A survey on the concentration of the radioactive cesium contained in Japanese milk samples (2021)

-Investigating the origin of the contamination and assessing the risks of cases where the radioactive cesium level is below the Standard Limits for Radionuclides in Foods

（March 2022）

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Food products were contaminated with radioactive materials due to the Fukushima Daiichi Nuclear Power Station accident in March 2011. The government responded by setting standard limits of the radioactive cesium contained in food products on April 2012 so that annual radiation exposure to food products would be below 1mSv. For the time being, although almost no food products which are above the standard limits are in circulation, some consumers, in pursuit of safer food, make choices regarding the area where the food was produced.

In this research, we analyzed the small amount of radioactive cesium contained in milk samples from all over Japan, using the AMP method. Radioactive cesium that was below the standard limits, was detected in all commercial products, including the ones made in western Japan. On the basis of the results of the analysis, comparing health risks of radiation exposure due to consuming food below the standard limits and health risks of carcinogenic materials contained in substances such as tap water, we raised the question: Can choosing food by its area of production be considered 'reputational damage'?

1. Introduction

Radioactive contamination of food due to the Fukushima Daiichi Nuclear Power Station accident

In March 2011, the Fukushima Daiichi Nuclear Power Station accident occurred and as a result, the environment and food were contaminated by radioactive materials. At that time, no standard limit of radiation levels of food circulating in Japan existed, and therefore the government set a temporary standard. In April 2012, about one year after the accident, new standard limits were established, in which radiation exposure due to consuming food was limited to 1mSv per year so the standard value of radioactive cesium is 100Bq/kg for common food, 50Bq/kg for milk and baby/infant food and 10Bq/kg for drinking water. This was supposed to regulate contaminated food, however, there were many reports of food that was over the standard limit being found in circulation. In the fiscal year of 2020, twenty-two cases of food over the standard limit were recognized.

Due to inadequate food contamination checks and circulation management systems and refusing to be unnecessarily exposed to radiation, some people decided to select food according to the area in which it was produced in order to avoid risks of radiation exposure. The Japanese government terms this action- avoiding the food produced in the affected area of the Great East Japan Earthquake- as 'reputational damage,' something which must be eliminated if reconstruction is to proceed.

Risks of radiation and risks of chemical materials

However, is making a choice of avoiding a health risk an unfair act that prevents the reconstruction of the affected areas?

The cancer risk of chemicals in tap water and other substances is regulated to a level of 1 in 100,000 per substance, which is usually the risk for a lifetime of ingestion of such substances. But those who don't want

to take the risk of developing cancer are able to buy organic products at some additional cost. This consumer action is not criticized by the government as 'reputational damage.'

The standard for radiation exposure from consuming food is set at 1mSv/year. According to the ICRP (International Commission on Radiological Protection) the fatality risk is about 5% per 1Sv and 'the standard is based on the hypothesis that the probability of radiation-induced cancer or hereditary effects increases in direct proportion to the increase in dose.' In addition, it estimates that the fatality risk is 0.4% if a person continues to be exposed to radiation at 1 mSv per year throughout his/her lifetime.

Adding five hundred-thousandths per year means that assuming a person's life is 80 years, four hundred people's deaths are added per hundred thousand people. This risk is two digits higher than that of chemical materials.

It is sometimes claimed that as the risk of radiation exposure is a total amount of the risks of various radionuclides, comparing regulation of one chemical material to regulation of radiation exposure is improper, because there are thousands of chemical materials that we may come in contact with in daily life. However, the radiation exposure standard of 1mSv per year is not the 'total' amount of exposure but 'additional' exposure. The radiation dose limit for the public from nuclear facilities under pre-accident conditions, the new standard for radioactive cesium in food, the standard of 8,000 Bq/kg for "designated waste" introduced to handle the large amount of radioactive waste generated by the Fukushima nuclear accident (exposure of workers disposing of the waste), and the standard for the disposal of radioactively contaminated water, which has been the focus of attention at the Fukushima nuclear power plant now undergoing decommissioning, are all conveniently used to explain that exposure of 1 mSv or less is safe. The fact that these exposure risks add up as well as the effects of this have not been explained to civil society by the regulators, and of course have not been discussed.

