Environmental Science & Technology

Article

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Analysis of Japanese Radionuclide Monitoring Data of Food Before and After the Fukushima Nuclear Accident

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Supporting Information

ABSTRACT: In an unprecedented food monitoring campaign for radionuclides, the Japanese government took action to secure food safety after the Fukushima nuclear accident (Mar. 11, 2011). In this work we analyze a part of the immense data set, in particular radiocesium contaminations in food from the first year after the accident. Activity concentrations in vegetables peaked immediately after the campaign had commenced, but they decreased quickly, so that by early summer 2011 only a few samples exceeded the regulatory limits. Later, accumulating mushrooms and dried produce led to several exceedances of the limits again. Monitoring of meat started with significant delay, especially outside Fukushima prefecture. After a buildup period, contamination levels of meat peaked by July 2011 (beef). Levels then decreased quickly, but peaked again in September 2011, which was primarily due to boar meat (a known accumulator of radiocesium). Tap water was less contaminated; any restrictions for tap water were canceled by April 1, 2011. Pre-Fukushima ^{137}Cs and ^{90}Sr levels (resulting from atmospheric nuclear explosions) in food were typically lower than 0.5 Bq/kg, whereby meat was typically higher in ^{137}Cs and vegetarian produce was usually higher in ^{90}Sr. The correlation of background radiostrontium and radiocesium indicated that the regulatory assumption after the Fukushima accident of a maximum activity of ^{90}Sr being 10% of the respective ^{137}Cs concentrations may soon be at risk, as the ^{90}Sr/^{137}Cs ratio increases with time. This should be taken into account for the current Japanese food policy as the current regulation will soon underestimate the ^{90}Sr content of Japanese foods.

INTRODUCTION

The Fukushima nuclear accident (Mar. 11, 2011) is regarded as one of the severest environmental disasters in the 21st century.[1,2] The release of radionuclides into the environment in the course of this accident has only been exceeded by the Chernobyl nuclear accident and the cumulative release from atmospheric nuclear explosions.[3,4]

The Fukushima accident caused the contamination of large areas that were and partly still are used for agricultural purposes.[5−7] It has been shown previously[8] that exposure through incorporation of contaminated food potentially affects a large fraction of the population and poses the most relevant radiological risks once evacuation measures have been completed.[9−11]

In the course of the Fukushima nuclear accident, both short- and long-lived activation and fission products have been released into the environment, most of which were nuclides of volatile fission products (noble gases, iodine, cesium, and tellurium). Less volatile elements/radionuclides (e.g., strontium, ruthenium, barium, lanthanides, and actinides) were released to a much lower extent and were monitored only on a few occasions. Fortunately, the most relevant radionuclides, i.e., ^{131}I, ^{132}Te, ^{134}Cs, and ^{137}Cs, are strong γ-emitters, which allows for their fast, straightforward, and reliable detection and quantification using γ-spectroscopy. Determination of pure β- and α-emitters is much more laborious, as it requires chemical treatment and separation. Thus, it does not come as a surprise that it took several months after the accident, before the first monitoring data on pure β-emitters such as ^{90}Sr and α-emitting actinides were published in the scientific literature.[12−16] In order to account for the presence of these "inconvenient" radionuclides, Japanese authorities assumed that ^{90}Sr occurs in a constant ratio together with γ-emitting ^{137}Cs, which can be measured easily. The initial assumption that was reflected in the regulatory limits was based on the experiences of the Chernobyl accident and the fallout of the atmospheric nuclear explosions,[17] assuming an activity correlation of

Received: November 28, 2014
Revised: January 19, 2015
Accepted: January 26, 2015
Published: January 26, 2015

DOI: 10.1021/es5057648
Environ. Sci. Technol. 2015, 49, 2875−2885

^{90}Sr:^{106}Ru:^{134}Cs:^{137}Cs = 5.2:4.3:25.9:64.6. This led to the conservative assumption for ^{90}Sr being 10% of the activity concentration of the respective ^{137}Cs concentration in foods.[8] From Apr. 1, 2012 on, the correlation was adjusted to the specifics of Fukushima, namely, an activity ratio of $^{238+239+240+241}$Pu:^{90}Sr:^{106}Ru:^{134}Cs:^{137}Cs = 0.000002:0.003:0.02:0.92:1.[18] According to these ratios, the maximum ^{90}Sr concentration was assumed to be 0.3% of the respective ^{137}Cs concentration after April 2012.

The radionuclide food monitoring campaign that followed the nuclear accident proved to be unprecedented in human history. Tens of thousands of samples were analyzed in the weeks and months after March 2011,[8,19] adding up to almost one million measurements by the end of 2014. Basically all radiation detection capacities of the country, including those of universities and research laboratories, were used to gather crucial information on the radionuclide contamination levels in various regions and prefectures in Japan. However, the amount of data stockpiled in this monitoring campaign was focused solely with respect to compliance with the regulatory limits, but often the data were published on the web without sufficient interpretation and analysis. In this study, we launch a first attempt to analyze the enormous data set of food monitoring data after the Fukushima accident. We will also discuss the lessons learned from pre-Fukushima food monitoring campaigns, which have interesting implications on the challenges Japan is currently facing.

■ MATERIALS AND METHODS

For a general classification of the food, we distinguished between animal products (not including seafood) and vegetarian produce. Three main categories of post-Fukushima data are discussed herein: meat/egg (see the caption of Figure 3 for subcategories), vegetables (see Table S1 in the Supporting Information for subcategories), and potable water.

The data set gathered and published by the Japanese Ministry of Health, Labour and Welfare (MHLW) after the Fukushima nuclear accident[20] comprises the stunning number of 877,635 samples (from Mar. 11, 2011 until Aug. 31, 2014) and includes activity concentrations of ^{134}Cs ($T_{1/2}$ = 2.1 a) and ^{137}Cs ($T_{1/2}$ = 30.1 a) and, in the early stage after the accident, short-lived ^{131}I ($T_{1/2}$ = 8.0 days). For this study, we focused on radiocesium contamination levels in food from the first year (Mar. 11, 2011 until Mar. 31, 2012), which includes 133,778 samples (see Supporting Information, Tables S1 (categories), S2 (summary), and S3 (individual measurements)). The samples represent the Japanese food basket; 80.9% were obtained from the "premarket" (farmers, producers, and so on), 8.8% were bought in grocer's shops and so forth ("post-market"); 10.3% were not specified. We focus on Fukushima prefecture and surrounding/affected prefectures (see Figure S1 in the Supporting Information for the geographical setting). In the discussion, the sampling dates are used as the reference time. On few occasions, no sampling date is given in the data pool; in this case, the measurement date (or date of the press release) was used instead. Concerning the origin of the food samples, Japanese authorities (MHLW) distinguish between "producers" and "processors" of foodstuffs. Hence, a food is attributed to the place of agricultural production, not the place of procession or sale. The provisional regulatory limit for $^{134+137}$Cs in vegetables, cereals, meats, eggs, seafood, and other foodstuffs was 500 Bq/kg until Mar. 31, 2012; the new regulatory limit is 100 Bq/kg (valid from Apr. 1, 2012).[8,21]

Data on contaminations (^{131}I and $^{134+137}$Cs) in potable water were obtained from refs 22−24. The regulatory limits for liquid foodstuffs (water) were 300 Bq/kg (^{131}I) and 200 Bq/kg ($^{134+137}$Cs), respectively, until Mar. 31, 2012; the new regulatory limit (valid from Apr. 1, 2012) is 10 Bq/kg for radiocesium.

