Micronutrients in favorite beverages and radioactivity in food samples associated with potential health risk in Hokkaido, Japan

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Abstract: Five favorite drinks namely green tea, apple juice, ginger ale, Japanese tea and pot/bottled water were analyzed to determine the micronutrient elements and three food samples namely, rice, small fish and milk for radioactive determination, collected from different super store of Sapporo, Japan. Inductively coupled plasma atomic emission spectroscopy (ICP-AES) have been applied for different micronutrients like sodium, magnesium, calcium, potassium, manganese, iron, copper and zinc determination from these drinks, and a radiation detector (Gama ray spectrometer) was used for determination of ^{134}Cs and ^{137}Cs in the above mentioned food samples. The ICP-AES data showed that the contents of the micronutrients were low and did not exceed the tolerable upper intake level. Therefore, the results indicated that these beverages have no harmful effect on human health in terms of major and minor mineral constituents. Moreover, radiation detector data explained that ^{134}Cs and ^{137}Cs concentration was below the detection limit and prescribed safe limit regulated by Ministry of Health, Labor and Welfare, Japan. The results indicated that the food samples collected from Hokkaido were not contaminated with ^{134}Cs and ^{137}Cs, and thus safe for consumption.
Key words: Cesium, Drinks, Foods, Microutrients, Radiation.

Introduction

Food is essential for our body. Several functions of foods are to develop, replace and repair cells and tissues; produce energy to keep warm, move and work; carry out chemical processes such as the digestion of food; and protect against, resist and fight infection and recover from sickness (FAO, WHO, 2002). Food contains both macronutrient (such as carbohydrate, protein and fat) and micronutrient (such as vitamin and mineral). Due to their significances, consumption of food with the right amounts of nutrients is important to assure the public health. Beverages are the common menu in our dining table and most of the people choose non-alcoholic drinks like coca-cola, apple juice, orange juice, ginger ale, green tea, sprite and *Ocha* (one kind of tea used in Japan). It is also considered as one of micronutrient sources on the diet. The important micronutrients are vitamins, minerals of sodium (Na), magnesium (Mg), calcium (Ca), potassium (K), iron (Fe), copper (Cu), zinc (Zn), manganese (Mn), phosphor (P), iodine (I) and fluorine (F) (WHO, FAO, 2006). Although it is only needed in small amount, micronutrients play important roles for preventing diseases, regulating metabolism, etc. The lack of micronutrients might leads human suffer from diseases. On the other hand, excess soft drink consumption is associated with increased energy intake, increased body weight, displacement of nutrients, and increased risk of chronic diseases (Vartanian *et al*., 2007). Therefore, an effort to avoid micronutrient deficiency or more sufficiency is evaluated by measuring the amount of micronutrients on several favorite beverages. Such information is important for the public health.

After the Fukushima nuclear accident (March 11, 2011) in Japan, releasing of different radionuclides into the environment was one of the severe environmental catastrophes in the 21st century (Povinec *et al*., 2013). After accident, both short and long-lived activation and nuclear-fission products have been released into the environment (Fig. 1), where most of nuclides were of the volatile fission products (noble gases, iodine, cesium, and tellurium) and causes different health hazards like cancer, etc. The accident introduced ^{134}Cs and ^{137}Cs into the coastal waters which was subsequently transferred to the local coastal biota thereby elevating the concentration of this radionuclide in coastal organisms (Tateda *et al*., 2013) and other environment. Internal exposure occurs, when radionuclides enter and were absorbed into the body. Ingestion of food is the most significant route of radionuclide intake for peoples (ICRP, 1992). After the Fukushima nuclear accident, radionuclides disseminate into the soil, ocean and air and contaminate the plant, fish, water, meat and milk. Hokkaido is the second largest island in Japan and largest and northernmost island among 47 prefectures in Japan (Wikipedia, 2015). Hokkaido is about 1070 km far from Fukushima prefecture (Fig. 1). However, the contamination might occur and the evaluation and monitoring of the radioactivity in some main Hokkaido foods like rice, milk and small fish should be conducted.

