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Author(s)	Sato, Osamu
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[FUKUSHIMA RADIATION SYMPOSIUM 2013]

ESTIMATION OF DIETARY INTAKE OF RADIOACTIVE  
MATERIALS BY DUPLICATE DIET METHOD

OSAMU SATO

*Co-op Fukushima and Faculty of Human Development and Culture, Fukushima University*

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**Abstract :** The radioactive materials released in the accident at the Fukushima Dai'ichi Nuclear Power Plant have forced many residents in Fukushima Prefecture, including members of Co-op Fukushima, currently living as they do in an environment exposed to radiation, to live in the shadow of anxiety that they may be subject to internal radiation exposure. A dietary survey was therefore conducted using the duplicate diet method on 300 participating Co-op member families over three periods for the purpose of investigating the intake of radioactive materials through everyday meals and thereby providing accurate information about existing conditions of internal radiation exposure. Neither  $^{134}\text{Cs}$  nor  $^{137}\text{Cs}$  was detected in excess of the lower detection limit of 1 Bq/kg in the meals of 283 of the 300 families (i.e., 94%). In the meals of 17 families,  $^{134}\text{Cs}$  or  $^{137}\text{Cs}$  was detected in excess of 1 Bq/kg. Of these, the greatest internal exposure dose for both of the radionuclides was observed in the period "I winter" (November 2011 thru April 2012) of the three-period survey. This coincided with the implementation period of the Provisional Regulation Values, which set a maximum annual exposure dose of 5 mSv. On the supposition that the family in question had continued to eat the sample for a whole year, the resulting annual exposure dose would have amounted to 0.135 mSv. This is far below the current annual radiation dose standard of 1 mSv. Accordingly, we conclude that a sufficient level of dietary safety has been attained and that there are no grounds for concern that the consumption of foodstuffs currently on the market will have any adverse effects on health as a result of internal radiation exposure.

## 1. INTRODUCTION

The accident that struck the Fukushima Dai'ichi Nuclear Power Plant (F-1 NPP) on March 15, 2011 resulted in the release of a large quantity of radioactive materials, which contaminated a wide area of Fukushima Prefecture. As far as external exposure is concerned, monitoring with dosimeters from a relatively early stage after the accident has enabled estimates to be made of accumulated exposure doses. Evacuation has helped avoid excessive exposure and decontamination operations have contributed to the reduction of radiation levels. On the other hand, it has not been possible adequately to determine the status of internal exposure. Reasons for this include technological difficulties with assessment methods, including measurement with whole body counters, and testing systems incapable of keeping up with the screening needs of the resi-

dents. Given such inadequacies, concern and anxiety have been mounting among homemakers, responsible as they are for preparing daily meals, and particularly among parents with young children. Their main source of concern is the radioactive materials contained in the food they purchase and any internal exposure that may arise from the ingestion of this through their meals. A survey conducted on the residents of "City D" in the northern part of the prefecture in December 2011 (2,700 respondents) revealed the following. In response to the question (with multiple answers) "What particular countermeasures do you want the city to take to protect the citizens from radioactivity?" 46.1% gave the answer "Surveys on internal exposure" and 47.3%, the answer "Radioactivity testing of foodstuffs." In other words, the results revealed a significant level of demand for countermeasures to address internal exposure. Incidentally, the

response rate for “Surveys on external exposure” was 14.7%.

Co-op Fukushima members also began to voice their concerns and anxiety concerning the safety of foodstuffs possibly contaminated by radioactivity. Accordingly, Co-op Fukushima decided to measure its members’ intake of radioactive materials through the meals they were consuming by using the duplicate diet method to analyze samples of meals actually consumed at home. These were to be voluntarily provided by Co-op members resident in Fukushima Prefecture. The results were then to be made widely available so as to give information as to the status of existing exposure, thereby providing objective criteria that would allow people to organize their diets with less anxiety.

## 2. PARTICIPANTS AND METHODS

### 2.1. Participant families and areas surveyed

Participants of the survey were recruited from among Co-op Fukushima members when they attended study sessions, seminars and the like on radioactive materials and their health effects. The survey was conducted in three stages: November 2011 through April 2012 (I winter), May 2012 through September 2012 (II summer) and October 2012 through February 2013 (III winter). As can be seen from Table 1, for each of the three survey periods, participants comprised 100 families (Co-op Fukushima members) resident in six administrative districts of Fukushima Prefecture. The participating families were located outside the 20 km evacuation zone surrounding F-1 NPP and the “emergency-evacuation-preparation” and “deliberate-evacuation” zones between 20 km and 30 km around F-1 NPP (during the Winter period between

November 2011 and April 2012). Levels of contamination by radioactive materials in the six districts are estimated at: 10–30 kBq/m<sup>2</sup> in Aizu and 30–300 kBq/m<sup>2</sup> in the remaining districts (as assessed by the fourth aerial monitoring survey published by the Ministry of Education, Culture, Sports, Science and Technology (MEXT) on December 16, 2011). In all the survey periods, families from Ken-poku and Ken-chu accounted for approximately 80% of the survey participants. In these districts, radiation doses were particularly high, compared with other locations in the prefecture. Survey results can be seen as reflecting conditions in districts of Fukushima where levels of radiation were relatively high.