For the discussion of the background activities, we analyzed data published by the Nuclear Regulation Authority.[25] This data set comprises as many as 776 γ-spectroscopic measurements of various foods (342 animal products and 434 vegetarian products) from several Japanese prefectures (see Supporting Information). In this database also the remarkable number of 418 radiochemical measurements of ^{90}Sr ($T_{1/2}$ = 28.9 a; 184 animal products and 234 vegetarian products) is included. The data set comprises measurement data for ^{90}Sr and ^{137}Cs from 1987 until 2004; however, most data were collected between 1999 and 2004. Some of the samples did not exhibit detectable activities of one of the two or both radionuclides. Data on ^{90}Sr and ^{137}Cs concentrations in polished rice[26] and wheat[27] were used to discuss the time span from 1959 to 1995. For rice, 516 ^{90}Sr measurements and 513 ^{137}Cs measurements were reported; for wheat, 363 ^{90}Sr measurements and 355 ^{137}Cs measurements.

Any scientific analysis is potentially prone to biases. Especially in the case of the measurements of post-Fukushima food, human biases in sampling (e.g., areas of origin) or the choice of investigated foods cannot be excluded. However, the sheer amount of measurements allow for the identification of trends and the identification of sentinel foodstuffs despite potential biases. It is unclear, whether or not corrections for peak summing of ^{134}Cs γ-photons were performed in the γ-spectroscopy, so some uncertainty may have been introduced here. A lacking correction may underestimate the ^{134}Cs activities by up to 20%.

■ RESULTS AND DISCUSSION

Food Monitoring after the Fukushima Nuclear Accident. A summary of the samples measured after the Fukushima accident and the fraction exceeding the regulatory limits is given in the Supporting Information, Table S2, which is based on the information provided by MHLW.[20] Naturally, the number of samples *measured* was greatest for Fukushima prefecture (see Supporting Information Table S2). Also the highest number of samples *exceeding* the regulatory limits was found there. With the increase of radioanalytical facilities, the number of samples measured increased from 21,549 samples in the first year to 34,857 in the second year and 40,759 samples in the third year after the accident. By laying focus on "suspicious" or sensitive food types, the fraction exceeding the limits in food from Fukushima increased from 3.3% in the first year to 4.0% in the second year; however, it dropped to 1.5% in the third year and 0.6% in the final period of observation (Apr. 1, 2014 until Aug. 31, 2014).

Other prefectures were significantly less affected (e.g., in Gunma 0.2% or in Ibaraki 0.6% exceeding limits in the first year), with the notable exception of Saitama prefecture south of Fukushima, which reported 3.6% of all measured samples exceeding the limits in the first year, thus a higher percentage even than for Fukushima prefecture. From a radioecological point of view, it is interesting to note that it was exclusively samples of Japanese tea that exceeded the limits in Saitama in

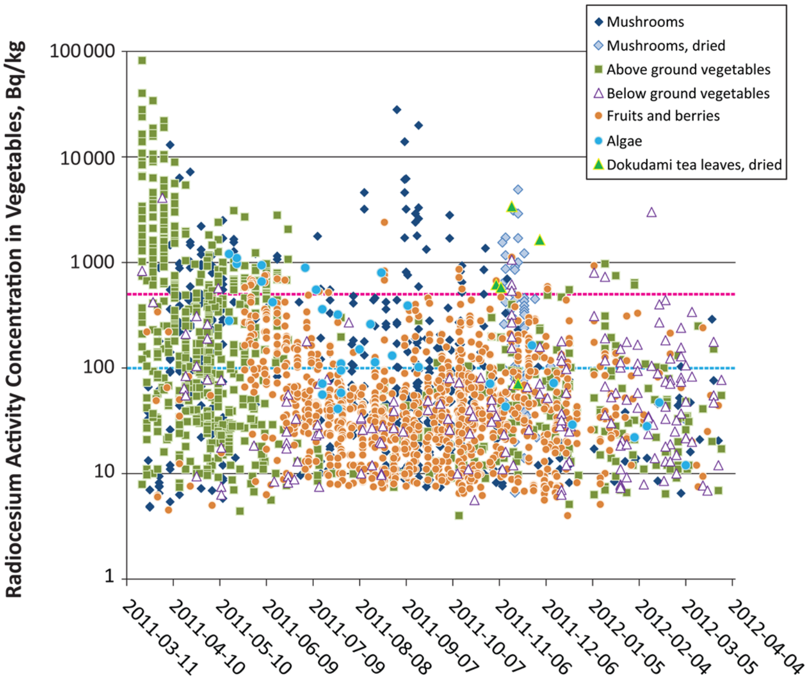

Figure 1. Radiocesium (^{134}Cs + ^{137}Cs) activity concentrations in vegetables and vegetarian produce from Fukushima prefecture sampled over the period Mar. 11, 2011 until Mar. 31, 2012. The provisional regulatory limit for vegetables, cereals, meats, eggs, seafood, and other foodstuffs (500 Bq/kg; valid until Mar. 31, 2012) is indicated by the dotted magenta line. For information purposes, the new regulatory limit (100 Bq/kg; valid from Apr. 1, 2012) is indicated by the dotted light blue line.