According to the previous explanation, this study was carried out to evaluate the micronutrient content of several favorite beverages like green tea, apple juice, ginger ale, ocha and water in Sapporo, Hokkaido. The micronutrients examined included Na, Mg, Ca, K, Mn, Fe, Cu and Zn. In connection with Fukushima accident, we would also like to evaluate the radioactivity of ^{134}Cs and ^{137}Cs on the popular Hokkaido foods such as rice, fish and milk.

Materials and Methods

Beverage Sampling and micronutrients analysis: Green tea, apple juice (KIRIN-Koiwai Family Products), ginger ale, Japanese tea and pot water were randomly collected from a supermarket in Sapporo city, Hokkaido, Japan. These beverages samples were then filtered using a 0.45-µm syringe-driven filter unit (Millipore, Billerica, USA) and stored in 15 mL polyethylene bottles. Na, Mg, Ca, K, Mn, Fe, Cu and Zn were analyzed from these samples using Inductively Coupled Plasma-Atomic Emission Spectrometry (ICP-AES) (Seiko SPS7700) followed by the method described by Van de Wiel (2003). Background correction was done for trace element determination. ICP-AES is calibrated by standard reference materials.

Standard deviation (SD), Limit of detection (LOD) and Limit of quantitation (LOQ): SD, LOD or detection limit and LOQ were determined (Table 1) by using seven standard blank, standard major and standard minor samples with the help of Gaussian curve for each of above eight analytes like Na, Mg, Ca, K, Mn, Fe, Cu and Zn.

Table 1. Standard deviation (SD), limit of detection (LOD) or detection limit and limit of quantitation (LOQ) (mg L^{-1}) by using seven standard blank

Std. blank	Na	Na	Mg	Mg	Ca	Ca	K	Mn	Mn	Fe	Fe	Cu	Cu	Zn	Zn
Blank 0	0	0	0	0	0	0	0	0	0	0	0	0	0	0	0
Blank 1	-0.0267	-0.0196	-0.0088	-0.0098	-0.0002	0	-0.0095	-0.0022	-0.0032	-0.0199	-0.0207	-0.0088	-0.004	-0.0059	-0.0012
Blank 2	-0.0341	-0.0196	-0.0125	-0.0114	-0.0007	-0.0003	-0.0156	-0.002	-0.0046	-0.0259	-0.0211	-0.0099	-0.0044	-0.0065	-0.0057
Blank 3	-0.0323	-0.0201	-0.0127	-0.0128	-0.0003	-0.0001	-0.0036	-0.002	-0.0035	-0.0269	-0.0269	-0.0119	-0.0048	-0.0067	-0.0034
Blank 4	-0.0295	-0.0189	-0.0109	-0.0111	-0.0004	-0.0001	-0.0202	-0.0018	-0.003	-0.0262	-0.0217	-0.0077	0.0104	-0.0066	-0.0024
Blank 5	-0.0289	-0.0186	-0.0123	-0.0112	-0.0004	-0.0001	-0.0108	-0.0019	-0.0025	-0.0175	-0.0318	-0.0085	-0.0089	-0.0047	-0.0053
Blank 6	-0.0319	-0.0219	-0.0135	-0.0142	-0.0005	-0.0001	0.0222	-0.0019	-0.0022	-0.0255	-0.0282	-0.0095	0.0049	-0.0046	-0.0079
SD	0.0118	0.0076	0.0047	0.0047	0.0002	0.0001	0.0139	0.0007	0.0014	0.0096	0.0104	0.0038	0.0066	0.0024	0.0028
LOD	0.035	0.023	0.014	0.014	0.0007	0.0003	0.042	0.0023	0.0043	0.029	0.031	0.011	0.020	0.007	0.008
LOQ	0.1181	0.0755	0.0471	0.0466	0.0022	0.001	0.1392	0.0075	0.0142	0.0963	0.1035	0.0379	0.0663	0.0237	0.0277

LOD (Eq. 1) is defined as the lowest concentration level that is statistically different from a blank and correspond to a 99% confidence level (Yang *et al*., 2012). LOD = 3SD/m ...... Eq. 1; Where, m = the slope of the addition graph, SD = the within-run standard deviation of blank determination. LOQ (Eq. 2) is defined as the level above which quantitative results can be obtained with 99% confidence level (Yang et al., 2012). LOQ = 10SD/m ...... (Eq. 2).