In addition, the risk assessment of carcinogenic chemical materials is based on 'causing cancer', but the risk assessment of radiation exposure is based on 'deaths from cancer'; it is impossible to compare the risk of the two. ICRP estimates that the risk of 'developing cancer' is twice as high as that of 'death from cancer'.

Also, the user decides whether or not to use chemical materials, weighing the advantages and disadvantages of its use, but in the case of exposure due to nuclear power plant accidents, there are no direct advantages to anyone.

Research and objective

We would like to support people's right of choice to avoid the risk of radiation exposure, by measuring even low levels of radioactive cesium contained in milk and disclosing its areas of production and levels of contamination.

Last fiscal year's research revealed an unanticipated fact regarding the milk produced in regions other than Fukushima. It is thought that many people avoided milk produced in Tohoku district and chose milk produced in Hokkaido, but it was revealed that the milk produced in Hokkaido is also contaminated by the Fukushima accident. In the same way, there were many people who selected the food produced in western Japan, but is this food really contamination-free? Are there any effects of radioactive contamination caused by nuclear bomb tests or the atomic bombing of Hiroshima and Nagasaki? In FY2021, the survey was expanded to include western Japan products in order to compare contamination on a national scale.

2. Method of analysis

Each measurement sample was 22kg of commercial milk which had an identifiable production location. In FY2021, the milk produced in 11 areas was included in the survey: Iwate (K), Miyagi (L), Ibaraki (M), Tokyo (N), Shizuoka (O), Ehime and Kochi (P), Miyazaki and Kagoshima (Q), Nagasaki (R), Oita (S), Shimane (T), and Ishikawa (U). (See Fig. 1. Areas A to J indicate areas surveyed in FY2020).

In measuring the concentration of radioactive cesium, 2kg of the sample was used as a direct measurement sample and the rest (20kg) was used as a concentrated measurement sample. In order to detect small amounts of radioactive cesium, we performed a concentration process, the method is described below.

After 5kg of milk was heated to 80℃, 350 ml solution of acetate (grain vinegar commercially available) was poured into it, causing an acid coagulation reaction of protein. This separated the milk into white sediment and slightly yellow liquid (whey). About 90% of cesium in milk exists in this whey. The mixture of white sediment and whey was stirred lightly and left to stand for 15 minutes, then it was filtered through non-woven cloth. This procedure was performed on 5kg of milk and repeated four times for a total of one 20kg sample. 18kg of whey was obtained from 20kg milk (including the acetate solution). The whey was left to stand for a night, and was filtered again to remove a little amount of white sediment. Then, concentrated hydrochloric acid was added to make its ion exponent pH2. After that 10.00g ammonium phosphomolybdate (AMP) per 20kg milk was added. After stirring it for 30 minutes, it was left to stand for 20 hours. Then the AMP was collected using suction filtration (ADVANTEC TOYO, No.5B). The collected AMP was dried naturally for 3 days and then put into a U8 container (screw-top type U container 100mL, φ55mm×H67mm).