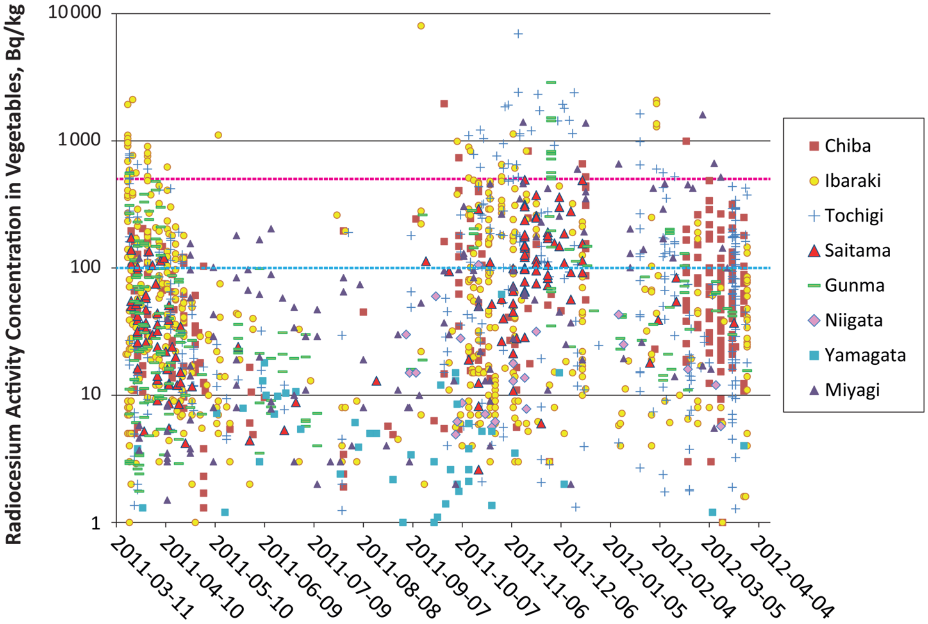

Figure 2. Radiocesium (^{134}Cs + ^{137}Cs) activity concentrations in vegetables and vegetarian produce from selected and affected prefectures around Fukushima prefecture sampled over the period Mar. 11, 2011 until Mar. 31, 2012. The provisional regulatory limit for vegetables, cereals, meats, eggs, seafood, and other foodstuffs (500 Bq/kg; valid until Mar. 31, 2012) is indicated by the dotted magenta line. For information purposes, the new regulatory limit (100 Bq/kg; valid from Apr. 1, 2012) is indicated by the dotted light blue line.

the first year. The Japanese tea plant is known to absorb deposited cesium by foliar uptake and to translocate the cesium from older leaves to younger leaves, which are then harvested and used for the production of tea.[28] Since this mechanism is only relevant in the first year after the accident, the number of tea samples exceeding the limits dropped from 127 (3.6%) in the first year after the accident to 0 in all following periods of observation. The 13 samples (0.3%) from Saitama exceeding limits in the second year were mostly deer meat and mushrooms; so were the 6 (0.1%) in the third year. In the latest period of observation no exceedances were reported from Saitama. Outside the Tohoku area, only very few samples exceeded the limits, as shown in Supporting Information Table S2. In these remote prefectures, it was mostly radioecologically sensitive organisms (such as fungi) that caused the exceedances.

Vegetarian Produce after the Fukushima Nuclear Accident. The database for vegetarian produce commences with monitoring data obtained on Mar. 21, 2011 in Fukushima prefecture and Mar. 17, 2011 in other prefectures outside Fukushima (Ibaraki). It is likely, though, that some scattered measurements were already conducted prior to these dates but not included into the data set. In Fukushima, exceedances of the provisional regulatory limits were reported right on March 21; in other prefectures, on March 18. Naturally, in the initial phase, ^{131}I was the main cause for exceedances of the limit. The maximum radiocesium activity concentrations dropped within a month by more than an order of magnitude, from 82 kBq/kg on March 21 to less than 8 kBq one month later (Figure 1). However, still a significant number of samples exceeded the provisional regulatory limit of 500 Bq/kg (indicated by the magenta line in Figure 1). For comparison, also the new regulatory limit of 100 Bq/kg (valid after Apr. 1, 2012) is included in the following figures in the form of the light blue dotted line.

By the beginning of August 2011, hardly any samples violated the regulatory limit, until the trend was reversed by mid-August due to high radiocesium found primarily in mushrooms (other foods occasionally exceeding the limits were, e.g., seaweed or (citrus) fruits). This trend peaked in early September 2011, when mushrooms containing high amounts of radiocesium (28 kBq/kg in coral fungi) were reported. Such high values have been observed in non-mushroom-vegetables only until the beginning of April 2011. Later in the mushroom season, a second distinct peak was observed in November which was mainly due to dried mushrooms. Also dried tea leaves contributed to the high activity levels. After this second peak, activity concentrations dropped again, until in January 2012 a third, much less pronounced peak was observed, not involving any mushrooms but primarily dried produce (dried yacon (leaves), dried taro, but also citrus fruits (yuzu) and Japanese radish and horseradish leaves). It is obvious from Figures 1 and 2 that the measurement density was much lower over the holiday season in late December 2011, so that a gap can be observed here.

A very similar or even more pronounced picture is shown in Fukushima's neighboring (or affected) prefectures (Figure 2).

In the early aftermath of the accident, mainly samples from Ibaraki (and some from Tochigi as well as one from Gunma) exceeded the regulatory limits; however, radiocesium concentrations were significantly lower than what was observed in Fukushima. After mid-April only very few samples (parsley from Ibaraki in mid-May) exceeded the limits, but from September 2011 on many violations occurred again. In this case, the maximum contamination levels in this period were even significantly higher than those found in the early period after the accident. Again, this peak was mainly due to (dried) mushrooms (e.g., Shiitake) and lasted until the end of March 2012. It is interesting to note that although mostly samples from Ibaraki caused the majority of exceedances in the early period, it was mostly samples from Tochigi that were responsible for some of the high activity concentrations (>1 kBq/kg) in the fall−winter period of 2011. However, also samples from Gunma, Miyagi, Chiba, and Ibaraki had relatively high contamination levels. Yamagata and Niigata were less affected.

Meat and Eggs after the Fukushima Nuclear Accident. According to the database (Supporting Information Table S3), the first two samples of beef from Fukushima prefecture were taken already on Mar. 15, 2011, but they did not reveal any detectable activities. The main monitoring campaign of meat/eggs from Fukushima, however, started on Mar. 26, 2011 and revealed detectable ^{131}I in chicken eggs right on the very first day. One day later, radiocesium was detected in chicken eggs. In contrast to vegetarian produce, the peak activity concentration was not observed in the very beginning with meat/eggs. After a constant increase and buildup of radiocesium activity concentrations, the provisional regulatory limits were exceeded for the first time on Jun. 10, 2011 in beef with a total radiocesium activity of almost 2 kBq/kg (Figure 3). At the end of June/beginning of July several beef samples clearly exceeded the regulatory limits; then the maximum levels dropped again. The maximum activity concentrations, however, were observed in boar meat on Sep. 5, 2011 and Dec. 26, 2011 (14600 and 13300 Bq/kg, respectively). Although the "summer peak" that was well-observed with vegetables (mainly due to the fact that mushrooms are remarkable cesium accumulators[29−31]) was not as clearly pronounced with meat (see Figure 3), the mechanisms for these peak activities are similar. Boars are well-known for feeding on mushrooms and other hyper-accumulators, thus accumulating high activities of radiocesium.[32]

