

Estimated environmental radionuclide transfer and deposition into outdoor swimming pools

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Title: Estimated environmental radionuclide transfer and deposition into outdoor swimming pools

Running title: Deposition of radionuclide into swimming pools

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Abstract

In 2011, a large radioactive discharge occurred at the Fukushima Daiichi nuclear power plant. This plant is located within a climatically temperate region where outdoor swimming pools are popular. Although it is relatively easy to decontaminate pools by refilling them with fresh water, it is difficult to maintain safe conditions given highly contaminated diurnal dust falls from the surrounding contaminated ground. Our objectives in this paper were to conduct daily radioactivity measurements, to determine the quantity of radioactive contaminants from the surrounding environment that invade outdoor pools, and to investigate the efficacy of traditional pool cleaners in removing radioactive contaminants. The depositions in the paper filterable particulates ranged from 0 to 72Bq/m²/day, with the highest levels found in the southern Tohoku District containing Fukushima Prefecture and in the Kanto District containing Tokyo Metro. They were approximately correlated with the ground contamination. Traditional pool cleaners eliminated 99% of contaminants at the bottom of the pool, reducing the concentration to 41Bq/m² after cleaning. Authors recommended the deposition or the blown radionuclides into outdoor swimming pools must be considered into pool regulations when the environments exactly polluted with radionuclides.

Key words; swimming, water, radionuclide, contamination, trap

Introduction

Swimming pools in elementary, junior-high, and high schools throughout Japan are important facilities for physical education. Studies by the Japanese Ministry of Education, Culture, Sports, Science, and Technology (MEXT) indicate that 28,171 outdoor pools affiliated with schools were active in the summer of 2008 (School Health Education Division, Sports and Youth Bureau, MEXT, 2008 and March 2011). Following the 2011 Fukushima Daiichi nuclear power plant (FDNPP) accident, MEXT outlined a national guideline whereby radiation levels (total Cesium) were provisionally required to be below 200Bq/l in school pools in Fukushima Prefecture (School Health Education Division, Sports and Youth Bureau, MEXT, 16. June 2011; School Health Education Division, Sports and Youth Bureau and Nuclear Safety Division, Science and Technology Policy Bureau, MEXT, 16. June 2011). This guideline was set based on the advices of the Food Sanitation Law in Japan (1947), Health Life Division, Health Service Bureau, the Ministry of Health, Labor, and Welfare (23. June 2011), Water Environmental Division, Environmental Management Bureau, the Ministry of Environment of Japan (24. June 2012), and the Food Safety Commission (27. October 2011). In response, the Nuclear Emergency Response Headquarters and the Board of Education (School Life Health Division) in Fukushima Prefecture began monitoring 144 outdoor pools not far from the site of the nuclear accident (Fukushima Prefecture, 4. July 2011). Thus, water quality in these pools remained at safe levels, and no specific health problems were reported.

Authors could not find actual radionuclide contamination of swimming pools preceding the accident. No radioactivity was detected from drinking water in Shizuoka Prefecture, the usual source of swimming pools, throughout 2006-2011 (Environmental Radioactivity Database). The government ^{137}Cs monitoring of ground surface soils in grass land or uncultivated places, showed less than 10Bq/kg in all Japan prefectures in 2010. However, in 2011 when the FDNPP failed, the values of three prefectures in Tohoku District, and all seven prefectures in Kanto District grew 3.0 to 98 times more ^{137}Cs (Environmental Radioactivity Database).

One year later, the tentative radioactivity guidelines were revised and restored to the drinking water standard preceding the accident (School Health Education Division, Sports and Youth Bureau, MEXT, 10. April 2012, Water Environmental Division, Environmental Management Bureau, the Ministry of Environment, 8. June 2012, WHO, 2008, and WHO, 2011). Fresh water supplied to pools by authorized plants was not contaminated after filtration; however, pool water must be partially filtered. Moreover, circular filter apparatuses typically installed in swimming pools are relatively ineffective at eliminating sediment (e. g., soils). Because school swimming pools are open, it is possible that radioactive dust blown from surrounding environments could settle into these pools. It is thus critical to investigate the quantities of radionuclide that accumulate in swimming pools located in highly contaminated areas. There are no recommendations how to treat or prevent radionuclides contaminations of bathing water (WHO, 2006).

