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**Radioecological data evaluated from recent  
international meetings in Japan:**

**The 13<sup>th</sup> International Conference on the  
Biogeochemistry of Trace Elements**

**Fukuoka International Congress Centre,  
Fukuoka, Japan, 12<sup>th</sup> – 16<sup>th</sup> July 2015**

**Fukushima COMET Workshop**

**Yoshikawaya inn, Iizaka,  
Fukushima, Japan, 18<sup>th</sup> – 19<sup>th</sup> July 2015**

Editor(s): Brenda J. Howard, Hirofumi Tsukada, Christine Balmer

Authors: B.J. Howard, H. Tsukada, A. Takeda, C. Adam, P. Hurtevent, G. Shaw, S. Uchida, K. Tagami, S. Eguchi.

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(D-N°:5.4) – ICOBTE 2015 & Fukushima COMET Workshop

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## Executive Summary

The second COMET workshop comprised three events in July 2015. There was a session in the 13<sup>th</sup> International Conference on the Biogeochemistry of Trace Elements (ICOBTE 2015), held at Fukuoka, an associated excursion with 26 participants to contaminated areas near the Fukushima Daiichi NPP and a COMET Fukushima Workshop was held at Iizaka. The Symposium was on “Understanding and mitigating the environmental behaviour of radiocaesium after the Fukushima accident”. There were 73 presentations, including 25 posters, from 9 countries. The COMET Fukushima workshop, organised by the Institute for Environmental Radioactivity, Fukushima University, had 42 participants from 6 countries. The speakers covered a wide range of topics and summarised current key findings and issues.

A very large research effort is ongoing with many different research institutes and universities involved. A great diversity in applied and developed experimental and modelling methodology is evident which will lead to significant advances in understanding of radiocaesium behaviour. Comparisons of contrasting data between studies need to consider whether fundamental behavioural differences are being observed or if differences are due to different methodologies. Important field observations, often at early stages after the accident, could be complemented by laboratory studies to elucidate mechanisms and will facilitate the development and validation of mechanistically based predictive models.

Information on radionuclide deposition levels alone is not enough for accurate predictions and dose assessment. Data on speciation in fallout, rates of transformation processes and site-specific environmental characteristics determining these rates, such as RIP, mineralogy and competing ion concentrations, are needed. Comparison of RIP with reference to contrasting mineral characteristics between Japanese and European soils would deepen our understanding of radiocaesium behaviour in forest and agricultural soils, including irrigated paddy fields. A long-term monitoring program should include these variables as well as measurements of radionuclide activity concentrations to enable improvements to be made in quantifying radiocaesium mobility and making generically applicable predictions.

The potential for contaminated sediment to act as a long term source for Cs in rivers, catchments, coastal areas, coastal marine systems and Open Ocean will need to be evaluated over a long time period. This will underpin long-term consideration and development of remediation strategies and decision making.

Transfer modelling is sensitive to ecophysiological parameters and appropriate parameterisation and characterization is crucial to reasonably constrain the associated uncertainty of modelling outputs. Therefore, the key factors in each type of contaminated terrestrial (agricultural land, residential areas, forest et.) and aquatic (freshwater, coastal, marine) environment need to be well characterised. The controlling transfer processes need to be quantified and predicted models validated for processes such as soil-to-plant systems, irrigation water, sediment to aquatic organisms and diet-to-animals. Information on how radiocaesium mobility and bioavailability for uptake changes with time should be taken into account when rehabilitation and decontamination strategies are developed on a local or regional scale.

Comparison with Chernobyl shows similarities and also differences which will need to be documented and incorporated into international literature, understanding and models.

## List of Acronyms and Abbreviations

CEC	Cation Exchange Capacity
[ <sup>137</sup> Cs]	Activity concentration of <sup>137</sup> Cs
FES	Frayed Edge Sites
FDNPP	Fukushima Daiichi Nuclear Power Plant
ICOBTE 2015	International Conference on the Biogeochemistry of Trace Elements, 2015
ICSA	Intensive Contamination Survey Area
LL	Litter layer
OM	Organic Matter
[RCs]	Activity concentration of radiocaesium, may include <sup>134</sup> Cs + <sup>137</sup> Cs
RIP	Radiocaesium Interception Potential
SDA	Special Decontamination Area
TF	Transfer factor soil-plant
ThF	Throughfall

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## 1. Introduction

The overarching objective of Work Package 5 of COMET is to enhance and maintain European capacity and skills in radioecology by establishing a dynamic interaction promoting effective collaboration between researchers, tool developers, regulators and industry. This Work Package ensures that COMET has measures in place to achieve effective communication with the wider community.

Task 5.2 is devoted to running a series of workshops on key radioecology related issues and, in doing so, bring together the different communities involved in radiation protection.

The second in this series of workshops constituted three different activities. These were (i) A special session in the 13<sup>th</sup> International Conference on the Biogeochemistry of Trace Elements, held at the Fukuoka International Congress Centre on 12<sup>th</sup> – 16<sup>th</sup> July 2015 (ICOBTE 2015) (ii) an associated excursion on 17<sup>th</sup> July 2015 in the region affected by the Fukushima accident, and (iii) the COMET Fukushima Workshop at Iizaka, Fukushima, on the 18<sup>th</sup> – 19<sup>th</sup> July 2015.

This deliverable from the meetings in Japan describes each of the three activities, summarises their content and considers the main messages for both Japan and the wider international community. For brevity, abbreviations of common terms - radiocaesium (RCs) <sup>137</sup>Cs activity concentration, transfer factor (TF) throughfall (ThF) litter layer (LL) etc. - are used throughout most of the document. A very large number of scientists contributed to the talks and posters summarised in this document. The first author only is given and the full authorship lists can be found either on the ICOBTE web site and/or in this document.

## 2. The 13th International Conference on the Biogeochemistry of Trace Elements

### 2.1. Introduction to ICOBTE 2015

The 13th ICOBTE took place at the International Convention Centre in Fukuoka, Japan, and included invited Special and Plenary Speakers who delivered talks about mercury, cadmium and radiocaesium research. The conference included thirteen Special Symposia, and Technical Sessions on all aspects of trace element research, organised by leading experts in their fields. There were special sessions on trace element issues of particular relevance to Japan, including mercury and radioactive element contamination and a special session on radioactive contaminants from the nuclear accident at the Fukushima nuclear power plant.

As with past conferences, ICOBTE 2015 offered an opportunity for professionals and students in academic and industrial fields to come together and exchange current information on topics related to the biogeochemistry of trace elements. The main themes of the conference were:

- Trace element biogeochemistry and environmental sustainability
- Natural abundance and behaviour of trace elements in the environments
- Trace element contamination, environmental and human health impacts
- Advances in toxicology and risk assessment
- Regulatory and policy dimensions of trace element contamination



- Advances in remediation technologies for trace element contaminated sites
- New techniques to study the fate of trace elements in the environment
- Heavy metal and radioactive element contamination
- Transnational transport of trace elements and pollutants

The conference was attended by 461 delegates representing a large number of countries and organisations. Detailed documents showing the full agenda for ICOBTE 2015 are in Annex 1.

There were 396 presentations, including four Special and Plenary Lectures and two Luncheon Seminars. The majority of the presentations were made as part of the special symposia, listed with associated PDF documents in Annex 2. Two sessions included presentation on the Fukushima Daiichi accident.

#### [Understanding and mitigating the environmental behaviour of radiocaesium after the Fukushima accident](#)

Chair: Brenda J Howard and Takayuki Takahashi. This session will be discussed in detail below.

#### [Behaviour of NORM and artificial radionuclides in the environment](#)

Chair: Robin L. Brigmon, Cristina Negri and Daniel I. Kaplan

This session was organised by R. Brigmon (SRNL) and C. Negri (ANL) and D. Kaplan (SRNL). Several reports on the Fukushima Daiichi accident were included in this session. The remit for the special session was as follows:

*Naturally-Occurring Radioactive Materials (NORM) and artificial radionuclides in the environment are a concern due to the potential for human exposure. This exposure may originate from contaminated air, water, or soil, and the pathway may be direct (“shine”), inhalation, or ingestion. As a result of man’s activities, NORM becomes more concentrated and is brought into the biosphere. Similarly, as a result of energy generation and industry, more artificial radionuclides are being created that eventually enter the biosphere. Remediation is commonly required to mitigate exposure. However, a key knowledge-gap in the implementation of remediation is our understanding of the fate and transport of radionuclides and their interaction with biological systems. This knowledge-gap hinders our ability to predict accurately risk posed by contaminated systems. It also hinders our ability to determine to what degree a contaminated site must be remediated: to pristine levels, background levels, or some acceptable risk level. This knowledge-gap became especially evident while trying to make informed decisions related to the aftermath of the Fukushima Nuclear Power Plant incident. Risk calculations must be based on detailed understand of the fate and transport of radionuclides and their interaction with biological systems.*

*This symposium dealing with NORM and artificial radionuclides covered: 1) biogeochemistry, 2) mobility, 3) characterization, 4) risk and transport modelling, and 5) remediation. Particular attention was directed at discussions related to applying scientific results to engineered solutions and informed decision making. Presentations dealt with low-cost, low-impact remediation approaches, and data or approaches to support Monitored Natural Attenuation and long-term stewardship of subsurface radiological waste-disposal facilities.*

The main themes were:

1. Source characterization of radionuclides in the environment
2. Dynamics of radionuclides in soil, water, microbial, and plant systems
3. Prediction and modelling of radionuclide fate in natural environment
4. Analytical techniques and speciation studies
5. Impact of radionuclides on agriculture and the food chain
6. Radionuclides in contaminated areas: case studies
7. Remediation and management of radionuclide contaminated soils and groundwater
8. Stewardship of radionuclide-contaminated systems or subsurface waste-disposal sites

## **2.2. Special Symposium 5 - Understanding and mitigating the environmental behaviour of radiocaesium after the Fukushima accident**

A special session was accepted on “Understanding and mitigating the environmental behaviour of radiocaesium after the Fukushima accident” (Special Symposium 5). This session was organised by B. J. Howard (CEH) and T. Takahashi (Fukushima University). The remit for the special session was as follows:

*Radiocaesium is one of the most important, environmentally mobile radionuclides. It has been released into aquatic and terrestrial environments during the operations of nuclear power plants, atmospheric weapons testing, nuclear accidents and other incidents. The importance of radiocaesium as the key, mid-long term radionuclide after the Chernobyl accident stimulated an intensive research effort to improve understanding of its environmental behaviour. Key features of the environment leading to high rates of transfer and elevated doses include the high bioavailability of radiocaesium in some soils, and the high rates of transfer to game animals and mushrooms. Methods to quantify radiocaesium mobility in soils have been developed with the Radiocaesium Interception Potential approach now being widely adopted.*

*Based on the extensive research, models of radiocaesium behaviour, its transfer in food chain and food web, and the consequent doses to humans and other organisms, have been greatly improved and partially validated. The quantification of radiocaesium behaviour has progressed from the use of empirical ratios to describing the mechanisms determining transfer in some models. In addition, an extensive suite of countermeasures for the emergency phase and for remediation in the post-accident recovery phase have been developed.*

*In 2011 and 2012 many Japanese scientists (with the assistance of other nationalities) were intensively involved in mitigation after the TEPCO Fukushima Daiichi accident. It was clear that there are some transfer pathways leading to significant contamination of certain foodstuffs which were not anticipated. It was also evident that although radioecological data largely collected in European countries was relevant for the situation in Japan, there were site specific features which had not previously been considered. From 2013 more focus has been possible on studying the environmental behaviour of radiocaesium in Japan to develop appropriate understanding to underpin the sustainable use of contaminated ecosystems and to assist remediation efforts. Particular challenges have been identified for contaminated forest*

*ecosystems, which dominate the contaminated areas, and the marine environment near the Fukushima Daiichi site.*

*.....The timing of the 13<sup>th</sup> ICOBTE conference, and its location, is an ideal opportunity to discuss progress in our understanding of the particular features of the environmental behaviour of radiocaesium and to identify lessons learned for the international community after the Fukushima accident. It will also provide the opportunity to consider if the Strategic Research Agenda for radioecology developed by the European Radioecology Alliance and research priorities in different countries adequately cover identified research challenges.”*

The main themes of Special Symposium 5 were:

1. Estimation of radionuclide deposition and redistribution in Japan after the Fukushima accident and associated external doses
2. Analytical techniques and speciation studies of radionuclides
3. Dynamics of radiocaesium in soil, water, plants and animals
4. Quantification and modelling of the short- and long-term fate of radiocaesium in agricultural, forest and aquatic ecosystems
5. Impact of radiocaesium on agriculture and the food chain and associated internal doses
6. Estimation of radiation doses to, and associated impact on, biota
7. Countermeasures and remediation after the Fukushima accident

There were 73 abstracts submitted for the session which was the greatest number for any of the special sessions. These were evaluated by the COMET organising committee (B Howard, H. Tsukada, H. Vandenhove, A. Liland and V. Kashparov) and revisions requested for about half of the abstracts. Due to the large number of abstracts, the session lasted over the entire duration of the conference rather than being limited to one day as originally expected. This meant that 44 oral presentations were delivered over 3.5 days plus 25 poster presentations. The main aim was to provide an international forum for a comprehensive consideration of radioecological data arising out of the Fukushima accident, to encourage good discussion, to identify important issues arising and to consider possible papers for a special issue.

There were 73 presentations, including 25 posters, from 10 countries (UK, Ukraine, Norway, Sweden, Russian Federation, Japan, Germany, France, Belgium and Australia). Contributors included leading environmental radioactivity researchers as well as scientists at earlier stages of their career. A list of participants is provided in Annex 3. Professor George Shaw of Nottingham University, UK, gave Plenary Lecture 2 entitled “Radiocaesium behaviour and impact – what have we learnt from atmospheric weapons fallout, Chernobyl and Fukushima?” (Annex 4).

#### 2.2.1. Synopsis of session 5 presentations

The six morning and afternoon sections of session 5 were each chaired by two invited chairpersons (Table 1).

Table 1. Chairpersons for Special Symposium 5

Date	Morning Session	Afternoon Session
Monday July 13 <sup>th</sup> 2015		<ul style="list-style-type: none"> <li>• Brenda J. Howard</li> <li>• Hirofumi Tsukada</li> </ul>
Tuesday July 14 <sup>th</sup> 2015	<ul style="list-style-type: none"> <li>• Aleksei Konoplev</li> <li>• Akira Takeda</li> </ul>	<ul style="list-style-type: none"> <li>• Frederic Coppin</li> <li>• Sadao Eguchi</li> </ul>
Wednesday July 15 <sup>th</sup> 2015	<ul style="list-style-type: none"> <li>• George Shaw</li> <li>• Atushi Nakao</li> </ul>	<ul style="list-style-type: none"> <li>• Christelle Adam-Guillermin</li> <li>• Seiya Nagao</li> </ul>
Thursday July 16 <sup>th</sup> 2015	<ul style="list-style-type: none"> <li>• Thomas G. Hinton</li> <li>• Yuichi Onda</li> </ul>	

The sessions were generally focused on one to three topic areas so, for clarity of presentation, summaries of the different topic areas, prepared with inputs from the chairpersons, are given below (with only the first author listed).

#### 2.2.2.1. Radiocaesium in soil – bioavailability, RIP, K<sub>d</sub>, distribution

In an invited review paper, **Yamaguchi** (Japan) noted that the vertical migration rate of RCs in Fukushima soils, which include Andosols, seem to be higher than in Chernobyl soils for the early phase of contamination. The RIP, of which measurement seems to be becoming an essential requirement for the study on RCs transfer in soil and to plant, could be a strong parameter to explain the observed large variations in the distribution coefficient (K<sub>d</sub>) and transfer factor (TF) based on positive RIP-K<sub>d</sub> and negative RIP-TF correlations. The link between RIP and gravimetric clay fraction in soil is not clear, except for a narrow range of soils with similar clay mineralogy studied so far. Empirical models or equations to estimate RIP from basic soil parameters would be necessary for wide application of RIP.

**Nakao** dealt with RIP measurements for 97 paddy soils from three major regions of Fukushima Prefecture looking for relationships of RIP with basic soil characteristics, i.e. total C content, clay content, pH, CEC, phosphate absorption coefficient, and the clay fraction K content. Correlation analysis revealed that RIP<sub>soil</sub> positively correlated best with clay-K<sub>soil</sub> and negatively correlated with total C content and the phosphate absorption coefficient. The data confirmed that RIP for paddy soils in Fukushima was mainly controlled by micaceous clay minerals in soil.

**Vandenhove** measured RIP in 88 surface soils from the International Soil reference Information Centre. RIP varied from 1.8 to 13300 mmol kg<sup>-1</sup>, with the lowest values for andosols, podzols and ferrasols. RIP was only loosely correlated with other soil characteristics but was linked to the acid-extractable fraction. An estimation of plant available RCs in soil from an easy acid extraction seems to be possible.

