reproduced with permission from Hashimoto *et al* (2020a). © IAEA, Vienna.

described by exponential functions with two components. The normalised, time-dependent Cs-137 activity concentrations[13] in litterfall (DM), throughfall and stemflow are presented for the period 2011–2018 in figure 3. The time-dependence can be approximated by an exponential function with two components where half-lives for the first component were less than one year, for the second component, they ranged from 0.9 to 11 years. Stemflow represents a long-term loss process for radiocaesium from tree canopies, which is reflected in particular by the second component of the loss function (Hashimoto *et al* 2020b).

Aggregated transfer factors (T_{ag})[14] determined for wood for cedar, cypress, pine, and oak ranged from 10^{-4} to 10^{-3} m^2 kg^{-1} in 2011–2015 and T_{ag} values were time dependent. The T_{ag} values for needles, bark, and branches were a somewhat higher than those for wood. The distribution of radiocaesium in heart- and sapwood varies with time and depends on tree species (Ohashi *et al* 2020).

The decline of radiocaesium activity concentrations of coniferous needles and pine branches followed exponential functions with half-lives of 1–3 years and 1–4 years, respectively. Two years after deposition, the forest soil became the largest radiocaesium reservoir in the forest. The vertical distribution of radiocaesium in soils established in the first year changed slowly thereafter (Imamura *et al* 2020).

The T_{ag} and f values as well as other parameters used to describe the distribution of radiocaesium in forest trees are by and large similar to those observed after the Chernobyl accident, if similar periods after the deposition are compared. However, the range of T_{ag} values for wood of coniferous trees affected by the FDNPP accident are lower than those reported for the Chernobyl accident. The processes related to the interception factor, f and loss could not be studied after the Chernobyl accident, since the sampling programme did not cover the early period when these processes are relevant.

[13] Activity concentration in litterfall (Bq kg^{-1} DM), throughfall (Bq l^{-1}) and stemflow (Bq l^{-1}) normalised to the total Cs-137 activity deposited per unit area (Bq m^{-2}).

[14] Aggregated transfer factor (T_{ag}): Activity concentration (Bq kg^{-1}) in a specified component per activity per unit area (Bq m^{-2}).

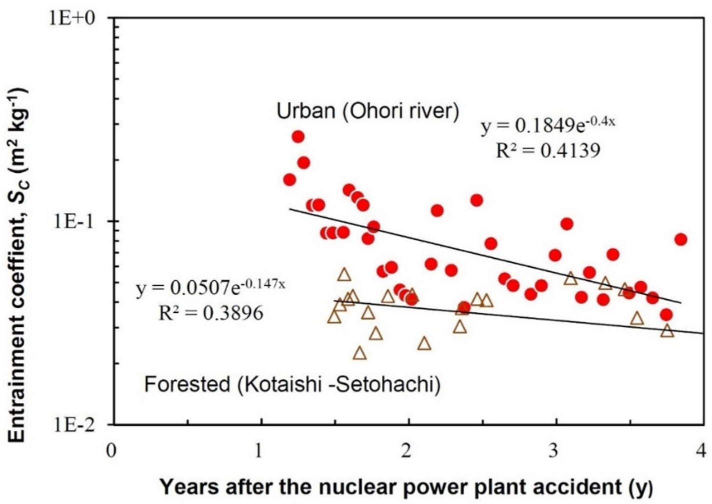

Figure 4. Comparison of the time-dependence of suspended sediments in run-off water in urban and forested catchments (Entrainment coefficient: ratio of Cs-137 activity concentration in suspended sediments of run-off water and Cs-137 deposition density) (as published in Onda *et al* 2020a). Reproduced with permission from Onda *et al* (2020a). © IAEA, Vienna.

Radiocaesium monitoring also included a wide range of mushrooms and edible wild plants. The T_{ag} values varied by about 100 times between species, although for most species these values ranged from 0.01 to 0.2 m^2 kg^{-1} DM. In general, mycorrhizal species had higher T_{ag} values than other types of mushrooms. The T_{ag} values for edible wild plants ranged from 0.0008 to 0.03 m^2 kg^{-1} DM. No clear time trend was observed for Cs-137 concentration in both mushrooms and wild plants. Effective half-lives of radiocaesium after the Chernobyl accident were much longer in forest products, such as mushrooms, game, and berries than those for agricultural products (Pröhl *et al* 2006). It is not yet clear whether the same will be found for Fukushima forests.

