

Contamination of Japanese foodstuffs of terrestrial origin after the Fukushima nuclear accident and related dose assessments Part 1: foodstuff contamination

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ABSTRACT During and after the Fukushima accident, the IRSN collected and interpreted the results of radiological measurements performed on foodstuffs of terrestrial origin published by Japan's Ministry of Health between mid-March 2011 and July 2012. Analysis of the findings shows that the accident's date, livestock-rearing practices and the deposits' characteristics had a decisive influence. The fact that radioactive fallout occurred very early in the growing and breeding season largely explains the moderate contamination of most foodstuffs of terrestrial origin, notably in the areas with the largest deposits. In the case of dairy products and meat, feeding imported fodder to livestock in stables, a common practice in Japan, compounded the calendar effect. Measurements published in Japan have also borne out the particular sensitivity of mushrooms, including cultivated species, and game.

Keywords: Fukushima NPP accident / foodstuff contamination

Introduction

Since mid-March 2011, the MHLW (the Japanese Ministry of Health, Labour and Welfare) has published data on ^{131}I , ^{134}Cs and ^{137}Cs activity measured in approximately 330,000 samples of farmed and wild foodstuffs on its website (www.mhlw.go.jp/english).

This article comments on these findings, on the contamination levels reached in the various categories of foodstuffs, and on their variability and changes over time. Among the items explaining the situation observed in Japan, the decisive influence of the accident's date, animal husbandry practices and the deposits' characteristics, notably their heterogeneity, are particularly stressed.

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1. Period from 15 March to 15 May 2011: the acute leaf vegetable and cow's milk contamination phase

Most of the radioactive deposits after the Fukushima accident occurred between 12 and 23 March, on the cusp of winter and spring (Champion *et al.*, 2013); the heaviest deposits were northwest of the stricken site and mainly formed by rain or snowfall when the radioactive discharges were dispersed. Figure 1 shows the spatial distribution of ¹³⁷Cs deposits northwest of the plant; these deposits were generally very heterogeneous, even within a single town.

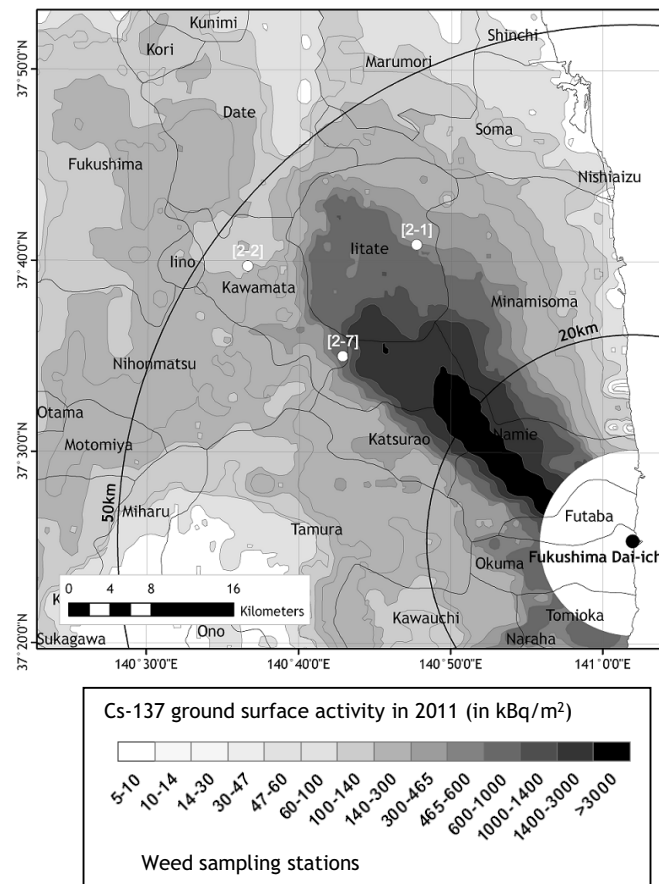


Figure 1 – Map of ¹³⁷Cs deposits measured by helicopter (according to MEXT, 2011a, 2011b). Amounts are expressed in kBq/m² at 31 March 2011. The sampling stations of adventitious leaves and calculation points chosen for the simulations performed with the IRSN's pX and SYMBIOSE software are also shown.

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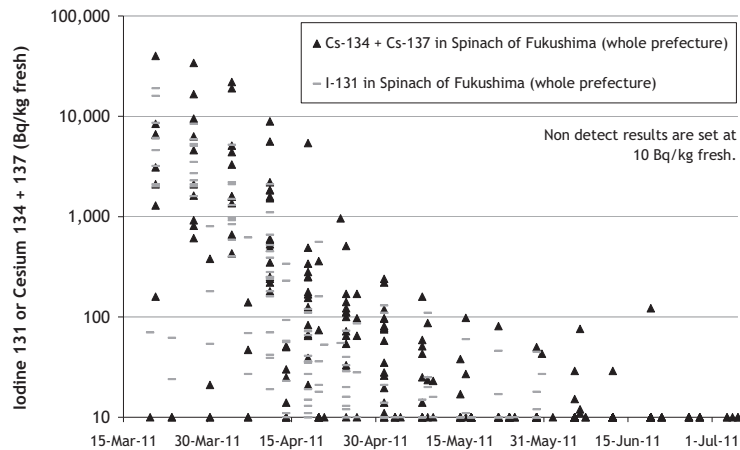


Figure 2 – Change in ^{131}I and $^{134}\text{Cs} + ^{137}\text{Cs}$ contamination of spinach in Fukushima prefecture (MHLW data).