**Uematsu** measured the RIP and other characteristics of 51 surface soils from arable fields in the areas contaminated by the Fukushima accident. A model was then developed to describe the data. Pot trials with 22 of these soils using paddy rice or rye grass are now underway. RIP ranged from 80 to 4000 mmol kg<sup>-1</sup> and the linear regression with clay content was 3.3 fold lower than that for Belgian soils. This suggests a lower <sup>137</sup>Cs selectivity on amorphous minerals

typical of Japanese andosols compared with micaceous minerals. Therefore, the Absalom model may overestimate RIP and thereby underestimate RCs uptake by plants in Japan. Soil OM and CEC explained 64% of the variance in RIP for the Japanese soils.

**Koshikawa** gave data on the extractability of RCs using CsCl which extracted less RCs from soil than ammonium acetate/nitrate. The extraction yield did not change from using 0.1 and 1 mol L<sup>-1</sup> CsCl suggesting that a large excess of stable Cs ions may inhibit the release of some forms of <sup>137</sup>Cs which are removed by ammonium acetate or nitrate. If the stable Cs collapses frayed edge sites (FES) thereby trapping <sup>137</sup>Cs ions, and therefore only extracts <sup>137</sup>Cs on planar sites, the method may therefore be of use in quantifying the planar sites capacity for “readily available <sup>137</sup>Cs”.

**Stepina** considered a flow-through method of measuring radioactivity in the solid phase that has been used to study fixation of <sup>137</sup>Cs in Fithian illite. The fixed fraction of <sup>137</sup>Cs was a function of [K+], aging time and temperature. At low [K+] the rate of <sup>137</sup>Cs fixation decreases but <sup>137</sup>Cs binding strength increases. It is important to consider realistic [K+] in the environment when determining the efficacy of K amendment of soils to reduce <sup>137</sup>Cs uptake to plants.

**Takahashi** looked at temporal changes in <sup>137</sup>Cs distribution in soil profiles from different land use types after the Fukushima accident on four occasions at eight sites. They reported that migration of <sup>137</sup>Cs from litter to soil was faster at grassland sites than forest sites, but seemed to be strongly bound in the soil thereafter. In mixed and cedar forest, the proportion in litter declined from c. 90% to 18-41% over two years; in mature cedar additional inputs occurred from the canopy which led to greater spatial variability. In agricultural fields only a small percentage was in litter or plants. Vertical migration occurred in the paddy field with c. 25% in the 5-10 cm depth after two years. The initial distribution was influenced by soil Cs fixation and penetration via water and soil particles

#### 2.2.2.2. Radiocaesium uptake in paddy fields

**Eguchi** reported on the application of a simplified process-based model of RCs soil-plant transfer by Absalom et al 1999 for paddy rice in Japan for short to long-term. Without calibrating any soil and crop parameters, the model underestimated the brown rice TF by one or two orders of magnitude. Calibrating model crop and soil parameters allowed substantially improved predictions. Furthermore, by modifying the model to take into account soil clay mineralogy, the model prediction was considerably improved, with an R<sup>2</sup> value of 0.89.

**Saito** quantified the effect of K fertilizer application (using KCl or K<sub>2</sub>SiO<sub>3</sub> which is a slow release fertilizer) on <sup>137</sup>Cs in soil solution and brown rice from a fluvisol in the north of Fukushima prefecture. KCl suppressed <sup>137</sup>Cs uptake more effectively than the slow release fertilizer. [K] in the soil solution declined considerably over the growing season for controls and slightly less so for treatments. KCl applied after puddling resulted in higher [K] in soil solution than when applied before puddling, possibly because in the latter case K on the soil surface is more readily removed via paddy water.

**Kondo** compared the dynamics of RCs and potassium in soil – plant in rice. The variation of RCs accumulation in rice could be partly explained by RCs supplying capacity of soil reflecting the competitive manner of the uptake of RCs against K. Exchangeable RCs /K ratio was suggested as a potential soil index to estimate RCs accumulation in rice. The distribution of

RCs into grain increased when the [K] in shoots was low indicating that transfer of RCs is competitive with K for both soil to plant and shoot to grain. Indica varieties of rice tended to have higher RCs accumulation compared with japonica varieties. These varietal differences tended to magnify in soils with low exchangeable K.

**Yoshikawa** considered the RCs balance in mountainous paddy fields in Fukushima prefecture where [RCs] in brown rice exceeded permissible levels in 2011. Two-year monitoring of three paddy fields, measuring inflows and outflows, suggested that the RCs inventory will decline by 50% and 80% by 5.5 and 29.6 years respectively, assuming a constant net flow of RCs from soil. As RCs is immobilised with time on clay minerals the levels in brown rice is expected to decline further with time.

### 2.2.2.3. Radiocaesium in forest ecosystems

The invited paper by **Gonze** was aimed at development of comprehensive understanding of dose rate monitoring data in Fukushima forests. Airborne and car-borne surveys in evergreen coniferous regions of the area revealed that average radiation levels decreased much faster than expected from physical decay. Coniferous forest in the southwestern part of the Abukuma Mountains have been predominantly contaminated by dry deposition (60 to 100%). As a consequence of the biological and physical depuration of forest canopies, simulated dose rates were shown to decrease by 25-40% during first 18 months, in close agreement with observations.

**Nishina** presented the modelling of  $^{137}\text{Cs}$  and C dynamics in an artificial forest ecosystem using the FoRothCs open-source model. This model simulates  $^{137}\text{Cs}$  and C:  $^{137}\text{Cs}$  in leaves and litter, which enables to link it with herbivores and soil decomposer diet. The model couples a carbon cycling model and a biomass growth model,  $^{137}\text{Cs}$  dynamics are integrated into C cycling using transfer parameters. The process-based model will help to plan optimal forest management. First results showed that the effect of thinning and harvesting after 5 years of fallout was limited on the RCs inventory because of rapid  $^{137}\text{Cs}$  migration from trees to soils.

**Coppin** talked about the distribution and availability of RCs in Fukushima forest andosols at two locations in Kawamata town. The aim is to estimate the residence time of  $^{137}\text{Cs}$ , stable Cs and K in different profile layers to feed into a forest model. In Nov 2013, 90% of the  $^{137}\text{Cs}$  was in the forest floor, 70% of which was in the upper organic layer and the 0-3cm soil layer. Kd data suggest that part of the  $^{137}\text{Cs}$  is available for root uptake and vertical water migration. Much of the sorbed fraction of  $^{137}\text{Cs}$  is exchangeable (15-50%) reflecting the low clay mineral content and high OM content of the soil.

**Yoshenko** presented a research program aiming at characterizing  $^{137}\text{Cs}$  distribution and fluxes in typical forest ecosystems (coniferous) in Chernobyl and Fukushima. In Ukraine, the biogenic fluxes of radionuclides are comparable with geochemical migration fluxes in the root layer, leading to a gradual decrease of the apparent vertical migration rates. In Fukushima, after an initial phase of important foliar interception of  $^{137}\text{Cs}$ , root uptake is becoming important.

**Takada** presented a compartment model used to fit data on  $^{137}\text{Cs}$  migration from the litter layer to the soil layer for 6 mixed deciduous forests. The  $^{137}\text{Cs}$  migration rate from litter layer was 10 times higher than the migration in soil layers, and was correlated with litter decomposition rate. Using the compartment model, it was estimated that half of the  $^{137}\text{Cs}$  would remain between the litter layer and surface soil layer for the next five years.

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**Takeuchi** presented the total RCs inventory in a mountainous forested catchment using 2 methods: continuous measurement of air dose rate during mountain-walking and a grid-based measurement of  $^{137}\text{Cs}$  inventory based on litter and top-soil samplings. Monthly rate of fluvial discharge of  $^{137}\text{Cs}$  was also estimated using continuous measurement of dissolved and particulate  $^{137}\text{Cs}$ . Air dose rates were higher on ridges and from east-facing slopes. Dissolved  $^{137}\text{Cs}$  accounted for 26 % of total fluvial discharge, and accounted for 0.03 % of the total inventory in the forest catchment.

**Kato** investigated the transfer of canopy-intercepted  $^{137}\text{Cs}$  to the forest floor in coniferous and deciduous stands. While  $^{137}\text{Cs}$  deposition on forest floor with throughfall was associated with an increase of ambient dose rate, the effect of litter-fall on changing of ambient dose rate still needs to be quantified.

**Loffredo** presented a 2 compartment model of  $^{137}\text{Cs}$  leaching dynamics from evergreen canopies, adjusted using *in situ* and literature data. While no spatial variability of kinetics of leachable  $^{137}\text{Cs}$  loss was observed, a high spatial variability of the leachable stock was observed, due to initial deposit and canopy closure. This latter parameter was added to the model to improve predictions.

**Hurtevent** presented  $^{137}\text{Cs}$  distribution and recycling in two even-aged cedar stands. Based on  $^{137}\text{Cs}$  determination in various tree compartments, litter and soil, calculated fluxes showed that litter fall remains a major pathway for  $^{137}\text{Cs}$  redistribution compared with internal translocation in crown. Thus, the rapid redistribution of  $^{137}\text{Cs}$  between tree and soil still prevails 3 years after the accident. Future studies will be focused on  $^{137}\text{Cs}/\text{K}$  ratios to characterize the stage of recycling of each element between the tree elements to quantify the dynamics of tree contamination.

**Itoh** reported on initial RCs deposition in forest ecosystems after the Fukushima Daiichi accident. They made use of bulk precipitation and throughfall samples collected before, during and after deposition for other purposes mainly in the Kanto region (100-250km from the NPP). The ratio of RCs in throughfall to that deposited in bulk precipitation to the end of March 2011 was highly variable at 0.13-0.66, then decreased rapidly. Thereafter, the ratio increased and eventually exceeded 1 suggesting that the forest canopy was gradually releasing intercepted RCs.

#### 2.2.2.4. Radiocaesium transport in catchments and through river systems.

**Eyrolle-Boyer** showed suspended load versus discharge relationships for five catchments in the Minami-Soma region of Fukushima and concluded that their behaviour was similar. There is some transfer of contaminated sediment from forest litter to rivers but this is 'rather conservative'. There is an inverse relationship between suspended sediment and  $K_d$  due to the higher reactivity of the finest particles. Dissolved (non-particulate) fluxes of RCs range from 30-90% of total flux under mean hydrological conditions.

**Konoplev** reported that RCs dispersion in undisturbed forest and grassland soils in the Fukushima contaminated area was significantly faster compared with the Chernobyl 30-km zone during the first three years. Effective dispersion coefficients of RCs migration for Fukushima soils are several times higher than that for Chernobyl, partially explained by the higher precipitation in Fukushima than for Chernobyl. Soils in Fukushima are characterized by greater bioturbation in the top soil layer. For both Fukushima and Chernobyl, forest soils are

characterized by fast RCs migration in the upper soil layer. The RCs Kd in Fukushima rivers was 1-2 orders of magnitude higher than corresponding values for rivers and surface runoff of Chernobyl suggesting higher ability of Fukushima soils and sediments to bind RCs. The normalized dissolved wash-off coefficients for Fukushima river watersheds are 1-2 orders of magnitude lower than corresponding values for the Chernobyl zone.

**Yamakawa** showed that the use of a stevia fermented product as an amendment seems to decrease RCs transfer to plant although it also seems to increase the RCs transfer in soil and increase RCs extraction from vermiculite compared with that due to water or K amendment.

**Wakiyama** quantified RCs discharge in the Niidagawa river Basin using instruments capable of collecting sediment discharge during intensive rainfall events. They installed sets of integrated suspended sediment samplers, turbidity sensors and water level sensors at three sites in summer 2014. Total  $^{137}\text{Cs}$  discharge in the basin was very small compared with total  $^{137}\text{Cs}$  deposition. Most  $^{137}\text{Cs}$  was discharged from smaller catchments during intensive rainfall events.

**Ogura** showed that the water and suspended solid (SS) dynamics could be simulated using a the Soil and Water Assessment Tool (SWAT). For the Hiso River catchment, model performance for daily SS load was less good than that for streamflow possibly due to some very high peaks in daily SS load, which occurred only a few times a year. The results indicate that there is a source for high peaks of SS load somewhere in the watershed. The source is also a hotspot of RCs. Continuous monitoring of stream load and investigating land conditions are strongly required.

**Delmas** combined prior uncertainty on wash-off parameters from the literature and *in situ* measured RCs concentrations for rivers in 5 contaminated watersheds in the region of Minamisoma to estimate average wash-off parameters. Bayesian inference on wash-off parameters from a double exponential transfer function was conducted by a Markov Chain Monte Carlo algorithm (MCMC) using the GNU-MCSim software. Only a small fraction of deposited RCs is available for wash-off.

**Nagao** described data on  $^{137}\text{Cs}$  for 2011-2014 in the Abukuma River, which is the largest in Fukushima area during 2011 – 2014. Measurements of total  $^{137}\text{Cs}$  versus turbidity showed mixed results at different sites, but there was a clear increase in [RCs] in river waters after rainfall. Kd values were also derived, but these values were obtained under non-equilibrium condition so is difficult to compare these Kd values with Kds obtained under equilibrium condition.

**Tsuji** also showed data for  $^{137}\text{Cs}$  in particulate and dissolved forms in storm runoff in rivers in the Fukushima area. He showed evidence for elution of  $^{137}\text{Cs}$  from forest litter in the riparian zone and he hypothesised that this part of the catchment could be important in supplying RCs to rivers since Bq/kg values in litter are similar to that in suspended sediment after rainfall.

**Evrard** stated that 70% of area affected by the initial deposition from Fukushima is drained by 3 rivers. He showed that paddy fields are the most connected parts of the rivers and that most significant erosion occurs on farmland. There has been progressive dispersion of RCs towards the Pacific Ocean, based on 4 sampling campaigns over 2 years. So far ~1% of the initial deposited RCs has been transported from catchments towards the ocean.



**Onda** tested the hypothesis based on post-Chernobyl observations of rivers that there are discrete phases to RCs release from soils to waters over the years following initial deposition. He described results from a \$10M programme conducted from 2011-14 in which, among other things, soil erosion plots were studied. Most suspended sediment comes via outflow from rice paddies. Different temporal trends were evident in the decrease of RCs carried by suspended sediment, depending on the system being studied. There was a faster decrease in sediment-associated RCs from paddy fields than from forests. A key message was that the temporal change in runoff of RCs from the land to surface waters changes with land use.

**Pratama** modelled RCs washoff into rivers and then flow into the ocean. He made simulations that predicted future discharges of RCs for 100 years. Areas with forest discharge less RCs than urban areas. His model showed that RCs discharge rates can be decreased if remediation activities are increased significantly.

#### 2.2.2.5. Radiocaesium in marine ecosystems

**Kitamura** presented the temporal and vertical trends of RCs in zooplankton. Four years post-accident RCs levels have returned to background levels within zooplankton. Coastal trends of RCs within zooplankton differed from oceanic zooplankton, with increase RCs evident along the coast due to river discharges of RCs. No positive relationship was observed between RCs in seawater and zooplankton.

**Bezhenar** presented results from a box model (Poseidon) that simulated RCs migration from bottom sediments through ocean food chains. Model predictions were in good agreement with data and showed strong correlations between RCs in sediments and benthic organisms.

**Duffa** spoke about several marine modelling efforts within the European COMET project. They are using biokinetic, sediment exchange, and trophic transfer models. Models have good agreement with plankton data, and with RCs in sardines, but only after fish migration was included in the model.

#### 2.2.2.6. Radiocaesium transfer to animals and wildlife doses

**Hinton** (invited talk) presented a new tool for quantifying external dose rate to free ranging organisms, merging GPS and electronic dosimeter systems. The replicability of these systems was tested on young boars in Savannah River site and on wild wolves in Belorussia. The results show that measured external dose rate was higher than the ambient dose rate. Internal self-irradiation due to wolf contamination has to be estimated from whole body measurements.

**Johansen** presented data from the IAEA MODARIA project. Doses to fish were calculated from measured data. The highest doses to fish were within the port of the Fukushima DNPP. Dose to these fish are remaining elevated with little decrease over time. Dose to other marine fish are declining with time, at a rate that is similar to what Kitamura presented for zooplankton. Near the FDNPP, the highest RCs concentrations were in fish with trophic levels of about 3.5-3.8 (not in fish with higher trophic levels of +4.0). Upper trophic level fish are not following the same RCs rate of decrease as lower trophic chains. Fish behaviour, foraging area differences and methods of food consumption that result in oral sediment intake were hypothesized as plausible reasons.

**Gjelsvik** described long-term measurements of transfers of  $^{137}\text{Cs}$  to animals in Norway and she showed that these transfer are higher in air-polluted affected areas. Higher TF to animals are seen in coastal areas than inland due to marine aerosol deposition to soils on the coast. Southern soils in Norway are also affected by acid precipitation and have shown faster loss of RCs and greater uptake of  $^{137}\text{Cs}$  by animals than other areas since 1986. The increased levels of  $^{137}\text{Cs}$  measured in big predators such as lynx, wolf, brown bear and wolverine in central Norway reflect the higher contamination in this region as well as RCs accumulation in these animals' food chains. The long-term use of Prussian blue and clean feeding continues to ensure that the intervention limit of 600 Bq/kg in sheep is met.

**Adam-Guillermin** (invited talk) began by describing studies of radiation exposures and effects in birds and frogs in the Fukushima area. Currently, there is no consensus on ecological effects of radioactivity released from Chernobyl. Conflicting results on measures of biological impact such as biodiversity might be due to inappropriate dosimetry when constructing dose-effect curves. Dr Adam-Guillermin demonstrated that ambient dose measurements for two very different organisms (birds and frogs) were an order of magnitude lower than doses assessed with respect to the specific niches occupied by the organisms. For both organisms dose-related effects were observed, but at higher dose rates than previous publications would suggest. Ambient dose negatively correlated with singing activity of frogs.