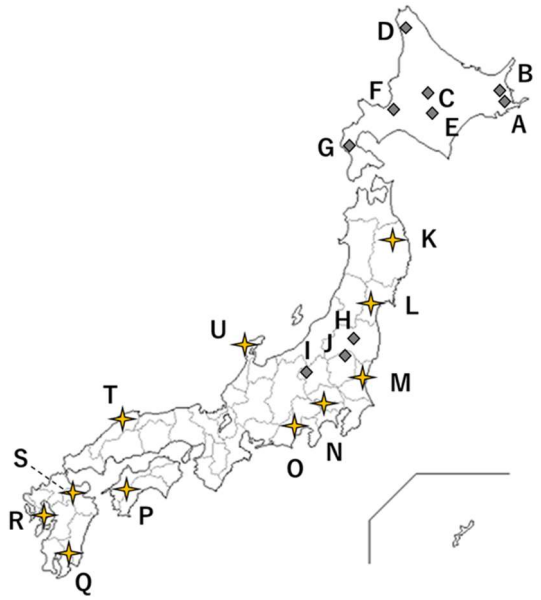

Figure 1 Production area of milk samples

The germanium semiconductor detector (BSI Co. GCD70-200) was used for gamma ray detection in this radioactivity measurement. 2kg of both the milk for the direct measurement and the white sediment obtained through the acid coagulation reaction were each put into 2 liter Marinelli containers and they underwent a gamma-ray measurement for 20-40 hours. AMP obtained through the concentration processing, which chemisorbed cesium, was measured in the U8 container for 96 hours. The concentration of radioactive cesium in the milk state was calculated in proportion to the AMP collection rate. The detection limit of the milk equivalent was 20-30mBq/kg in the direct measurement, and 0.4mBq/kg in the concentrated measurement.

3. Results

The results of the measurement are shown in Table 1. Of K-U measured in this fiscal year, the production areas in which cesium 137 was detected in the direct measurement were Iwate (K) and Miyagi (L). No cesium 134 was detected in any areas in the direct measurement. In the concentrated measurement, cesium 137 was detected in all areas including the Kyushu district. The most contaminated area was Miyagi (L) with 152mBq/kg. This figure was higher than

135mBq/kg of Fukushima (H). The second most contaminated was the milk produced in Iwate (K), 79mBq/kg. They were followed by Shizuoka (O)(16mBq/kg), Ibaraki (M) (11mBq/kg), Tokyo (N) (7.3mBq/kg), Miyazaki and Kagoshima (Q)(7.0Bq/kg), Oita (S) (5.7mBq/kg), Shimane (T)(5.4mBq/kg), Nagasaki (R) (5.2mBq), Ehime and Kochi (P) (5.1mBq.kg) and Ishikawa (U) (3.9mBq/kg). Cesium 134 was detected only in Miyagi (L) (4.4mBq.kg) and Iwate (K) (2.0mBq/kg).

We note that about 90% of cesium in milk exists in whey, and therefore adopting this cesium concentrated method indicates 10% lower figures in the condensation measurement than in the direct measurement. Comparing the samples in which cesium 137 was detected both in the direct measurement and in the concentrated measurement, Iwate's result for the direct measurement was 83mBq/kg and for the concentrated measurement, 79mBq/kg. Miyagi's result for the direct measurement was 181mBq/kg and for the concentrated measurement, 152mBq/kg. The ratios were 0.95 and 0.84 respectively.

4. Consideration

Origin of cesium 137

At the time the Fukushima Daiichi Nuclear Power Station accident occurred, cesium 134 and cesium 137 were emitted into the environment at the ratio of about 1:1. The half-life of cesium 137 is about 30 years, and that of cesium 134 is about 2.1 years. By taking into consideration each cesium's half-life and the length of time from the disaster to the measurement, it is possible to calculate a ratio (cesium 134/cesium 137) of cesium that was emitted as a result of the accident at the time of the measurement. Five years after the accident (March in 2016), the cesium ratio became 0.21, Ten years afterwards (March in 2021), it decreased to 0.046.