Cultivated plant foodstuffs

The only agricultural foodstuffs harvested in this period were of market garden origin, including leafy vegetables (spinach, lettuce, broccoli, kukitachina, komatsuna, etc.), which intercepted radioactive deposits and, consequently, were the foodstuffs most sensitive to atmospheric fallout.

As expected, the highest activity was observed in samples of leafy vegetables collected from the Fukushima, Ibaraki, Chiba, Tochigi and Gunma prefectures in the first days after the radioactive deposits formed. The highest amounts the MHLW measured and published were 54,100 Bq/kg of fresh weight of ^{131}I in a spinach sample taken on 18 March in Hitashi, 120 kilometres south of the plant, and 82,000 Bq/kg of fresh weight of $^{134}\text{Cs} + ^{137}\text{Cs}$ in a kukitachina sample taken on 21 March in Motomiya, 70 kilometres west of the plant.

Figure 2 shows the changes in ^{131}I and $^{134}\text{Cs} + ^{137}\text{Cs}$ activity in spinach produced in Fukushima prefecture, which fell very quickly in the weeks after the radioactive deposits due to radioactive decay (for ^{131}I), biological dilution caused by the plants' growth and, in lesser degree, leaf leaching by rainwater. The effective period of activity decay in this kind of foodstuff from Fukushima prefecture ranges from 8 to 10 days for caesium and 4 to 5 days for ^{131}I (Gonze *et al.*, 2012). These values are the same as those found in France in leafy vegetables harvested in May 1986 after the Chernobyl accident (Renaud *et al.*,

2007). Thus, in less than two months the contamination of leafy vegetables fell 100- to 1,000-fold, reaching as early as the end of April levels below the consumption threshold for caesium from 500 Bq/kg in effect at the time. The detection limits of the measurement methods used in Japan to monitor foodstuffs, when specified, were approximately 10 Bq/kg of fresh weight. Since June, ^{131}I has not been detected in leafy vegetables from Fukushima prefecture; significant amounts of ^{134}Cs and ^{137}Cs have been measured very occasionally.

The high variability of amounts measured in Fukushima prefecture owes partly to varying surface deposit activity, deposit conditions (dry or wet) and agricultural practices (fields or greenhouses). In order to limit the effect of spatial variability in the prefecture, the leafy vegetable contamination study focused on two of the towns most exposed to radioactive fallout (outside the 20-km perimeter): Kawamata-Machi and Iitate-Mura. Figures 3a and 3b show ^{131}I and ^{137}Cs activities measured in leaf vegetables (spinach, broccoli, cabbage, kukitachina, komatsuna, lettuce, parsley and shinobufuyuna) from both towns. Data on ^{134}Cs do not appear but are very similar to the figures for ^{137}Cs . In these charts, the iodine and caesium activities observed in these vegetables are compared with the theoretical ones as estimated by SYMBIOSE simulation software (Gonze *et al.*, 2012). The estimates are based on hypotheses regarding the atmospheric fallout's characteristics and precipitation in Kawamata-Machi and Iitate-Mura, taking into account data collected and simulations performed by the IRSN. By performing a geostatistical analysis of the cartographic data resulting from the first soil sampling campaign carried out by teams of Japanese scientists coordinated by MEXT¹, it is possible to obtain the statistical distribution of the deposits measured on the scale of this area. The 10th percentile (the value below which 10% of the distribution is located) and the 90th percentile (the value above which 10% of the distribution is located) thus obtained range from 2,000 to 7,000 kBq/m² for ^{131}I and 110 to 1,100 kBq/m² for ^{137}Cs , respectively. In other words, 80% of these towns' area appears to have received ^{131}I deposits, with activity ranging between 2,000 and 7,000 kBq/m², and ^{137}Cs deposits, with activity ranging between 110 and 1,100 kBq/m². Simulations of the atmospheric dispersion of discharges from the accident and related radioactive fallout, based on the C³X platform's pX model (Champion *et al.*, 2013), have made it possible to estimate that dry deposits accounted for 20% of ^{131}I deposits and 10% of ^{137}Cs deposits in both towns. Estimated precipitation (rain and snow) during the main deposit episode (15 March 2011) has been put at seven millimetres a day, based on radar observation data. SYMBIOSE's default values were retained for the other modelling parameters.

