Radiocaesium contamination of wild boars in Fukushima and surrounding regions after the Fukushima nuclear accident

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Abstract

Analysis of radioactivity data obtained under the food monitoring campaign in Japan indicates that elevated $^{134}\text{Cs}+^{137}\text{Cs}$ activity concentrations in wild boar meat remained constant or slowly decreased in Fukushima and surrounding prefectures from 2011 to 2015. The activity concentrations in some samples are still over the regulatory limit of 100 Bq kg^{-1} fresh weight, even in 2015. Activity concentrations of ^{137}Cs in muscle of wild boars we captured in 2011 were higher than those in kidney, liver, spleen, heart and lung. A food processing retention factor, F_r , was 0.5 or 0.6 for ^{137}Cs when the wild boar meat was boiled, suggesting that a parboiling process is effective for reduction of radiocaesium intake from wild boar meat.

1. Introduction

On 11 March 2011, a catastrophic earthquake (M 9.0) occurred in the northwest Pacific, about 130 km off the coast of northeastern Japan, followed by a gigantic tsunami, which caused serious damage to the electrical system of the Fukushima Dai-ichi Nuclear Power Plant (F1-NPP). Therefore, the nuclear reactor cooling systems failed, and the reactors suffered damage as the fuel overheated and melted. This damage at the F1-NPP led to the release of radionuclides into the atmosphere. The released high volatility fission products including $^{129\text{m}}\text{Te}$, ^{132}Te , ^{131}I , ^{134}Cs , ^{136}Cs , and ^{137}Cs were dispersed in the atmosphere, and subsequent wet and dry depositions led to their accumulation on the ground. As a result, large areas in the eastern part of Japan, from the Kanto to Tohoku districts, especially the Fukushima Prefecture, were contaminated with radionuclides.

Wild boar, *Sus scrofa*, is an omnivorous mammal inhabiting natural or semi-natural ecosystems. In such ecosystems, especially forest ecosystems, radiocaesium remains more available for uptake by plants and fungi as compared to agricultural areas due to differences in soil characteristics (IAEA,

2006). As a result, activity concentrations of radiocaesium in wild boars have been relatively high around Chernobyl and in some European countries contaminated by the Chernobyl Nuclear Power Plant accident (Gulakov, 2014; Kapala et al., 2015; Semizhon et al., 2009; Strebl and Tataruch, 2007; Vilic et al., 2005). Since people consume wild boar meat, it is a potential source of radioactive contamination for people, especially for critical populations such as hunters and their family members.

In Japan, radioactivity monitoring has been conducted for wild boar meat since the F1-NPP accident, and $^{134}\text{Cs}+^{137}\text{Cs}$ activity concentrations in some samples have exceeded the regulatory values (500 Bq kg⁻¹ fresh weight (FW) until 31 March 2012 and 100 Bq kg⁻¹ FW thereafter). Based on these results, distribution and/or consumption of wild boar meat is restricted in some regions such as Fukushima and surrounding prefectures. The monitoring data obtained from 2011 to 2012 were analysed to clarify regional and temporal characteristics of radiocaesium contamination of wild boar meat (Merz et al., 2015; Steinhauser and Saey, 2016; Tagami and Uchida, 2013), but the monitoring data obtained thereafter were not analysed. This study therefore aims to clarify regional differences and temporal changes in radiocaesium contamination of wild boar meat from 2011 to 2015. This study also aims to clarify radiocaesium distribution characteristics in organs of wild boars and to obtain radiocaesium reduction rates by cooking (parboiling process). Results of this study will contribute to evaluation of current and future radiation doses to humans due to intake of wild boar meat.

2. Materials and methods

2.1. Regional and temporal characteristics of radiocaesium contamination

Regional and temporal characteristics of radiocaesium contamination of wild boar meat were

analysed based on radioactivity monitoring data obtained by local governments and gathered by the Japanese Ministry of Health, Labour and Welfare (MHLW) under the post-Fukushima food monitoring campaign. We focus on Fukushima and surrounding prefectures Miyagi, Ibaraki, Tochigi and Gunma (Fig. 1), which were radioactively contaminated by the F1-NPP accident. On 2 March 2016, $^{134}\text{Cs} + ^{137}\text{Cs}$ activity concentration data on wild boar meat were isolated from the monthly and weekly reports of MHLW (2016) for each prefecture and year (from 2011 to 2015). When “lower than detection limit (DL)” was reported as a measurement of the $^{134}\text{Cs} + ^{137}\text{Cs}$ activity concentration, a value of half the DL was used for data analysis.

Significance of differences in the $^{134}\text{Cs} + ^{137}\text{Cs}$ activity concentrations between years was tested in each prefecture by one-way ANOVA and post-hoc Tukey-Kramer’s test. This test was performed using the StatView software, version 5.0 (SAS Institute Inc., Cary, NC, USA) at the significance level of 0.05. The log-transformed activity concentration data were used for this statistical test.