#### 2.2.2.7. Remediation

The invited paper by **Howard** (UK) was some background on remediation and a brief summary of remediation activities after the Fukushima accident based on two recent IAEA reports and work done for the imminent IAEA Fukushima report. The presentation also gave a comparison of the remediation after the Chernobyl and Fukushima accidents.

**Unno** considered the ability of different metal phytates which are a major component of soil organic phosphorous, to remove  $^{137}\text{Cs}$  from solution including soil solution. Al and Fe phytate had a high  $^{137}\text{Cs}$  adsorption potential in slightly acidic soil which is widespread in northeast Japan. Phytates may play a role in adsorbing RCs early after deposition. As ammonium acetate removes the RCs, the adsorbed fraction may be available for plant uptake.

**Djedidi** discussed transfer of RCs to Brassica spp. after the application of different bacterial isolates (rhizobacteria) which promote plant growth as a possible phytoremediation option. Many isolates increased  $^{137}\text{Cs}$  uptake by the plants, particularly into the roots. The pot experiments now need to be supplemented by field trials.

**Noda** showed that the expression level of HAK5 and transcription factor RAP2.11 of legume could be used to select plants with high TF for remediation process or conversely with low TF to reduce RCs transfer.

**Popov** presented laboratory studies of RCs penetration and washoff from urban building materials (e.g. concrete, brick, asphalt). The data were acquired from laboratory equipment that physically simulated weathering of RCs that had been applied to building materials, as well as simulated rainfall and washoff of RCs. Washoff was greatest for asphalt > granite > concrete = brick.

### 2.2.2. Session posters

Session 5 also included 25 poster presentations, listed below giving the first author only. Three of the posters received awards for “best of conference” posters: (Tashiro et al. (1), Tsukada et al. (11) and Noda et al. (21)). The posters are summarised below under the name of the first author.

1. Tashiro et al, Radiocaesium retention ability of soil clay fraction of paddy soils in Japan in relation to clay mineralogy.

After the Fukushima Daiichi nuclear power plant accident in March 2011, RCs ( $^{137}\text{Cs}$ ) retention ability was shown to be largely dependent on micaceous minerals in the clay fraction in Fukushima. However, it was not clear whether this result can be extended to wider areas in Japan with a higher variety in clay mineralogy. The objective of this study was to understand the general trend in the relationship between  $^{137}\text{Cs}$  retention ability and clay minerals in Japan. Eighty-four upper soil layer (0-15 cm) samples were collected from paddy fields sampled from all over Japan from Hokkaido to Okinawa. The clay fraction ( $< 2\mu\text{m}$ ) was collected from each soil sample by sedimentation and the RIP measured. RIP was largely controlled by the amount of micaceous minerals rather than vermiculite, consistent with the result obtained for the soil clays from Fukushima paddy soils. In conclusion, not only in Fukushima but also for wider areas in Japan, we found that the amount of mica was the most important factor controlling  $^{137}\text{Cs}$  retention ability.

2. Nanzyo et al, Vertical Distribution of Radiocaesium in Tsunami-Affected Farmland Soils in Miyagi Prefecture, Japan.

The tsunami deposits on farmlands in Miyagi prefecture, and underlying two soil layers were collected during May 2011 at 344 sites in Miyagi Prefecture. The tsunami-deposits were mostly differentiated from the underlying soil by the presence of coarse sand. A thin muddy layer, which was the most highly contaminated by RCs, was deposited over the coarse sand. Imaging Plate analysis on vertical sections showed two distribution patterns of radioactivity. The distribution of imaging spots suggests that a significant fraction of RCs had been deposited as particulate material, and deposition of the radioactive particulate material occurred after the sedimentation of the muddy layer. The other pattern was a very faint single, layer distribution at the upper part of the vertical section which, if due to RCs, may have deposited as a soluble form, and become fixed by suspended soil materials in the inundation water during sedimentation.

3. Ito et al, Distribution of Radioactive Caesium in Classified Particles for Decontamination in Fukushima.

Volume reductions are being developed in Fukushima to cope with the vast amount of waste associated with remediation. Two soils from Namie and Okuma were wet sieved and evaluated. The fractions with the highest RCs contamination was that associated with clay particles, but the size fraction of the clay minerals differed substantially between the two soils. Removal of RCs by wet sieving would be effective for clay fractions  $< 2\mu\text{m}$ , but the large particle clay fraction needs an improved classification method.

4. Kowata et al, Replanting of mulberry: is it effective in reducing the risk of radiocaesium contamination in leaves?.

[RCs] uptake in soil and leaves was compared in three mulberry plantations of different ages (1, 2 and  $> 25$  years after replanting). The RCs TF to the oldest tree was highest. Ammonium acetate exchangeable K concentrations in soil  $> 50\text{ mg-K}_2\text{O}/100\text{g-dw}$  reduced TFs in the younger trees, but not the oldest. Water-soluble K exceeding  $20\text{ mg-K}_2\text{O}/100\text{g}$  resulted in a

similar reduction. Replanting of mulberry can be effective in reducing the transfer of RCs to leaves, if soil amendment with K fertilizers is performed at the same time.

5. Suzuki et al, The effect of irrigation on  $^{137}\text{Cs}$  uptake in rice.

From 2011 to 2013, we found that brown rice and/or straw collected near the water inlet of paddy fields contained significantly higher [ $^{137}\text{Cs}$ ] than those collected at the centre of the rice fields and near the water outlet. Further field examinations of the impact of irrigation were carried out in three rice fields in Fukushima Prefecture. Soil samples were collected from the surface 0-15 cm layer at 1, 2, 3, 5, 7, 11, 15 and 21 m apart from the inlet at harvest. Rice samples were harvested at the same positions and divided into brown rice and straw. No relationship between soil clay contents and distance from the water inlet was found. For all three fields, the highest [ $^{137}\text{Cs}$ ] were often found in soil and rice or straw, and also the highest TF values, at sample sites which were within three m from an inlet. [ $^{137}\text{Cs}$ ] and TF were lower at more distant sample sites. The data suggest that irrigation water may take an important role to the elevated RCs uptake in rice grown near water inlets.

6. Suzuki et al, Effect of radiocaesium fractions in irrigation water on radiocaesium uptake in brown rice.

The study investigated  $^{137}\text{Cs}$  uptake from irrigation water into brown rice by determining [ $^{137}\text{Cs}$ ] in brown rice grown in three gray lowland soils. The plants were grown in pot experiments in water containing  $^{137}\text{Cs}$  (1.0 or 10 Bq L<sup>-1</sup>) in the dissolved fraction, bound to the soil particle fraction, or bound to the organic matter fraction. For each  $^{137}\text{Cs}$  fraction, the [ $^{137}\text{Cs}$ ] in the brown rice increased in proportion to that in the water. The uptake rate for the dissolved fraction was higher than that for the organic-matter-bound and soil-particle-bound fractions. The uptake rates for the three different soils used varied. Furthermore, the uptake into brown rice from the dissolved and organic-matter-bound fractions were similar for two of the soils, but that from the soil-particle-bound fraction differed for these two soils.

7. Shinano et al, Varietal difference in the radioactive caesium uptake in *Amaranthus* and Soybean.

Introducing plant varieties or lines which have a relatively lower ability to accumulate RCs from the soil, can be one possible phytoremediation measure for application in the contaminated area. Using *Amaranthus* or soybean it would require more than 400 years to decontaminate the soil (ignoring the half-time and leaching from the soil), thus actual usage of plant for phytoremediation is not acceptable. Based on ionome analysis, it seems that the uptake of non-essential elements including Cs is carried out not only by K transporter but also by other mechanisms.

8. Fujimura Difference in  $^{137}\text{Cs}$  uptake between two paddy rice cultivars

To understand the interactive effect of uptake of K<sup>+</sup> and Cs<sup>+</sup>, we examined the difference in  $^{137}\text{Cs}$  uptake using two lowland rice cultivars (Hokuriku 193 and Koshihikari) which differ in ability to accumulate K<sup>+</sup> (Hokuriku 193 > Koshihikari). In one field experiment, the [ $^{137}\text{Cs}$ ] in brown rice and the plant was four-fold and two fold higher respectively in Hokuriku 193 than in Koshihikari with similar dry weights. In the second experiment using rice seedlings, [ $^{137}\text{Cs}$ ] and the amount of  $^{137}\text{Cs}$  in shoot was significantly higher in Hokuriku 193 than in Koshihikari. The accumulation of K also tended to be greater in Hokuriku 193 than in Koshihikari. However, in a hydroponic experiment, [ $^{137}\text{Cs}$ ] in shoot and root was similar in both cultivars in two K treatments. The amount of  $^{137}\text{Cs}$  in shoot and root was significantly greater in Hokuriku 193 than in Koshihikari due to higher dry weight in the former in both K treatments.  $^{137}\text{Cs}$  uptake in the hydroponic experiment was contrary to those in the field suggesting that experimental conditions in the soil need to be carefully considered.

9. Saito et al, Distribution of radioactive caesium in soil and its uptake by herbaceous plants in temperate pastures after the Fukushima Daiichi Nuclear Power Station accident.

The status of RCs contamination in soil and plants at Tohoku University, Miyagi prefecture, 150 km north of the NPS was evaluated. Pasture management greatly influenced the RCs content of herbaceous plants whereas plant species had less influence. In manure-treated soils the soil-to-plant transfer factor was negatively correlated to pH(H<sub>2</sub>O) and exchangeable K content of root mat soils. The RCs content of plants decreased with plant maturity: the decrease (early May to early June) was smaller in legumes than in grasses and forb.

10. Ishikawa et al, Profiles of potassium and cation transporters in rice plants: Investigation on possible caesium transporters.

When rice plants were grown by hydroponic culture ([K] 256 μM) for 20 days some high-affinity K transporters, *HAK1*, *HAK7* and *HAK10* were highly expressed, suggesting that they may play dominant roles in root uptake of K. When the 20-day-old seedlings were treated for 24 h with hydroponic solution of different [K] (from 2 to 51 μM), gene expression of most transporters except *HKT2;1*, another high-affinity transporter, showed little change. However, *HKT2;1* was up-regulated below 10 μM of K suggesting that *HKT2;1* may contribute to absorb K when present in very low [K] in the soil. Differences of gene expression were analysed using root samples from different cultivars grown in a paddy field. Gene expression of the high-affinity potassium transporter *HAK1* and a potassium channel *K1.1* were higher in Indica than in Japonica cultivars, which can be compared with data showing that Indica cultivars have higher RCs uptake than Japonica cultivars.

11. Tsukada et al, Relationship between soil-to-plant Transfer Factor of <sup>137</sup>Cs in agricultural plants and Radiocaesium Interception Potential.

RIP was determined and compared with the transfer factor of RCs in agricultural plants collected from fields. The TF values in polished rice, potato and cabbage in Aomori decreased with increasing the RIP values, and they showed good correlation. The TF value in polished rice collected from Fukushima was also well correlated with the RIP value, and it shows a similar range with the results in Aomori. This indicates that RIP is the important factor for controlling root uptake of RCs by plant.

12. Kojima et al, Comparison of Radiocaesium Accumulation among Soybean Cultivars.

To elucidate the factors which control the accumulation of RCs in the grain of soybean the characteristics of RCs accumulation among soybean cultivars containing various genetic backgrounds was compared. When grown in soil with a mean [RCs] of 3,675 Bq/kg DW no cultivar accumulated more than 100 Bq/kg in the grain sampled in 2012. The histogram of [<sup>137</sup>Cs] had a Gaussian distribution, and the modal value was 15~20 Bq/kg DW. The Takiya cultivar grain had the lowest [<sup>137</sup>Cs] of 6.9 Bq/kg DW. The highest was found for the Kurakake cultivar at 31.5 Bq/kg DW, so the [<sup>137</sup>Cs] varied by a factor of 4.5. The varietal ranking of [<sup>137</sup>Cs] in the cultivars differed between 2012 and 2013, for instance, cultivars that were ranked top in [<sup>137</sup>Cs] in 2012 were not in 2013. The average value of [<sup>137</sup>Cs] in cultivars in 2013 was nearly 50% of that in 2012.

13. Rai et al, Development of low caesium uptake *oryza sativa* by a chemical mutagen.

Chemical mutants of rice were made with sodium azide and Nmethyl-N-nitrosourea (MNU). Three low Cs uptake mutants were selected from 8027 M3 plants after a <sup>133</sup>Cs uptake trial. In the first experiment, the [<sup>133</sup>Cs] concentrations in brown rice of the three mutant strains grown in soil with 0 ppm, 1 ppm, 2.5 ppm <sup>133</sup>CsCl added were reduced by about four fold in Andosol and > 10 fold for Fluvisol in the treatments compared with the controls. In a hydroponics experiment, two weeks old seedlings in Kimura B liquid medium were transplanted into a

medium with 1.0, 2.5, 5, 10, 25 ppb stable Cs added. The Cs concentrations of the roots of the three strains were low compared with the control for all Cs levels, but that of the shoots were not. Similar experiments were then set up with Na, K, Ca, Mg concentrations of x 0, 5, x 2, x 3 times the concentration of the original medium. Only the K enhancement had an inhibitory effect on Cs uptake. In the low K treatment (x 0.5), Cs uptake of the mutants was much lower than the control in both the shoots and roots. However, the Cs uptake of the control was reduced to a similar extent as the mutant strains with increasing K concentration. The data suggested that the selected three strains were mutations of one of the minor K transporters.

14. Dohi et al, <sup>137</sup>Cs in lichens after the Fukushima Nuclear Accident.

There was a significant positive correlation between [<sup>137</sup>Cs] in lichens (n=48) in Japan from Fukushima Prefecture and <sup>137</sup>Cs inventories in soil. Ratios of <sup>134</sup>Cs: <sup>137</sup>Cs were consistent with those from the Fukushima Daiichi accident in Fukushima prefecture and lower for those further away.

15. Iijima et al, Transport behaviour of radioactive caesium through water system in the coastal area of Fukushima.

This paper provides an overview of the results of field observations on the distribution and evolution of [<sup>137</sup>Cs] in the Ukedo River system in 2013 and 2014. Deposition of RCs after the accident was higher upstream than downstream. Therefore, RCs bound to suspended particles originating from the catchment of the river downstream of the dam dominates the transport behaviour of RCs in the river downstream. Ogaki dam is located in the middle of Ukedo River 17 km from the river mouth. Bottom sediments of the Ogaki dam lake were cored at ten locations and each depth profile of [<sup>137</sup>Cs] was evaluated. The dam was shown to have a significant ability to constrain transport of RCs from upstream to downstream.

16. Lepage et al, Modelling the source of sediment-bound RCs in two coastal catchments in the Fukushima Prefecture with sediment tracing techniques.

A sediment fingerprinting technique was applied to determine the location and relative contributions of different sediment sources in the Mano and Niida Rivers in Fukushima prefecture. In the Mano River ~20% of the RCs sampled was modelled to be derived from the upstream area compared with ~30% in the Niida River. The highest contribution of upstream RCs was modelled after the typhoon seasons in 2011 and 2013. Fluvisols were the dominant source of sediment and RCs indicative of sediments derived from sources that are highly connected to the river network. Understanding the relative contributions of these different sediment sources will allow for more direct management of sediment, and thus RCs transfers in these Fukushima coastal catchments.

17. Ueda et al, Discharge of Fukushima-derived radiocaesium from mountainous river in the contaminated watershed, Japan.

Transport rates of RCs through the small Hiso River and Wariki River that traverse mountainous areas in Fukushima Prefecture were estimated. The RCs concentrations strongly depended on the rates of water discharge. Particulate RCs during periods of flooding contributed over 90% of the total Cs in the samples. The discharge of RCs from the catchments during 2011 was estimated to be 0.5% and 0.3% of the total amount of RCs deposited on the catchments of the Hiso River and Wariki River, respectively. Particulate RCs during periods of flooding contributed > 90% of the total RCs in the samples. The discharge of RCs from the catchments during 2011 was estimated to be 0.5% and 0.3% of the total deposited on the catchments of the Hiso River and Wariki River, respectively. RCs discharge (c. 0.2 %) in 2012 was lower than 2011 showing its relatively higher mobility just after the deposition on the ground.

18. Miyazu et al, Development of a method for monitoring radiocaesium in throughfall using a prussian blue impregnated nonwoven fabric.

A simplified method was developed for monitoring RCs in throughfall using a prussian blue impregnated nonwoven fabric. The method has the advantages that: (i) large volumes of throughfall water are not required for analysis, saving labour resources; (ii) the time required to pretreat samples prior to analysis is considerably reduced; and (iii) a 10-fold increase in analytical sensitivity over that of the conventional method is achievable.