¹ http://www.mext.go.jp/b_menu/shingi/chousa/gijyutu/017/shiryo/_icsFiles/afieldfile/2011/09/02/1310688_1.pdf
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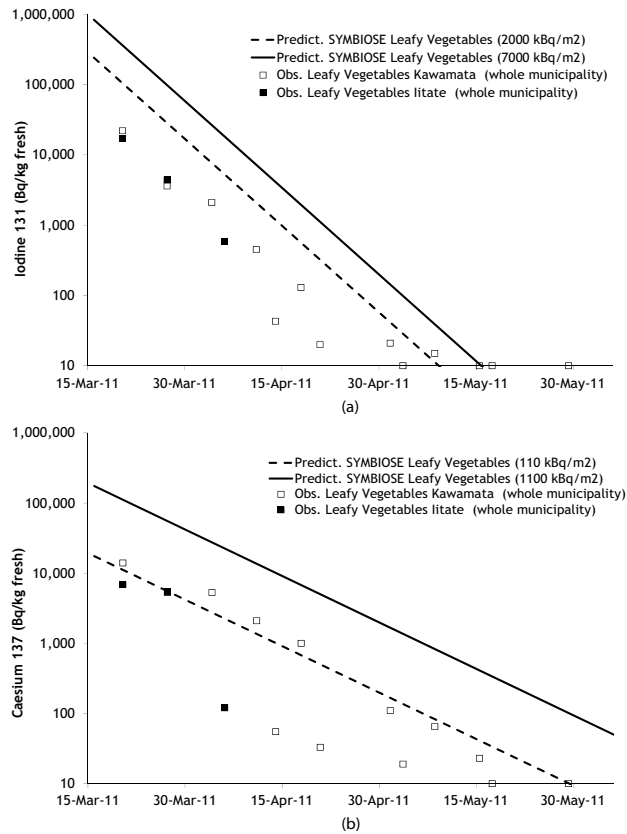


Figure 3 – Change in ^{131}I and ^{137}Cs contamination of leafy vegetables (lettuce, spinach, broccoli, cabbage, kukitachina, komatsuna, parsley, shinobufuyuna) sampled in Kawamata-Machi and Iitate-Mura. The simulations for leafy vegetables were made with SYMBIOSE (10th percentile = 2,000 kBq/m² of total ^{131}I deposits and 110 kBq/m² of ^{137}Cs deposits; 90th percentile = 7,000 kBq/m² of total ^{131}I deposits and 1,100 kBq/m² of total ^{137}Cs deposits). The hypotheses chosen for all the simulations are: 20% of dry deposit for ^{131}I and 10% of dry deposit for ^{137}Cs ; deposit on 15 March with 7 mm of precipitation per day; default values for all the other parameters used by SYMBIOSE.

The results this model obtained accurately reconstruct the kinetics of the decrease in iodine and caesium activity in broadleaf vegetation, with an effective decrease period (radioactive decay, biological dilution and leaf leaching) of approximately four days for ^{131}I and seven days for ^{137}Cs .

The highest activity observed for ^{137}Cs is fairly close to the level evaluated based on the 10th percentile of the deposit amounts observed in both towns

(110 kBq/m²). However, in the case of ¹³¹I, the simulated activity was much higher than the level observed, even taking the smallest deposits into consideration. Several factors may explain this. First of all, uncertainties linked to parameter values must be associated with evaluations given by foliar transfer modelling:

- an overestimation of leafy-vegetable contamination can be the result of an underestimation of wet deposits' contribution to the total deposits, notably in most contaminated areas;
- the amount of rainfall considered is a daily one; if rainfall events are more intensive in a shorter period, the foliar captation would be lower;
- only a single value of the foliar biomass is fixed in SYMBIOSE despite the variability between the different kinds of vegetables; spinach *vs.* cabbage, for example.

Finally, the proportion of different chemical forms of iodine in the air (particulate iodine, iodine gas), which has an influence on the rate of dry deposition and foliar transfer factor, may explain such differences between the model / measurements of the most significant iodine.

Moreover, the SYMBIOSE-based evaluations do not take account of the protective role greenhouses play in shielding plants from precipitation-borne (snow or rain) deposits².

Figures 4a and 4b show ¹³¹I and ¹³⁷Cs activities in the leaves of weeds (or adventitious flora³) measured in Kawamata-Machi and Iitate-Mura. Unlike the leafy vegetable samples, whose provenance in the town is unknown, the adventitious plant samples were taken from three precisely located stations (see Fig. 1). Like the deposits measured in these stations, adventitious flora from the centre of Kawamata-Machi (station [2-2]) was 10 times less contaminated than that from the southern part of the town (station [2-7]) and from Iitate-Mura (station [2-1]). Activity measured in the leaves of adventitious flora from a given station nevertheless shows a rather wide range of variability, particularly for caesium, which probably stems from the diversity of the species sampled. The average iodine and caesium activity measured in adventitious flora is higher than that of the vegetable leaves. A smaller amount of deposits taken up by leafy vegetables than by adventitious plants (a smaller leaf area) may partly account for the difference, which may also be explained by the above-mentioned reasons involving differences between contamination measured in leafy vegetables and that evaluated by SYMBIOSE; in

² The words "in a greenhouse" or "outside" sometimes accompany the measurements the MHLW published but not systematically enough to take account of it in the model.

³ Non-cultivated herbaceous or woody plants commonly called weeds. Adventitious flora can be perennials that reproduce vegetatively or stay in the same place for several years (quackgrass, sow thistle, wild buckwheat, etc.) or, more usually, annuals which reproduce by seeds, often with a high reproduction potential (wild oat, speedwell, chickweed, etc.).