Food sampling and radio cesium analysis: Hokkaido cow's milk, small fish and rice (*Oryza sativa* subsp. *Japonica)* samples (produced from Hokkaido) are collected from *AEON* super shop, Sapporo, Hokkaido to analyze radioactive ^{134}Cs and ^{137}Cs. Samples were processed according to USEPA *et al*. (2004) and Department of Food Safety, Japan (2012) and then analyzed by Gamma-ray spectrometer (GEM20P4) with an electric cooler and automatic sample changer (Seiko EG&G ORTEC Co.). The measurement life time was 3000 seconds/sample. Radioactivity measurements cannot be made without consideration of the background. Background radiation, is the radiation that enters the detector concurrently with the radiation emitted from the samples being analyzed. So background count rate was determined by a separate operation and subtracted from the total activity or gross count rate (DOE-HDBK, 2009). Parameters used in Gamma-ray spectrometer analysis for radio cesium were given in Table 3. ^{134}Cs and ^{137}Cs radiation detection limit (DL) (Bq/Kg) and standard deviation (SD) from the samples were calculated (modified from DOE-HDBK, 2009) using the following formula. Counts (N) = 3.3 $\sqrt{B \times 2.5}$ .... (Eq. 3)

$$\text{Radiation DL} = \frac{N(Counts)}{Life\ Time\ (Sec.)} \times \frac{1}{E.P.} \times \frac{1}{D.E} \times \frac{1000g}{Weight\ (g)} \text{ .... (Eq.4)}$$

Net Counts (N) = [{Gross Channel Counts (GCC)} – {Background Channel Counts (BCC)}] ....(Eq. 5). Net Counts ± Standard deviation (SD) = N $\pm \sqrt{N + 2.5B}$ .... (Eq. 6); Where, B = the Background Channel Count, E.P. = Emission probability
D.E. = Detection efficiency, Weight = sample weight in grams.

Results and Discussion

Micronutrients in beverages and potential health risk: Five popular drinks namely green tea, apple juice, ginger ale, *ocha* and water were analyzed for Na, Mg, Ca, K, Mn, Fe, Cu and Zn micronutrients (Table 2). Among the five popular drinks, apple juice contains higher amount of Na (95.70 mg L^{-1}) and K (102.33 mg L^{-1}) whereas lowest amount was found in ginger ale (Table 2). The amounts of micronutrients in *ocha* (one kind of tea used in Japan) were similar than those of green tea (Table 2). Moreover, water contained small amount of all elements but higher than the ginger ale. Green tea and *Ocha* contained higher microelements than the ginger ale and apple juice (Table 2). These micronutrients in addition with polyphenol contents in tea much more helpful to human body like increase enzyme activities, cellular metabolism and immune function (Chacko et al., 2010).

Table 2. Elemental concentration (mg L^{-1}) in different beverage samples using ICP-AES

Samples	Na	Mg	Ca	K	Mn	Fe	Cu	Zn
Std. major 1	50.03	25.22	25.07	49.8	-	-	-	-
Std. major 2	48.18	24.51	23.97	51.56	-	-	-	-
Std. minor 1	-	-	-	-	20.02	20.1	20.16	19.98
Std. minor 2	-	-	-	-	19.74	19.77	20.1	19.54
LOD	0.023, 0.035	0.014, 0.014	0.0007, 0.0003	0.042	0.0023, 0.0043	0.029, 0.031	0.011, 0.020	0.007, 0.008
Green tea	71.93	4.46	1.46	72.60	1.35	ND	ND	0.04
Apple juice	95.71	3.93	3.90	102.33	0.02	ND	ND	0.01
Ginger ale	1.07	ND	0.01	ND	ND	ND	ND	ND
Ocha	74.34	4.42	1.44	74.83	1.32	0.03	0.01	0.04
water	7.62	1.43	7.00	53.38	ND	ND	0.04	0.03
RDA/AI*	1,500*	350	1,000	4,700*	2.3*	8	0.9	11
UL	2,300	350	2,500	ND	11	45	10	40