19. Hashimoto, Modelling dynamics of Fukushima-derived radiocaesium in forest ecosystem using RIFE1 mod.

A forest ecosystem (RIFE1 model), was parameterized and tested using data observed in the first and second year after the Fukushima accident. Over a period of 1-2 years, the RCs is predicted to move from the tree and surface organic soil to the mineral soil. The total inventory within the mineral soil compartment peaks during the first year (deciduous broadleaf forests) or second year (evergreen conifer forests) and then decreases slowly, primarily due to radioactive decay. The  $^{134}\text{Cs}$  inventory in the mineral soil compartment starts decreasing approximately one year after the initial fallout whereas that for  $^{137}\text{Cs}$  in the mineral soil compartment peaks during the fifth/sixth year.

20. Ishii et al, Effect of Faeces of Rhinoceros Beetle Larvae on the  $^{137}\text{Cs}$  Transfer to Komatsuna (*Brassica rapa var. perviridis*).

The effect of faeces from larvae feeding on leaf litter on the transfer of  $^{137}\text{Cs}$  to a Komatsuna plant was determined. The TF for faeces-treated soil was similar to the control. However, the wet weight of plant produced was significantly higher on faeces-treated soil than the control suggesting a stimulatory effect on plant growth by faeces of the larvae. The  $^{137}\text{Cs}$  which was eluted from faeces would be sorbed on soil particles before being taken up by plant roots.

21. Noda et al, Radiocaesium Behaviour and Expression Analysis of Potassium Channels in Poplar.

Poplars grown with stable Cs (0 to 20  $\mu\text{M}$  for a month) showed no visible symptoms. Cs concentration in root, stem, petiole and leaf increased depending on applied Cs concentration and there was no specific organ for Cs accumulation. Detailed Cs localization analysis using  $^{137}\text{Cs}$  and autoradiography showed that after root application,  $^{137}\text{Cs}$  accumulated in shoot apices and upper leaves whereas after foliar application,  $^{137}\text{Cs}$  was detected in the treated leaf and the stem below that leaf. These results indicated root and foliar absorption might have different Cs transport pathways in poplar so Cs absorbed by root was transported by transpiration in xylem and foliar absorbed Cs by phloem transport. Gene expression analysis indicated that PttSKOR-like2 localization suggested PttSKOR-like2 might transport Cs/K into xylem sap in the poplar root. Furthermore, PttSKOR-like2 expression was regulated by environmental factors such as day length and/or temperature

22. Kobayashi et al, Transport of radiocaesium by water flow through canopy, litter layer, and mineral soils in forests.

In the first 1.5 months after the Fukushima accident, the total (dissolved + particulate) [RCs] in throughfall (ThF) collected at a site in Ibaraki prefecture ranged from 14 - 60  $\text{Bq L}^{-1}$  and RCs in ThF was in the dissolved phase. Litter layer (LL) sampled in the same period had [RCs] < 10  $\text{Bq L}^{-1}$ , indicating that a large proportion of deposited RCs was retained in the LL soon after the accident. In the following months, the [RCs] in ThF and LL gradually decreased and the proportion of RCs in the particulate phase increased. In the summer period, the amount of RCs present as suspended particulate matter in ThF and LL often increased and resulted in high particulate [RCs]. The [RCs] in ThF and LL both decreased in winter. The summer increase in particulate RCs in ThF and LL was also observed at a conifer plantation and deciduous forest in Fukushima prefecture, sampled from 2012. The temporal increase in suspended particulate matter seems to be composed mainly of organic C and may have been due to a comprised of insect herbivory and litter decomposition. The [RCs] of dissolved in soil solution was very low

at both sites indicating that RCs is captured strongly in soil, and that runoff from forests will be limited.

23. Aono et al, The distribution and behaviour of radiocaesium in marine environment around off the Fukushima in 2012-2014.

After the accident, the [ $^{137}\text{Cs}$ ] in seawater had decreased more than 10 fold compared with those before the accident. However, [ $^{137}\text{Cs}$ ] in sediment decreased more slowly than those in seawater and had large fluctuations. The sedimentary environment depends on several factors such as the seabed topography and freshwater and terrestrial material input via rivers. Sediment-seawater distribution coefficients  $K_d$  (L/kg) varied from 1,900 to 25,000 in the same area in 2013-2014. Decreases of [ $^{137}\text{Cs}$ ] and [ $^{110\text{m}}\text{Ag}$ ] in marine biota will depend on differences in the mechanisms of accumulation and metabolism. The estimated CR of  $^{137}\text{Cs}$  (CR-Cs) in marine organisms ranged from 26 in muscle of squid to 10,000 in clam viscera. Large differences in CR-Cs in different parts of marine organisms were not observed. For CR-Cs values, it is necessary to consider the influence of sediment and particles on marine biota, as these organisms often contain particulate matter and sediment in the coastal area. Sr-90 was not detected in the bony parts of fish and [ $^{239+240}\text{Pu}$ ] in visceral parts were the same as before the accident. [ $^{137}\text{Cs}$ ] in seawater decreased ten-fold over 6 months, and consequently that in the marine biota also dropped.

24. Yamasaki et al, Chemical form of radioactive Cs in the estuary sediments of the Kuma River, Fukushima, Japan.

After heavy precipitation or storm events, certain fractions of soil minerals can be remobilized, suspended and transported via surface water (i.e., river system) to the distance depending on the mineral size. Those minerals eventually reach the ocean and become a source of contamination for estuarine and marine organisms. This study investigated the nature of transported mineral particles associated with RCs and the chemical state of RCs in estuary sediments near the FNDPP. A full characterization of the estuary sediment was carried out as was part of the transport process of RCs via surface water. Sequential extraction revealed that ~90% of RCs was bound to layered alumino silicate. ~2% of the RCs was desorbed within 5 sessions of leaching the sediment with simulated seawater, indicating a small amounts of RCs can be desorbed in seawater within 10 h and be present in a soluble form if the sediment is transported to the ocean. Mobile clay aggregates associated with RCs play a key role in RCs migration in the surface environment and can be a potential source of RCs either in the form of soluble or fine colloidal particle.

25. Ueno et al, Caddisfly Watch – A monitoring approach for radioactive caesium in river environment of Fukushima using the caddisfly larvae.

Fish have previously been used as a bioindicator. However, the radioactivity detected from fish samples have showed high variation possibly due to migrating long distance and reflecting the contaminant presence in a wide area of the river environment.. In this study, “caddisfly larvae” was selected as a possible bioindicator for monitoring the radioactivity in rivers. Caddisfly larvae is one of the sessile organisms in river ecosystem which makes a nest under stones in the river bed. This species has many advantages as a bioindicator, namely that it does not move, its life span is short (six month), it can be found in all seasons, and it can be collected by hand.



### 3. Fukushima Excursion

The ICOBTE meeting was followed by a one-day excursion to the contaminated areas in Fukushima prefecture. Twenty seven ICOBTE participants attended the excursion accompanied by three organisers from Fukushima University and a professional guide (Annex 5). Table 2 shows the itinerary for the excursion.

Table 2. Itinerary for the excursion of contaminated areas around the Fukushima Daiichi NPP.

Michi-no-Eki Soma
Lone Pine Tree that survived tsunami (Minamisoma City) <Floral tribute to victims of tsunami>
Lunch at Kifune Rest House, Namie Town (Evacuation-order-lift- preparation zone)
Experimental Field, Okuma Town (Difficult-to-return zone)
Community Revitalization Base, Okuma Town (Habitation-restricted zone) <Talk by Okuma Town employees>
Radiation screening
Passing through litate Village

Figure 1 is a map of the excursion route. The first stop, Michi-no-Eki Soma, was used as an investigation base and a relief supply relay station after the tsunami. At the Lone Pine Tree at Minamisoma City, the group paid a floral tribute to the victims of the tsunami.

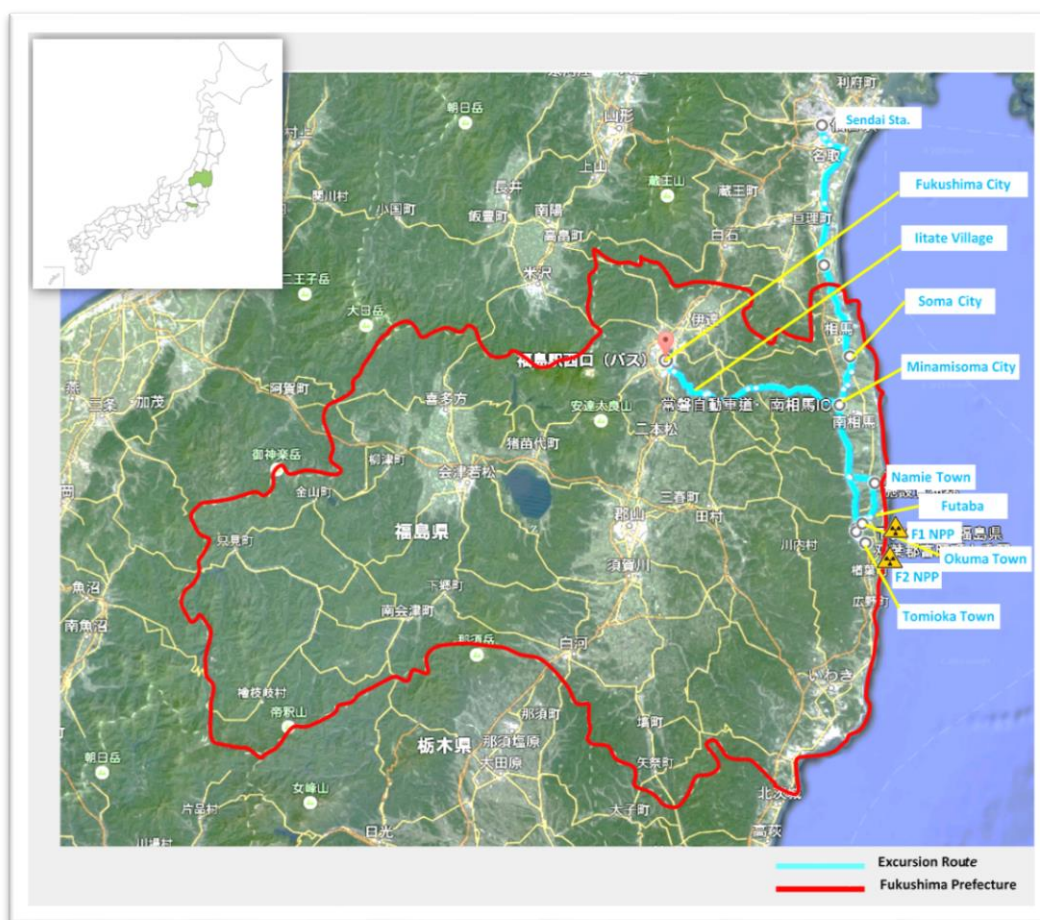


Figure 1 The Fukushima excursion route

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Firstly we arrived at Minamisoma to place a floral tribute to victims of the tsunami (Figure 2).



Figure 2 Lone Pine Tree which survived the tsunami (Minamisoma).

Thereafter, we visited Namie Town which is in the evacuation-order-lift-preparation zone. The entire population was evacuated in the wake of the Fukushima Daiichi nuclear disaster in March 2011. Namie town was devastated by the earthquake. However, during the excursion it was clear that a huge and impressive reconstruction effort had resulted in the renovation of infrastructure such as roads, and in all residential housing and other buildings.

At Okuma, which includes land adjacent to the Fukushima NPP, and has land (including forests in SDA 1, 2 and 3. We visited the experimental fields to examine ways to reduce RCs uptake by crops operated by Okuma Town and Fukushima University (Figure 3).



Figure 3 Experimental plots in Okuma town.

At the Community Revitalization Base, in the Habitation-restricted zone of Okuma town, we received a briefing on current plans for remediation from local people who were heavily involved in revitalisation efforts in this area (Figure 4).



Figure 4 Community Revitalization Base in Okuma town.

## 4. Fukushima COMET Workshop

### 4.1 Introduction

Following on from ICOBTE 2015 and the Fukushima excursion, there was an associated workshop held at the Yoshikawayama inn, Iizaka near Fukushima on 18-19th July followed by a tour of the facilities at Fukushima University. This meeting differed from the ICOBTE meeting in that the speakers were deliberately selected to cover a range of topic areas and able to summarise the current key findings and issues. The workshop included local Fukushima University and prefecture scientists (Figure 5) and also scientists from national Japanese organisations. A full list of the participants is given in Table 3.



Figure 5. Fukushima COMET Workshop attendees.

Table 3. Participants of the COMET Fukushima workshop.

<b>Name</b>	<b>Affiliation</b>
<b>Japanese Participants</b>	
Kenji Nanba	Institute of Environmental Radioactivity, Fukushima University
Takayuki Takahashi	Institute of Environmental Radioactivity, Fukushima University
Hirofumi Tsukada	Institute of Environmental Radioactivity, Fukushima University
Kei Okuda	Institute of Environmental Radioactivity, Fukushima University
Toshihiro Wada	Institute of Environmental Radioactivity, Fukushima University
Yoshifumi Wakiyama	Institute of Environmental Radioactivity, Fukushima University
Izumi Mizushima	Institute of Environmental Radioactivity, Fukushima University
Rie Kanno	Institute of Environmental Radioactivity, Fukushima University
Kimiaki Saito	Japan Atomic Energy Agency
Hisatomi Harada	Agriculture, Forestry and Fisheries Research Council Secretariat
Hiromi Yamazawa	Nagoya University
Sadao Eguchi	National Institute for Agro-Environmental Sciences
Keiko Tagami	National Institute of Radiological Sciences
Daisuke Kobayashi	Fukushima Medical University
Masahiro Kobayashi	Forestry and Forest Products Research Institute
Tetsuo Yasutaka	National Institute of Advanced Industrial Science and Technology
Toshio Mizoguchi	Fukushima Prefecture
Jian Zheng	National Institute of Radiological Sciences
Yuichi Onda	Tsukuba University
Shinji Kaneko	Forestry and Forest Products Research Institute
Shoji Hashimoto	Forestry and Forest Products Research Institute
Tatsuo Aono	National Institute of Radiological Sciences
Tetsuo Sekiguchi	National Agriculture and Food Research Organization
Masashi Kusakabe	Marine Ecology Research Institute
Akira Takeda	Institute for Environmental Sciences
Noriko Yamaguchi	Soil Environment Division, National Institute for Agro-environmental Sciences
Mutsuo Sato	Fukushima Agricultural Technology Centre
Takeshi Saito	Fukushima Agricultural Technology Centre
Takuro Shinano	National Agriculture and Food Research Organization
Jun Nishikawa	Tokai University
<b>Overseas Participants</b>	
Brenda Howard	Centre for Ecology and Hydrology
Christelle Adam-Guillermin	Institut de Radioprotection et de Sûreté Nucléaire (IRSN)
Frédéric Coppin	Institut de Radioprotection et de Sûreté Nucléaire (IRSN)

Marc André Gonze	Institut de Radioprotection et de Sûreté Nucléaire (IRSN)
Rodolphe Gilbin	Institut de Radioprotection et de Sûreté Nucléaire (IRSN)
Pierre Hurtevent	Institut de Radioprotection et de Sûreté Nucléaire (IRSN)
Shinichiro Uematsu	Belgian Nuclear Research Centre (SCK•CEN)
Åste Sjøvik	Norwegian Radiation Protection Authority (NRPA)
Mat Johansen	Australian Nuclear Science and Technology Organization
George Shaw	University of Nottingham
Yves Thiry	French National radioactive Waste Management Agency (ANDRA)

#### **4.2 Programme of the Fukushima COMET Workshop**

The Program of the COMET Fukushima Workshop, 18-19<sup>th</sup> July 2015 is given below.

##### **July 18, 2015**

##### **(Chairperson: H. Tsukada)**

- 09:00 - 09:10 Opening remarks - Takayuki Takahashi (Fukushima Univ.)
- 09:10 - 09:35 (18-1) Source term and atmospheric dispersion of Fukushima Daiichi NPP radioactivity - Hiromi Yamazawa (Nagoya Univ.)
- 09:35- 10:00 (18-2) Distribution of radionuclide deposition densities and air dose rates in Fukushima - Kimiaki Saito (JAEA)
- 10:20- 10:45 (18-3) Factors affecting distribution coefficient of radiocaesium in various agricultural environments of Fukushima - Sadao Eguchi (NIAES)
- 10:45- 11:10 (18-4) Reducing radioactive Cs concentrations of forage crops after Fukushima Daiichi NPP accident in Japan - Hisatomi Harada (MAFF)
- 11:10- 11:35 (18-5) Reduction of radioactive caesium concentration in soybean seeds by potassium application in Japan - Tetsuo Sekiguchi (NARO)
- 11:35- 12:00 (18-6) Radiocaesium reduction rates in some fruit trees in Japan - Keiko Tagami (NIRS)

##### **(Chairperson: N. Yamaguchi)**

- 13:10- 13:25 (18-7) Linking radiocaesium sorption to soil chemical properties for Japanese soils: A comparison to European soils - Shinichiro Uematsu (SCK•CEN)
- 13:25- 13:50 (18-8) Cs dynamics in plants - Daisuke Kobayashi (Fukushima Medical Univ.)
- 13:50- 14:15 (18-9) Time Changes in radiocaesium wash-off from various land uses and transport through river networks after the Fukushima Daiichi accident - Yuichi Onda (University of Tsukuba and Fukushima University.)
- 14:15- 14:40 (18-10) Deposition and dynamics of radiocaesium in forest ecosystem for four years after Fukushima Daiichi NPP accident - Shinji Kaneko (FFPRI)
- 14:40- 15:05 (18-11) Transport of dissolved and particulate radiocaesium in Japanese forested ecosystems - Hasahiro Kobayashi (FFPRI)
- 15:20- 15:45 (18-12) Modelling spatio-temporal dynamics of radiocaesium deposited onto forests following the Fukushima nuclear accident - Shoji Hashimoto (FFPRI)
- 15:45- 16:10 (18-13) Radiocaesium in river and lake systems - Kenji Nanba (Fukushima Univ.)