### 2.2. Sample collection

Participant families were asked to prepare one extra set of dishes identical to those eaten by family members at each meal, and these meals were collected as samples to be assessed under the duplicate diet method. Each family provided two days’ worth of meals, i.e. six meals, as well as drinks and snacks, all of which they put into plastic bags and sent by home delivery service to the inspection centre.

### 2.3. Survey forms

Survey forms took the form of a questionnaire survey comprising (1) a questionnaire about the details of each meal: the type of meal (i.e. lunch, breakfast, dinner or snack), the menu, the quantity, ingredients, and area of production for each ingredient) and (2) a questionnaire requiring answers about participants’ knowledge of radioactive potassium-40 and naturally existing radionuclides (i.e. choice of (1) ACQUAINTED WITH and (2) UNFAMILIAR WITH), in addition to their levels of awareness of where the ingredients were produced (i.e. choice of four possibilities ranging from (1) ALWAYS to (4) NEVER).

### 2.4. Measurement and radionuclides measured

Measurements of radionuclide concentration were conducted at the inspection centres of JCCU, Tokai Co-op, Co-op Kobe, Kyushu Co-op, Co-op Net and You Co-op. At these centres, the collected sets of six meals, beverages and snacks were all homogenized, of which a 1 kg sample was transferred to a Marinelli beaker for measurement. Each sample was measured for 50,000 seconds with a germanium detector in such a way as to obtain a detection limit of no more than 1 Bq/kg. The radionuclides measured included <sup>134</sup>Cs,

Table 1. Distribution of Participants (Families) in Fukushima Prefecture

Area	Period	I winter	II summer	III winter
Ken-poku		42	37	44
Ken-chu		38	40	38
Ken-nan		5	5	5
Aizu		10	10	8
Iwaki		3	3	5
Sohso		2	5	0

\*The total number of each period is 100

Period for Investigation

I winter: November 2011 thru April 2012

II summer: May 2012 thru September 2012

III winter: October 2012 thru February 2013

$^{137}\text{Cs}$ ,  $^{131}\text{I}$  and  $^{40}\text{K}$ .

### 3. RESULTS AND DISCUSSION

#### 3.1. Attitude of the participants

##### (1) Knowledge of radiation and radioactive materials

Participants' knowledge of radioactive potassium-40 ( $^{40}\text{K}$ ) and naturally existing radionuclides was assessed with the two questionnaire options "ACQUAINTED WITH" and "UNFAMILIAR WITH." Participants' level of knowledge of  $^{40}\text{K}$  is shown in table 2. Participants replying "ACQUAINTED WITH" in the survey I winter, approximately one year after the F-1 NPP accident, amounted to 79%. This figure rose to approximately 90% between II summer and the end of III winter.

The questionnaire item regarding naturally existing radionuclides was added from III winter. Shown in table 3, the results reveal that almost all of the respondents were "ACQUAINTED WITH" these.

The level of knowledge concerning radioactive materials and radiation was seen to be high. However, it remains unclear whether or not respondents had an understanding or awareness of the fact that  $^{40}\text{K}$  is a radionuclide present in the foodstuffs they had regularly been eating even before the accident and that it had been emitting radiation responsible for internal exposure. Ever since the days shortly after the accident, when very little was known about the degree of exposure, one has continued to hear in circulation comments to the effect that internal exposure is even more to be feared than external exposure. Indeed, as pointed out in the Introduction, anxiety toward internal exposure

Table 2. Knowledge on  $^{40}\text{K}$   $n=100$

Period Answer	I winter	II summer	III winter
Acquainted	79	88	89
Unfamiliar	21	10	10
No answer	0	2	1

Table 3. Knowledge on Natural radiation  $n=100$

Period Answer	I winter	II summer	III winter
Acquainted	—	—	97
Unfamiliar	—	—	0
No answer	—	—	3

remains at a high level. Now that internal and external radiation doses can actually be measured on an individual basis, knowledge and awareness about  $^{40}\text{K}$  and naturally existing radionuclides can constitute effective criteria in assessing the extent of exposure doses. We are faced with the task of ascertaining the precise nature and level of such knowledge and awareness so as to bring it to bear in more effective initiatives to allay anxiety.