The cesium ratio differs slightly according to each reactor. Therefore the cesium ratios in the fallout in each place due to the accident are different. The initial cesium

Table 1 Results of radioactive cesium concentration in milk

Year of measurement	Symbol	Production location	Direct measurement Concentration (+1σ) / mBq kg^{-1}		Concentrated measurement Concentration (+1σ) / mBq kg^{-1}		Date of measurement
			Cs137	Cs134	Cs137	Cs134	
2020	A	Nemuro, Hokkaido	69 ± 7.0	< 17	69 ± 0.7	0.7 ± 0.1	September 14, 2020
	B	Nemuro, Hokkaido	49 ± 10	< 28	66 ± 0.7	< 0.3	November 6, 2020
	C	Kamikawa, Hokkaido	< 27	< 27	15 ± 0.5	< 0.2	November 30, 2020
	D	Soya, Hokkaido	75 ± 11	< 27	57 ± 0.6	< 0.4	December 4, 2020
	E	Tokachi, Hokkaido	< 29	< 27	18 ± 0.4	< 0.4	December 15, 2020
	F	Ishikari, Hokkaido	38 ± 10	< 27	28 ± 0.3	< 0.3	December 25, 2020
	G	Oshima, Hokkaido	57 ± 10	< 27	52 ± 0.5	< 0.3	December 31, 2020
	H	Fukushima	147 ± 12	< 28	135 ± 1.3	5.2 ± 0.2	December 22, 2020
	I	Gunma	102 ± 11	< 28	85 ± 0.8	3.0 ± 0.2	February 22, 2021
	J	Tochigi	62 ± 11	< 28	60 ± 0.6	2.4 ± 0.2	February 9, 2021
2021	K	Iwate	83 ± 11	< 27	79 ± 0.8	2.0 ± 0.1	November 19, 2021
	L	Miyagi	181 ± 13	< 27	152 ± 1.5	4.4 ± 0.2	October 29, 2021
	M	Ibaraki	< 29	< 27	11 ± 0.2	< 0.4	September 10, 2021
	N	Tokyo	< 26	< 25	7.3 ± 0.2	< 0.4	July 16, 2021
	O	Shizuoka	< 29	< 27	16 ± 0.3	< 0.4	July 30, 2021
	P	Ehime・ Kochi	< 27	< 28	5.1 ± 0.2	< 0.4	September 17, 2021
	Q	Miyazaki・Kagoshima	< 26	< 25	7.0 ± 0.2	< 0.4	September 24, 2021
	R	Nagasaki	< 26	< 25	5.2 ± 0.2	< 0.4	August 13, 2021
	S	Oita	< 25	< 25	5.7 ± 0.2	< 0.4	August 21, 2021
	T	Shimane	< 28	< 27	5.4 ± 0.2	< 0.4	April 27, 2021
	U	Ishikawa	< 29	< 27	3.9 ± 0.2	< 0.4	August 28, 2021

Table 2 The rate of cesium 137's contribution to the Fukushima nuclear disaster in the samples in which cesium 134 was detected

Area of production	Cs137(Whole) Concentration (±1σ) mBq kg^{-1}	Cs134 Concentration (±1σ) mBq kg^{-1}	Date of measurement	Elapsed years from the disaster to the measurement	Atmospheric fallout (March to May, 2011) (MBq/km^2・For three months) Cs-137	Cs-134	Cs134/Cs137 Ratio Time of the disaster	Time of the measurement	Cs137 (Derived from the disaster) Concentration (±1σ) mBq kg^{-1}	Contributing rate (±1σ)
【A】 Hokkaido	69 ± 0.7	0.7 ± 0.1	September 14, 2020	9.5	8.0×10^0	8.4×10^0	1.05	0.053	13 ± 1.3	0.19 ± 0.02
【H】 Fukushima	135 ± 1.3	5.2 ± 0.2	December 22, 2020	9.8	3.5×10^6	3.3×10^6	0.94	0.044	116 ± 4.6	0.86 ± 0.04
【I】 Gunma	85 ± 0.8	3.0 ± 0.2	February 22, 2021	9.9	5.2×10^3	5.2×10^3	1.00	0.045	66 ± 4.0	0.78 ± 0.05
【J】 Tochigi	60 ± 0.6	2.4 ± 0.2	February 9, 2021	9.9	7.2×10^3	7.3×10^3	1.01	0.045	53 ± 3.7	0.89 ± 0.06
【K】 Iwate	79 ± 0.8	2.0 ± 0.1	November 19, 2021	10.7	1.5×10^3	1.5×10^3	1.00	0.035	56 ± 3.9	0.71 ± 0.05
【L】 Miyagi	152 ± 1.5	4.4 ± 0.2	October 29, 2021	10.6	2.2×10^4	2.2×10^4	1.00	0.036	121 ± 4.8	0.79 ± 0.03