- 16:10- 16:35 (18-14) Time series and spatial distribution of <sup>137</sup>Cs concentration in dissolved and suspended fractions in irrigation water collected within 80 km zone around TEPCO's Fukushima Daiichi NPP - Hirofumi Tsukada (Fukushima Univ.)
- 16:35- 17:00 (18-15) Cost and effectiveness of decontamination against radiocaesium contamination in Fukushima-How do we manage decontamination waste more effectively? - Tetsuo Yasutaka (AIST)
- 17:00- 17:25 (18-16) New geochemical tracers for source identification and long-term environmental behaviour studies of radionuclides released - Jian Zheng (NIRS)
- 19:00- 21:00 Research exchange discussion

### **July 19, 2015**

#### **(Chairperson: A. Takeda)**

- 09:00- 09:25 (19-1) Survey on Radionuclide Movement in wild boars (Fukushima Initiative IAEA Cooperation Project) - Toshio Mizoguchi (Fukushima Pref.)
- 10:05- 10:30 (19-3) Radionuclides transfer in marine organisms around Japan - Tatsuo Aono (NIRS)
- 10:30- 10:55 (19-4) Effects of the nuclear power plant accident on marine products and fisheries in Fukushima - Toshihiro Wada (Fukushima Pref.)
- 10:55- 11:20 (19-5) Distributions of Fukushima-derived <sup>137</sup>Cs in sediments and their temporal change: Results from four-year monitoring in the waters off Fukushima and nearby prefectures, Japan - Masashi Kusakabe (MERI)
- 11:20- 11:30 Learning for the Fukushima accident and Closing remarks - Brenda Howard (Centre for Ecology and Hydrology)
- 12:30- 14:30 Excursion to Fukushima University
- 14:30- 16:00 COMET meeting – COMET participants

#### **4.3 Presentation summaries**

Some presentational material given at the workshop has not yet been published. However, most participants have kindly agreed to provide a summary which are compiled below.

## 18-2: Distribution of radionuclide deposition densities and air dose rates in Fukushima

Fukushima Environmental Safety Center, Japan Atomic Energy Agency

Kimiaki Saito

### Introduction

Distribution of radionuclide deposition densities and air dose rates in Fukushima have been investigated by large-scale environmental monitoring repeatedly implemented in mapping projects commissioned by the national government. According to statistical analyses of the obtained data, time-dependent features of the contamination conditions were elucidated. An empirical model was developed for the prediction of air dose rate distribution within the 80 km zone.

### Conclusions

- Deposition density maps were constructed for  $^{134}\text{Cs}$ ,  $^{137}\text{Cs}$ ,  $^{131}\text{I}$ ,  $^{129\text{m}}\text{Te}$ ,  $^{110\text{m}}\text{Ag}$ ,  $^{238}\text{Pu}$ ,  $^{239+240}\text{Pu}$ ,  $^{89}\text{Sr}$  and  $^{90}\text{Sr}$ ; cesium was confirmed to be most important from a viewpoint of long-term exposure doses.
- The deposition density of radiocesium in undisturbed flat fields ( $\text{Bq}/\text{m}^2$ ) has decreased according to the physical decay; the horizontal movement of radiocesium is small.
- Radiocesium in the ground has gradually penetrated into deeper parts; however, most of radiocesium is still contained within 5 cm from the soil surface.
- A decreasing tendency in air dose rates drastically differs depending on the condition: the average air dose rate reduction declining in the order of; above roads > in various living environments > in undisturbed flat fields > in forests.
- Various human activities as well as decontamination are considered to accelerate the air dose rate reduction.
- On the basis of statistical analyses for a large amount of environmental monitoring data, air dose rate distributions were predicted up to 30 y after the accident; the results indicated faster reduction of contamination conditions in living environment than the physical decay.

## 18-3: Factors affecting distribution coefficient of radiocesium in various agricultural environments of Fukushima

National Institute for Agro-Environmental Sciences

Sadao Eguchi

### Introduction

The distribution coefficient ( $K_d$ ) of radiocesium (rad-Cs) controls the dissolved concentration of rad-Cs, which strongly affects the rad-Cs transport in soil and aquatic environments as well as the absorption by crops. Understanding of the mechanisms of the spatiotemporal variation of the  $K_d$  value under various agricultural field conditions is a prerequisite for predicting the rad-Cs dynamics in contaminated agro-ecosystems in Fukushima from short to long-term perspective.

### Conclusions

- The measured freshwater ecosystem  $K_d$  values for  $^{137}\text{Cs}$  in various farmlands in Fukushima varied over 5 orders of magnitude ( $7 \times 10^2 \sim 5 \times 10^6 \text{ L}/\text{kg}$ ) and covered the range of all the published freshwater ecosystem  $K_d$  data before and after the Tokyo Electric Power Company Fukushima Daiichi Nuclear Power Plant accident.
- The  $K_d$  values largely fluctuated with the dynamic water quality conditions (electrical conductivity, dissolved organic carbon concentration,  $\text{K}^+$  concentration,  $\text{NH}_4^+$  concentration) as well as the radiocesium interception potential (RIP) value, showing significant seasonal (intra-annual) changes with a less significant long-term trend.
- The frayed edge site  $K_d$  values, calculated from the RIP and water quality data, were generally one order of magnitude or much more smaller than the measured  $K_d$  values, suggesting the importance of other  $^{137}\text{Cs}$  retention sites (fixed at collapsed interlayer sites, contained in dead or alive biological cells, occluded in iron precipitates, etc.).

#### 18-4: Reducing radioactive Cs concentrations of forage crops after TEPCO's Fukushima Daiichi NPP accident in Japan

NARO Institute of Livestock and Grassland Science

Hisatomi Harada

##### Introduction

The development of remediation measures against food contamination by radioactive Cs became a pressing issue in northeastern Japan after Tokyo Electric Company's Fukushima Daiichi Nuclear Power Plant accident in 2011. Several effective and practical methods for reducing radioactive Cs transfer in forage production have been elucidated and improved by researchers in NILGS and prefectural research stations.

##### Conclusions

- For perennial grasses, the modified grassland renovation is effective to reduce radioactive Cs concentration. The modifications are as following; the surface grassland layer with higher radioactive Cs and organic matter contents should be mixed with underlying soil thoroughly and deeply, and K fertilizer should be applied to reach soil exchangeable  $K_2O$  content of 30-40 mg/100g in the depth of 0-15cm.
- A part of volcanic ash soil with high carbon content, showed lower RIP and higher radioactive Cs of grasses in the survey for soil factor affecting radioactive Cs transfer in 2012.
- For forage corn, continuous cattle manure application at the rate of 3 kgFM/m<sup>2</sup> has a reducing effect on <sup>137</sup>Cs concentration. Application of the contaminated manure of 3900 Bq/kgDM at the rate of 4.3 kgDM/m<sup>2</sup> increased the <sup>137</sup>Cs concentration in the soil by 64 Bq/kg, and in the forage corn by 9.2 Bq/kgDM. The transfer factor was lower than that observed for soil to which uncontaminated manure had been applied as a control. Thus, cattle manure can be safely and effectively used for reducing radioactive Cs transfer from soil.

#### 18-5: Reduction of radioactive caesium concentration in soybean seeds by potassium application in Japan

National Agriculture and Food Research Organization(NARO)

Tetsuo Sekiguchi

##### Introduction

Concerns have been raised about the risk for radioactive Cs contamination of soybean seeds caused by TEPCO Nuclear Power Plant accident. There has been no practical remediation measures to reduce radioactive Cs concentration (RCs) in soybean seeds in Japan. We examined the effectiveness of potassium(K) application on decreasing RCs in soybean seeds in Japanese environmental conditions and developed practical countermeasures to reduce RCs below the Japanese allowance level (100 kq/kg).

##### Conclusions

- TF and RCs in soybean seeds was negatively correlated with the Ex-K content of the topsoil during the harvest season.
- Potassium application to decrease RCs in soybean seeds is significantly effective in Japanese environmental conditions .
- It is necessary to keep Ex-K<sub>2</sub>O content above 25 mg /100g during growth to decrease RCs below the allowance level .
- To enrich Ex-K<sub>2</sub>O content up to 25 mg /100g by applying the usual amount of K fertilizer with readily available K fertilizer before sowing is more effective to reduce RCs in seeds than using slow released K fertilizer.
- Satisfactory results have been received from product monitoring in which samples contained RCs exceeded the allowance level decreased from 0.5% in 2013 to 0.1% in 2014.
- There are still some cases that cannot be explained based on the Ex-K content, so further studies on the factors that affect the absorption of Cs from the soil to the plant and the uptake mechanism of Cs are required.

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## 18-6: Radiocesium reduction rates in fruit trees in Japan

National Institute of Radiological Sciences

Keiko Tagami

### Introduction

The purpose of this study is to answer the following FAQs, (1) How fast do Cs concentrations decrease in fruits? and (2) Does the decreasing rate differ with tree species? Thus, we measured effective half-lives ( $T_{eff}$ ) of radiocesium in persimmon and loquat trees collected in Chiba City. To estimate  $T_{eff}$  for other fruit trees in Fukushima Prefecture, open source  $^{137}\text{Cs}$  concentration data in edible part of fruits collected from local government (cities, towns and villages) since 2011 were used.

### Conclusions

- The observed  $T_{eff}$  of radiocesium ranged from 183-473 d, and these values were similar to those observed after the Chernobyl accident.
- The rates of decline of radiocesium concentration has slowed in the most recent year of measurement (2014) compared with the period of 2011-2013.
- Aboveground, directly deposited radiocesium was likely the main source of contamination in the first three years. However, the slowly decreasing rates suggested that root uptake may become the major transfer pathway to fruits in the near future.

<One comment from floor> The through bark Cs uptake process in trees seems probable but is not clear. Some more evidence including radiotracer experiment data are necessary to support this field-based observation.

## 18-7: Linking radiocaesium sorption to soil chemical properties for Japanese soils: A comparison to European soils

SCK•CEN & K.U.Leuven, Belgium

Shinichiro Uematsu (suematsu@SCKCEN.BE)

### Introduction

The radiocaesium interception potential (RIP), a specific factor determining radiocaesium ( $^{137}\text{Cs}$ ) sorption in soil, and other major soil physicochemical properties were characterised for soils collected from the Fukushima accident affected area. A model to predict the RIP with soil properties was developed for Japanese soils.

### Conclusions

- The contribution of the soil clay fraction to selective radiocaesium sorption was three times ( $P < 0.001$ ) lower for the Japanese soils than Belgian soils, suggesting that amorphous minerals in Japanese Andosols contribute to the lower affinity of the clay minerals to  $^{137}\text{Cs}$ .
- The soil RIP decreased with increasing organic matter content, suggesting that highly selective sorption sites of  $^{137}\text{Cs}$  in the soil clay fraction are masked by soil organic matter.
- Andosols exhibited significantly lower soil RIP than other Japanese soil types, indicating that Andosols may be more vulnerable than other soils for  $^{137}\text{Cs}$  contamination.
- Soil organic matter and cation exchange capacity (CEC) explained 64% of the variation in the RIP, allowing to map the soil RIP based on existing soil map information.

## 18-8: Cs dynamics in plants

Dept. of Cellular Integrative Physiology, Fukushima Medical Uni., Daisuke Kobayashi

### Introduction

Stable Cs-133 is present in the soil in the order of ppm and Cs can be detected in plant roots and leaves. However, Cs is not an essential for plant growth. The Cs transport pathway of plant is not well known. If we know the transporter's character, we can understand cesium dynamics in plant.

### Conclusions

In a molecular biological study, characterization of K-channel and K-transporter revealed one of these transporter proteins was inhibited by Cs in a competitive manner, and that one of those proteins was able to transport Cs.

- Cs is the most toxic element of the alkaline metals
- Cs in xylem sap behaves as Cs<sup>+</sup>
- The protein AtHAK/KUP/KT9 is a candidate to be a Cs transporter
- Cs inhibits chlorophyll synthesis

## 18-9: Time Changes in radiocesium wash-off from various land uses and transport through river networks after the Fukushima Daiichi NPP accident

Center for Research in Isotopes and Environmental Dynamics, University of Tsukuba Yuichi Onda

### Introduction

Due to the Fukushima Daiichi Nuclear Power Plant accident, radioactive materials including Cs-134 and Cs-137 were widely distributed in the surrounding area. The radiocesium has been transported in river networks. The monitoring started at 6 sites from June 2011. Subsequently, an additional 24 monitoring sites were installed between October 2012 and January 2013. Flow and turbidity (for calculation of suspended sediment concentration) were measured at each site, while suspended sediments and river water were collected every one or half month to measure Cs-134 and Cs-137 activity concentrations by gamma spectrometry.

### Conclusions

- The Cs-137 activity concentration in eroded sediment from the runoff-erosion plot has been almost constant for the past 3 years, however the Cs-137 activity concentration of suspended sediment from the forested catchment showed a slight decrease through time.
- The suspended sediment from paddy fields and those in river water from large catchments exhibited a rapid decrease in Cs-137 activity concentration with time.
- The decreasing trend in Cs-137 activity concentration was fitted by a two-component exponential model. Differences in the decreasing rate of the model were compared and discussed among various land uses and catchment scales.
- Such analysis can provide important insights into the future predictions of the radiocesium wash-off from catchments from different land uses.
- The decreasing trend of the river system varied with catchments. Our analysis suggests that these differences can be explained by upstream land use with different decreasing trends.

## 18-10: Deposition and dynamics of radiocaesium in forest ecosystem for four years after the Fukushima Daiichi NPP accident

Forestry and Forest Products Research Institute

Shinji Kaneko

### Introduction

Forest covers 70% of the territory of Fukushima Prefecture, so radioactive contamination due to the Fukushima Daiichi NPP accident is a great issue for inhabitant in this area, especially foresters. In order to recognize the distribution and behavior of radiocaesium in forest ecosystems, we established study plots in three sites with different contamination levels in Fukushima prefecture.

### Conclusions

- The amount of radiocaesium deposition differed correspondingly to the contamination level in study forest.
- Radiocaesium distribution pattern was different between Japanese cedar forest and mixed forest of red pine and broad leaved forest.
- The existence of radiocaesium in wood suggested uptake by tree occurred immediately after deposition. Radiocaesium distribution in the forest ecosystem largely changed from 2011 to 2012 as a result of the migration of radiocaesium from canopy to forest floor.
- Change of radiocaesium distribution was small after 2012.
- Total radiocaesium inventory was almost constant in the forest ecosystem during the observation periods.
- More study should be focused in radiocaesium uptake in subsequent years.

## 18-11: Transport of dissolved and particulate radiocesium in Japanese forested ecosystems

Forestry and Forest Products Research Institute

Masahiro Kobayashi

### Introduction

To clarify transport of Fukushima-derived radiocesium by water flow through the forest canopy, litter layer, and mineral soil, we determined the concentration of dissolved and particulate radiocesium in throughfall (TF), litter leachate (LL), and soil water (SW) sampled at forested catchments in Ibaraki and Fukushima prefectures.

### Conclusions

- In the period just after the accident, most radiocesium in TF was detected as dissolved.
- The concentration of dissolved and particulate radiocesium in TF and LL gradually decreased with time.
- In summer, particulate radiocesium in TF and LL temporally increased and enhanced the transport from the trees to the soil.
- The suspended solid seemed to be organic carbon with a small amount of mineral particles.

## 18-12: Modelling spatio-temporal dynamics of radiocesium deposited onto forests following the Fukushima nuclear accident

Forestry and Forest Products Research Institute

Shoji Hashimoto

### Introduction

About 70 % of the contaminated area is covered by forests. A holistic view (synthetic approach) is necessary to facilitate effective remediation measure strategies. The present study predicted the temporal dynamics of the radiocesium deposited in forests in Fukushima using a model for Radionuclides In a Forest Ecosystem (RIFE1) with new data and new root uptake assumptions.

### Conclusions

- Prediction of inventories did not differ between root uptake assumptions.
- Compared with an earlier modelling study (Hashimoto et al. 2013), migration to the soil compartment from the tree and surface litter compartments slightly slowed but the trends were basically the same.
- Over a period of one to two years, radiocesium is predicted to move from the tree and surface organic soil to the mineral soil, which eventually becomes the largest radiocesium reservoir within forest ecosystems.
- Prediction of  $^{137}\text{Cs}$  in stem wood differed depending on assumptions of root uptake but orders of magnitude increases in  $^{137}\text{Cs}$  concentration in stem wood seem less likely based on the new simulation.

