

April 2011.¹ The occurrence of milk powder contamination with ^{134}Cs and ^{137}Cs (22–31 Bq/kg) was announced by Meiji Holdings on 6 December 2011 (cited on 6 August 2012; available from <http://www.meiji.co.jp/notice/2011/detail/20111206.html>). This contamination was concluded to be derived from atmospheric air during the process of drying of milk powder, and not from water or dairy ingredients. Thus, environmental pollution with radioactive materials occurred and reached pregnant women after the FNP accident.

The placentas of women living in Hiroshima, Osaka, Tokyo and Canada in the 1960s contained detectable levels of ^{137}Cs ^{3–5} due to environmental pollution with ^{137}Cs after the repeated nuclear tests conducted by several countries, such as the USA and the former USSR. As the estimate of ^{137}Cs deposition at the Meteorological Research Institute, Tsukuba, after the FNP accident far exceeded that in the 1960s in Japan (Fig. 1),⁶ the placentas of women living near FNP may contain higher levels of ^{134}Cs and ^{137}Cs than those in the 1960s in Japan. However, the degree of placental contamination with radioactive Cs has not been studied. Therefore, the present study was performed to investigate the ^{134}Cs and ^{137}Cs contents in the placentas of women living within 300 km of FNP.

Materials and Methods

This study was conducted with the approval of the institutional review boards of Kameda General Hospital and Japan National Institute of Public Health.

Women who provided placentas

Placentas were obtained from 10 women: five (cases 1–5) living within 50 km (neighbor group) and five (cases 6–10) living within 210–290 km (distant group) of FNP until delivery after the FNP accident (Table 1). All 10 women gave birth to a healthy term singleton infant during the period between October 2011 and August 2012. The five women in the neighbor group gave birth earlier by the end of February 2012, while the five women in the distant group gave birth later in or after July 2012.

Measurement of radionuclides

Each whole placenta with a wet weight varying 0.418–0.672 kg was ashed to 4.13–7.40 g (Table 1) by muffle furnace at 450°C for 24 h after lyophilizing according to the preparation method recommended in the USA (<http://www.epa.gov/rpdweb00/docs/marlap/402-b-04-001b-12-final.pdf>). These ashed samples were placed individually into cylindrical plastic containers

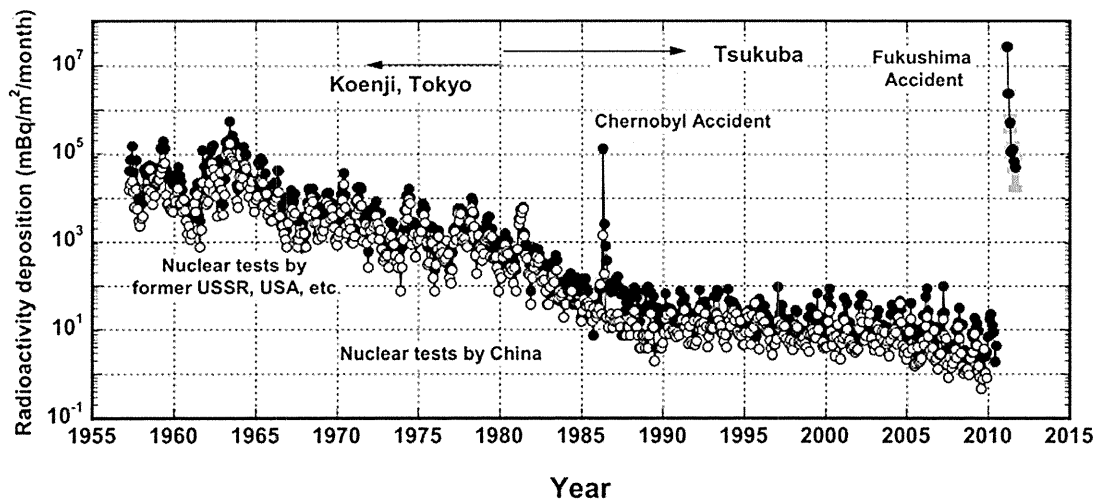


Figure 1 Estimates of ^{137}Cs deposition at the Meteorological Research Institute, Tsukuba, are presented for several months after the accident in March 2011. The estimate was computed based on the value obtained by measuring aliquots of the sample water (wet + dry depositions). As cesium is distributed between the liquid and the solid phases, the accurate value is not obtained unless the concentration of the whole sample by evaporation is achieved. Probably, current values are underestimates. Moreover, as ^{134}Cs was deposited in comparable amounts, the total radioactive cesium had mostly doubled. —●—, ^{137}Cs ; —○—, ^{90}Sr . (Adopted from ⁶).