ND = Not detected as the value was below the LOD, RDA = Recommended dietary allowance, *AI = adequate intake, UL = Tolerable upper intake levels (https://www.nal.usda.gov/fnic/DRI/DRI_Tables/recommended_intakes_individuals.pdf)
http://iom.nationalacademies.org/Activities/Nutrition/SummaryDRIs/~/media/Files/Activity%20Files/Nutrition/DRIs/ULs%20for%20Vitamins%20and%20Elements.pdf)

In order to evaluate the obtained values, we compared the results with recommended dietary allowances RDA (or adequate intake AI) and tolerable upper intake level (UL) for life stage group of males 19-30 years old (Table 2), according to USDA. Recommended dietary allowance could be defined as the average daily intake level. It describes the sufficient value to meet the requirements of nutrient for nearly 97-98% of healthy individuals in a life stage group. If scientific evidence is not sufficient enough, the adequate intake (AI) is used instead of RDA. AI could be considered as the value to cover the needs of all healthy individuals in a life stage group. On the other hand, the definition of UL is the highest level of daily nutrient intake which might pose no risk of adverse health effects to almost all individuals in a group (Food and Nutrition Board, Institute of Medicine).

The results showed that all the amounts of micronutrients on all tested beverages were lower than both RDA and UL (Table 2). It indicated that those beverages were safe for daily consumption. In addition, the beverages (except ginger ale) might contribute to the daily requirement of microelements.

Radio cesium content in food and associated health hazards: Monitoring for the food safety after Fukushima nuclear power plant disaster was carried out by determining the radioactivity of several main foods such as rice, fish and milk. The parameter analyzed were ^{134}Cs and ^{137}Cs radionuclides as they have long half-life of 2 and 30 years, respectively (Table 3). Therefore, these radionuclides might exist on the environment for several years, contaminate the food and threat the human health.

As displayed on Table 4, the determined values for rice, fish and milk were markedly low and below the detection limit. In addition, these values did not exceed the safe limit regulated by Ministry of Health, Labor and Welfare, Japan (2011) (Matsuyama, 2011). The results indicated that these foods were safe for consumption in the sense of radiation contamination.

Table 3. Parameters used in Gamma-ray spectrometer for radio cesium analysis

Radioactive elements	Half life (Years)	Detection Efficiency (D.E.)	Emission Probability (E.P.)	Channel	Strength
^{134}Cs	2	10.5%	97.6%	1208	604 keV
^{137}Cs	30	10%	85%	1324	662 keV

Table 4. ^{134}Cs and ^{137}Cs determination in rice, small fish and milk samples

Radio cesium	Sample weight (g)	BCC	DL (Bq/Kg)	LT (Sec.)	GCC	N=GCC-BCC	N±SD	Radiation count (Bq/Kg)	Remark	Safe limit (Bq/kg)*
Rice										
^{134}Cs	53.06	112	3.4	3000	107	-5	-5±16.583	-0.307	NC	500
^{137}Cs	53.06	101	3.9	3000	80	-21	-21±15.215	-1.305	NC	
Fish										
^{134}Cs	7.937	112	23	3000	91	-21	-21±16.04	-8.606	NC	500
^{137}Cs	7.937	101	26	3000	84	-17	-17±15.35	-8.399	NC	
Milk										
^{134}Cs	64.2	112	2.8	3000	90	-22	-22±16.06	-1.11	NC	200
^{137}Cs	64.2	101	3.2	3000	107	6	6±16.07	0.362	NC	