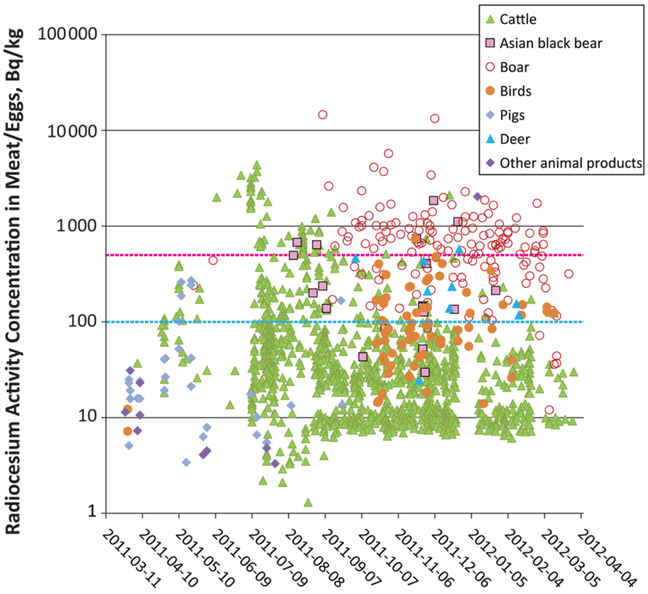

Figure 3. Radiocesium (^{134}Cs + ^{137}Cs) activity concentrations in meat/eggs from Fukushima prefecture sampled over the period Mar. 11, 2011 until Mar. 31, 2012. The provisional regulatory limit for vegetables, cereals, meats, eggs, seafood, and other foodstuffs (500 Bq/kg; valid until Mar. 31, 2012) is indicated by the dotted magenta line. For information purposes, the new regulatory limit (100 Bq/kg; valid from Apr. 1, 2012) is indicated by the dotted light blue line. "Birds" include the meat of chicken, common teal, copper pheasant, green pheasant, crossbred mallard and domestic duck, mallard, and spot-billed duck. "Other animal products" include chicken eggs, chicken liver, pig liver, horse meat, and hare meat.

Similarly, the specific diet of deer and Asian black bears (including berries and lichen) also leads to higher activity concentrations in the meat of bears (e.g., Oct. 13, 2011) and deer (e.g., Dec. 26, 2011) (see Supporting Information Table S3).[33,34] Although the category of beef dominated the violations of the provisional regulatory limit in the early phase after the accident, with very few exceptions, most cattle

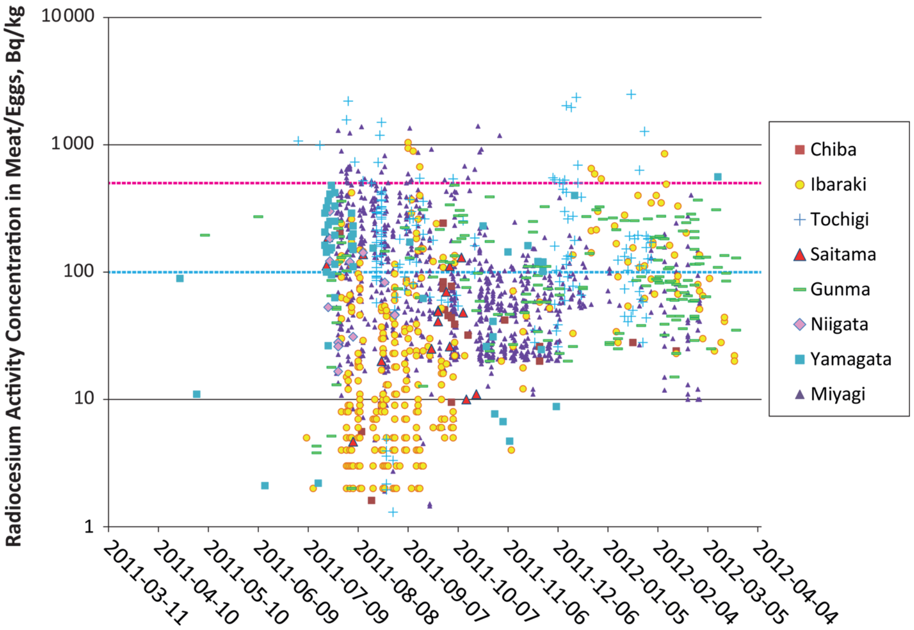

Figure 4. Radiocesium (^{134}Cs + ^{137}Cs) activity concentrations in meat/eggs from selected and affected prefectures around Fukushima prefecture sampled over the period Mar. 11, 2011 until Mar. 31, 2012. The provisional regulatory limit for vegetables, cereals, meats, eggs, seafood, and other foodstuffs (500 Bq/kg; valid until Mar. 31, 2012) is indicated by the dotted magenta line. For information purposes, the new regulatory limit (100 Bq/kg; valid from Apr. 1, 2012) is indicated by the dotted light blue line.

did not exhibit high activities from late summer 2011 onward. The violations of the limit were clearly dominated by boar meat. Also in the meat/eggs monitoring campaign, a significant gap can be observed during the holiday season in December 2011.

In Japanese prefectures other than Fukushima, monitoring of meat/eggs started with a significant delay (Figure 4). Although some samples were taken and measured as soon as March 20, the systematic survey of meat/eggs started only by the middle/end of July 2011. Due to the delay it seems likely that some above-limit meat/eggs may have made it into the markets and may have been consumed. The first sample we are aware of that exceeded the radiocesium regulatory limit was deer meat from Tochigi (Jul. 3, 2011; 1069 Bq/kg). Soon, boar meat from Tochigi also exceeded the limit (Jul. 16, 2011; 990 Bq/kg). Beef also caused several exceedances, with the highest value found in Iwate (not shown in Figure 4) with 2430 Bq/kg on Aug. 18, 2011. Several beef samples from Miyagi also exceeded the regulatory limits in late summer and fall 2011. Again, it was primarily boar, beef, and deer meat that caused the violations in the affected prefectures around Fukushima. From December 2011 to February 2012, it was mainly boar and deer meat in Tochigi and Ibaraki that exceeded the radiocesium limits.

Contaminations (^{131}I and $^{134+137}$Cs) of Potable Water after the Fukushima Nuclear Accident. Relatively little has been published in English scientific literature about radionuclide contamination levels in potable water[35−38] and its treatment in response to the Fukushima accident.[39] The databases,[22−24] however, allow an assessment of the activity concentrations of ^{131}I and radiocesium ($^{134+137}$Cs) in tap water (see Figure 5).