2.2. Radiocaesium distribution in organs

Six individuals of wild boars captured in Fukushima Prefecture in summer and autumn of 2011 were used to determine the comparative Cs activity concentrations between the different tissues sampled. Details of boar samples are shown in Table 1. Muscle, kidney, heart, liver, spleen and lung were separately dissected.

Each organ sample (33-102 g) was homogenised with scissors, and was transferred to a plastic bottle (U8 bottle, Yamayu Plastic Medical Products, Osaka, Japan). The preparation of this counting sample was carried out in duplicate for each organ. Activity concentrations of ^{137}Cs were measured by gamma spectrometry using a Ge semiconductor detector (GC2018, Canberra Industries Inc., Oak Ridge, TN, USA) with 20 % relative efficiency. The gamma-ray counting efficiency of this detector

was calibrated by constructing a relative gamma-ray counting efficiency curve using a certified volume standard source (MXO33U8PP, the Japan Radioisotope Association, Tokyo, Japan). A gamma-ray peak of 661.64 keV was analysed. The counting time was set at 1,800-3,600 s. Under these measurement conditions, lower detection limits for ^{137}Cs ranged from 4.6 to 22.1 Bq kg⁻¹ FW.

The relative ^{137}Cs activity concentration in each organ was calculated as the ratio of ^{137}Cs activity concentration in the organ to that in muscle of the same individual.

2.3. Radiocaesium removal by parboiling

Two individuals of wild boars were used for this experiment. One (boar sample A; female; weight: 40 kg) was captured in Iwaki City, Fukushima Prefecture, on 8 October 2011, and the other (boar sample B; male; weight: 60 kg) was captured in Tamura City, Fukushima Prefecture, on 24 August 2011.

Meat isolated from each wild boar was sliced in a thickness of approximately 3 mm. For boar samples A and B, 130 and 50 g of the sliced meat were boiled in 650 and 250 mL of tap water, respectively. Because this experiment mimicked parboiling process in cooking, the boiling time was 3 min. After boiling, the resulting hot water was discarded, and the boiled meat was used for determination of ^{137}Cs activity concentrations. For boar samples A and B, 130 and 50 g of the raw sliced meat were also used for radioactivity determination, respectively.

Boiled or raw meat samples were ashed using a method adopted by MEXT (1982). The resulting ash was transferred to U8 bottles containing 1 % (w/v) agarose powder in distilled water. The U8 bottles were heated in a microwave oven to dissolve the agarose and mixed well by swirling. They were placed on ice to promote gelation. The radioactivity of ^{137}Cs in these counting samples was measured by gamma spectrometry using Ge semiconductor detectors (GEM45-76-LB, Seiko EG &

G Co., Ltd., Tokyo, Japan) with 45 % relative efficiency. The gamma-ray counting efficiency of this detector was calibrated by constructing a relative gamma-ray counting efficiency curve using a certified volume standard source (MXO33U8PP, the Japan Radioisotope Association, Tokyo, Japan). A gamma-ray peak of 661.64 keV was analysed. The counting time was set at 3,600-10,000 s. Under these measurement conditions, lower detection limits for ^{137}Cs ranged from 3.6 to 13.7 Bq kg⁻¹ FW. There were three replicates for each boar sample and treatment (boiled or raw).

In accordance with definition by IAEA (1994), the food processing retention factor, F_r , was calculated by dividing the total radioactivity of ^{137}Cs in the boiled boar meat by that in the original raw meat.

3. Results and discussion

The F1-NPP accident occurred in March 2011, and soon local governments in Japan started radioactivity monitoring of food including wild boar meat. Figure 2, which was prepared from such monitoring data, shows activity concentrations of $^{134}\text{Cs}+^{137}\text{Cs}$ in meat of wild boar captured in Fukushima and surrounding prefectures (Miyagi, Ibaraki, Tochigi and Gunma) from 2011 to 2015, and Table S1 shows statistical data of the activity concentrations. Sample numbers in each prefecture vary widely between years. For example, in Miyagi Prefecture, sample numbers ranged from 8 to 102 in each year. Representativeness of the activity concentration data is therefore different between years, and the smaller sampling numbers (e.g. eight in 2011) cannot be considered as representative.