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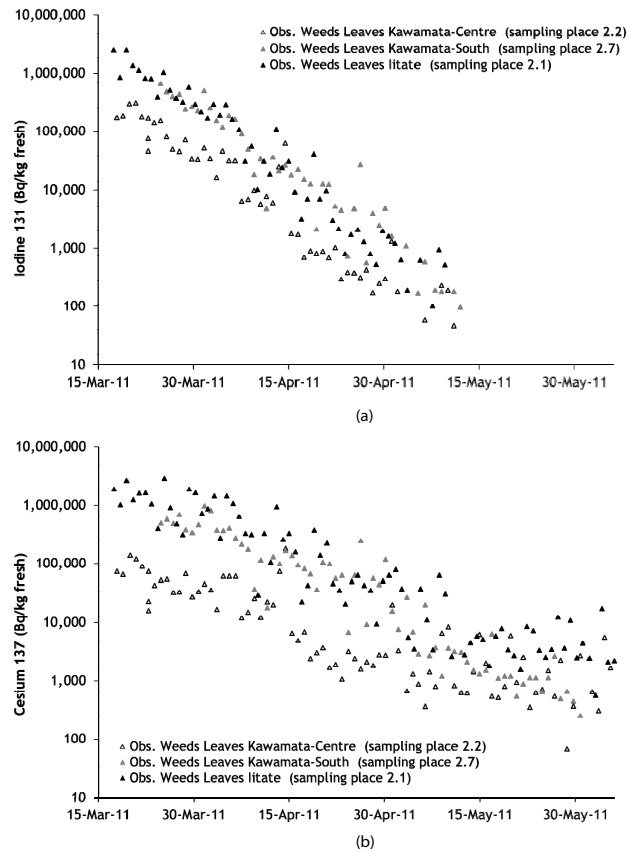


Figure 4 – Change in ^{131}I and ^{137}Cs contamination of weed leaves (adventitious plants) sampled in Kawamata-Machi and Iitate-Mura.

other words, the location of market gardens in areas less contaminated than the places where adventitious plants grow or greenhouses are used.

The parallels between changes in iodine and caesium contamination observed and predicted in leafy vegetables and those observed in adventitious plants mean that the effective contamination decrease period seems to be the same for both categories of plants.

As expected, fruit vegetables such as tomatoes or melons and root vegetables such as turnips and wasabi were much less contaminated than leaf vegetables. Only a fraction of the activity retained by the leaves was transferred to the

part consumed. The highest amounts were found in a turnip sample taken on 3 April 2011 (Fukushima prefecture): 1,000 Bq of ^{131}I /kg of fresh weight and 4,100 Bq of $^{134}\text{Cs} + ^{137}\text{Cs}$ /kg of fresh weight. The highest readings for tomatoes were in March 2011: approximately 20 Bq of I/kg of fresh weight and around 100 Bq of caesium/kg of fresh weight. After the first two months, only a few samples of wasabi (the root part) taken in Fukushima prefecture still contained amounts of caesium exceeding consumption limits.

Cow's milk

Environmental contamination also had an impact on cow's milk in the process of being produced when the accident occurred: the animals very quickly incorporated radionuclides, which entered the mammary glands. However, the contamination seems to have been moderate compared with the deposits and plant contamination in the most severely affected areas. The highest activity measured ranged between 10 and 100 Bq/L, peaking at 210 Bq/L for each of the caesium isotopes (Iitate-Mura, 19 March 2011) and 100 to 1,000 Bq/L, with a maximum of 5,300 Bq/L for ^{131}I (Kawamata-Machi on 20 March). Between March 2011 and early February 2012 iodine or caesium activity exceeded consumption limits in just 23 of approximately 2,000 milk and dairy product samples.

The fact that the accident occurred in late winter probably limited contamination of cows, which at that time of year consume local fodder harvested the previous season or imported fodder (importation of animal feed seems to be common in Japan). Figure 5 shows that if the cows had consumed plants as contaminated as the adventitious leaves sampled in Iitate-Mura, milk contamination could have reached 100,000 Bq/L in the first days after 15-16 March, when the deposits occurred linked to rain.

Activities measured in milk are therefore not compatible with a high intake and regular local fodder directly impacted by fallout. Rather, they reflect contamination of animals due to the ambient contamination in these territories: incorporation of surface contamination, ingestion of fodder or drinking water exposed to a contaminated environment.

Figure 5 also shows that milk activities observed in the same town varied greatly (see the batch of ^{131}I readings in Kawamata-Machi), which may stem from the variability of fodder contamination, itself due more to its nature and origin than to that of the radioactive deposits in the town (see also the section below on meat).

Iodine activity measured in milk plummeted, with an effective period of approximately 4.5 to five days, the same as that observed in France in May 1986 after

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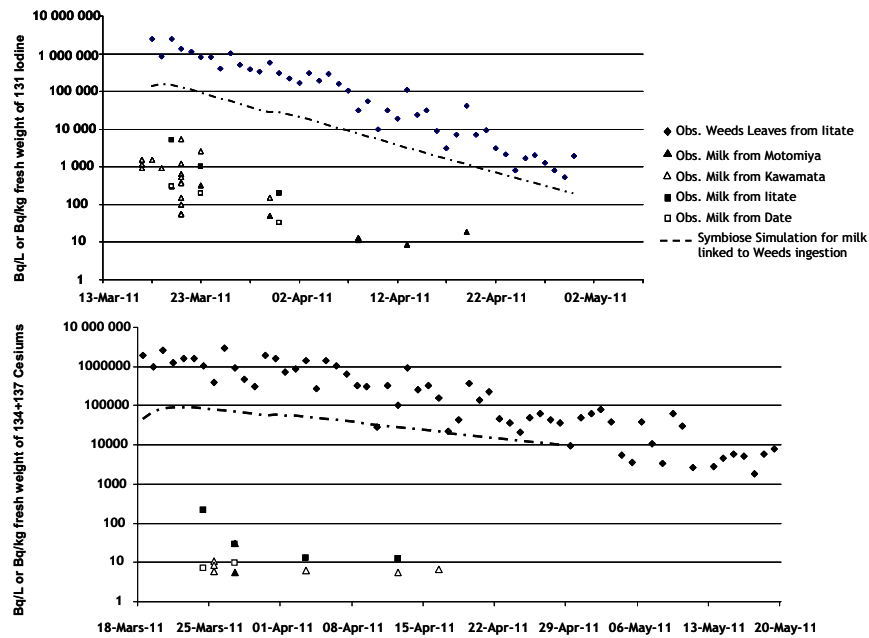