Values for T_{ag} for Sika deer and wild boar in Japan, were similar to those reported for other terrestrial game mammals in Europe. The T_{ag} geometric mean values for Sika deer, wild boar, and Asian black bear are all in the range of 0.003–0.008 m^2 kg^{-1} FM. Values for T_{ag} for green pheasant and wild ducks are in the range 0.0001–0.001 m^2 kg^{-1} FM, for green pheasant T_{ag} values in the range of 0.0016–0.005 m^2 kg^{-1} FM were observed (Hashimoto *et al* 2020a).

4.9. Key finding 8: radiocaesium transport in catchments

Run-off and erosion process cause a continuous redistribution of radionuclides in terrestrial and freshwater environments. Run-off depends on topography, land-use, and the amount precipitation.

Erosion losses increase with increasing fractions of rice paddies, farmland, and residential areas in a catchment. The activity loss with erosion decreases in the order bare farmland > vegetated farmland > grassland and forest. Except for continuously bare farmland, in general, the removal of radiocaesium is a process of minor importance.

Immediately after the radionuclide deposition, radiocaesium run-off is most pronounced. In a catchment with predominantly residential areas (more than 50%), run-off was initially a factor of 3 higher than in forested areas due to the retarding effect of the vegetation cover (figure 4).

In an urban catchment, from 2011 to 2015, the loss of Cs-137 due to run-off declined with an effective half-life of 1.5–2 years. Decontamination activities in catchments areas may cause a higher run-off of soil and the attached Cs-137. On forested steep slopes, erosion losses ranged from 0.001% to 0.2% per year, which is at least a factor of 10 lower than the decline of activity due to physical decay (2.3% per year) (Iwagami *et al* 2017).

In general, the loss of radiocaesium from a catchment does not modify substantially the radiocaesium inventory in catchments. However, the loss is not homogeneous over the catchment; in some parts of a catchment, the loss may be considerable, whereas other parts may not be affected by erosion at all. The same is true for the landscape element that receives the activity lost. The total inventory of the receiving landscape element may not change significantly, but locally, activity concentrations may be modified considerably.

4.10. Key finding 9: radiocaesium in rivers

Radiocaesium activities in rivers and the water column of lakes declined quickly due to dilution and sedimentation of radiocaesium attached to suspended sediments. One consequence of this phenomenon was that irrigation water used in paddy fields was not a major source of radiocaesium to rice.

The time-dependence of Cs-137 in river water can be described by exponential functions with one to three components representing a fast, intermediate, and long-term phase after deposition. The fast component could not be determined as the measurements started only on 2012 in the tributaries. From 2012 to 2016, the effective half-lives of the Cs-137 decline in suspended sediments of 48 rivers ranged from 0.7 to 16 years. Forty-two values were between 1.1–4.6 years (Taniguchi *et al* 2020, Onda *et al* 2020b).

By and large, the time trends observed in Japan agree reasonably well with pre-2011 experience (Onda *et al* 2020a).

4.11. Key finding 10: radiocaesium in freshwater fish

A clear decline of radiocaesium activity concentrations in fish, aquatic biota and water has been observed since the FDNPP accident in 2011. The GM of CR[15] values in 2015–2017 ranged from 650 to1800 $\mathrm{l\,kg^{-1}}$ for river fish and 1000–4000 $\mathrm{l\,kg^{-1}}$ for lake fish. In lakes, radiocaesium activity concentrations in fish increased with the trophic level (Ishii *et al* 2020).

The time-dependence of Cs-137 in aquatic organism can be approximated by exponential functions with effective half-lives that range from 1.1 to 2.5 years, depending on the species. The GM of CR values for freshwater fish are in general agreement with global experience and observations made in Japan before the FDNPP accident (Onda *et al* 2020a).

4.12. Key finding 11: modification of radiocaesium levels in food during processing and culinary preparation

The modification of radiocaesium levels in crops and food during processing and cooking was studied in detail after the accident in FDNPP. The results are in general agreement with pre-2011 data. The modification of radiocaesium levels during preparation of bamboo, wild plants, and some mushroom species as well as the production of sake was studied for the first time (Tagami *et al* 2020b). By comparison with leafy vegetables, more radiocaesium is retained after washing and boiling thick pieces of wild plants, such as bamboo or fern shoots. Making tempura from edible plants does not remove caesium from the product. For sake (Japanese rice wine), more than 90% of radiocaesium was removed from its raw product, that is, brown rice.