## 18-13: Radiocaesium in Abukuma river water measured at Kuroiwa and Harai in Fukushima city

Fukushima University

Kenji Nanba

### Introduction

The Abukuma river originates from the southern end of Naka-dori Fukushima prefecture, flows through the major cities of Fukushima prefecture including Shirakawa, Sukagawa, Koriyama, Motomiya, Nihonmatsu, Fukushima and Date, and enters the Pacific ocean from the Miyagi Prefecture. From April 2012, weekly observation of radiocaesium in the river water at Kuroiwa in Fukushima city started. The measurements included quantification of radiocaesium in suspended substance (SS), dissolved forms of radiocaesium, and organic forms of radiocaesium. From 2014 similar observations were started for the Harai river.

### Conclusions

- The concentration of radiocaesium in SS often exceeded 10,000 (Bq/kg DW) in 2012 and 2013. After 2014 it did not exceed 10,000 (Bq/kg DW).
- Dissolved radiocaesium showed a decreasing trend with seasonal variation, high in summer and low in winter. Dissolved radiocaesium was 30-50 % of the total radiocaesium in the water.
- Particulate organic radiocaesium was a few percent of the total particle radiocaesium.
- Most of  $K_d$  values in Abukuma river water were in the range  $6 \times 10^5 - 1 \times 10^6$  L/kg.
- The  $K_d$  values did not show a relationship with ammonium or potassium concentration. They showed a negative influence of DOC concentration.
- Radiocaesium concentrations in the Harai river were higher for SS (Bq/kg DW), dissolved (Bq/L), organic (Bq/kg DW) and total (Bq/L) than in Abukuma river at Kuroiwa whereas they were similar for the  $K_d$ .

18-14: Time series and spatial distribution of <sup>137</sup>Cs in dissolved and suspended fractions in irrigation water collected within 80 km zone around TEPCO's Fukushima Daiichi Nuclear Power Stations

Institute of Environmental Radioactivity, Fukushima University

Hirofumi Tsukada

**Introduction**

There are more than 3,700 irrigation ponds in Fukushima Prefecture, and they are mostly used in association with rice paddies although carp were farmed in some ponds. Migration of radionuclides in the environment was different according to their physicochemical fractions. The present study determined seasonal variation and spatial distribution of <sup>137</sup>Cs concentration in suspended and dissolved fractions of irrigation water collected within 80 km zone.

**Conclusions**

- The concentration of radiocesium in dissolved fraction of irrigation water is decreasing with the time elapsed.
- 95% of radiocesium in suspended matter collected from Oguni was in the strongly bounded to particles.
- Most of radiocesium in the dissolved fraction existed as Cs<sup>+</sup> (cation).
- The K<sub>d</sub> value in Oguni decreased with increasing content of suspended matter. However, the K<sub>d</sub> value in Ohta had a relatively constant value
- K<sub>d</sub> values collected from the 80 km zone varied by three orders of magnitude and the geometric mean was 110,000, which is higher than the IAEA reported values (TRS472).
- The K<sub>d</sub> individual values depended on the areas and there was a negative correlation with the RIP values.

18-15: Cost and effectiveness of decontamination against radio Cs contamination in Fukushima  
-How do we manage decontamination waste more effectively?

National Institute of Advanced Industrial Science and Technology

Tetsuo Yasutaka

**Introduction**

The first objective of this study was to evaluate the cost and effectiveness of radiation decontamination in Fukushima Prefecture in its entirety. The second was to evaluate the cost and effectiveness of volume reduction technology employed for the radioCs contaminated waste.

**Conclusions**

Evaluation of decontamination cost (published by Yasutaka et al.,2015)

- The total cost of decontamination is estimated to be 13 - 38 Billion EURO. The cost differs for each scenario.
- This result indicates that the decontamination cost can be reduced by selecting the appropriate decontamination scenarios and methods.
- In the special decontamination zone, the decontamination costs for storage of contaminated soil account for the largest part of the total cost.
- In the Intensive Contamination Survey Areas, the decontamination costs for removal account for the largest part of the total cost.

Evaluate the effectiveness of volume reduction technology (VRT).

- Volume reduction technologies is sometimes effective but not always.
- The total cost is mainly dependent on the "Reuse and final dispose scenario"
- We must evaluate and discuss the cost and reuse possibility before apply VRT

18-16: New geochemical tracers for source identification and long-term environmental behavior studies of radionuclides released from the Fukushima nuclear accident

National Institute of Radiological Sciences

Jian Zheng

**Introduction**

Source identification of radioactive contamination and long-term environmental behavior of released radioactive materials are important issues to study after the FDNPP accident. We discuss new geochemical tracers, such as Pu isotope ratios ( $^{240}\text{Pu}/^{239}\text{Pu}$  and  $^{241}\text{Pu}/^{239}\text{Pu}$ ) and radioactive Cs isotope ratios ( $^{135}\text{Cs}/^{137}\text{Cs}$  and  $^{135}\text{Cs}/^{133}\text{Cs}$ ) for contamination source identification and long-term environmental studies of Pu and Cs released from the FDNPP accident.

**Conclusions**

- Pu was released from Unit 1-3 reactors, there was no release from the spent fuel pools
- Our results highlight the necessity for long-term dose estimation for  $^{241}\text{Pu}$  and  $^{241}\text{Am}$
- The amounts of Pu isotopes released were four orders of magnitude lower than that of the Chernobyl accident, and no immediate Pu contamination was detected in the marine environment 5-220 km off Fukushima
- The Fukushima nuclear accident-sourced  $^{135}\text{Cs}/^{137}\text{Cs}$  ratio was identified to be 0.341, which is very close to that of the Chernobyl accident, but significantly different from global fallout
- We demonstrated that radiocaesium was mainly released from the Unit 2 reactor due to the breach of the Unit 2 containment vessel. The possible release of FPs from the SFPs was negligible.
- The  $^{135}\text{Cs}/^{137}\text{Cs}$  ratio is a useful fingerprint and tracer for contamination source identification, and potential geochemical tracer to study the long-term environmental behavior of Fukushima accident-sourced radioactive Cs.

19-1: Survey on Radionuclide Movement in Wild Boars and some Wild Animals in Fukushima

Fukushima Prefectural Centre for Environmental Creation Development Unit

Toshio Mizoguchi

**Introduction**

The issue of wild boars in Fukushima has been becoming more and more serious since the accident of Fukushima Daiichi Nuclear Power Plant. Some boars were found every year which radiocaesium activity in the muscle of more than a thousand Bq/kg. All wild boars in Fukushima are subject to regulation because their radiocaesium values exceed the food regulatory values. Measurement of radionuclide activities in wild animals other than wild boars has been conducted on gamma-emitting nuclide by using germanium semiconductor detector since 2011, includes Japanese black bear and Japanese shika-deer.

**Conclusions**

- The time series change of Cs-137 concentration in the muscle of Asian Black Bears generally shows a downward trend and also the fluctuation in the range for each fiscal year has remarkably decreased in size.
- Regarding Japanese sika-deer, In 2011 and 2012, concentrations exceeding 100 Bq/kg were frequently detected, but in 2013 and 2014 the concentrations in muscles were lower than 100 Bq/kg and seem to be stabilizing.
- These clear features can be found in the results of wild boars. First, there is no decreasing trend, but a stagnant trend. Second, the range of fluctuation in each fiscal year is not decreasing in size, but strongly fluctuating. Third, there are many areas where contamination in the muscle are high, for example, exceeding 1,000 Bq/kg. These areas were intensively observed in the evacuation areas and surrounding areas, where the soil contamination levels are distinctively higher than other areas.

### 19-3: Radionuclides transfer in marine organisms around Japan

Fukushima Project Headquarters, National Institute of Radiological Sciences,

Tatsuo Aono

#### Introduction

Artificial radionuclides such as  $^{137}\text{Cs}$  and  $^{239}\text{Pu}$ ,  $^{240}\text{Pu}$  have entered terrestrial and marine environments through radioactive fallout, and liquid and solid-state radioactive wastes before the Chernobyl and Fukushima accident. However, the Fukushima Daiichi Nuclear Power Station (FDNPS) accident completely changed the marine environment immediately. It is important to make a comparison between the pre-accident levels and the levels after and since the accident. This study aimed to examine the temporal and spatial variation of radionuclide activities in marine environment and to discuss the Concentration Ratio (CR) of marine biota around Japan.

#### Conclusions

- The seawater around/off Fukushima
  - The Cs activities in seawater are gradually decreasing to the background level before this accident.
- The sediment around/off Fukushima
  - Fukushima derived Cs in sediments have tended to transfer far out at sea.
  - Fukushima derived Pu were not observed in sediments.
- The marine biota around/off Fukushima
  - Fukushima derived Cs in marine biota has gradually decreased.
  - The estimated CR-Cs values in marine biota around Fukushima were higher than the reported values in TRS-422, and similar to  $K_d$ -Cs value in TRS-422. It seemed that the particles such as suspended matter, settling particles and sediment caused the high CRs-Cs after this accident.

### 19-4: Effects of the nuclear power plant accident on marine products and fisheries in Fukushima

Institute of Environmental Radioactivity, Fukushima University

Toshihiro Wada

#### Introduction

After the release of huge amount of radionuclides into the ocean from the devastated Fukushima Dai-ichi Nuclear Power Plant (FDNPP), safety concerns have arisen for marine products in Fukushima Prefecture. As of December 2014, the Fukushima Prefectural Government has measured the radiocesium ( $^{134}\text{Cs}$ ,  $^{137}\text{Cs}$ ) concentrations from 23,850 specimens within 177 marine species. In this report, I show the monitoring results overview and recovery process of Fukushima's fishery, mainly focused on the trial fishing operations.

#### Conclusions

- Radiocesium concentration in marine products have decreased significantly.
- Radiocesium contamination of fishes (especially in demersal fish) is still continuing, but the levels are low, except for those in the port of the FDNPP.
- Fish biomass has increased since the accident.
- Fishery facilities have been restored gradually.
- Target areas for trial fishing operations have expanded and target species has increased up to 64 species as of July 2015.
- Long-term and careful monitoring of marine products, especially around the FDNPP are necessary to lift restrictions on shipment, to expand the trial fishing operations reliably, and to prevent harmful rumours in the future.

19-5: Distributions of Fukushima-derived  $^{137}\text{Cs}$  in sediments and their temporal change: Results from four-year monitoring in the waters off Fukushima and nearby prefectures, Japan

Marine Ecology Research Institute

Masashi Kusakabe

**Introduction**

Soon after the Fukushima Daiichi nuclear power plant accident, the Marine Ecology Research Institute launched a monitoring project in the waters off Fukushima and nearby prefectures for radioactivity in seawater and bottom sediments under contract with the Ministry of Education, Culture, Sports, Science and Technology (currently with the Nuclear Regulation Authority). Bottom sediments are collected at 32 monitoring sites four times a year. The possible contributing factors for spatio-temporal changes of  $^{137}\text{Cs}$  in the sediment will be discussed.

**Conclusions**

The concentrations of  $^{137}\text{Cs}$  in the surface sediments shows wide variation ranging from 0.8 to 540 Bq/kg-dry during the entire monitoring period. Almost all of the data exceeded the level of pre-accident 5yr-average (0.87 Bq/kg) in the waters off Fukushima Prefecture. Proximity of the sampling site to the accident site did not necessarily correspond to the higher values.

- Cs-137 in the surface sediments decrease with a half life of ca. 2 yrs. Sediments rich in coarse/medium sand lost more  $^{137}\text{Cs}$  than clayey sediments.
- No evidence for downward migration of  $^{137}\text{Cs}$  due to biological activity.
- Resuspension and subsequent lateral transportation of the sediment may play an important role for the redistribution of  $^{137}\text{Cs}$  in the surface sediments.
- Cs dissolution/desorption from the sediment may be also important and needs to be evaluated.

## 5. Summary of the workshops

Contributing authors: B Howard, H. Tsukada, A. Takeda, C. Adam, P. Hurtevent, G. Shaw, S. Uchida, K. Tagami, S. Eguchi

The range of topic areas covered by the oral and poster presentations given in the two workshops was very broad. The majority of the information was focused on soil, paddy fields, forests and catchments, and marine areas. Some of the key scientific outcomes are summarized below with the names of some of the relevant first authors whose inputs are outlined above.

There was a very large amount of information on radiocaesium behaviour obtained after the Chernobyl accident, which have a European bias, and we need to consider whether the data and models have general applicability worldwide. Some differences, discussed below, are becoming evident between the environmental behaviour of radiocaesium after the accidents at Chernobyl and Fukushima. Evaluation of recently acquired Fukushima data allow initial identification of where environmental data from Chernobyl were applicable and where they were not. Such reflections could be incorporated into the current consideration of suitable topics for MODARIA II to be discussed in Nov 2015 at IAEA Vienna.

### 5.1. *Source term and deposition - characterisation, estimation and external doses*

The source term after the Chernobyl accident was associated with a single reactor emitting a semi-continuous atmospheric release for 10 days deposited largely over terrestrial



ecosystems. Deposition of released radionuclides occurred in the far field in many other countries as well as the near field. Hot particles were an important feature of deposition close to the reactor. That for the Fukushima Daiichi accident was complex with multiple sources, discharging at different times into marine, freshwater (or irrigated) and terrestrial environments. Significant deposition was confined to Japan. Characterisation and quantification of the Fukushima releases is still ongoing with current possible overestimation of the release of radioiodine and radiocaesium from Unit 3 being investigated (Yamazawa).

Detailed deposition maps for  $^{134}\text{Cs}$ ,  $^{137}\text{Cs}$ ,  $^{131}\text{I}$ ,  $^{129\text{m}}\text{Te}$ ,  $^{110\text{m}}\text{Ag}$ ,  $^{238}\text{Pu}$ ,  $^{239+240}\text{Pu}$ ,  $^{89}\text{Sr}$  and  $^{90}\text{Sr}$  have been created for a series of aerial, above ground and soil sample measurements. The deposition density of radiocaesium in undisturbed flat fields ( $\text{Bq}/\text{m}^2$ ) has decreased according to physical decay with small lateral movement. The reduction in air dose rates with time differs with the type of surface, with the average air dose rate reduction declining in the order of; above roads > in various living environments > in undisturbed flat fields > in forests. Various human activities also accelerate the air dose rate reduction (Saito).

Radiocaesium deposited on the ground has gradually moved down the soil profile but most is retained within 5 cm of the soil surface. Statistical analyses of the environmental monitoring data have allowed predictions to be made of the spatial variation in the air dose rate for up to 30 y after the accident. A faster reduction of external doses in the living environment is anticipated than that due to physical decay alone (Saito) due to soil migration, and natural and man-made decontamination.

The tsunami deposit onto terrestrial areas was characterised as a coarse sand layer overlain by a muddy, layer which was contaminated with radiocaesium, some of which was in particulate form (Nanzyo).

## 5.2. *Radiocaesium in soil*

After Chernobyl, soil type differences were important for radiocaesium especially over the longer term. Most radiocaesium in soil exists in a strongly bound fraction, adsorbed into weathered fronts in the mineral interlayers expanding into wedge shapes as frayed-edge sites (FES). FES fixation limits desorption and uptake by plants and was quantified by the development of the radiocaesium interception potential (RIP). The RIP method, which represents the selective adsorption capacity of caesium in soil, was widely adopted, as were mechanistic models which used exchangeable K, clay mineral, organic matter and pH as influencing parameters. Radiocaesium availability in the soil was high for sandy and highly organic soils. RIP measured in soils from an international data bank was only loosely correlated with other soil characteristics, but was linked to the acid-extractable fraction.

After Fukushima, the behaviour of radiocaesium in andosols has been a key issue and consideration is being given regarding whether it is useful to adapt the RIP methodology to improve estimates of availability for plant uptake in Japan. Many of the studies in Japan have systematically measured a wide variety of soil characteristics, including exchangeable ion concentrations, RIP and detailed mineralogy as well as radiocaesium uptake by plants. Linking such studies will provide valuable validation of radiocaesium models which use the RIP concept. For example, TF values in polished rice, potato and cabbage from Aomori and Fukushima (rice only) prefectures significantly decreased with RIP values (Tsukada).

A large amount of new data on RIP and its relationship with soil characteristics such as OM and CEC for Japanese soils has been reported. The reduction in soil RIP with OM suggests that the OM is masking selective sorption sites (Yamaguchi). RIP in Japanese soil appears to be mainly controlled by the amount of micaceous clay minerals in soils from the Fukushima region as well as from different areas in Japan (Tashiro, Uematsu). Andosols, rich in OM and in non-crystalline (amorphous) clay minerals, tended to show lower RIP than other soil types in Japan (Takeda, Harada, Uematsu). The contribution of the clay fraction for 51 Japanese surface soils was three times lower than that for Belgian soils possibly due to the presence of amorphous minerals (Uematsu). As the Absalom (1999) model is based on European soils, it may overestimate RIP and thereby underestimate radiocaesium uptake by plants in Japan. Some Andosols showed high RIP, possibly caused by the presence of other micaceous minerals from aeolian dust (wind borne material) (Takeda). Such complexities in mineralogy suggest that it might be difficult to predict RIP of all Japanese soils from only a few simple chemical properties.

Radiocaesium vertical migration in undisturbed forest and grassland soils in the Fukushima contaminated area was significantly faster compared with the Chernobyl 30-km zone during the first three years. The differences can be partially explained by the higher precipitation in Fukushima prefecture than for the area around Chernobyl (Konoplev).