Figure 5 – ^{131}I and ^{137}Cs activity by volume in milk samples with the highest contamination levels from the Iitate-Kawamata-Date region and from Mitomiya. ^{131}I activity measured in weed samples taken in Iitate-Mura and modelling of ^{131}I activity in milk produced by the cows that would have consumed them, using SYMBIOSE software.

the Chernobyl accident. By mid-April 2011 the milk samples' ^{131}I activity had fallen below detection limits, which are often between 10 and 50 Bq/L. By late May 2011 radioactive decay of ^{131}I brought the radionuclide's activity in milk to below one Bq/L. The theoretical decrease in caesium contamination is due to the drop in fodder contamination and the metabolic elimination in the animal of the absorbed caesium. After some months, lower detection limits allow one to show the decrease in caesium activities which only exceptionally exceed 5 Bq/L from April 2012.

^{131}I and $^{134}\text{Cs} + ^{137}\text{Cs}$ activity levels measured in milk samples from Ibaraki, south of the stricken power plant, were similar to those presented above. This area also had significant radioactive deposits, with the proportion in dry form higher than in the northwest of Fukushima (Champion *et al.*, 2013), which may have brought about greater contamination of vegetation.

2. After 15 May 2011: persistent, often low-level contamination of some foodstuffs

Fruits

Most fruit trees were still leafless in mid-March 2011, considerably limiting their contamination by radioactive deposits, which explains why the activity of fruits harvested later in the year (apples, pears, peaches, plums, etc.) – several tens of Bq/kg of caesium – did not exceed consumption limits, in contrast with some early-blossoming fruit trees: Japanese apricots, yususu, kiwis and figs. A similar observation was made in France after the 1986 Chernobyl accident, when radioactive caesium activity was higher in early-growing “griotte” cherries produced in the country’s eastern part than in other fruit harvested the same year.

After the Fukushima accident, the first fruits showing significant activity were Japanese apricots (ume), which were probably already blossoming in mid-March. The highest activity measured ranged from 137 to 700 Bq/kg of fresh weight for $^{134}\text{Cs} + ^{137}\text{Cs}$ and came from areas with the biggest deposits. The most contaminated fruits were the yusu⁴ (up to 2,400 Bq/kg of fresh weight in Minamisoma-Shi on 26 August and 860 Bq/kg of fresh weight on 11 October), kiwis harvested in autumn (the peak activity measured was 1,100 Bq/kg of fresh weight in Minamisoma-Shi on 14 November) and figs (the highest level was 520 Bq/kg of fresh weight in Minamisoma-Shi on 19 July, which fell to below 10 Bq/kg of fresh weight in October 2011).

The results of the analyses of the 2012 harvest of yususu and Japanese apricots show that most of the $^{134}\text{Cs} + ^{137}\text{Cs}$ activities are lower than detection limits (a few Bq/kg of fresh weight). The highest activities observed are 10 times lower than those of the 2011 harvest and rarely exceed 50 Bq/kg of fresh weight. It seems that the effect of the very early date of the accident, which led to a relative sparing of the first harvest of fruits and also permitted a moderate contamination of the trees, led to saving the following harvests, including yususu and apricots. This is still to be verified in the following fruits and coming years.

The case of chestnuts, measured in a sample taken in Minamisoma-Shi on 6 September, whose $^{134}\text{Cs} + ^{137}\text{Cs}$ activity reached 2,000 Bq/kg of fresh weight, is similar to that of nuts (hazelnuts, almonds, walnuts, etc.). What these foodstuffs have in common is their low water content and a high amount of potassium, which

⁴ The yusu resembles a small grapefruit with a colour ranging from green to yellow depending on its degree of ripeness. The peel is used in Japanese cookery. The yusu tree, which belongs to the same family as the lemon tree, blossoms and produces fruits year round.

is chemically similar to caesium. The relatively higher contamination of nuts had already been observed in France in 1986, when the activity of hazelnuts and almonds reached several hundred Bq/kg despite considerably smaller radioactive deposits (Renaud *et al.*, 2007). At the time, contamination was passed on in derived products (50 to 150 Bq/kg of fresh weight measured in pralines, chocolate with hazelnuts and other confectioneries in 1986).

Grains

By mid-March 2011, the annual great crops such as grains and oilseeds probably already had foliage that intercepted radioactive deposits but were very far from the flowering stage, which determines how many radionuclides are transferred from leaves to seeds. That explains why the activities in these foodstuffs have only very occasionally exceeded the consumption limits.