4.13. Key finding 12: radiocaesium in marine systems

The scientific value of long-term observations is demonstrated by the results of the seawater monitoring programmes that have been carried out since the 1960s. Before 2011, under equilibrium conditions, concentration ratios of Cs-137 for marine organisms ($\mathrm{l\,kg^{-1}}$ FM) were determined for a wide spectrum of species in a range of approximately 10–100 $\mathrm{l\,kg^{-1}}$ FM (Takata *et al* 2020). Ongoing input of Cs-137, an inhomogeneous distribution of Cs-137 in seawater and the different morphological and physiological characteristics of marine organism are reasons for the variation of CR.

Due to the short-term input of radiocaesium during the accident in FDNPP, equilibrium in sea water was distorted and the apparent activity concentration ratio[16] (CR_a) values for Cs-137 increased rapidly (figure 5). The peak $\mathrm{CR_a}$ values were observed in 2012–2013 and the decline to pre-2011 $\mathrm{CR_a}$ values followed exponential functions with effective half-lives in the range of 0.5–3.3 years. The currently observed CR-values are in good agreement with pre-accident data; only in Areas I several values are significantly enhanced as a consequence of the continued releases to the sea (figure 5).

Before 2011, equilibrium was established for values of the distribution coefficient K_d for marine bottom sediments. In the coastal waters around Japan, the median and geometric mean K_d values of 720 and 920 $\mathrm{l\,kg^{-1}}$, respectively were determined (Kusakabe and Takata 2020).

Pre-2011 activity concentrations of Cs-137 in seawater were achieved approximately 8 years after the accident in 2019, except in the area close (∼30 km or less) to the accident site. The decline of Cs-137 activity concentrations in bottom sediments is slower and the concentrations are still significantly higher than before the accident. It is expected that the Cs-137 level in the bottom sediments will continue to decline due to re-suspension and subsequent lateral transport of the sediment, dissolution of Cs-137 from the sediment, and vertical migration of radiocaesium in the sediment. The geometric mean of the apparent $K_{\mathrm{d(a)}}$[17] values determined in 2018/2019 were in the range of 5000–7000 $\mathrm{l\,kg^{-1}}$.

[15] Concentration ratio for aquatic biota, CR: ratio of activity concentrations in aquatic biota (FM) and surrounding water (unit: $\mathrm{l\,kg^{-1}}$ FM).

[16] Apparent activity concentration ratio water-aquatic organism under non-equilibrium conditions (unit: $\mathrm{l\,kg^{-1}}$ FM).

[17] Apparent distribution coefficient: K_d-value water bottom-sediment under non-equilibrium conditions (unit: $\mathrm{l\,kg^{-1}}$).