It is interesting to note that in tap water ^{131}I activity concentrations (Figure 5a) were not only much higher than the respective radiocesium concentrations but also that the maximum contamination levels were roughly in the same order of magnitude in all 4 of the most affected prefectures (Fukushima, Chiba, Ibaraki, Tokyo). The levels dropped quickly after the accident: after Mar. 23, 2011, our databases did not report any exceedances of the limits of ^{131}I (300 Bq/kg). Restrictions for tap water were canceled by Apr. 1, 2011. Figure 5a also shows that the ^{131}I levels dropped faster than just due to physical decay (gray diagonal lines), suggesting a shorter effective half-life in tap water than 8 days. Data on radiocesium in tap water are rather sparse (Figure 5b). The data compiled in our databases did not indicate any exceedances of the early regulatory limit for radiocesium (200 Bq/kg). Since later monitoring (late summer of 2011) did not show any detectable radiocesium in tap water,[22] it appears unlikely that the new regulatory limit of 10 Bq/kg (blue dotted line) was exceeded.

Cesium-137 Background Prior to the Fukushima Nuclear Accident. Certain background levels of radiocesium exist in Japan due to the fallout from atmospheric nuclear explosions of the 20th century. Currently, the impact of the Fukushima accident can easily be distinguished from the background by the presence of the relatively short-lived reactor nuclide ^{134}Cs.[40] The average ^{134}Cs/^{137}Cs activity ratio at the time of the accident was 0.98 in food.[21] After some years of decay, however, it will become increasingly difficult to determine residual activities of ^{134}Cs. As an alternative, the ratio of ^{137}Cs to long-lived ^{135}Cs ($T_{1/2} = 2 \times 10^6$ a) can be used instead;[41−44] however, this is laborious and requires special instrumentation. Cesium-135, therefore, is unlikely to become a tracer for routine monitoring campaigns. Instead, the contribution of the pre-Fukushima background can only be estimated based on previous monitoring data (see Table S4 in the Supporting Information). See Supporting Information Figure S2 for the temporal evolution of the ^{134}Cs/^{137}Cs activity ratio in food over the first year.

The ^{137}Cs background data (Figure 6) from Japan reveal that most samples were rather low in radiocesium, most of which with less than 0.5 Bq/kg (with the exception the 1992 samples of mutton and chocolate). Generally, more samples of meat/

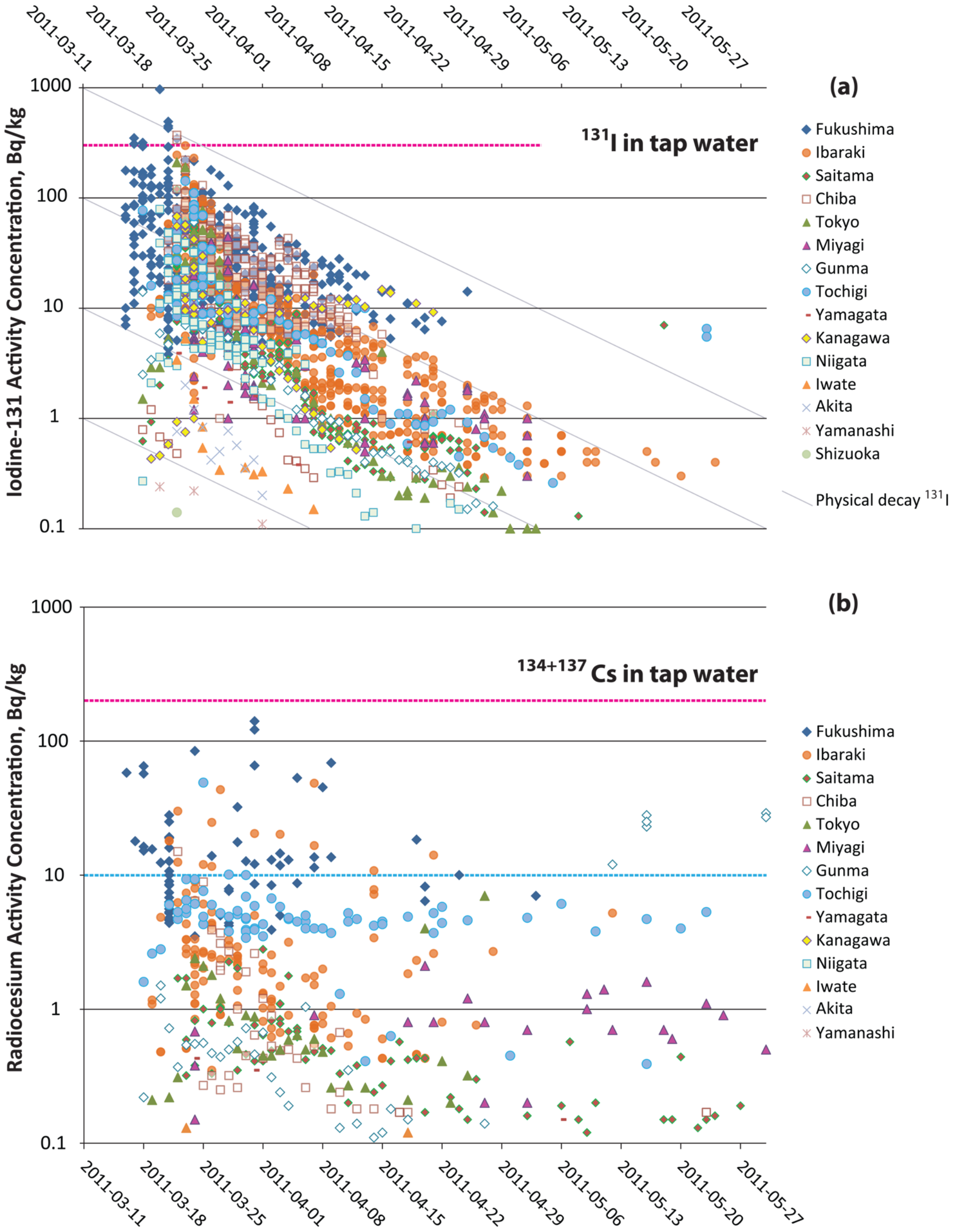

Figure 5. Iodine-131 (a) and radiocesium (^{134}Cs + ^{137}Cs) (b) activity concentrations in tap water from affected prefectures sampled over the period Mar. 18, 2011 until May 27, 2012. The provisional regulatory limit for liquid foodstuffs (300 Bq/kg for ^{131}I and 200 Bq/kg for $^{134+137}$Cs; valid until Mar. 31, 2012) is indicated by the dotted magenta line. For information purposes, the new regulatory limit (10 Bq/kg for $^{134+137}$Cs; valid from Apr. 1, 2012) is indicated by the dotted light blue line. Gray diagonal lines in (a) indicate the physical decay of ^{131}I. Data taken from refs 22−24.

eggs than vegetarian produce exhibited detectable ^{137}Cs activities. Interestingly, several samples of meat/eggs from Fukushima prefecture were higher in ^{137}Cs activities than samples from other prefectures (Figure 6). It is unlikely, however, that the background will contribute significantly to the total post-Fukushima contamination of foods.