“High” radiocaesium uptake into crops or agricultural animals in Japan may not be considered so from a wider perspective, as the food permissible limits applied in Japan are low compared with those in other parts of the world. Therefore, the comparison with radiocaesium in crops is with a low permissible radiocaesium value. Similarly, an OM content in Japanese soils is considered to be *ca.* 10-15%, whereas soils in Western Europe with persistent restriction on food production often have OM contents exceeding 30%. This emphasises the importance of comparing measured activity concentrations in crops with those deposited so that transfer can be compared with data from other countries more readily and put into an appropriate context.

### 5.3. *Caesium in plants – gene expression, cultivars, fruit*

Stable Cs (<sup>133</sup>Cs) is present in soil in ppm quantities and also in plant roots and leaves in ppb quantities, even though Cs is not an essential for plant growth. Cs is the most toxic element of the alkaline metals and inhibits chlorophyll synthesis. The Cs transport pathway of plant is not well known and efforts have been ongoing in Japan to identify both K and Cs transporters’ characteristics, to help understand caesium dynamics in plant.

Gene expression analysis has been ongoing to identify genes and proteins associated with K and Cs uptake in Japan. In a molecular biological study, characterization of a K-channel and K-transporter has shown that one of the K transporter proteins is inhibited by Cs in a competitive manner (Kobayashi).

High affinity K transporters have also been identified in rice. Rice cultivars which have relatively high radiocaesium uptake (indica cultivars) also have relatively high gene expression of one of these K transporter genes at very low [K] in soil (Ishikawa). Further studies are required to clarify the possible role of Cs transport proteins.

In poplars, root and foliar absorption of Cs involves different transport pathways with Cs absorbed by root transported by transpiration in xylem and foliar absorbed Cs transferred by phloem (Noda). However, Cs from both pathways behaves similarly in the tree recycling

system after absorption. Gene expression analysis in poplar indicated that PttSKOR-like2 localization suggested PttSKOR-like2 might transport Cs/K into xylem sap in the poplar root. Furthermore, PttSKOR-like2 expression was regulated by environmental factors such as day length and/or temperature.

#### 5.4. *Radiocaesium in paddy fields (incl. Kd)*

There are no paddy fields at Chernobyl whereas paddy rice is the most important crop in Japan. Many paddy fields in mountainous areas in Fukushima Prefecture, Japan, are irrigated with water that flows from nearby forests growing in mountainous terrain. Rice plants grown in paddy fields take up radiocaesium both from the soil and from irrigation water. In 2011, the [ $^{134}\text{Cs}+^{137}\text{Cs}$ ] in some brown rice grown in such mountainous areas in the prefecture exceeded the provisional permissible limit at that time of  $500 \text{ Bq kg}^{-1}$ . Rice planting generally starts from early May-June thus the contribution of radiocaesium direct deposited onto rice plants was thought to be negligible to the total amount in rice grains.

After the Fukushima accident many studies have looked at the rate of inflows and outflows from paddy fields to estimate the changes with time in the radiocaesium inventory and physico-chemical form. The data are highly variable depending on factors such as: (i) methodology used to sample and measure the radiocaesium, season (eg. Nanba), river, catchment characteristics and water flow in these dynamic systems.

A large database on Kd for  $^{137}\text{Cs}$  in freshwater systems including paddy fields has been compiled (Eguchi). The Kd values vary by over 5 orders of magnitude covering the previously published ranges. The Kd values vary with water quality conditions (electrical conductivity, DOC,  $[\text{K}^+]$ ,  $[\text{NH}_4^+]$ ) which vary considerably with time, as does the RIP, showing significant seasonal changes with a less significant long-term trend.

The frayed edge site Kd values, (calculated from RIP,  $\text{NH}_4^+$ ,  $\text{K}^+$  data), were generally at least one order of magnitude smaller than the measured Kd values. The discrepancy suggests the presence of other  $^{137}\text{Cs}$  fixation sites such as collapsed interlayer sites, undecomposed biological material and iron precipitates [Eguchi].

At one intensively surveyed site at Oguni, 95% of radiocaesium in suspended matter was strongly bound to particles with the [RCs] in the dissolved fraction (as  $\text{Cs}^+$ ) of irrigation water decreasing with time. The Kd value in Oguni decreased with increasing content of suspended matter, but that in a different site at Ohta was relatively constant. Surveys of paddy field inflow irrigation water within the 80 km zone showed Kd varying by three orders of magnitude with a geometric mean of  $110,000 \text{ L/kg}$ , which is higher than that given in TRS 472. The Kd values varied between areas and were negatively correlated with RIP (Tsukada).

The physico-chemical form of the radiocaesium and proportion of radiocaesium in the dissolved form matters as it affects mobility and transfer to rice. In controlled lab experiments, the uptake rate for radiocaesium in a dissolved form was higher than that for the organic-matter-bound and soil-particle-bound fractions (Suzuki).

The application of a simplified process-based model of radiocaesium soil-plant transfer by Absalom et al (1999) for paddy rice showed that, without calibrating any soil and crop parameters, the model underestimated brown rice TF by one or two orders of magnitude. However, calibrating model crop and soil parameters allowed substantially improved

predictions. Furthermore, modifying the model to take into account the soil clay mineralogy considerably improved the model predictions giving an  $R^2$  value of 0.89 (Eguchi).

A comparison of the impact of rice harvest in several paddy fields at different distances from irrigation water inlets has shown that [ $^{137}\text{Cs}$ ] for soil, rice and rice straw and the TF values tend to be higher within 3 km of the inlet (Suzuki).

Data from Japan for transfer of radiocaesium from soil to rice are provided in TRS 472, but are relatively sparse for both radiocaesium and many other radionuclides compared with other crops. Therefore, the data now being acquired at Fukushima will be invaluable in providing a detailed understanding of the behaviour of radiocaesium in such agricultural systems. From a worldwide perspective this is crucial given the huge importance of rice as a staple component of the diet in large parts of the world.

### 5.5. *Radiocaesium in forest ecosystems*

Forests are important components of the landscape around both the Chernobyl and Fukushima Daiichi NPPs. Forest covers 70% of Fukushima Prefecture, so radioactive contamination due to the Fukushima Daiichi NPP accident is an important issue for inhabitants, especially foresters.

After Chernobyl, data collection in forests was not an initial priority so the early phase redistribution between parts of the trees where initial interception occurred and other parts of the ecosystem was not fully characterised. However, there is a large amount of long-term data on radiocaesium behaviour in forests after the Chernobyl accident which can be compared with that obtained from the Fukushima data (Yoshenko). After Fukushima, a considerable amount of research and associated modelling on forested catchments has been carried out.

Airborne and car-borne surveys in evergreen coniferous regions show that average radiation levels decreased much faster than expected from physical decay with decreases of 25-40% during the first 18 months (Gonze). Measurement of ambient dose rates in forests have been shown to be higher on ridges and from east-facing slopes (Takeuchi). Enhanced  $^{137}\text{Cs}$  deposition on forest floors via throughfall has been associated with an increase of ambient dose rate (Kato).

Valuable early data was collected enabling better quantification of interception of deposition, and the considerable spatial and temporal variation in radiocaesium flux in throughfall and litter fall with relevant driving variables (Kobayashi). This study showed that in the first 1.5 months after the Fukushima accident, the total (dissolved + particulate) [RCs] in throughfall was measured. A large proportion of deposited radiocaesium was retained in the litter layer soon after the accident. In the following months, the [RCs] in throughfall and litter layer gradually decreased and the proportion of radiocaesium in the particulate phase increased. In the summer, the amount of radiocaesium present as suspended particulate matter in throughfall and litter layer often increased and resulted in higher particulate [RCs]. The [RCs] in throughfall and litter layer both decreased in winter. The temporal increase in suspended particulate matter seems to be composed mainly of organic C and may have been due to a combination of insect herbivory and litter decomposition.

Throughfall and litter fall was also identified as a major pathway for  $^{137}\text{Cs}$  redistribution leading to the rapid transfer of  $^{137}\text{Cs}$  between tree and soil 3 years after the accident. For example, In Nov 2013, on forest andosols 90% of the  $^{137}\text{Cs}$  was in the forest floor, 70% of which was in the upper organic layer and the upper soil layer (Coppin). Much of the sorbed  $^{137}\text{Cs}$  is exchangeable (15-50%) reflecting the low clay mineral content and high OM content of the andosol.

Differences between the radiocaesium transfer between compartments between different stands of tree species and due to climate variables, especially typhoons, are becoming evident in forests. The radiocaesium distribution in forests changed from 2011 to 2012 largely as a result of the migration of radiocaesium from the tree canopy to the forest floor (Itoh). Most of the redistribution occurred in 2011 and 2012, although it is still significant three years after the accident (Hurtevent). The pattern of radiocaesium distribution in forest components differed between Japanese cedar forest and mixed forests of red pine and broad leaved forest (Kaneko). During the initial phase, when interception of radiocaesium by coniferous leaves and bark occurred, there was early evidence of the transfer of radiocaesium into wood. The role of radiocaesium interception by bark and subsequent translocation seems to be important from field data, but needs to be clarified and quantified under controlled experimental conditions. As radiocaesium moved to the forest floor root uptake became more important as a source of radiocaesium in wood (Yoshenko).

A high accumulator of radiocaesium identified after Chernobyl was the fruiting bodies of mushrooms. After Chernobyl, the importance of mushrooms collected in forests and pastures was clearly documented and relationships given between the rate of mushroom consumption and whole body content of humans. Seasonal variation in radiocaesium content of roe deer and wild boar has been attributed to mushroom and truffle consumption by wild animals. In Japan many consumed mushrooms are grown commercially on logs (eg. Shitake) for which permissible limits have been applied. The relative importance of forest collection is less than that in Europe although the pathway is still potentially important for those people who collect wild mushrooms.

Quantification of the impact of site-specific ecophysiological parameters is critical to model radiocaesium behaviour in forests and requires long term monitoring in connection with forest ecosystem growth. Various models of forest ecosystems have been developed and are being gradually improved and validated in Japan (eg. Takada, Loffredo)

#### 5.6. *Radiocaesium transport in catchments and through river systems*

After deposition onto a soil surface, radiocaesium from the FDNPP, as discussed above, is primarily bound to fine soil particles. Subsequently, rainfall and snow melt run-off events will lead to the downstream transfer of radiocaesium, much of which is in forest soils, via river and irrigation systems, through coastal catchments of the Fukushima region, and ultimately to the Pacific Ocean. Sediment-bound radiocaesium may also be deposited in the populated coastal floodplains or in sediments of dams when present (Iijima). Understanding the location, origin and mobility of radiocaesium associated with these sediment sources, is essential to the management of radiocaesium within these coastal catchments.

For both Fukushima and Chernobyl, forest soils are characterized by fast radiocaesium migration in the upper soil layer which is consistent with high bioturbation in the top soil layer (Konoplev).

About 70% of the area affected by the initial deposition from Fukushima is drained by 3 rivers. The radiocaesium  $K_d$  in Fukushima rivers was 1-2 orders of magnitude higher than corresponding values for rivers and surface runoff of Chernobyl (Konoplev) which is consistent with the low dissolved [RCs] in soil solution measured in forest soil (Kobayashi). The data suggest higher binding of radiocaesium in Fukushima soils and sediments (Konoplev).

Radiocaesium activity concentrations in river waters increase after rainfall, as does the suspended sediment load. Most  $^{137}\text{Cs}$  seems to be discharged during intensive rainfall events (Onda, Lepage). After heavy precipitation or storm events, certain fractions of soil minerals can be remobilized, suspended and transported laterally via freshwater systems to differing extents depending on mineral size. Those minerals eventually reach the ocean and become a source of contamination for estuarine and marine organisms. However, the availability of radiocaesium for desorption seems to be limited (Yamasaki).

Fluvisols were identified as the dominant source of sediment and radiocaesium indicative of sediments derived from sources that are highly connected to the river network (Lepage). This is consistent with Onda et al who reported that most suspended sediment in rivers comes via outflow from rice paddies in Japan.

The temporal change in runoff of radiocaesium from the land to surface waters changes with land use. Different temporal trends were evident in the decrease of radiocaesium carried by suspended sediment, with a faster decrease in sediment-associated radiocaesium from paddy fields than from forests.

The normalized dissolved wash-off coefficients for Fukushima river watersheds are 1-2 orders of magnitude lower than corresponding values for the Chernobyl zone (Konoplev). Similarly, models indicate that only a small fraction of deposited radiocaesium is available for uptake into wood or catchment wash-off (Hashimoto, Delmas). Current estimates mostly show estimated losses of radiocaesium from contaminated catchments to be < 1% in 2011 of that deposited (Ueda, Eyrolle-Boyer). Overall, various studies have shown that the total  $^{137}\text{Cs}$  discharge via the catchments has been small compared with total  $^{137}\text{Cs}$  deposition (eg. Wakiyama).

### 5.7. *Radiocaesium in marine ecosystems*

The importance of marine systems after the Chernobyl accident was relatively low although studies were conducted on the input into the Black sea via the Dnieper River, and into the Barents Sea from associated catchments. There is also a large amount of radioecological literature associated with discharges such as the Sellafield reprocessing plant and activities such as dumping of nuclear material at sea and nuclear submarine accidents. The transfer of a wide range of different radionuclides was reported in TRS 422, including CR and  $K_d$  values, in some cases derived using a number of assumptions. In contrast, after the Fukushima accident, a large amount of data has been collected due to the large release of radioactive elements.

After the main discharges from the Fukushima Daiichi accident ceased, the [ $^{137}\text{Cs}$ ] in seawater had decreased by more than 10-fold compared with that before the accident. However, [ $^{137}\text{Cs}$ ] in sediment decreased more slowly than those in seawater and had large fluctuations. Almost all of the surface bottom sediment [ $^{137}\text{Cs}$ ] data exceeded that of the pre-accident 5yr-average (0.87 Bq/kg) in the waters off Fukushima Prefecture. The proximity of the sampling site to the accident site did not necessarily correspond to the higher values which were clearly influenced by currents, resuspension and lateral movement of sediment and other factors (Kusukabe). The sedimentary environment depends on several factors such as the seabed topography and freshwater and terrestrial material input via rivers. In one study, apparent sediment-seawater distribution coefficients  $K_d$  varied from 1,900 to 25,000 L/kg in the same area in 2013-2014 (Aono).

Data have been reported for many species consumed in Japan which had not previously been documented. High transfer to some marine organisms has been reported compared with that given in TRS 422. CR and  $K_d$  data being collected will test the validity of the CR concept and allow other approaches to be developed where needed. The data will enhance current databases such as MARiS and allow the assumptions used in TRS 422 to be tested.

The highest [RCs] have been recorded for demersal fish in the port area. The estimated apparent CR of radiocaesium (CR-Cs) in marine organisms in one study ranged from 26 in muscle of squid to 10,000 in clam viscera – the latter due to inclusion of gut material. Generally large differences in CR-Cs in different parts of marine organisms were not observed. For CR-Cs values, the influence of sediment and particles on marine biota is evident, as these organisms often contain particulate matter and sediment in the coastal area (Aono).

There are some data showing that temporal variation in radiocaesium transfer is not consistent between trophic levels (Johansen). [RCs] in marine products has decreased significantly since 2011, but high levels remain in some species in the port area. Decreases of [ $^{137}\text{Cs}$ ] and [ $^{110\text{m}}\text{Ag}$ ] in marine biota will depend on differences in the mechanisms of accumulation and metabolism.

Fish biomass has increased since the accident, probably due to the lack of commercial fishing. Long term, comprehensive monitoring of marine products (so far nearly 24,000 samples from 177 species), especially close to Fukushima Daiichi, has guided the lifting of restrictions and the expansion of trial fishing whilst maintaining public confidence. Fishery facilities are gradually being restored and areas for trial fishing operations expanded. The number of selected species for potential resumption of fishing was 64 species as of July 2015 (Wada).

Significant improvements in marine models are being made for both the coastal and Pacific area using biokinetic, sediment exchange, animal migration and trophic transfer data.

#### 5.8. **Radiation transfer to animals and wildlife doses**

After Chernobyl, there was high, long-term transfer of radiocaesium to many wild animals including wild boar, roe deer, reindeer and grouse which was largely explained by soil types and dietary preferences. The value of long term data for wild game, such as that from Norway (Gjelsvik) has been emphasized. After Fukushima, persistently high radiocaesium transfer to wild boar has also been observed (> 1000 Bq/kg in wild boar in the more highly contaminated areas) but is lower in other species such as black bear and sika deer (Mizoguchi). However, a long term prognosis cannot yet be made as more data is needed at specific locations to better

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understand the relevant pathways. All wild boars in Fukushima are subject to regulation because their radiocaesium values exceed the food regulatory values, leading to problems of population control and damage to farmland.

Ongoing studies are also considering the external and internal doses to game animals in forests in both Chernobyl and Fukushima (Hinton) using attached dosimeters and tracking systems. This will enable improved quantification of the impact of the utilisation of the contaminated habitat by these species.

In the marine systems, the highest doses to fish are to those within the port of the Fukushima DNPP. Doses to these fish are remaining elevated with little decrease over time. Doses to other marine fish are declining with time (Johansen).