Although MEXT began monitoring pool water and water source contamination immediately following the nuclear accident, additional counter-measures may be required to avoid further pool contamination from pollutants introduced through human activities, wind, and rain. Previous studies of historical radioactive discharges (e.g., Hiroshima, Nagasaki, Nevada, Chernobyl, and Three-mile Island) did not examine the levels of radioactive contamination in swimming pools, and data on this subject will be of practical importance for the safe use of many outdoor school pools located in Fukushima Prefecture.

The radionuclide resuspensions of deposited ground surfaces from contaminated nuclear test or inhabiting areas have documented or studied relatively well (Sehmel, 1977; Nicholson, 2009; IAEA, 2010), however, the most of them were model or mathematical simulation study. Actual field data obtained from a pond showed that a huge volume of radioactivity flown or brown into the pond from the surrounding environment (e.g., by fast-moving streams, creeks, or wind) accumulated in sediments and decayed more slowly (Kryshev, 1995). Outdoor swimming pools are artificial ponds in inhabiting areas, so, the volume of deposited radionuclides blown into must be realized whether the value exceeds more or not the WHO's drinking water guideline (10 Bq/l) when the sediments are

resuspended, in the pool water (WHO, 2006).

Although the monitored outdoor swimming pool (25m long × 25m width × 5m depth) has not been used after the FDNPP accident, it has been refilled and maintained regularly. The radioactivity of the water and the sediments was tested before seasonal use of the pool. The total cesium radioactivity of the water was below the background level and that of the sediments was 33,000 Bq/m². As a case of contaminated pond in the Chernobyl nuclear power plant failure, the outdoor swimming pool sediments also affected by contaminated surrounding radionuclide (Kryshev, 1995, Yoshida and Kanda, 2012). It is already known that distributed or contaminated long-life radionuclide will redistribute and considerably accumulate. The sediments collected in July 2011, four months after the FDNPP accident, exhibit no traces of ¹³¹I, which has the most radiotoxic effect on children.

Almost all swimming pool users such as swimmers, instructors/teachers, and maintenance mechanics have no information on the radioactive contamination of swimming pools. Unfortunately, the authors experienced this first hand, and the results of the experiments on the swimming pool are presented in this paper.

Materials and Methods

We distributed plastic tanks (53cm long×36cm width×9cm depth) filled with water at 35 sites from 270km north to 1,100km southwest of the FDNPP (Figure 1), and the detail information listed in Table 1.. The tanks remained exposed at these sites for 30 days, and we collected all materials blown into the tank through filtration (Type 101, 15cm ϕ , Toyo Roshi Co., Ltd., Tokyo). Collected specimens and filtration papers were sealed in plastic bags, mailed back to the laboratory, and wrapped into filter paper to form a 5cm×5cm square for further analyses.

Table 1. Characteristics of the collection sites.

District	City	Prefecture	Distance (km) from FDNPP	Direction from FDNPP	Pool side
Tohoku (northern Honsyu Island)	Morioka	Iwate	270	N	Yes
	Yokote	Akita	200	N	No
	Yokote	Akita	200	N	No
	Yamagata	Yamagata	100	NW	No
	Sendai	Miyagi	100	N	No
	Fukushima	Fukushima	50	NW	No
	Iwaki	Fukushima	40	S	Yes
Kanto (southeast of Honsyu Island)	Mouka	Tochigi	150	SW	No
	Tsukuba	Ibaraki	180	SSW	Yes
	Tsuchiura	Ibaraki	180	SSW	No
	Satte	Saitama	180	SSW	No
	Itabashi	Tokyo	220	SSW	No
	Itabashi	Tokyo	220	SSW	Yes
	Nakano	Tokyo	220	SSW	No
	Meguro	Tokyo	220	SSW	No
	Kunitachi	Tokyo	250	SSW	No
	Yokohama	Kanagawa	270	SSW	No
Tyubu (center of Honsyu Island)	Yamanashi	Yamanashi	300	SW	No
	Shizuoka	Shizuoka	370	SW	No
	Niigata	Niigata	100	WNW	No
	Nagoya	Aichi	450	SW	No
West Japan (west Honsyu, Shikoku, and Kyusyu Islands)	Osaka	Osaka	600	SW	No
	Tokushima	Tokushima	700	SW	No
	Nagasaki	Nagasaki	1,100	SW	No