The figures of each municipality's radiation measurement research were used in this calculation. Because there were no such data in Miyagi, data of the environmental radiation monitoring such as the nuclear power plant were used. (There are cases in which figures don't match due to round-off.).

ratios in the atmospheric fallouts in each area from March to May in 2011 were calculated using data from the environmental radiation database (https://www.kankyo-hoshano.go.jp/data/database/).

The proportions of cesium-137 (derived from the Fukushima nuclear reactor / total in the milk sample) were derived by calculating the measured value of cesium 134 concentration and calculated cesium 134/137 ratio at the time of measurement.

In FY2020's survey, the production areas where cesium 134 was detected were Hokkaido, Fukushima, Gunma, Tochigi, Iwate and Miyagi. The cesium 134/137 ratios in the atmospheric fallouts soon after the Fukushima nuclear disaster were as follows: Hokkaido 1.05, Fukushima 0.94, Gunma 1.00, Tochigi 1.01, Iwate 1.00 and Miyagi 1.00. The cesium 134/137 ratios (at the time of the measurement) were calculated by using the figures of each cesium's half-life and the number of years from the disaster to when the measurements were taken. The following figures were obtained: Hokkaido 0.053, Fukushima 0.044, Gunma 0.045, Tochigi 0.045, Iwate 0.035 and Miyagi 0.036. Dividing the cesium 134 concentration of the measurement by the cesium 134/137 ratio (at the time of the measurement) of the sample production area, the results of cesium 137 concentrations that derived from the Fukushima nuclear disaster were obtained and are shown in Table 2.

The following is one calculation example of Iwate. The cesium 134/137 ratio measured on November 19 in 2021 which was derived from the Fukushima nuclear disaster was calculated as 0.035 in Iwate. As the result of the measurement, 2.0± 0.1mBq/kg of cesium 134 was detected and therefore cesium 137 derived from Fukushima reactor should be 56±3.9mBq/kg on the basis of the cesium 134/137 ratio. However, the measurement result of the concentration of cesium 137 in the sample was 79±0.8mBq/kg. The reason it is higher than expected is because it contained cesium 137 that traces back to nuclear weapon tests and so on. Thus, out of the total cesium 137 contained in the milk produced in Iwate, it was concluded that the ratio of 0.71±0.05 was derived from the Fukushima nuclear accident.

The origins and concentrations of cesium 137 in the samples were compared based on the production areas (Figure 2). In western Japan (P-U), which is supposed to be hardly influenced by the Fukushima nuclear disaster, the concentrations of cesium 137 were below 7mBq/kg; this was thought to be derived from various nuclear weapon tests and the Chernobyl disaster.

By contrast, in eastern Japan (except Hokkaido) H-O, the cesium concentrations derived from nuclear weapon tests and the Chernobyl accident were 7-19mBq/kg, and in Hokkaido the cesium concentration tends to be higher