Table 1 Radioactivity of ^{134}Cs , ^{137}Cs and ^{40}K in the placentas of 10 women

Case	City† (distance/direction from FNP)	Month/year‡	Placental weight		Radioactivity (Bq/kg)			^{137}Cs to K ratio (Bq/mmol)
			Wet (kg)	Ashed (g)	^{134}Cs	^{137}Cs	^{40}K	
1	Minami-soma (25 km, N)	Oct. 2011	0.520	6.45	0.742	0.922	46.5	0.024
2	Iwaki (45 km, S)	Nov. 2011	0.549	6.88	0.549	0.648	59.3	0.013
3	Iwaki (45 km, S)	Dec. 2011	0.532	6.23	0.090	0.207	46.9	0.005
4	Iwaki (45 km, S)	Dec. 2011	0.651	6.44	0.268	0.302	51.9	0.007
5	Iwaki (45 km, S)	Feb. 2012	0.590	7.40	0.373	0.563	50.5	0.014
6	Mobara (230 km, S)	Jul. 2012	0.418	4.13	0.462	0.694	47.6	0.018
7	Kamogawa (270 km, S)	Jul. 2012	0.627	5.92	0.064	0.121	49.5	0.003
8	Tokyo (230 km, SW)	Aug. 2012	0.543	5.53	<0.054	0.109	49.8	<0.003
9	Yotsukaido (210 km, S)	Aug. 2012	0.672	6.49	0.042	0.078	50.9	0.002
10	Tateyama (290 km, S)	Aug. 2012	0.436	4.35	0.061	0.093	52.1	0.002

†Cities where women were living. ‡Month and year when women gave birth. The distance and direction from the Fukushima nuclear power plant is indicated in parentheses. N, north; S, south; SW, southwest.

(100-mL capacity). To determine the gamma-emitting nuclides in the samples, gamma ray spectrometry was performed for more than 80 000 s with high-purity germanium detectors (GEM40-76; Ortec, Oak Ridge, TN, USA) connected to a multichannel analyzer and analytical software, and the activity concentrations of the radionuclides were corrected to the delivery dates. Each measured radioactivity was multiplied by $2^{(N/T)}$: N and T were intervals until the measurement after delivery of the placenta (year) and half-life of each radionuclide (year), respectively. The energy and efficiency calibrations were performed using the nine nuclides mixed activity standard volume sources (MX033U8; Japan Radioisotope Association, Tokyo, Japan) composed of ^{109}Cd , ^{57}Co , ^{139}Ce , ^{51}Cr , ^{85}Sr , ^{137}Cs , ^{54}Mn , ^{88}Y and ^{60}Co . These sources, contained in the same containers as the samples, had five different heights (0.5, 1, 2, 3 and 5 cm, respectively) to determine the detection efficiency of the detector as a function of sample height.

Results

As expected, ^{134}Cs and ^{137}Cs were detected in nine and 10 of the 10 placentas with varying activities ranging 0.042–0.742 Bq/kg for ^{134}Cs and 0.078–0.922 Bq/kg for ^{137}Cs , respectively (Table 1), while relatively constant levels of ^{40}K were detected, ranging 46.5–59.3 Bq/kg, regardless of the differences in cities where they were living after the FNP accident. If we assumed that ^{134}Cs content was 0.050 Bq/kg for case 8, median ^{134}Cs content, 0.373 Bq/kg (range, 0.090–0.742) in the five placentas of the neighbor group was relatively higher than that of 0.061 Bq/kg (range, 0.042–0.462) in the five placentas of the distant group, but difference did not reach a significant level ($P = 0.05556$, Mann–Whitney

U-test). Median ^{137}Cs content was 0.563 Bq/kg (range, 0.207–0.922) for the neighbor group and 0.109 Bq/kg (range, 0.078–0.694) for the distant group ($P = 0.09524$).