Strontium-90 Background Prior to the Fukushima Nuclear Accident. In contrast to ^{137}Cs, more vegetarian produce revealed detectable ^{90}Sr activity concentrations than animal product samples (Figure 7). This is probably due to the fact that ^{90}Sr is a bone-seeking radionuclide, so that any ^{90}Sr taken up by animals is hardly transferred to the muscle but to the bones instead. A notable exception may be milk, which is naturally rich in Ca and thus a good carrier for ^{90}Sr. Note that only one sample of (condensed) milk was included in the data set of the ^{90}Sr background. In any case, activity concentrations usually did not exceed 0.5 Bq/kg (with the exception of one green pepper sample from 2004). Samples from Fukushima prefecture proved to exhibit similar ^{90}Sr activity concentrations like other prefectures.

Correlation of ^{90}Sr and ^{137}Cs in Foodstuffs. Determination of radiostrontium is rather laborious, making it one of the understudied radionuclides after the Fukushima nuclear accident.[45] In order to account for the environmental presence of ^{90}Sr, Japanese authorities assumed a constant ratio between ^{90}Sr and ^{137}Cs, the latter of which can be measured rapidly using γ-spectrometry.[8,13] According to this assumption, the ^{90}Sr activity concentration should not exceed 10% (March 2011 to April 2012) or 0.3% (after April 2012), respectively, of the respective ^{137}Cs activity concentration. This assumption is

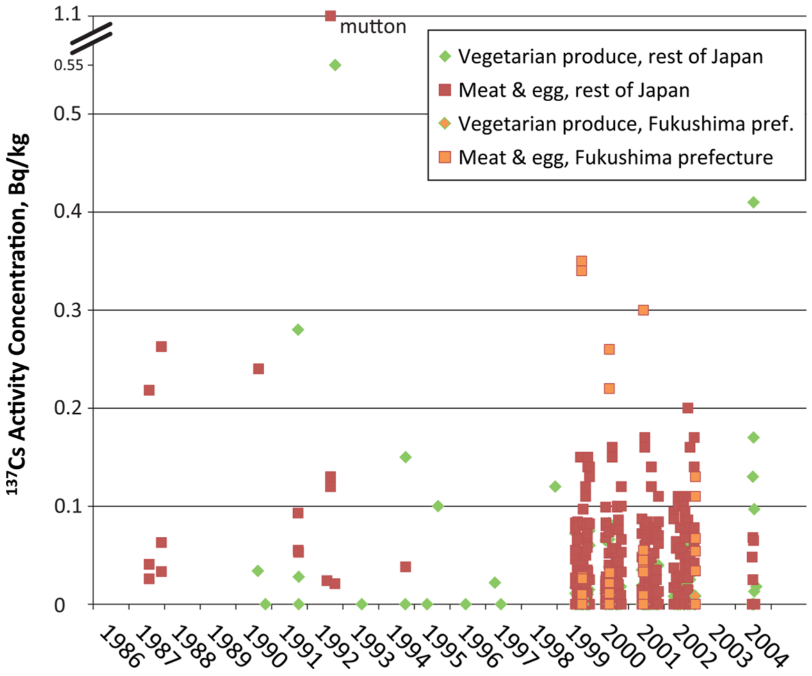

Figure 6. Cesium-137 activity concentrations in vegetarian produce and meat/eggs from Fukushima prefecture and other prefectures in Japan sampled over the period 1987 until 2004.

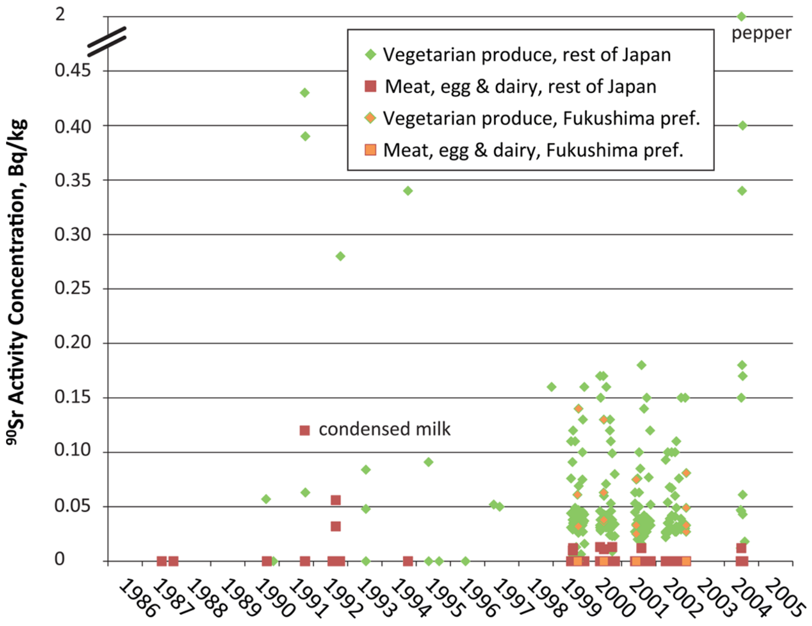

Figure 7. Strontium-90 activity concentrations in vegetarian produce and meat/eggs from Fukushima prefecture and other prefectures in Japan sampled over the period 1987 until 2004.

routed in observations following the Chernobyl accident and atmospheric nuclear explosion fallout, e.g., ref 17. Since the Fukushima nuclear accident was a much more powerful source of radiocesium than of less volatile radiostrontium than Chernobyl or the nuclear weapons fallout,[3,46] it was a reasonable and conservative approach to implement the same ratio as the maximum ^{90}Sr content in Fukushima's contaminations.

Our analysis of the background activities, however, shows that this assumption is at risk (Figure 8). The (rather low) number of samples that exhibited detectable activities of both ^{137}Cs and ^{90}Sr showed that the vast majority of the food samples exceeded the ^{90}Sr/^{137}Cs ratio of 0.1, and all were higher than 0.003. Most samples even showed a ratio of ≥ 2. Only the one sample of mutton that was discussed previously with its unusually high ^{137}Cs activity concentration had a ^{90}Sr/^{137}Cs ratio of <0.1. Generally, meat/eggs proved to show a slightly lower ^{90}Sr/^{137}Cs ratio than vegetarian produce, owing to the generally greater activity of ^{137}Cs in meat/eggs compared with ^{90}Sr.