Rice had not yet been planted when the deposits were formed, so contamination of this grain could stem only from the soil via the roots, at maximum predictable levels on the order of several hundred Bq/kg, even in the unevacuated areas most severely affected by the deposits. However, the Japanese government allowed its cultivation only in soil whose maximum caesium content was below 5,000 Bq/kg of fresh weight in order to keep within the standards in effect in 2011 (500 Bq/kg of fresh weight). Approximately 3,900 analyses on uncooked or refined rice took place between August 2011 and January 2012. The ^{137}Cs and ^{134}Cs activity measured was often below the measuring devices' detection limits; fewer than 10% of the analyses showed detectable caesium contamination (most of the samples were from Fukushima prefecture); only three tests revealed activity above standards, the highest amount being 630 Bq/kg of fresh weight of uncooked rice.

Farmed meat

Like milk, and for the same reasons, the contamination of farmed meat was moderate considering the radioactive deposits in the most severely affected areas: it was late winter, and livestock were consuming fodder that had been imported or harvested the previous year. ^{131}I in meat was almost always below detection limits. So, caesium activity exceeding standards was detected in just 141 of the 65,000 meat samples tested between March 2011 and February 2012. By way of comparison, had the livestock consumed weeds exhibiting the activity levels measured in Iitate-Mura, ^{137}Cs contamination could have reached 200,000 Bq/kg of fresh meat by mid-April 2011.

Figure 6 shows the $^{134}\text{Cs} + ^{137}\text{Cs}$ activity measured in beef samples taken in localities with the highest readings. With rare exceptions, caesium activity in beef did not regularly exceed 20 Bq/kg after late April. Unlike milk, where the transfer

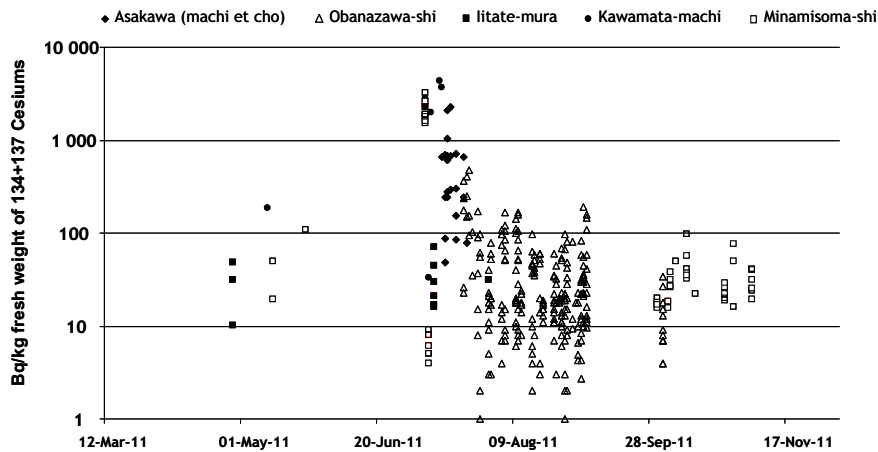


Figure 6 – $^{134}\text{Cs} + ^{137}\text{Cs}$ activity measured in beef samples taken between March and November 2011 in localities where the most tests were performed because they were among the places with the highest activity.

of caesium occurs very fast, caesium builds up only gradually in meat. $^{134}\text{Cs} + ^{137}\text{Cs}$ activity ranged from 1,000 to 4,350 Bq/kg of fresh meat (Kawamata-Machi), peaking in early July.

Feeding practices were more decisive than the location and size of deposits. On 8 and 9 July tests were performed on two batches of meat samples from a farm just outside the 20-km exclusion zone in Minamisoma-Shi north of the Fukushima Daiichi plant. The findings were drastically different from each other: 1,500 to 4,200 Bq/kg for the first batch and four to 10 Bq/kg for the second. As mentioned in the Mainichi Daily News on 12 July, the farmer fed the first batch of cows with rice straw that had been harvested the previous autumn but stored outdoors and, consequently, was contaminated by radioactive fallout. Beef from Obanasawa, located over 150 km northwest of the Fukushima plant, where deposits should not have exceeded 30 kBq/m^2 , was as, if not more, contaminated than meat from areas showing the highest readings of radioactive fallout, showing the importance of livestock feeding practices.

In some cases where activity in meat exceeded 1,000 Bq/kg, on the 14 April 2011, it is possible that the decision by Japan's Ministry of Agriculture, Forests and Fisheries to limit activity levels in fodder to below 300 Bq/kg ($^{134}\text{Cs} + ^{137}\text{Cs}$) was not respected. Some measurement records suggest that after these cases, which involve very few farms, decisions were taken on livestock-rearing practices to lower the meat's contamination levels. However, the decrease was slow: it takes approximately three months for caesium to be biologically eliminated from a

cow's body. For other farms, which do not seem to have taken special precautions because of their distance from the accident site and moderate contamination levels, measurement records show relatively stable activity observed in meat. An example is a farm in Fujinomya (Shizuoka prefecture, over 100 km southwest of Tokyo), where several tens of Bq/kg of activity were observed on a regular basis from July 2011 until recently.

Radioactive caesium activity measured in meat has remained below 100 Bq/kg since early 2012; most findings are lower than the measuring instruments' detection limits, which range from 10 to 50 Bq/kg of fresh weight. The readings seem to show a thorough control of livestock feed and reflect the fact that most farmers keep their cows in stables. However, regular monitoring is still necessary to detect increases. If cattle consume moderately contaminated fodder (approximately 100 Bq/kg of fresh weight) on a regular basis, the concentration of caesium in meat could gradually rise again.