BCC-Background Channel Count, DL-Detection Limit, LT-Life Time (Seconds), GCC-Gross Channel Count, N-Net Count, SD-Standard Deviation, NC-Not contaminated with ^{134}Cs and ^{137}Cs because ^{134}Cs and ^{137}Cs concentration in these samples below the detection limit.
* http://www.mhlw.go.jp/english/topics/2011eq/dl/food-120821_1.pdf
*Matsuyama K, 2011 http://www.bloomberg.com/news/articles/2011-03-21/japan-sets-safe-limits-for-consuming-radiation-contaminated-food-table-

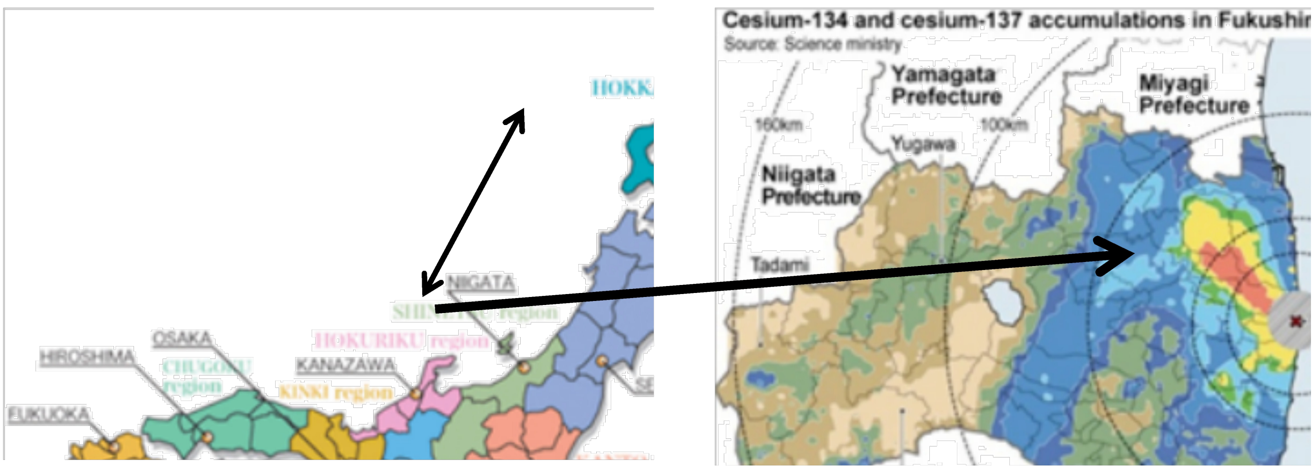

Fig.1. Japan map (left) and enlarge view of Fukushima nuclear plant (right) indicating ^{134}Cs and ^{137}Cs accumulation.
⟷ Indicates Fukushima to Sapporo distance about 1070 km (Courtesy from Google map).

Government of Hokkaido also frequently monitored radiation materials in tap water at the filtration plant originated from different rivers of Sapporo city and found that ^{131}I, ^{134}Cs and ^{137}Cs were below the detection limit and were not contaminated (City of Sapporo, 2013). Ministry of Health, Labor and Welfare, Japan (2015) also determined the total Cs level in different food products of different prefectures and found that the total Cs level was less than 25 Bq/kg in cattle meat (pre marketed) from Hokkaido, thus it was safe for consumption. This was probably due to the Hokkaido is far away from Fukushima power plant disaster place (Fig. 1). The very small amounts of radioactive materials were transported to Hokkaido Island as they spread near power plant and became diluted in Pacific Ocean and Japan Sea.

The common commercial beverages in Japan (green tea, apple juice, ginger ale, water and *ocha*) contained low amount of mineral micronutrients. The content of the micronutrients did not exceed the tolerable upper intake level. Therefore, the results indicated that these beverages have no harmful effects on human health in terms of major and minor mineral constituents. In the connection of Fukushima nuclear power plant disaster, the levels of ^{134}Cs and ^{137}Cs radionuclides on rice, fish and milk were very low. Such low levels indicated that Hokkaido originated rice, fish and milk were safe for consumption in respect to radioactive contamination.

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