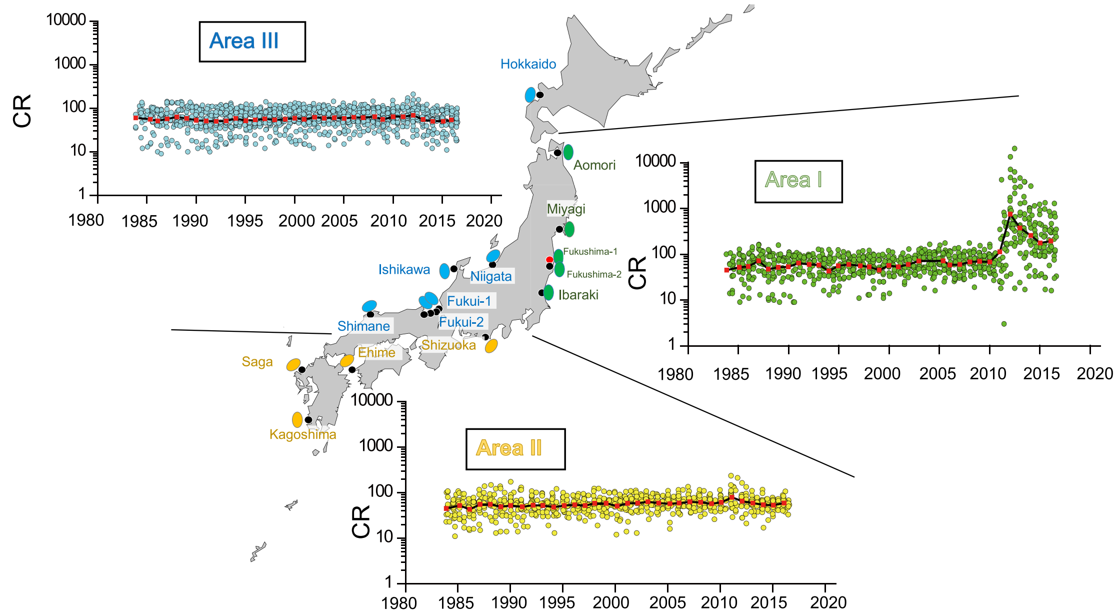

Figure 5. CR values (l kg^{-1} FM) of Cs-137 for marine species based on three areas classified according to prevailing seawater currents. Red squares represent annual geometric means of CR values in each area. Values prior to 2010 relate to global fallout and those after 2010 are CR_a values (as published in Takata *et al* 2020). Reproduced with permission from Takata *et al* (2020). © IAEA, Vienna.

5. Conclusions

Two main features of the marine and atmospheric releases resulting from the FDNPP accident were fundamentally important aspects of the overall impact of the accident. A large proportion of the releases drained into, or were deposited on, the marine waters around Japan. The activity concentration of radionuclides in the marine areas were greatly diluted by the receiving water mass. The interpretation and analysis of the behaviour of radiocaesium released into the Pacific Ocean was facilitated by the availability of previous long-term data from systematic monitoring that has been carried out for many years in offshore areas of Japan. The activity concentrations of Cs-137 in sea water had returned to approximately pre-accident values by 2019.

After the Chernobyl accident, in some part of Europe, the podzolic soils in parts of Belarus, Russia, Ukraine and highly organic soils in upland areas of Western Europe, the exchangeable potassium levels, pH-values and clay content of these soils were low causing a high caesium availability of soil (International Atomic Energy Agency (IAEA) 2006) allowing enhanced uptake rates of radiocaesium that persisted over decades. Fortunately, in Japan the high fertility of the soils in the affected areas and the intensive agriculture associated with a sufficient potassium supply prevented high uptake of radiocaesium from soil to crops.

Remediation of agricultural soils after both accidents included cultivation methods, such as ploughing, deep ploughing, and removal of the upper soil layer in some areas. After the Chernobyl accident, further reduction of radiocaesium activity concentrations in crops required based on a multi-factored approach, which took into account the pH, soil texture, and fraction of organic matter. The remediation strategies for agricultural crops in Japan involving soil treatments were successfully and mainly focused solely on the exchangeable potassium status of the soil.

In Japanese forests, as seen after the Chernobyl accident, trees effectively captured radionuclides deposited during the passage of the radioactive plumes; and radiocaesium rapidly migrated to soils in the first 2 years.

Radiocaesium activities in rivers, and in the water column of lakes declined quickly due to dilution and sedimentation of radiocaesium attached to suspended sediments. The concentration factors for water-fish for marine and freshwater fish agreed well with pre-2011 experience.

Immediately after a release, the flux of radionuclides between environmental compartments is rapid and intense. Typically, as occurred after the Chernobyl accident, early estimates of accident consequences are largely based on radioecological models, which are subject to considerable uncertainties. Data was collected in the first few days after the FDNPP accident, utilising previously installed monitoring equipment used for other purposes. The use of such facilities for monitoring purposes or experimental studies provided an invaluable advantage as it enabled the collection of data for the initial fast transfer processes after deposition such as the interception of radionuclides by plant canopies, and the loss of radionuclides from plants due to weathering processes.

In the aftermath of nuclear accidents, linking radioecological models with early monitoring data obtained under relatively well-controlled conditions from existing experimental setups is a powerful tool for improvement of the reliability of predicted consequences. In addition, such early data helps prioritise the use of limited monitoring resources.

We would like to emphasise that international collaboration was very important to deepen the understanding of the fate of radionuclides in the environment in the aftermath of the accident; it provided smooth and easy exchange of knowledge and experience. The comparison of the observation with the results elaborated after the Chernobyl accident provided an invaluable input to evaluate the behaviour of radionuclides in the environment after the Fukushima accident.

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ORCID iDs

Keiko Tagami https://orcid.org/0000-0002-9139-4441
Shoji Hashimoto https://orcid.org/0000-0003-3022-7495
Masashi Kusakabe https://orcid.org/0000-0001-5304-0944
Yuichi Onda https://orcid.org/0000-0001-8753-0378
Sergey Fesenko https://orcid.org/0000-0003-1238-3689
Alexander Ulanowski https://orcid.org/0000-0003-4863-4128

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