Due to very similar half-lives of both ^{90}Sr and ^{137}Cs, any of these ratio anomalies cannot be due to physical decay. Instead, analysis of 1959−1995 data on ^{90}Sr and ^{137}Cs in rice[26] and wheat[27] reveals that a low ^{90}Sr/^{137}Cs activity ratio is justified

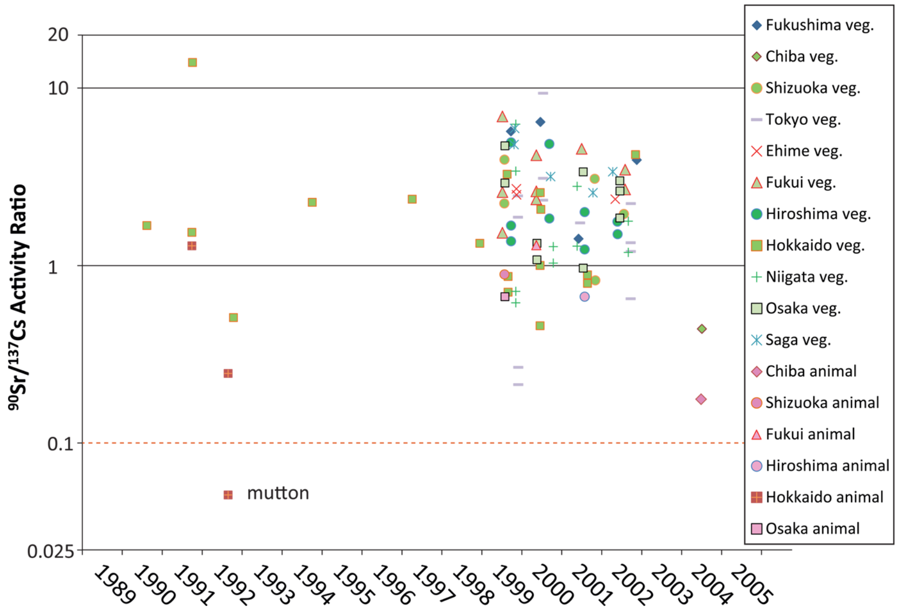

Figure 8. Activity ratios of ^{90}Sr/^{137}Cs in vegetarian produce and meat/egg products from Japan sampled and measured before the Fukushima nuclear accident. The dashed red line indicates the 10% limit that was assumed by Japanese authorities as the maximum ^{90}Sr content after the Fukushima accident.

only for the initial period of a couple of years, as the ratio rises over the years (Figure 9). The analysis shows that wheat has a higher ratio than rice, but both rise significantly over the following years and decades following the period of the main nuclear fallout in the 1960s. Only the Chernobyl accident caused a singular outlier in the series of wheat measurements. The Chernobyl accident was a significant source of ^{90}Sr only in the closer vicinity of the destroyed NPP, but in Japan only significant amounts of airborne radiocesium were observed.[47]

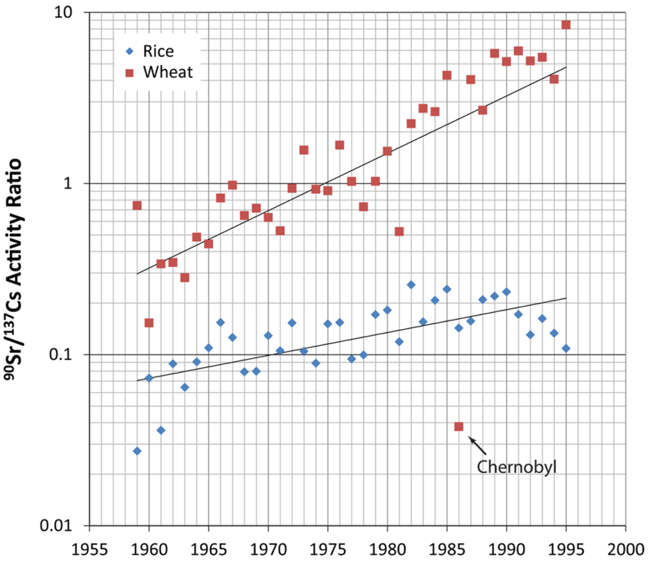

Figure 9. Activity ratios of ^{90}Sr/^{137}Cs in wheat and polished rice from Japan sampled and measured from 1959 until 1995. Data taken from refs 26 and 27.

This analysis reveals that ^{90}Sr exhibits a higher mobility and bioavailability than radiocesium, whereas ^{137}Cs is more readily adsorbed and immobilized on clay minerals, thus causing the distortion of the initial activity ratio in food. One can speculate that the data presented in Figure 8 represent the plateau of this distortion where the ^{90}Sr/^{137}Cs activity ratio of most foodstuffs remains at a rather constant level between 1 and 10. The reason for the slower increase of the ratio in rice is most probably due to a radioecological anomaly of paddy-cultivated rice. The paddy cultivation causes the formation of ammonia from putrefaction. The NH_3 dissolves in water and forms NH_4^+ ions which are very efficient ion exchangers for adsorbed Cs^+ ions on clay minerals. Compared with conventional cultivation methods, the paddy cultivation of rice thereby increases the mobility of Cs and hence makes ^{137}Cs more bioavailable.

The increasing ^{90}Sr/^{137}Cs activity ratio and its effects on the regulatory limit must be taken into account for the Fukushima nuclear accident and future radioecological considerations with respect to food safety and monitoring. The current assumption of the maximum ^{90}Sr/^{137}Cs activity ratio in food will be no longer true within a few years after the accident.

The impact of this erroneous assumption of a constant ^{90}Sr/^{137}Cs activity ratio can be quantified as shown in Figure 10. For this figure, a daily consumption of 1.7 kg of (solid) foods was assumed (National Nutrition Survey of Japan[48]). Also we assume no contribution from short-lived ^{134}Cs, because the effects will become critical after several years only. Consumption over 1 year of foods contaminated with 100 Bq/kg ^{137}Cs and 10 Bq/kg ^{90}Sr (authority-assumed ratio of ^{90}Sr/^{137}Cs of 0.1) will cause a committed dose of 1 mSv. With a higher ^{90}Sr/^{137}Cs ratio, the received dose increases as shown in Figure 10. In this figure, any white areas are "covered" by the Japanese regulations: foods with ^{137}Cs contaminations > 100 Bq/kg will be "rightfully" banned; foods with ^{137}Cs contaminations < 100 Bq/kg and low ^{90}Sr/^{137}Cs ratios will be "rightfully" permitted. However, any colored areas in Figure 10 are "blind spots" that remain uncovered by the regulations: Foods with activities falling into this area will be falsely permitted although their consumption may be critical. For example, when foods contaminated with 23 Bq/kg ^{137}Cs are consumed over the period of a year (clearly below the current