Discussion

The present study demonstrated that placentas of women living within 290 km of FNP contained detectable levels of ^{134}Cs and ^{137}Cs . The difference in degree of contamination of placentas with radioactive Cs may have reflected dietary habits, the degree of environmental pollution and the interval until delivery after the FNP accident. The shortened biological half-life of radioactive Cs from approximately 100 days for non-pregnant adults to approximately 60 days in pregnant women⁷ may have also contributed to the lesser contamination of the placenta in women who gave birth in and after July 2012. Although environmental pollution with radioactive Cs has been decreasing, daily ^{137}Cs activities of fallout exceeded 10 MBq/km² in 15 days in March 2012 in Fukushima City (Preliminary results of monitoring the environmental radioactivity level of fallout [File number 93], cited on 10 August 2012; available from http://radioactivity.mext.go.jp/old/ja/1285/2012/03/1285_033018.pdf). Surface soils contained more than 1000 Bq/kg of radioactive Cs in wide areas of Fukushima Prefecture where the five women of the neighbor group were living (cited on 10 August 2012; available from http://www.s.affrc.go.jp/docs/map/pdf/02_2_04bunpu_fukushima.pdf).

As shown in Figure 1, environmental pollution with radionuclides occurred after the repeated nuclear tests in the mid-20th century and after the Chernobyl accident in 1986. According to a study that examined ^{137}Cs content in the placenta and urine of inpatients at

Hiroshima University Hospital and in daily foods served for these inpatients over a 5-year period from 1966–1970,⁵ ¹³⁷Cs content in the placentas was approximately 35 pCi (1.3 Bq)/kg, ¹³⁷Cs daily dietary intake was approximately 30 pCi (1.1 Bq) and ¹³⁷Cs daily excretion in the urine was approximately 25 pCi (0.9 Bq) in 1966. Japanese and Canadian groups investigated ¹³⁷Cs content in the human placentas collected in the Tokyo and Osaka areas in Japan and in the Montreal area in Canada in the mid-1960s.^{3,4} The average content of ¹³⁷Cs was similar in Japanese and Canadian placentas, regardless of the differences in dietary habits (averages of 25.2 pCi [0.93 Bq]/kg and 24.8 pCi [0.92 Bq]/kg for Japanese and Canadian placentas, respectively).³ Thus, placentas of Japanese and Canadian women in the mid-1960s contained an average of 0.9–1.3 Bq/kg ¹³⁷Cs. Placentas contained less than 0.8 Bq/kg ¹³⁴Cs and less than 1.0 Bq/kg ¹³⁷Cs in this study. Although there may be a problem of direct data comparison between studies in which different assay methods were used, these results suggested that placentas of Japanese and Canadian women in the mid-1960s were more heavily contaminated with ¹³⁷Cs than the placentas examined in this study.

The Chernobyl accident occurred on 26 April 1986. According to the Japanese Ministry of Education, Culture, Sports, Science and Technology (released on 13 March 2012; cited on 6 August 2012; available from <http://radioactivity.mext.go.jp/ja/list/338/list-1.html>), total amounts of dispersed ¹³¹I and ¹³⁷Cs into the environment after the FNP accident were 1.3–1.6 × 10¹⁷ Bq and 1.1–1.5 × 10¹⁶ Bq, respectively, while corresponding values after the Chernobyl accident were 1.8 × 10¹⁸ Bq and 8.5 × 10¹⁶ Bq, respectively. Thus, the degree of environmental pollution is estimated to be 11–14-fold higher for ¹³¹I and 6–8-fold higher for ¹³⁷Cs after the Chernobyl accident than after the FNP accident. An Italian group examined ¹³⁴Cs and ¹³⁷Cs contents in the placentas of women who gave birth at the University of Bologna over a 13-month period from June 1986 to September 1987 after the Chernobyl accident.⁸ Mean placental ¹³⁷Cs content increased from 4.2 Bq/kg in June 1986, showing a peak of 11.5 Bq/kg in March 1987, and then decreased to 6.6 Bq/kg in September 1987.⁸ The Italian group also estimated dietary ¹³⁷Cs intake on the basis of the average diet in the region where study subjects lived,⁸ daily ¹³⁷Cs intake was estimated to be 15 Bq in the summer of 1986,⁸ which is approximately 14-fold higher than that of 1.1 Bq in the Hiroshima area, Japan, in 1966.⁵ An investigation conducted 4 months after the FNP acci-

dent in early July 2011 revealed that median values of daily dietary intake of ¹³⁴Cs and ¹³⁷Cs were 0.6 Bq and 0.9 Bq in Soma (neighboring city to the north of Minami-soma), and 0.4 Bq and 0.7 Bq in Iwaki, respectively.⁹ Thus, ¹³⁷Cs content per kg of the placenta well reflected daily ¹³⁷Cs intake and appeared to be 50–120% of the daily ¹³⁷Cs intake. Another Italian group reported daily urinary excretion of 13.5 Bq ¹³⁷Cs in people living in the Pordenone area of Italy in the latter half of 1987,¹⁰ which is more than 10-fold higher than that of 0.9 Bq in women living in the Hiroshima area in 1966.⁵ Thus, levels of exposure to radioactive Cs in Japanese pregnant women in the mid-1960s and after the FNP accident were much lower than those in women living in certain areas of Europe after the Chernobyl accident. In another report from Germany,¹¹ the radioactive Cs load in the placenta was shown to have increased by 10-fold compared with studies before the Chernobyl accident in western Germany.