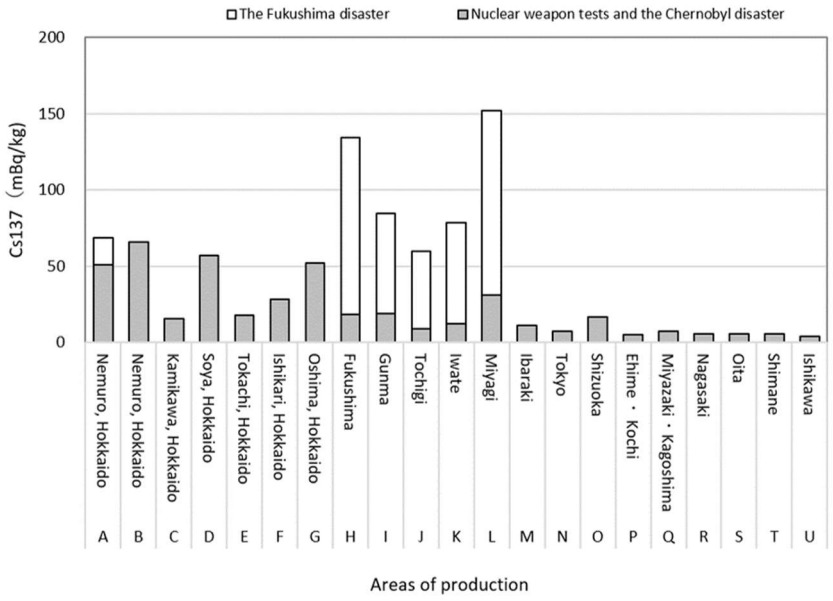

Figure 2 The origins and concentrations of cesium 137 in milk samples

(15-66mBq/kg) than those in eastern Japan. We would like to consider this reason.

Domestic radioactive contamination by the atmospheric nuclear weapons testing

Radioactivity measurements of atmospheric fallouts have continued throughout Japan since the days of nuclear weapons testing. Using continuous data of cesium 137 deposition since 1963, the regional trends in radioactive contamination were considered. The monthly depositions of cesium 137 fallout were added up for periods of five years and we considered periods 1 (1963-1967) to 9 (2003-2007) as the ones before the Fukushima nuclear disaster (the upper part of Figure. 3).

The amount of cesium 137 deposition in Japan is largest in period 1 during the years from 1963 to 2007, and it accounts for 74-91% of the total amount to 2007. The amount of accumulated cesium 137 gradually decreased from period 1 to period 4, but in period 5 (1983-1987), it increased. That increase is thought to be due to the Chernobyl nuclear accident. After period 6 (after 1988), there are no significant decreases of cesium 137.

The accumulated value of the amount of cesium 137 fallouts is the highest in Akita; the lowest is in Wakayama. The accumulated amount in Akita is four times larger than that in Wakayama. In order of the amount of cesium deposited, Akita is followed by Fukui, Ishikawa, Miyagi, Tokyo, and Tottori. Overall, the degree of contamination is high in the Sea of Japan side and low in western Japan (Wakayama, Hyogo and Okayama)

Focusing only on the period 5 (1983-1987: the time of the Chernobyl nuclear accident), the amount of cesium 137 is notably large in Fukui, Aichi, Akita and Ishikawa, in descending order. Except for Aichi, these three prefectures are included in the top 4 during the era of atmospheric nuclear weapons testing. There seem to be some geographic reasons why microparticles conveyed from abroad tend to descend in these areas.

Considering its decay rate, accumulated values of each period's cesium concentration after attenuation (as of