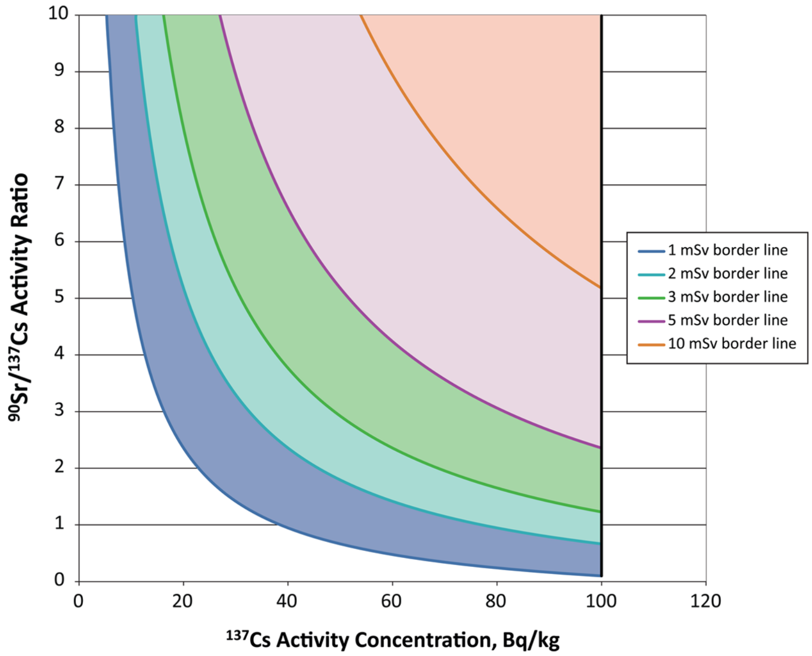

Figure 10. Dose relevance of the ^{90}Sr/^{137}Cs ratio at a given ^{137}Cs activity concentration in foods, assuming a consumption of such contaminated foods over the period of 1 year. The colored areas represent the "blind spots" that are not covered by the current regulation.

limit of 100 Bq/kg), a ^{90}Sr/^{137}Cs ratio of 2 will already cause a committed dose of 1 mSv. At this activity ratio, a contamination level of 46 Bq/kg ^{137}Cs (less than half of the current limit) will deliver 2 mSv. If we keep in mind that the pre-Fukushima samples often had ^{90}Sr/^{137}Cs activity ratios > 2 (up to 10), this scenario illustrates the potentially severe impact of this erroneous assumption of a constant ratio at 0.1 (or even below).

Lessons Learned. An ample set of food monitoring data allows for the observation of general radioecological trends, such as the mobility and bioavailability of radionuclides. In this particular case, the long series of pre-Fukushima monitoring data teach us that the ^{90}Sr/^{137}Cs activity ratio is not constant in foodstuffs but constantly increases with time, thus causing an underestimation of the internal exposure as long as a constant (and low) ratio is assumed by the regulatory bodies. Data from pre-Fukushima monitoring campaigns revealed that animal products tend to be higher contaminated with radiocesium, whereas vegetarian produce exhibited higher activity concentrations in radiostrontium. The stunning amount of post-Fukushima food monitoring data clearly allows the identification of radioecologically "sensitive" foodstuffs. For the vegetarian food sector, these sentinels are primarily mushrooms and to a lower extent yuzu (citrus fruits), berries and Japanese radish. Due to its special diet, the boar is a suitable sentinel in the animal product sector as it feeds on mushrooms and other hyperaccumulators.

In summary, the Fukushima nuclear accident triggered an unprecedented monitoring campaign for radionuclides in food. Vegetables from Fukushima prefecture exhibited high radiocesium activity concentrations soon after the accident. However, by late summer of 2011, it was mostly mushrooms or dried vegetarian foodstuffs that exceeded the provisional regulatory limit. A similar picture was observed in other affected prefectures: after an initial high, activity concentrations in vegetable dropped quickly, but peaked again due to mushrooms and dried vegetables. This confirms the necessity to monitor mushrooms as sentinel species for radiocesium.[30] Monitoring of meat/eggs started with significant delay after the accident, especially in prefectures other than Fukushima. Due to the constant intake of contaminated pasture, radiocesium concentrations in animal products from Fukushima built up relatively slowly and peaked for the first time in early July 2011. In this initial period, it was mainly beef responsible for exceedances of the provisional regulatory limits. After the peak, activity concentrations dropped again to rise back from September 2011. This time, it was mainly boar meat that was highly contaminated. Iodine-131 in tap water exhibited high levels shortly after the accident in several affected prefectures, but no exceedances of the limit were observed after March 2011. Radiocesium levels in tap water were rather low. Given the high monitoring density, the mostly rapid response of Japanese authorities and the rapid decrease of very high initial contamination levels of the most common foods, it seems very unlikely that more than very few members of the public in Japan exceeded the maximum permissible internal exposure of 1 mSv/year. This observation is in agreement with the results of previous studies.[9−11,21,49−51] A key finding of this study is that the correlation between ^{90}Sr and ^{137}Cs may soon no longer follow the assumption of a maximum ^{90}Sr/^{137}Cs activity ratio of 0.1 or even 0.003 in food. Background data from Japan suggested that after several years following the release into the environment, the ^{90}Sr/^{137}Cs activity ratio observed in food rises significantly (most of the samples showing a ratio > 2). This calls for an adaption of the current policy and also increased monitoring efforts with respect to ^{90}Sr. The diminution of the regulatory limit (^{90}Sr/^{137}Cs = 0.003) as of April 2012 was an adaption into the wrong direction. The Japanese authorities are urged to reimplement the "old" limit (^{90}Sr/^{137}Cs = 0.1), which probably will have to be raised further in the future. This observation fosters the need for continuous monitoring of both ^{137}Cs and ^{90}Sr; otherwise the ^{90}Sr content of food will soon be underestimated.

■ ASSOCIATED CONTENT

Ⓢ Supporting Information

Categories of vegetarian produce after the Fukushima accident (separate Table S1), summary of all monitoring activities in Japan from Mar. 11, 2011 until Aug. 31, 2014 (Table S2), individual samples and measurements of food after the Fukushima nuclear accident (separate Table S3) and before the accident (separate Table S4), location of the Fukushima Daiichi Nuclear Power Plant in Fukushima prefecture and other prefectures affected by the 2011 nuclear accident (Figure S1), and activity ratios of ^{134}Cs/^{137}Cs in food from Fukushima prefecture sampled over the period of the first year after the accident (Figure S2). This material is available free of charge via the Internet at http://pubs.acs.org.

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Notes

The authors declare no competing financial interest.

■ ACKNOWLEDGMENTS

This study was supported by JSPS KAKENHI (Grant No. 25870158) as well as Grant No. T42OH009229-07 from CDC NIOSH Mountain and Plains Education and Research Center. Its contents are solely the responsibility of the authors and do not necessarily represent the official views of the CDC NIOSH and MAP ERC. G.S. gratefully acknowledges funding by the U.S. Nuclear Regulatory Commission (NRC), Grant No. NRC-HQ-12-G-38-0044.

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