FDNPP: Fukushima Daiichi Nuclear Power Plant, N: North, S: South, NW: North West, SW: South West, SSW:

South South West, WNW: West North West. Yes: Pool side collection, No: Not poolside collection.

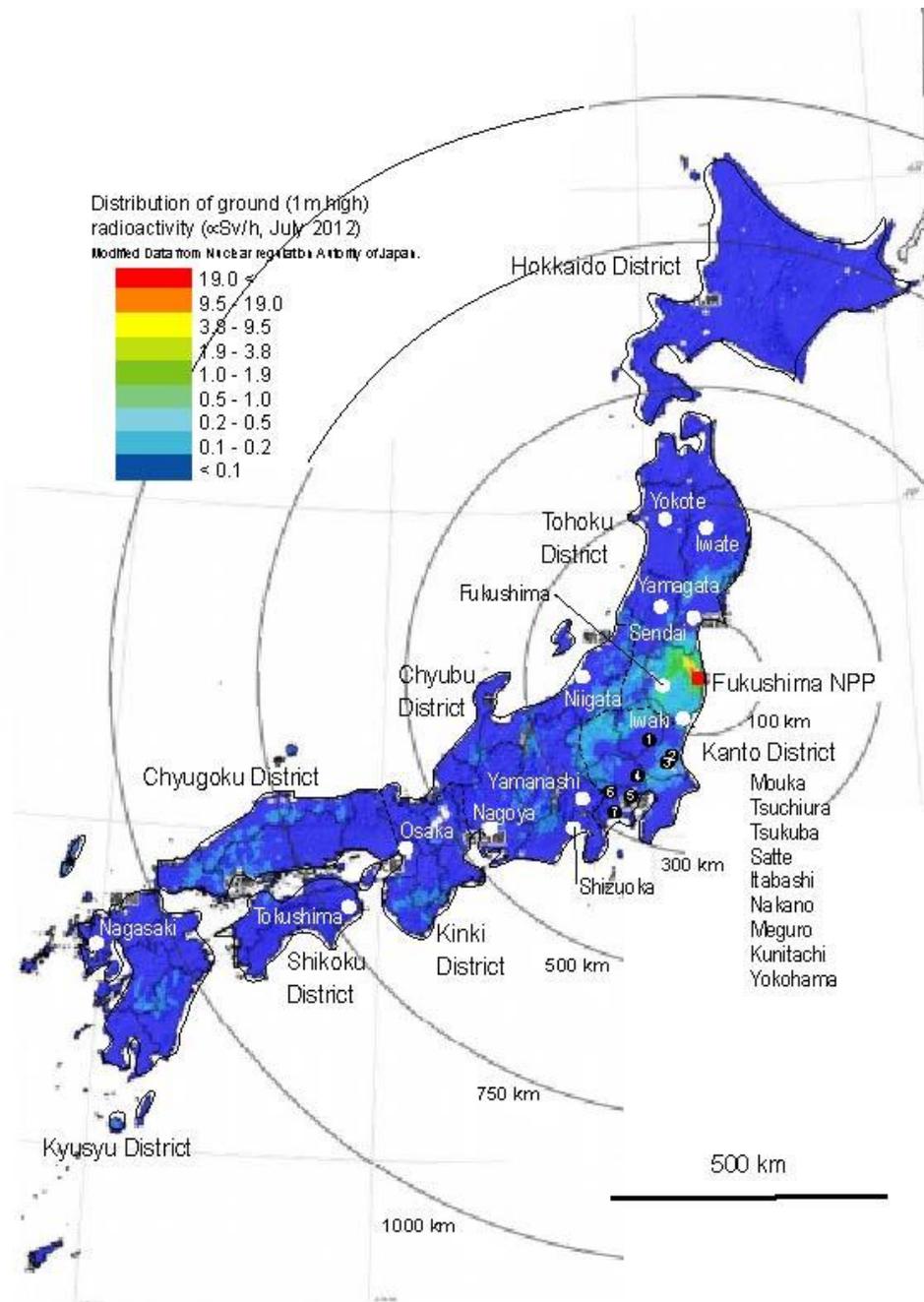


Figure 1. Estimated radioactivity of outdoor swimming pool water when daily migration, elimination by circulatory filtration, and occurred residual accumulation, and when stirred up the migrants by swimming.