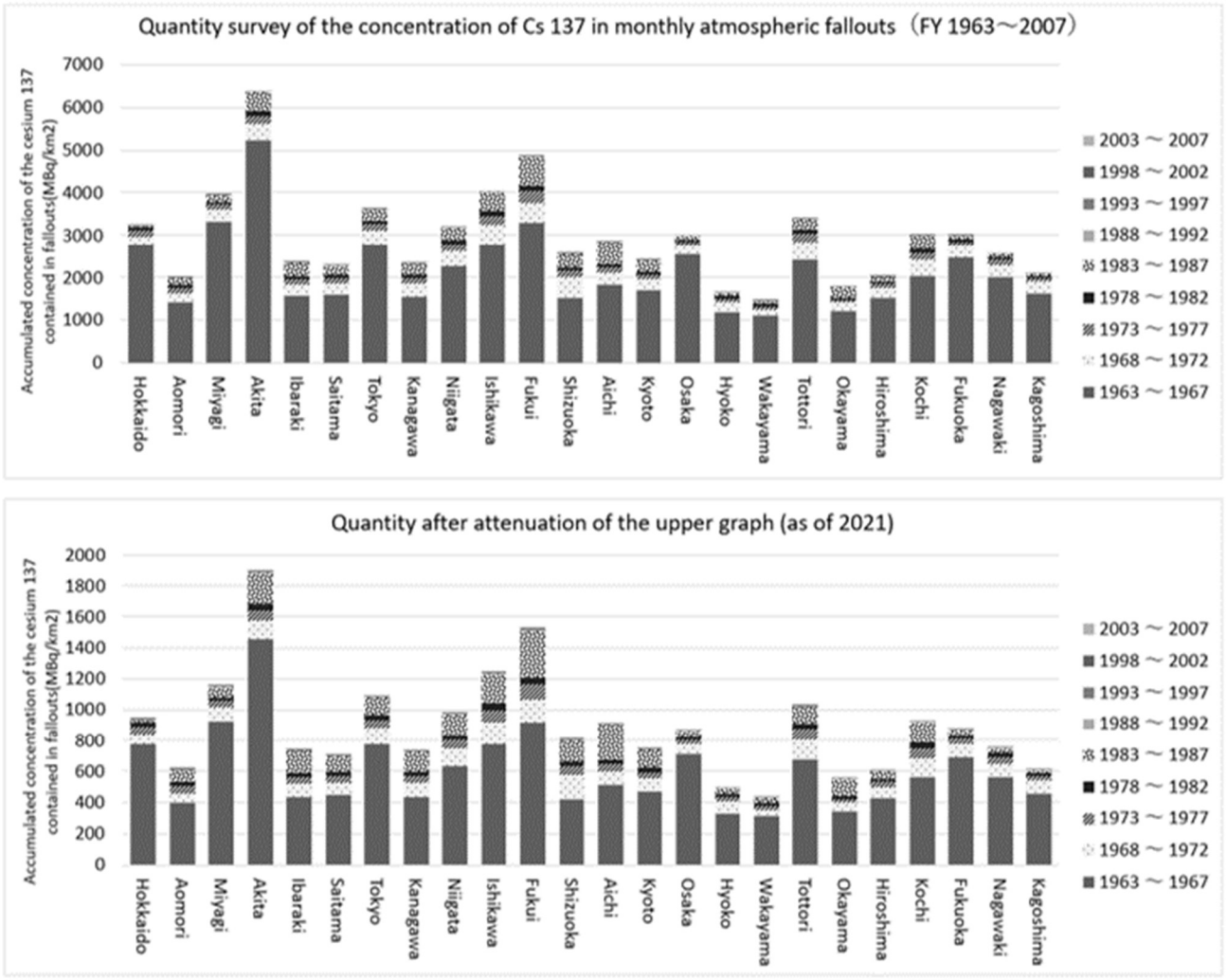

Fig. 3 Quantity survey of cesium 137 and the concentration of cesium 137 after attenuation contained in atmospheric fallouts in each area.

Based on environmental radiation database. Because the detection limit is unclear, 'not detected' is excluded from the calculation.

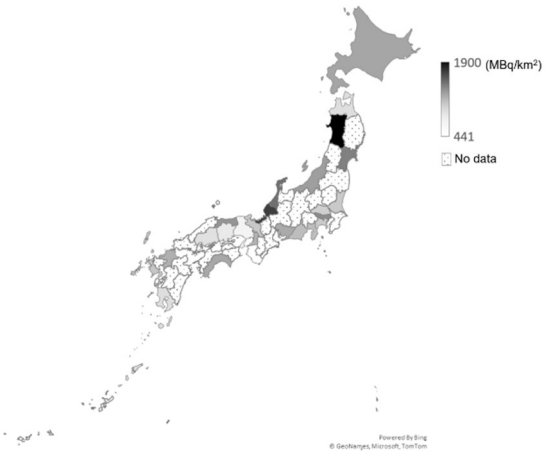

Figure 4 Evaluation of cesium 137 fallout in each area (after attenuation)

2021) of areas were calculated and shown in the lower part of Figure 3. In order to help comprehend geographically, the same information is shown in a map (Figure 4). About 60 years have passed since the era of the atmospheric nuclear weapons tests, but the effects of these tests, which caused an extraordinarily great amount of cesium fallout, are still significant. The amount of cesium 137 which derived from nuclear weapons testing has declined to only 1/4 of those days, and this contamination still accounts for a large part of the total contamination.