##### (2) Attitude when purchasing foodstuffs

Table 4 shows the results of questions as to how aware respondents were of the source of foodstuffs when shopping. The questionnaire presented four options for levels of awareness, ranging from "ALWAYS" to "NEVER." Combining response rates for the two options "ALWAYS" and "OFTEN" revealed the following figures for each period: I winter: 94% (excluding 27 cases of "No answer"), II summer: 98%, III winter: 89%. Up to period II, i.e., a point some 18 months after the accident, the proportion of households aware of the source of production when purchasing foodstuffs was in excess of 90%. Thereafter this proportion showed a gradual tendency to decline. Even so, the results show that approximately 90% of households remained referring to the source in choosing foodstuffs in period III, i.e., two years after the accident. As will be shown later, the survey results revealed that the amounts of radioactive materials were extremely small and that foodstuffs on sale throughout the prefecture pose absolutely no danger to the health of anyone eating them. To date, we have disseminated the results of this survey by making them publicly available in a variety of formats: press conferences, information sessions, posters, pamphlets, and websites. One of the tasks that we are currently faced with is how most effectively to use the survey results in promoting the kind of understanding that will bring about a change in public perceptions and behavior.

Table 4. Referring to the source in choosing foodstuffs  $n=100$

Period Answer	I winter	II summer	III winter
Always	22 (30)	39	42
Often	47 (64)	59	47
Not so often	2 (3)	0	9
Scarcely	2 (3)	2	2
No answer	27	0	0

\*I winter: The numerical value in parentheses is a percentage based on 73 people excluding those who gave 'No answer'.

### 3.2. Consumption of rice, water and other foodstuffs

#### (1) Rice and water

Table 5 shows the areas of production of the main staple, rice. From period I through period III, approximately 65% of households were eating rice grown in Fukushima Prefecture. As shown in table 6, tap water was basically the greatest source of water, while bottled mineral water was used in conjunction with tap water.

#### (2) Foodstuffs produced in Fukushima Prefecture

On the questionnaire relating to menu, respondents were asked about foodstuffs used and the area of production in accordance with the following categories: vegetables, fruit, meat, fish, and soy products. Both the overall number of items and the number of Fukushima products in each category were counted. Table 7 shows the ratio of Fukushima products of the total number and mean value of foodstuff items used in each of these categories. In I winter, the proportion of respondents choosing Fukushima products was 32%, revealing a tendency to avoid local products. However, the proportion of respondents choosing Fukushima products had a tendency to increase progressively. With the advance in provisions to screen foodstuffs produced in Fukushima for radioactive

materials, restrictions have been put in place to prevent foodstuffs that exceed standard values reaching the market. All of which points to the fact that trust in Fukushima products is showing signs of recovery, gradual though they may be. The summer periods showed higher consumption ratios for Fukushima products, which is to be expected, given the abundant availability of Fukushima-grown fruit and vegetables during this season.

#### (3) Results of measurements

Table 8 shows the results of families found to have the lower level of  $\geq 1$  Bq of  $^{134}\text{Cs}$  or  $^{137}\text{Cs}$  per 1 kg of foodstuff sample collected over two days. Of a total of 300 families taking part in the survey from I winter through III winter, some 17 (approximately 6%) were found to exhibit  $\geq 1$  Bq/kg of  $^{134}\text{Cs}$  or  $^{137}\text{Cs}$ . When broken down into the survey periods, this amounts to 10 families (A-J) in I winter, 2 families (K & L) in II summer, 7 families (M-S) in III winter. Thus the results showed that concentrations overall were higher for winter than summer. As seen above in "Foodstuffs produced in Fukushima Prefecture," summer was the period during which the ratio of use of Fukushima products was at its highest. According to these figures, there seems to be no correspondence between detected concentrations of  $^{134}\text{Cs}$  or  $^{137}\text{Cs}$  and the use of Fukushima products. Comparisons of the foodstuffs used (ingredient items) revealed no clear relationship. This is no doubt one of the limitations of the duplicate diet method, in that it carries out measurements on homogenized samples of collected foodstuffs. The greatest dose of both radioactive materials was D in I winter: levels of  $^{134}\text{Cs}$  and  $^{137}\text{Cs}$  stood at 5.0 Bq/kg and 6.7 Bq/kg respectively. Constituting as it does some 0.0117% of the essential nutrient potassium,  $^{40}\text{K}$  was detected in the meals of all the families, ranging from 13 to 58 Bq/kg, a mean value of 32 Bq/kg, which accounts for a large proportion of radioactive materials originating from food.  $^{131}\text{I}$  was not detected in the meals of any of the families, having as it does a short half-life of approximately eight days.