Germanium gamma counting was performed for more than four hours. Peak counts were subtracted from background activity, and those values were divided by the counting time (sec). Divided values were then corrected for specific radiation probability (0.976 for 604keV (^{134}Cs); 0.851 for 661keV (^{137}Cs)), detection efficiency (0.0350 for 604keV; 0.0364 for 661keV), and the tank opening area (0.1908m²). Gamma-ray measurements were performed with a Ge detector (Princeton Gamma-Tech, Type: IGC25190, SC, USA). The polarity: P type, the Ge crystal active volume: 110 cm³, and the relative efficiency: 1332 keV at 25.6%. ^{131}I , the most important and harmful radionuclide

for our health, was not detected from the specimens, presumably because they will be decayed below the instrumental sensitivity.

In addition, we performed radio-counts for five sandy soil samples (Soil ① - Soil ⑤) collected from asphalt-covered pavement, and six (Soil ⑥ - Soil ⑪) ground-surface soils (less than 5mm depth) surrounding one university outdoor swimming pool in Tsukuba. The soil sampling sites and an example water trap site are shown in Figure 2. The collected soil samples were dried, homogenized, weighed (1.0 g), and wrapped in filter paper, and gamma-ray counted for more than four hours using the Ge detector.

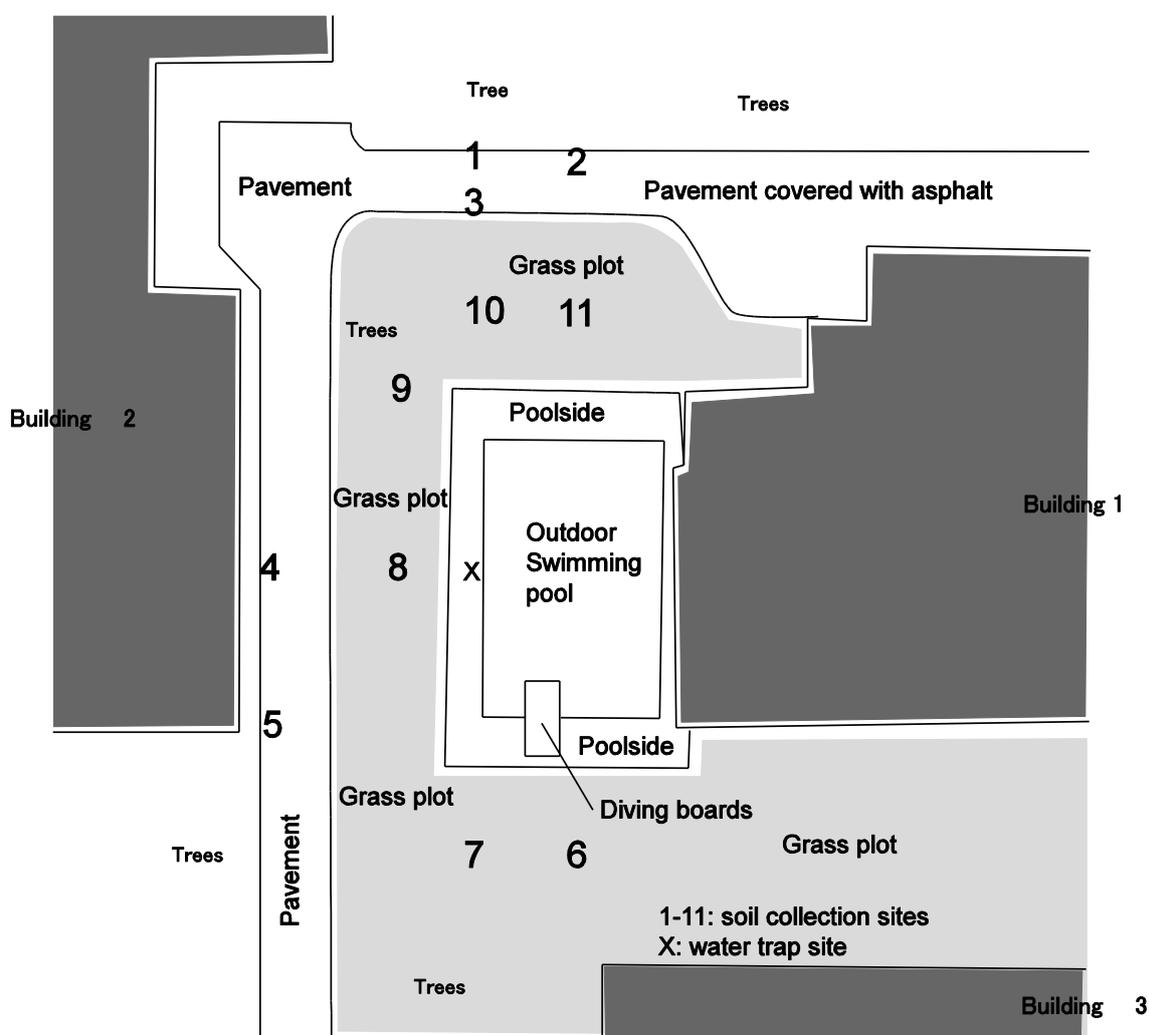


Figure 2. Soil and water-trap collection sites surrounding an outdoor swimming pool in Tsukuba University, Ibaraki Prefecture, 180 km SSW from an injured Fukushima Daiichi Nuclear Power Plant.