As stated above, we organized environmental radioactive contamination before the Fukushima

nuclear disaster on a regional basis, but the level of contamination is not especially high only in Hokkaido. We are not able to explain the reason why the cesium contamination in the milk produced in Hokkaido was high. And there wasn't any relationship between the surveyed cesium concentration from the atmospheric fallouts from 1967 to 2007 and the concentration of cesium137 in milk samples obtained in this survey.

Consideration of health risk

In an environmental contamination situation where the cesium concentration of milk is 50Bq/kg, assuming that radiation exposure from all food is 1mSv per year, what is the additional risk of cancer death caused by radiation exposure through food based on concentration of Cs137 in milk measured in this study?

The concentration of cesium in milk in this survey was 4-150mBq/kg, and when milk is at the contamination level of 150mBq/kg, this is equivalent to 0.003mSv per year. Under these conditions the lifetime risk of dying from cancer increases by 1.2 people per 100,000.

As said before, in general, carcinogenic chemical materials are regulated so that their concentration causes one person per 100,000 to develop cancer in his/her lifetime. If people are to face an equivalent risk of cancer death from consuming radioactively contaminated food, the detection limit must be 0.1Bq/kg (100mBq/kg). A sufficient number of detections are also required to support the choice of citizens who would like to avoid exposure.

5. Summary

The Fukushima Daiichi nuclear accident has caused serious environmental radiation contamination to Fukushima and surrounding areas. Since then, some people have selected food and purchased food that is produced in western Japan and Hokkaido in preference to the food produced in the affected area, in order to avoid radiation exposure through food. However, we need to evaluate in totality the influence of atmospheric nuclear weapons tests in addition to the influence of the Fukushima Daiichi nuclear accident in making a rational choice to avoid radiation exposure.

The concentrations of radioactive cesium in milk which was produced in specific areas across Japan were measured using the AMP method and the germanium semiconductor detector. This procedure made it possible to compare contamination in each area.

In all measurements, figures were considerably lower than the new standard limit of radioactive cesium contained in food (50Bq/kg), but the commercially available milk produced in Miyagi measured in 2021 was more highly contaminated than that produced in Fukushima measured in 2020, which suggests that the products of Fukushima are not necessarily the most contaminated. The milk produced in Hokkaido tended to contain more cesium than that produced in western Japan.

We were not able to find any relationship between the concentration of cesium in the atmospheric fallouts since 1963 and the concentration of cesium in milk surveyed this time. At least, the main cause of the environment radiation in all areas except Tohoku and North Kanto districts is the nuclear weapons tests, which were conducted more than 60 years ago.

The risk of dying from cancer caused through food intake 10 years after the Fukushima Daiichi nuclear accident in the contaminated area was calculated based on the measurement values obtained this time, and it was at the same level of the management standards of carcinogenic chemical substances.

Every person's sense of value, what he/she thinks is the highest priority and what risk he/she wants to avoid, should be respected. The option of avoiding the risk of exposure to radiation should be thought to be as important as the option of avoiding the risk of chemical substances and a mechanism must be established to

allow this.

Western Japan, which was not so much affected by the Fukushima nuclear disaster, has also been contaminated by radiation from a historical angle; the harsh fact is that the past contamination of the atmospheric nuclear weapons tests is still contained in food. We must become more aware that the mistake which the current generation has made by causing serious environmental radiation contamination from the Fukushima Daiichi nuclear accident will continue to affect generations in the future.