The ratio of radioactive Cs to total K (stable and radioactive) is conventionally taken as a measure of radioactive Cs contamination, independent of body size and sex.¹² Soft tissue ¹³⁷Cs content corrected for potassium did not differ between mother and fetus,¹³ suggesting that the placenta is not a barrier for radioactive Cs. Mean activities of placental ⁴⁰K were reported to be 770 pCi per placenta (57 Bq/kg) and 45 Bq/kg in Japanese⁴ and Italian⁸ studies, respectively, consistent with the values ranging 46.5–59.3 Bq/kg in this study. The heaviest contaminated placenta contained 0.922 Bq/kg ¹³⁷Cs and 46.5 Bq/kg ⁴⁰K. This ⁴⁰K activity was equivalent to a placental K level of 38.4 mmol/kg. Thus, this placenta exhibited a ¹³⁷Cs to K ratio of 0.024 Bq/mmol. According to a study in Glasgow by Watson,¹² whole-body ¹³⁷Cs to total body K was 0.109 Bq/mmol after the Chernobyl accident; this figure is several-fold higher than that of 0.037 Bq/mmol determined in mainland Scotland in 1978–1979,¹⁴ and that of 0.024 Bq/mmol in the placenta of case 1 in this study. The mean whole-body activity of naturally occurring ⁴⁰K was 2859 Bq for females (52 Bq/kg, if we assume that bodyweight was 55 kg),¹² falling between two figures (45 Bq/kg⁸ and 57 Bq/kg⁴) of placental ⁴⁰K activity. Thus, placental ⁴⁰K activity concentration appeared to be similar to whole-body ⁴⁰K activity concentration.

A study of the whole-body radioactive Cs¹⁵ showed another aspect of exposure to ¹³⁴Cs and ¹³⁷Cs in Minami-soma residents after the FNP accident. Although only one Minami-soma resident was included in our study population, this woman showed

less placental contamination than those reported in the published work.^{3-5,8} However, relatively heavy exposure to radioactive Cs occurred in residents in Minami-soma. According to a study that examined whole-body radioactive Cs (¹³⁴Cs and ¹³⁷Cs) in 9498 residents in Minami-soma during the period between 26 September 2011 and 31 March 2012,¹⁵ radioactive Cs (≥ 210 Bq for ¹³⁴Cs and ≥ 250 Bq for ¹³⁷Cs) was detected in 38% (3051/8066) of adults and 16% (235/1432) of children (6–15 years old), ranging 210–12 771 Bq (median, 744 Bq), with a concentration of 2.3–196.5 Bq/kg (median, 11.4) for adults and 210–2953 Bq (median, 590), with a concentration of 2.8–57.9 Bq/kg (median, 11.9) for children. Based on these data, we speculated that the pregnant Minami-soma woman in this study may have managed to avoid contaminated food materials. Available data on whole-body ¹³⁴Cs and ¹³⁷Cs activities are as follows: whole-body ¹³⁴Cs and ¹³⁷Cs activities were 172 Bq and 363 Bq, respectively, in non-pregnant adults living in the Glasgow area in June and July 1986 after the Chernobyl accident;¹² and that for ¹³⁷Cs activity was estimated to be 3 nCi (111 Bq) in 1966, with a gradual decline to less than 1 nCi (37 Bq) in 1969 in pregnant Japanese women living in the Hiroshima area.⁵

In conclusion, placentas from women living within 290 km of FNP contained detectable levels of ¹³⁴Cs and ¹³⁷Cs. However, the degree of contamination was lower than those in Japanese and Canadian women in the mid-1960s and in northern Italian women in 1986–1987 after the Chernobyl accident. It has not been elucidated how placental contamination with radioactive Cs occurring in the past affected fetuses adversely. Such adverse effects, if present, may be disclosed in follow-up studies that are being conducted in Fukushima Prefecture in future.

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