Currently, there is no consensus on ecological effects of radioactivity released from Chernobyl and more data is needed with high quality, detailed dosimetry estimation and adequately replicated studies for a variety of species. Direct comparison between the two accidents is difficult as it is possible that adaptation of organisms to the radioactive contamination may have occurred over the ~ 30 years since the Chernobyl accident whereas after the Fukushima accident this is unlikely within less than 5 years. For example, there may be a greater likelihood of transgenerational effects (e.g. mutations) near the Chernobyl NPP than near Fukushima Daiichi NPP.

More precise knowledge for the time period of the releases and thereafter has been possible after the Fukushima accident, in part due to a large amount of data being available on aspects such as bird population monitoring. Recent work has shown that prior estimates of dose to organisms at Chernobyl using ambient air dose rates at one metre above the ground surface may underestimate external doses by one or more orders of magnitude (Adam).

### 5.9. *Remediation*

Remediation after the Chernobyl accident included decontamination of residential areas but focused over the short- and longer-term on reducing internal doses largely from animal products (milk and meat). The most appropriate and effective measures used were directed at production of crops, fodder and pasture grass and on clean feeding to decontaminate meat-producing animals. A wide range of soil amelioration measures were applied including extensive renovation via ploughing, reseeding, additional K and other applications. After Fukushima, most remediation has been focused on reducing external dose by decontaminating surfaces. Internal dose can arise from crops and forest products but protection of the public has been ensured via comprehensive restrictions, monitoring and the application of remediation measures.

An important component of agricultural remediation has been the widespread supply of additional potassium which was effective in preventing Cs uptake from soil to crops, especially in Japanese paddy fields. Cost-effective remediation with K application is clearly important for long-term management in arable fields. For example, recommendations for perennial grassland with relatively high [RCs] and OM are that the surface soil should be mixed with underlying soil and the exchangeable K<sub>2</sub>O content should be 30-40 mg/100g in the 0-15 cm soil depth (Harada). Equivalent values for soybean (Sekiguchi) and rice are both 25 mg/100g. However, K fertilization above these amounts, which has occurred in some instances, will have limited, if any, additional benefit. It will be important that appropriate guidance on application

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rates is adhered to by the agricultural community. For forage corn, application of contaminated cattle manure has been shown to be safe and effective in reducing radiocaesium uptake. The availability of Cs in soil for plant uptake decreases with time, so application of K fertilizers can be reduced according to the conditions.

Phytoremediation has been considered for some plants which are known to accumulate Cs to a relatively high degree. However, phytoextraction has been shown to be ineffective and not appropriate as a remedial measure (Shinano). Also the difference in radiocaesium accumulation by cultivars of the same crop has been considered to enable selection of the lowest accumulator of radiocaesium. The variation in uptake into soybean grain was a factor of 4.5, but the ranking of the different varieties was different between 2012 and 2013 (Kojima).

Remediation of fruit trees has been a particular problem as fruit is a major export crop for Fukushima prefecture. Where problems of exceeding the permissible limit remain, replanting of trees may be one solution considered since older mulberry trees have a lower TF than newly planted trees. K fertilisation also reduces radiocaesium transfer (Kowata). However, this is disruptive for the farmer and will result in a gap in production. Information on the effective half-lives of radiocaesium in fruit trees will help to inform any decisions. For instance, observed  $T_{\text{eff}}$  of radiocaesium ranged from 183-473 d in persimmon and loquat trees, which were similar to those observed after the Chernobyl accident (Tagami). Aboveground, directly deposited radiocaesium was probably the main source of contamination in the first few years. However, the slowly decreasing rates suggested that root uptake may become the major transfer pathway to fruits in the near future.

Extensive decontamination operations have removed some of the radiocaesium contamination in soil, vegetation, road surfaces etc. Contaminated materials are being transferred to temporary storage facilities, originally intended to be used for three years. According to the roadmap of decontamination of the Japanese government, the waste is intended to be stored in interim storage facilities for 30 years. Much of the ICSA has been remediated and the costs of removal account for most of the associated decontamination waste-related costs (Yasutaka). Meanwhile, the waste generation and temporary storage is now focused largely in the SDA where the costs of storage of decontamination waste is relatively high (Yasutaka). The volume of waste could exceed the capacity for interim and long term disposal. Therefore, practical and effective methods for volume reductions need to be developed prior to interim storage.

# Annexes

# Annex 1: Agenda for ICOBTE 2015

## ICOBTE 2015 Programme



ICOBTE\_2015\_Program.pdf

## 13<sup>th</sup> ICOBTE Oral Program













Program\_oral.pdf




## 13<sup>th</sup> ICOBTE Poster Program



Program\_poster.pdf

## Annex 2: Special Radiocaesium Symposia Pdf Documents

Symposium	Title	Synopsis Document
1	Fate and transport of metals in contaminated sediments – new approaches in remediation	 special_symposia_1_0917.pdf
2	Principles and applications of trace element transport in soils and geological media	 special_symposia_2_1110.pdf
3	Physiological and molecular mechanisms of toxic trace element accumulation in plants	 special_symposia_3_0917.pdf
4	Trace element biogeochemistry at the soil-plant and soli-plant-microbial interfaces	 special_symposia_4_0917.pdf
5	Understanding and mitigating the environmental behaviour of radiocaesium after the Fukushima accident	 special_symposia_5_0917.pdf
6	Biogeochemistry of trace elements in coal combustion waste	 special_symposia_6_0917.pdf
7	Behaviour of NORM and artificial radionuclides in the environment	 special_symposia_7_0917.pdf
8	Recent progress in cadmium studies in Japan: in commemoration of 50 years after the identification of cadmium as the cause of Itai-itai disease	 special_symposia_8_0917.pdf
9	Gentle remediation of trace element-contaminated or enriched soils: optimising fundamental processes to obtain beneficial ecosystem services and renewable biomass	 special_symposia_9_0917.pdf
10	Remediation of heavy metals-contaminated soils: Novel practical approach based on state of the art science	 special_symposia_10_0917.pdf

11	Biochar as a sorbent for contaminant management in soil and water	 special_symposia_1 1_0917.pdf
12	Application of synchrotron radiation (SR) –based methods to biogeochemistry and environmental; geochemistry of trace elements	 special_symposia_1 2_0917.pdf
13	Environmental implications of manufactured nanoparticles	 special_symposia_1 3_0917.pdf

## Annex 3. List of participants in Session 5 of ICOBTE

Annex 3. Countries and organizations represented within Session 5.

Country	Organisations Represented
Australia	ANSTO
Belgium	SCK•CEN
France	IRSN Laboratoire des Sciences du Climat et de l'Environnement (LSCE) ANDRA
Germany	German Federal Office for Radiation Protection (BfS)
Japan	Fukushima University Kyoto University Tohoku University Hiroshima University Tsukuba University Institute for Environmental Sciences NARO Tohoku Agricultural Research Center Hachinohe Institute of technology Forestry and Forest Products Research Institute Japan Atomic Energy Agency National Institute of Radiological Sciences Tokyo University of Agriculture and Technology Kyushu University Niigata University Japan Agency for Marine-Earth Science and Technology Hokkaido University Kanazawa University Saga University Akita Prefectural University National Institute for Agro-Environmental Sciences Iwate University Kyoto Prefectural University
Norway	Norwegian Radiation Protection Authority
Russian Federation	RPA "Typhoon"
Sweden	Stockholm University
UK	Centre for Ecology and Hydrology University Of Nottingham
Ukraine	Institute of Mathematical Machine and System Problems Institute of Biophysics Siberian Branch of RAS

## Annex 4. G. Shaw plenary abstract

# Radiocaesium behaviour and impact – what have we learned from atmospheric weapons fallout, Chernobyl and Fukushima?

**G. Shaw**

*University of Nottingham, Sutton Bonington, Leicestershire, United Kingdom*

**ABSTRACT:** The accident at Fukushima Daiichi in 2011 has re-ignited international interest in the environmental fate and impact of radiocaesium as a result of the widespread dispersal of both  $^{134}\text{Cs}$  and  $^{137}\text{Cs}$  from the facility. The Fukushima source term consisted of approximately equal activities of  $^{134}\text{Cs}$  (half-life = 2.06 years) and  $^{137}\text{Cs}$  (half-life = 30.1 years); measurements of contaminated soils shortly after the accident indicated a  $^{134}\text{Cs}:^{137}\text{Cs}$  ratio of 0.9 – 1.0. In the short-term (1 – 5 years) both isotopes pose a radiological hazard in the environment. However, due to its longer radioactive half-life,  $^{137}\text{Cs}$  is a much more persistent environmental contaminant.

Cs-137 was first introduced into the environment long before the Fukushima accident. According to UNSCEAR, nuclear weapons testing from 1945 to 1980 injected  $950 \times 10^{15}$  Bq (Becquerels) of  $^{137}\text{Cs}$  into the global atmosphere. This source term is more than  $11\times$  greater than the release of  $^{137}\text{Cs}$  from Chernobyl (in 1986) and  $80\times$  greater than the best estimate of  $^{137}\text{Cs}$  released from Fukushima. The Chernobyl and Fukushima power plants were both point sources. In contrast, weapons-derived releases of  $^{137}\text{Cs}$  were more widely dispersed both geographically and temporally, though by far the largest peak in releases from weapons tests was in 1962 when devices with an explosive yield equivalent to 72 M tonnes of TNT were detonated. Much of this  $^{137}\text{Cs}$ , along with numerous other radionuclides, was injected into the troposphere and stratosphere where mean residence times can be greater than 4 years. As a consequence, radioactive particles from weapons were able to mix and circulate widely, before eventually depositing on soil and vegetation surfaces around the world. Peak deposition occurred in 1963 leading to a discrete radioactive ‘spike’ in soils, sediments and peats which can still be measured today. In fact, this radioactive marker has recently been proposed as a means of distinguishing the beginning of the most recent geological epoch, the Anthropocene.

As well as providing direct evidence of the global (inter-hemispheric) circulation of the products of nuclear weapons, measurements of the worldwide occurrence of  $^{137}\text{Cs}$  in crops and associated foodstuffs focussed attention on the physical and biogeochemical behaviour of radiocaesium in the environment. Transfer from atmosphere to soil and vegetation, migration and physico-chemical fixation in soils, and uptake by crops and non-food plants are all processes which dictate the degree of exposure of humans and other organisms to  $^{137}\text{Cs}$  in the short- and long-term. Surveys of weapons fallout in Europe prior to the Chernobyl accident showed quite clearly that rainfall was an enhancing factor which increased deposition of  $^{137}\text{Cs}$  from atmosphere to the soil. These observations, based on correlations between deposited activity and spatial rainfall patterns, were confirmed by measurements made in the UK during the deposition of the Chernobyl plume in which rainfall washout coefficients of ‘several thousand’ were obtained. The nature of the ground surface, and particularly vegetation type, was also observed to play a role in controlling atmospheric deposition of  $^{137}\text{Cs}$ . Comparisons between grassland, farmland and forest soils showed that accumulation of weapons fallout  $^{137}\text{Cs}$  under tree canopies could be 20 – 40% higher than on grassland and that long-term dry deposition rates were up to  $9\times$  higher for forests than for grassland. This has important implications for the relative vulnerabilities of agricultural and semi-natural ecosystems to radiocaesium deposition.

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Across the major part of Europe affected by the Chernobyl accident,  $^{134}\text{Cs}$  and  $^{137}\text{Cs}$  deposited to soil and vegetation surfaces in the form of fine particles, with local rainfall events providing the driving force for 'patchy' localized deposition. According to the IAEA,  $^{134}\text{Cs}$  and  $^{137}\text{Cs}$  were released from the Chernobyl reactor in the ratio of 0.50 – 0.52 and this isotopic signature was used to distinguish newly-deposited radiocaesium from Chernobyl from the  $^{137}\text{Cs}$  deposited from global nuclear weapons fallout more than 20 years previously. Measurements of depth distributions of weapons-derived  $^{137}\text{Cs}$  before and after the Chernobyl accident indicated that it had limited vertical mobility in soils and was largely restricted to the upper 10 cm of the soil profile. Based on measurements made in southern Germany in 1990, estimates of mean residence times for vertical migration ranged from 2 – 4.5 years  $\text{cm}^{-1}$ , with Chernobyl-derived  $^{137}\text{Cs}$  migrating 2 – 4 $\times$  faster than 'aged' weapons-derived  $^{137}\text{Cs}$ . These rate estimates were based on different averaging periods for the two sources of  $^{137}\text{Cs}$ , which underemphasized the role of time-dependent fixation by clay minerals for the more recent (1986) source. Illite, in particular, efficiently sequesters caesium isotopes in frayed edge and interlayer sites. The frayed edge sites of illite have a very high degree of selectivity for the  $\text{Cs}^+$  ion, much higher (10 – 1000 $\times$ ) than for the homologous ions  $\text{NH}_4^+$  and  $\text{K}^+$ . Nevertheless, because  $\text{NH}_4^+$  and  $\text{K}^+$  are much more abundant in soils than caesium, they both play an important role in controlling the mobility and bioavailability of caesium isotopes.

The concept of the Radiocaesium Interception Potential (RIP) was devised in the late 1980s to quantify the cation exchange capacity of a soil which is specifically able to interact with the  $\text{Cs}^+$  ion. Post-Chernobyl research on soils in semi-natural ecosystems, which are often highly organic and deficient in illite clays, showed that the 'immobilization capacity' of a soil is strongly controlled by the RIP (effectively, the illite content of the soil) as well as the potassium and ammonium status. Ecosystems which are more vulnerable to contamination with radiocaesium are those which have soils with low RIP, low potassium status, relatively high ammonium ion concentrations (perhaps due to waterlogging) and low pH. Many plant species growing in such soils have adapted to acquire and recycle nutrients such as potassium with very high efficiency. As a homologue of  $\text{K}^+$ ,  $\text{Cs}^+$  is also efficiently acquired and retained by such species if it is not 'fixed' within the soil. Measurements after Chernobyl showed that fixation rates were initially controlled by diffusion of  $^{137}\text{Cs}$  into the interlayer sites of illite though, over the longer-term (5 – 15 years), the reversibility of this process led to a steady state in which radioactive decay became the rate-limiting process that controlled the ecological half-time of  $^{137}\text{Cs}$ . A low degree of 'fixation' of  $^{137}\text{Cs}$  in organic mountain soils led to the long-lived contamination of livestock, such as sheep, which provide the basis for agricultural economies in upland areas of Europe. As an example of the long-lasting impact of  $^{137}\text{Cs}$  deposition from Chernobyl, government restrictions on the sale and movement of sheep from some UK farms contaminated in 1986 were only lifted in 2012, 26 years after the accident.

Next year (2016) will mark the 30th anniversary of the Chernobyl accident and the question has been raised whether radioecological knowledge and experience gained in this time can be applied to forecast and mitigate the impacts of radiocaesium releases from Fukushima. At the time of the Chernobyl accident a similar question was asked about the relevance and applicability of measurements made after global fallout from atmospheric nuclear weapon tests. The relatively high human exposures to  $^{137}\text{Cs}$  in semi-natural ecosystems came as an unwelcome surprise after Chernobyl. With this in mind, it should be remembered that approximately 70% of the Fukushima prefecture (~9650  $\text{km}^2$ ) is covered by forest, all of which is contaminated, to a greater or lesser degree, with  $^{137}\text{Cs}$ . Soil types and mineralogy in Japan differ substantially from those in the parts of Europe most affected by Chernobyl fallout, so understanding the long-term fate of weapons-derived  $^{137}\text{Cs}$  in Japanese soils (which is still measurable) may give more reliable clues on which to base forecasts of radiocaesium deposited in 2011. In addition, valuable information may be gained from analysing the very long-lived  $^{135}\text{Cs}$  (half-life =  $2.3 \times 10^6$  years) and stable  $^{133}\text{Cs}$  isotopes in semi-natural and agricultural ecosystems in the Fukushima-affected areas.



## Annex 5. Participants of the Fukushima excursion

Staff		Nationality
Hirofumi Tsukada (塚田 祥文)		Japanese
Izumi Mizushima (水嶋いづみ)		Japanese
Yoshifumi Wakiyama (脇山義史)		Japanese
Teruo Watanabe (渡辺光男)		Japanese
ICOBTE and Fukushima COMET Workshop 参加者	14	
Brenda Howard		British
Christelle Adam Guillermin		French
Frédéric Coppin		French
Marc André Gonze		French
Rodolphe Gilbin		French
Pierre Hurtevent		French
Shinichiro Uematsu (植松 慎一郎)		Japanese
Åste Søvik		Norwegian
Sindre Nelson		Norwegian
Mathew Johansen		American
George Shaw		British
Yves Thiry		Belgian
Shinano Takuro (信濃 卓郎)		Japanese
Akira Takeda (武田 晃)		Japanese
ICOBTE	11	
Ali Kanso		Lebanese
Emile Izry		French
Bernhard Markert		German
Simone Wünschmann		German
Peter Bossew		Austrian
Ying Ji		P.R. China
Frédéric Rees		French
Junpei Takano (高野 順平)		Japanese
Takehiko Tsuruta (鶴田 猛彦)		Japanese
Katsuhiro Kojima (小島 克洋)		Japanese
Masanori Saito (齋藤 雅典)		Japanese