In each prefecture and year, the activity concentrations exhibited a considerable variability, i.e., three and two orders of magnitude in Fukushima and the other prefectures concerned, respectively. The causes of this variability could be due to (i) different contamination levels within each prefecture and (ii) a relatively great variety of food habits of wild boars (herbaceous plants, wild

fruit, mushrooms, acorns, chestnuts, beech trees, roots, snails, frogs, mice, earthworms, insects etc., whose radioactive contamination levels are expected to be different one another). Similar high variability of radiocaesium contamination levels have been observed in wild boar meat around Chernobyl and in some European countries contaminated by the Chernobyl accident, and similar mechanisms have been proposed (Gulakov, 2014; Kapala et al., 2015; Semizhon et al., 2009; Strebl and Tataruch, 2007; Vilic et al., 2005).

The $^{134}\text{Cs}+^{137}\text{Cs}$ activity concentrations of some boar samples in Fukushima prefecture have been higher than $10,000 \text{ Bq kg}^{-1} \text{ FW}$, achieving the highest concentration of $61,000 \text{ Bq kg}^{-1} \text{ FW}$ (^{134}Cs : $21,100 \text{ Bq kg}^{-1} \text{ FW}$; ^{137}Cs : $40,200 \text{ Bq kg}^{-1} \text{ FW}$) in 2013. This most severe contamination level is not as high as that in the Chernobyl exclusion zone, e.g., $661,000 \text{ Bq kg}^{-1} \text{ FW}$ of ^{137}Cs (Gulakov, 2014). The $^{134}\text{Cs}+^{137}\text{Cs}$ activity concentrations in many samples captured in Fukushima Prefecture have exceeded the regulatory values for food in Japan ($500 \text{ Bq kg}^{-1} \text{ FW}$ until 31 March 2012 and $100 \text{ Bq kg}^{-1} \text{ FW}$ thereafter), even in 2015, when four years have passed since the F1-NPP accident. The highest activity concentrations have been around $1,000 \text{ Bq kg}^{-1} \text{ FW}$ in the prefectures surrounding Fukushima, and the activity concentrations in some samples have exceeded the regulatory values for food in Japan. These monitoring results have been a basis for restriction of distribution and/or consumption of wild boar meat in these prefectures.

In Fukushima Prefecture, the $^{134}\text{Cs}+^{137}\text{Cs}$ activity concentrations showed a slow but statistically significant decrease trend through the whole period, though they fluctuated between years (Fig. 2). In Miyagi Prefecture, $^{134}\text{Cs}+^{137}\text{Cs}$ activity concentration data obtained in 2011 cannot be regarded as representative due to small sample numbers ($n=8$) as mentioned above. The activity concentrations decreased from 2012 to 2013. Thereafter, they remained constant, though decrease in the activity concentrations due to physical decay of ^{134}Cs (half life: 2.1 y) is expected. In Ibaraki Prefecture,

$^{134}\text{Cs}+^{137}\text{Cs}$ activity concentrations decreased from 2011 to 2013, and then remained constant. In Tochigi and Gunma Prefectures, the activity concentrations slowly decreased in a time-dependent manner. Reasons for these differences in temporal trends between prefectures are unclear. The temporal trends of radiocaesium concentrations observed in this study are similar to those observed around Chernobyl and in some European countries contaminated by the Chernobyl accident. In these countries, the ^{137}Cs contamination of wild boar is remaining relatively high and exhibits a much slower decrease or is either remaining fairly constant or slightly increasing on average even in the 2010s, i.e., 25 years after the accident (Gulakov, 2014; Kapała et al., 2015; Semizhon et al., 2009; Strebl and Tataruch, 2007). Especially, the highest ^{137}Cs activity concentrations were found 10 years after the accident and not earlier around Chernobyl (Gulakov, 2014). On the other hand, these temporal trends of radiocaesium levels in wild boar meat are different from those in other many food such as vegetables and beef, whose radiocaesium levels decreased quickly in Fukushima and affected prefectures in Japan, so that only a few samples exceeded the regulatory limits by summer and autumn 2011, respectively (Merz et al., 2015; Nihei et al., 2016). A similar difference in radiocaesium decrease was observed between wild foods and farmed foods after the Chernobyl accident (Beresford et al., 2016). Concern has been expressed that persistence of high radiocaesium activity concentrations in wild boar meat reduce hunting wild boars in Fukushima and surrounding prefectures, and thus damage to crops caused by wild boars will increase in the future (Ihara, 2013).

In this study, ^{137}Cs activity concentrations were determined in muscle, kidney, heart, liver, spleen and lung of wild boars captured in Fukushima Prefecture. The determined concentrations and the resulting relative ^{137}Cs activity concentrations are shown in Table 1. The relative concentrations in muscle were higher than those in the other organs examined in this study. This is the same result reported by Gulakov (2014) and Tanoi et al. (2016) for wild boars in Chernobyl and Fukushima

Prefecture, respectively. The same result was also observed in other mammals such as abandoned cattle and wild mice in Fukushima Prefecture (Fukuda et al., 2013; Kubota et al., 2015) and pigs fed rice contaminated by the F1-NPP accident (Ohmori et al., 2014). These consistencies give some validity to results of this study, in which sample numbers were limited ($n=1-6$). In addition to meat (muscle), heart and liver of wild boars are consumed by humans in Japan, and other organs potentially could be. The higher radiocaesium concentration in muscle than other organs, which was observed in this and previous other studies, suggests that monitoring of wild boar meat, which is commonly carried out, would guarantee the radiological safety of possible intake of other organs as well as that of meat itself.