Analyses on chickens and eggs have remained negative (below detection limits except for very low, sporadically observed ^{131}I activity). This is normal considering the feed used, which is often derived from grain harvested the previous year, before the accident. The same goes for pork and pork offal, although activity reaching 100 and even 200 Bq/kg of fresh weight has occasionally been observed.

Mushrooms and game

After the Chernobyl accident it became clear that mushrooms, berries, game and thyme are particularly sensitive to radioactive fallout, exhibiting higher activities than those observed in production of agricultural products originating in the same territory and for a longer time. After several months, levels contrasted sharply with activities in major farmed foodstuffs and livestock. The Fukushima accident confirmed this feature.

Activity observed in April in mushrooms from Fukushima prefecture, from several tens to several thousand Bq/kg of fresh weight of ^{131}I and $^{134}\text{Cs} + ^{137}\text{Cs}$, show that radioactive deposits quickly affected the mycelium. The highest ^{131}I activity, 12,000 Bq/kg of fresh weight, was measured in a shiitake mushroom picked on 8 April 2011; $^{134}\text{Cs} + ^{137}\text{Cs}$ activity stood at 13,000 Bq/kg of fresh weight in this sample. The highest published $^{134}\text{Cs} + ^{137}\text{Cs}$ activity was 28,000 Bq/kg of fresh weight in a milk-cap picked on 1 September 2011.

This activity was found in areas of moderate deposits and it is likely that in the most severely affected areas the highest caesium contamination levels may be observed in the future. However, mushroom analyses quickly focused on cultivated

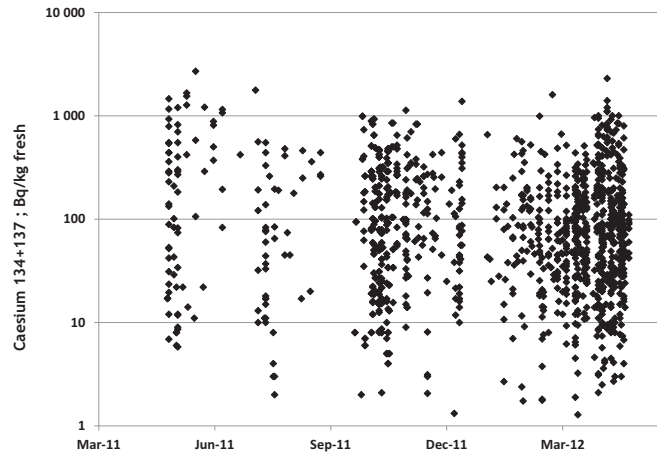


Figure 7 – Caesium activity (^{134}Cs and ^{137}Cs) in fresh shiitakes measured between April 2011 and May 2012 (source: MHLW, 2012). By convention, undetectable amounts are noted as 1 Bq/kg.

shiitakes (over 2,000 tests between March 2011 and June 2012), whose extremely variable activity (from below one to 2,000 Bq/kg) suggest that decrease will occur slowly. If it can be assumed that contamination of natural mushrooms will persist, further measurements should monitor cultivated species, especially shiitakes, to determine whether cultivation practices will lead to decreased contamination.

Nearly 420 tests on samples of game – boar (over 80% of the analyses), Asian black bear, pheasant and cervids – were performed in Japan between April 2011 and May 2012, most after autumn 2011.

The highest caesium activity in boar and bear meat was found in Fukushima prefecture (the highest amount recorded was 14,600 Bq/kg of fresh boar meat and 1,850 Bq/kg of fresh bear meat). Pheasant and cervid meat had the lowest levels: several hundred Bq/kg at the most.

The impossibility of controlling their food intake, the forest environment and the wild animals' lifestyle explain such wide-ranging variations in game contamination levels (from several to several thousand Bq/kg of fresh meat), which, on the whole, are much greater than those of livestock. In the case of boar, the environmental contamination's spatial heterogeneity combined with substantial differences in diet depending on the season: they are herbivorous/fructivorous in spring and summer (although boar are omnivorous and very opportunistic) and foragers (acorns, roots, tubers, earthworms and maggots) in autumn and winter. It is worth noting that after the Chernobyl accident this seasonal variability had been observed

in mushroom-eating deer: contamination levels in meat from an animal killed in autumn can be 10 times higher than in one killed in the spring.

Shoots and tea leaves

Evergreen plants such as bamboo and tea shrubs⁵ intercepted radioactive deposits in March 2011. The larger the leaves' surface area, the more activity they took up. Then, sap carried caesium to all the plants' tissues before it entered new shoots and leaves as long as several months later.

The resulting activity affected tea from the first harvest: 1,330 Bq/kg of fresh leaves in Kanagawa and 981 Bq/kg of fresh leaves in Shizuoka on 21 June (¹³⁴Cs and ¹³⁷Cs). Readings for bamboo shoots of 2,060 Bq/kg in Minamisoma-Shi and 1,070 Bq/kg in Souma were taken on 23 June. Relatively moderate deposits of caesium, estimated at several tens of thousands of Bq/m², which may have formed up to several hundred kilometres from the Fukushima Daiichi plant, may have been responsible for the tea contamination levels.

Many tests were performed on tea but only a few results exceeded detection limits (and even fewer surpassed sales standards). In May 2012 alone (the period of the first "flush", or harvest), 536 tea samples from Tochigi, Chiba and Ibaraki prefectures were analysed; just 11 results exceeded 10 Bq/kg and none were above 25 Bq/kg. However, activity of bamboo shoots harvested the same month still routinely ranged from a few dozen to several hundred Bq/fresh kg.