Results and Discussion

Radioactivity depositions ($\text{Bq/m}^2/\text{day}$) measured by water traps are shown in Table 2. The values varied by district, presumably because soil contamination was a function of a sample site's distance from the FDNPP, wind velocity, direction of contaminant release, and topography. The estimated radionuclide depositions for Kanto District, where more than 30% of the Japanese population resides, ranged from $0.02\text{Bq/m}^2/\text{day}$ (Satte, Saitama Pref.) to $4.60\text{Bq/m}^2/\text{day}$ (Tsukuba, Ibaraki Pref.). However, the majority of the values for this district ranged from 0.2 to $1.0\text{Bq/m}^2/\text{day}$. The values in Tohoku District, where Fukushima Prefecture and the FDNPP are located, ranged from 0.02 (Yokote , Akita Pref.) to $45.5\text{Bq/m}^2/\text{day}$ (Fukushima, Fukushima Pref.). Sites at Yokote (Akita Pref.) and Morioka (Iwate Pref.) had relatively low radioactivity deposition. These values were less than those of Kanto district Kanagawa Ibaraki, Tochigi and Tokyo Prefectures. Three locations in Fukushima Pref. also displayed relatively high levels of radioactivity deposition. Radionuclide deposition values obtained from other locations outside of Kanto and Tohoku Districts showed below $0.05\text{Bq/m}^2/\text{day}$, presumably because released radionuclide in the air was diluted owing to rain.

Table 2. Levels of radioactivity in water traps distributed throughout Japan.