In Japan, when wild boar meat with strong smell is cooked, parboiling is sometimes carried out to remove the smell. That is, after boar meat is boiled in water for several minutes, the resulting hot water is discarded and only the boiled meat is used for preparation of boar dish such as hot pot. In this study, sliced boar meat was boiled for 3 min, which mimicked this parboiling process, and a food processing retention factor, F_r , of 0.53 or 0.60 was obtained for ^{137}Cs (Table 2). This suggests that the parboiling process is effective for reduction of radiocaesium intake from wild boar meat. The F_r values obtained in this study are consistent with those (best estimation: 0.4; range: 0.2-0.7) summarised by IAEA (1994) for radiocaesium in boiling meat of mammals, suggesting some validity of this study, in which sample numbers were limited ($n=2$), though there might be differences in experimental methods between this study and studies which IAEA referred to – for example, in one of these studies, cubed beef was stewed for 1 h 30 min and the F_r of 0.47 was obtained (Meyer et al., 1962).

Conclusions

From 2011 to 2015, elevated $^{134}\text{Cs}+^{137}\text{Cs}$ activity concentrations in wild boar meat remained constant or decreased slowly in Fukushima and surrounding prefectures. The activity concentrations in some samples are still over the regulatory limit of $100 \text{ Bq kg}^{-1} \text{ FW}$. Activity concentrations of ^{137}Cs in muscle were higher than those in other organs. Almost half amount of ^{137}Cs was removed from wild boar meat when it was boiled, suggesting that a parboiling process is effective for reduction of radiocaesium intake from wild boar meat.

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Figure captions

Fig. 1. Prefectures concerned in this study.

Fig. 2. Activity concentrations of $^{134}\text{Cs}+^{137}\text{Cs}$ in wild boar meat from 2011 to 2015. Closed diamonds are geometric means. The other statistical data are shown in Table S1. Different letters (a, b and c) indicate statistically significant differences ($p < 0.05$, Tukey-Kramer's test). Dotted horizontal lines are regulatory limits for food in Japan (500 Bq kg^{-1} FW until 31 March 2012 and 100 Bq kg^{-1} FW thereafter).

Fig. 1

Fig. 2

Table 1 ¹³⁷Cs distribution in organs of wild boars captured in Fukushima Prefecture

Individual No.	Sampling place	Sampling date	Sex	Weight, kg	¹³⁷ Cs activity concentration ^a , Bq kg ⁻¹ FW						Relative ¹³⁷ Cs activity concentration ^b , %					
					Muscle	Kidney	Lung	Liver	Heart	Spleen	Muscle	Kidney	Lung	Liver	Heart	Spleen
1	Tamura City	5 June 2011	Female	75	136	-	-	67	-	-	100	-	-	49	-	-
2	Tamura City	24 August 2011	Male	60	217	-	114	163	-	-	100	-	53	75	-	-
3	Koriyama City	31 August 2011	Male	60	599	-	-	280	-	-	100	-	-	47	-	-
4	Koriyama City	8 September 2011	Male	50	312	-	-	204	-	-	100	-	-	65	-	-
5	Tenei Village	7 October 2011	Female	28	627	-	-	182	301	-	100	-	-	29	48	-
6	Ono Town	4 October 2011	Male	40	68	50	-	29	30	22	100	74	-	43	44	32
Mean±SD											100	74	53	51±16	46	32

^a Average value of two samples counted. Decay is corrected to the sampling date.

^b Percentage of ¹³⁷Cs activity concentration in each organ to that in muscle of the same individual

Table 2 Reduction of ¹³⁷Cs in wild boar meat by boiling

Boar sample	Meat before boiling		Meat after boiling		Food processing retention factor (Fr=(C × D/1000) / (A × B/1000))
	Fresh weight, g (A)	¹³⁷ Cs activity concentration ^a , Bq kg ⁻¹ FW (B)	Fresh weight, g (C)	¹³⁷ Cs activity concentration ^a , Bq kg ⁻¹ FW (D)	
A	130.3±0.3	383±66	95.2±1.9	280±52	0.53±0.11
B	49.9±0.2	152±13	35.0±1.6	130±14	0.60±0.05

Mean ± standard deviation (n=3)

^a Decay is corrected to the sampling date.