Table 5. Source of the staple food (rice)  $n=100$

Period	I winter	II summer	III winter
Fukushima	66	65	66
Other areas	33	35	34
Unknown	1	0	0

Table 6. Source of drinking water  $n=100$

Period	I winter	II summer	III winter
Tap water	37	52	56
Tap water & Bottled water	14	17	6
Bottled mineral water	9	7	7
Domestic well water	6	3	3
Output of water purifier	5	5	7
Distilled water	1	4	0
No answer	28	12	18

Table 7. Number of foods and ratio of Fukushima products (%)

Period	I winter	II summer	III winter
A : Total number of foodstuffs used in the two days' meals	22	19	21
B : Number of local foodstuffs used in the two days' meals	7	8	8
B/A (%)	32	42	38
Number of families that don't use Fukushima products	10	13	2

Table 8. Summary of 17 family dishes with either radiocaesium above 1 Bq/kg

Case code	weight of food per 2 days		<sup>134</sup> Cs		<sup>137</sup> Cs		<sup>40</sup> K		Cs/K ingested activity ratio
	kg		concentration Bq/kg	uptake by ingestion Bq/2 days	concentration Bq/kg	uptake by ingestion Bq/2 days	concentration Bq/kg	uptake by ingestion Bq/2 days	
A	3.4		2.0	6.8	2.4	8.2	31	105	0.14
B	3.4		0	0.0	1.4	4.8	38	129	0.06
C	3.9		0	0.0	1.1	4	37	144	0.35
D	1.7		5.0	8.5	6.7	11.0	33	056	0.05
E	8.8		2.4	21.1	3.0	26.0	38	334	0.11
F	4.0		0	0.0	1.2	4.8	35	140	0.13
G	3.9		0	0.0	1.0	3.9	27	105	0.06
H	3.6		1.6	5.8	2.2	7.9	35	126	0.14
I	2.9		1.2	3.6	1.7	4.9	22	064	0.06
J	3.0		0	0.0	1.2	3.6	35	105	0.05
K	4.1		1.3	5.3	1.9	7.8	23	094	0.14
L	7.7		0	0.0	1.1	8.5	37	285	0.05
M	5.4		1.3	7.0	2.2	11.9	34	184	0.10
N	4.9		0	0.0	1.6	7.8	30	147	0.05
O	4.5		0	0.0	1.4	6.3	36	162	0.04
P	4.8		0	0.0	1.1	5.3	43	206	0.03
Q	4.2		1.3	5.5	2.4	10.1	20	084	0.19
R	5.7		0	0.0	1.2	6.8	35	200	0.03
S	2.5		1.1	2.8	2.0	5.0	42	105	0.07

Intakes were converted into doses using effective dose coefficients

 $1.9 \times 10^{-2} \mu\text{Sv/Bq}$  for <sup>134</sup>Cs $1.3 \times 10^{-2} \mu\text{Sv/Bq}$  for <sup>137</sup>Cs\*Case code from **A** to **J** in **I** winter, **K** and **L** in **II** summer, from **M** to **S** in **III** winter

(4) Internal exposure doses resulting from ingestion of radioactive materials

As a means of ascertaining concentrations (Bq/kg) of  $^{134}\text{Cs}$ ,  $^{137}\text{Cs}$ , and  $^{40}\text{K}$  detected in the meals of each family, in addition to the quantity of radioactive materials ingested over two days from the weight (kg) of meals taken over two days, the dose conversion coefficient for each radionuclide (ICRP Publication 72) was used to calculate committed effective doses. The largest two-day values for  $^{134}\text{Cs}$  and  $^{137}\text{Cs}$  were  $0.40\ \mu\text{Sv}$  and  $0.34\ \mu\text{Sv}$  respectively, i.e., a total 2-day value of  $0.74\ \mu\text{Sv}$ . These were the estimated values for E, whose meals for two days weighed 8.8 kg, a particularly high figure. In view of the particularly large meal consumption of E, who had the highest internal exposure dose, it was decided to calculate a hypothetical value for E's committed effective dose on the suppo-

sition that E had kept consuming identical meals for one year, although this was likely to result in overestimation. The resulting value was  $0.135\ \text{mSv}$ . Even for an overestimation, this is in the order of a mere  $0.1\ \text{mSv}$  annually, falling far short of the annual radiation dose standard of  $1\ \text{mSv}$ . The column to the right shows the committed effective dose ratios of the radioactive cesium and  $^{40}\text{K}$  ingested from meals. It will be appreciated from this that the ratios are very low, even in terms of  $^{40}\text{K}$  internal exposure, the intake of which is inevitable anyway.

#### REFERENCES

- ICRP Publication 72 1996 Age-dependent doses to members of the public from intake of radionuclides : part 5 *Ann. ICRP* **26** (1)