Caesium activity in 2012 resulting from the leaves' initial interception of radioactive deposits was also responsible for contaminating other seasonal foodstuffs: aralia and koshiabura⁶ shoots and fiddleheads (ostrich ferns, royal ferns, etc.). The activity observed in these samples, collected between April and May 2012, ranged from several tens to several thousands of Bq/fresh kg.

Yacon powder and other products potentially concentrated by processing

Many food-processing methods, including the simplest ones, such as drying, can concentrate the radionuclides found in fresh foodstuffs and be responsible for the highest activity found in products on the market. Activity, for example in rehydrated freeze-dried products such as powdered milk, is often close to that of the initial product. Similarly, although 80% of the caesium in dried tea leaves can be found in brewed tea, activity is much lower than in the fresh leaves due to the low leaf-to-water ratio.

⁵ Tea can be harvested two to four times a year; in Japan bamboo shoots are harvested once a year around May.

⁶ The koshiabura is a tree whose shoots are usually fried in a tempura batter.

The high activity measured in yacon powder⁷, ranging from 15,000 Bq/kg to 20,000 Bq/kg in four of the 12 samples from Miyagi prefecture tested in 2012, resulted in large part from the process of grinding the tuber into a powder.

Conclusion

The date of the Fukushima accident, which occurred very early in the growing and breeding seasons, limited fallout contamination of most foodstuffs of terrestrial origin, especially compared with the heaviest deposits, even more than the date of the 1986 Chernobyl accident did. The highest activity – several tens of thousands of Bq/kg of ¹³¹I or ¹³⁴Cs and ¹³⁷Cs – showed up only in leafy vegetables. If the fallout had occurred three months later, most farm produce, fruit and grains, including rice, could have reached those same levels.

In the case of orchards, the low interception of the deposits due to the lack of leaves for most fruit trees will also limit the following harvests' contamination.

The persistence of the initial foliar transfer is responsible for several hundred thousand Bq/kg of caesium activity measured in 2011 and 2012 in tea leaves, bamboo shoots, aralia and other plants whose evergreen leaves intercepted radioactive deposits.

In the case of meat and dairy products, the common Japanese practice of feeding animals with imported fodder increased the effect of the accident's date. Farmers already had stocks of feed from established supply channels, so they did not use local feed. Some measurements published in Japan have shown that the fact alone of feeding livestock with fodder that was stored outdoors unprotected when the accident occurred can have a significant impact on meat contamination.

The Chernobyl accident showed that "natural" products such as mushrooms and game are particularly sensitive to radioactive fallout; Fukushima confirmed this observation, including for cultivated mushrooms, for which future measurements will determine whether steps can be taken to reduce the persistence of several years of high caesium contamination.

On the whole, knowledge of radioecology makes it possible to explain the observations of foodstuffs made after the Fukushima accident.

⁷ The yacon, or "earth pear", a tuber similar to the Jerusalem artichoke cultivated for its sweetening properties, is used to make "yacon syrup" or ground into a powder to make "yacon tea".

REFERENCES

- Champion D., Korsakissok I., Didier D., Mathieu A., Quelo D., Groell J., Quentric E., Tombette M., Benoit J.P., Saunier O., Parache V., Simon-Cornu M., Gonze M.A., Renaud PH., Cessac B., Navarro E., Servant-Perrier A.C. (2013) The IRSN's earliest assessments of the Fukushima accident's consequences for the terrestrial environment in Japan, *Radioprotection* **48** (1), 11-37.
- Gonze M.A., Simon-Cornu C., Métivier J.M., Garcia-Sanchez L., Mourlon C., Parache V. (2012) Landscape-level Model Predictions of ^{131}I , ^{134}Cs and ^{137}Cs transfer through terrestrial systems in the 80-km Fukushima-Daiichi area using the Symbiose Platform. IRPA, May 2012, Glasgow.
- Korsakissok I., Mathieu A., Didier D. (2013) Atmospheric dispersion and ground deposition induced by the Fukushima Nuclear power plant accident: a local-scale simulation and sensitivity study, *Atmospheric Environment* (accepted for publication).
- MEXT (2011a) Results of the third airborne monitoring by the Ministry of Education, Culture, Sports, Science and Technology. http://radioactivity.mext.go.jp/en/1840/2011/07/1304797_0708e.pdf last accessed on 19 December 2011.
- MEXT (2011b) Results of airborne monitoring Survey by the Ministry of Education, Culture, Sports, Science and Technology in the Western Part of Fukushima Prefecture. http://radioactivity.mext.go.jp/en/1270/2011/09/1270_0912_2.pdf last accessed on 19 December 2011.
- MHLW (2012) Trends in Food Intake by Food Group, 1975 to 2008 (Per capita per day, National Average), extrait du document <http://www.mhlw.go.jp/bunya/kenkou/eiyoudl/h20-houkoku-06.pdf>, page consultée le 10/11/2012.
- Renaud PH., Champion D., Brenot J. (2007) *Les retombés en France de l'accident de Tchernobyl sur le territoire français; conséquences environnementales et exposition des personnes*, TEC&DDOC Lavoisiers, collect. Sciences et Techniques, ISBN 978-2-7430-1027-0, 190 pages.