Collection Cites			$^{134}\text{Cs} \pm 1\sigma$	$^{137}\text{Cs} \pm 1\sigma$	$^{134+137}\text{Cs}$	Notes
District	City	Prefecture	Bq/m ² /day corrected at 1 July 2012			
Tohoku (northern Honsyu Island)	Morioka	Iwate	0.14 ± 0.03	0.15 ± 0.02	0.30	Pool side
	Yokote 1	Akita	0.00 ± 0.11	0.00 ± 0.09	0.00	Near pool side
	Yokote 2	Akita	0.01 ± 0.07	0.00 ± 0.07	0.01	Near pool side
	Yokote 3	Akita	0.00 ± 0.10	0.00 ± 0.09	0.00	Near pool side
	Yamagata	Yamagata	0.79 ± 0.01	0.84 ± 0.01	1.63	3F, roof
	Sendai 1	Miyagi	0.43 ± 0.02	0.48 ± 0.01	0.92	Private garden
	Sendai 2	Miyagi	0.82 ± 0.01	0.86 ± 0.01	1.69	Private garden
	Fukushima 1*	Fukushima	3.72 ± 0.01	3.34 ± 0.00	7.06	Private garden
	Fukushima 2**	Fukushima	31.8 ± 0.00	30.7 ± 0.00	62.5	Private garden
	Iwaki	Fukushima	9.02 ± 0.00	9.49 ± 0.00	18.5	Pool side
Kanto (south east of Honsyu Island)	Mooka 1	Tochigi	0.11 ± 0.03	0.13 ± 0.02	0.24	Private garden
	Mooka 2	Tochigi	0.10 ± 0.03	0.12 ± 0.02	0.22	Private garden
	U-Tsukuba 1	Ibaraki	0.46 ± 0.02	0.49 ± 0.01	0.96	2F roof
	U-Tsukuba 2	Ibaraki	2.14 ± 0.01	2.46 ± 0.01	4.60	Pool side
	Tsukuba-school 1	Ibaraki	0.29 ± 0.02	0.32 ± 0.02	0.61	Private Garden
	Tsukuba-school 2	Ibaraki	0.14 ± 0.03	0.14 ± 0.02	0.28	Private Garden
	Tsukuba-school 3	Ibaraki	0.30 ± 0.02	0.34 ± 0.01	0.64	Pool side
	Tsuchiura	Ibaraki	1.48 ± 0.01	1.71 ± 0.01	3.19	Private garden
	Satte	Saitama	0.01 ± 0.07	0.01 ± 0.06	0.02	Private garden
	Itabasi-ku 1	Tokyo Metro	0.10 ± 0.04	0.11 ± 0.03	0.21	Private garden

	Itabasi-ku 2	Tokyo Metro	0.11 ± 0.03	0.13 ± 0.02	0.24	Pool side
	Meguro-ku	Tokyo Metro	0.20 ± 0.02	0.23 ± 0.02	0.44	Private garden
	Nakano-ku	Tokyo Metro	0.36 ± 0.02	0.38 ± 0.01	0.74	Private garden
	Kunitachi	Tokyo	2.11 ± 0.01	2.22 ± 0.01	4.32	Private garden
	Yokohama 1	Kanagawa	0.19 ± 0.03	0.19 ± 0.02	0.38	Private garden
	Yokohama 2	Kanagawa	0.25 ± 0.02	0.29 ± 0.02	0.54	Private garden
	Yokohama 3	Kanagawa	0.63 ± 0.01	0.67 ± 0.01	1.30	Private garden
	Shibata	Niigata	0.01 ± 0.07	0.00 ± 0.07	0.01	Private garden
Tyubu (center of Honsyu Island)	Yamanashi	Yamanashi	0.01 ± 0.07	0.01 ± 0.06	0.02	Private garden
	Shizuoka	Shizuoka	0.01 ± 0.07	0.01 ± 0.06	0.02	Private garden
	Nagoya	Aichi	0.02 ± 0.06	0.01 ± 0.06	0.02	Private garden
	Osaka	Osaka	0.00 ± 0.04	0.00 ± 0.03	0.00	Private garden
West Japan (west Honsyu, Shikoku, and Kyusyu Islands)	Tokushima	Tokushima	0.01 ± 0.04	0.01 ± 0.03	0.02	Private garden
	Nagasaki-1	Nagasaki	0.01 ± 0.08	0.00 ± 0.07	0.01	Private garden
	Nagasaki 2	Nagasaki	0.01 ± 0.08	-0.01 ± 0.08	0.00	5F, veranda

*152 days collected due to large snowfalls. ** 218 days collected due to large snowfalls.

Our water trap collected radionuclide depositions, presumably resuspended from fallout in the residing areas, agreed well with official data of ground soil contamination (Environmental Radioactivity Database, Figure 3).

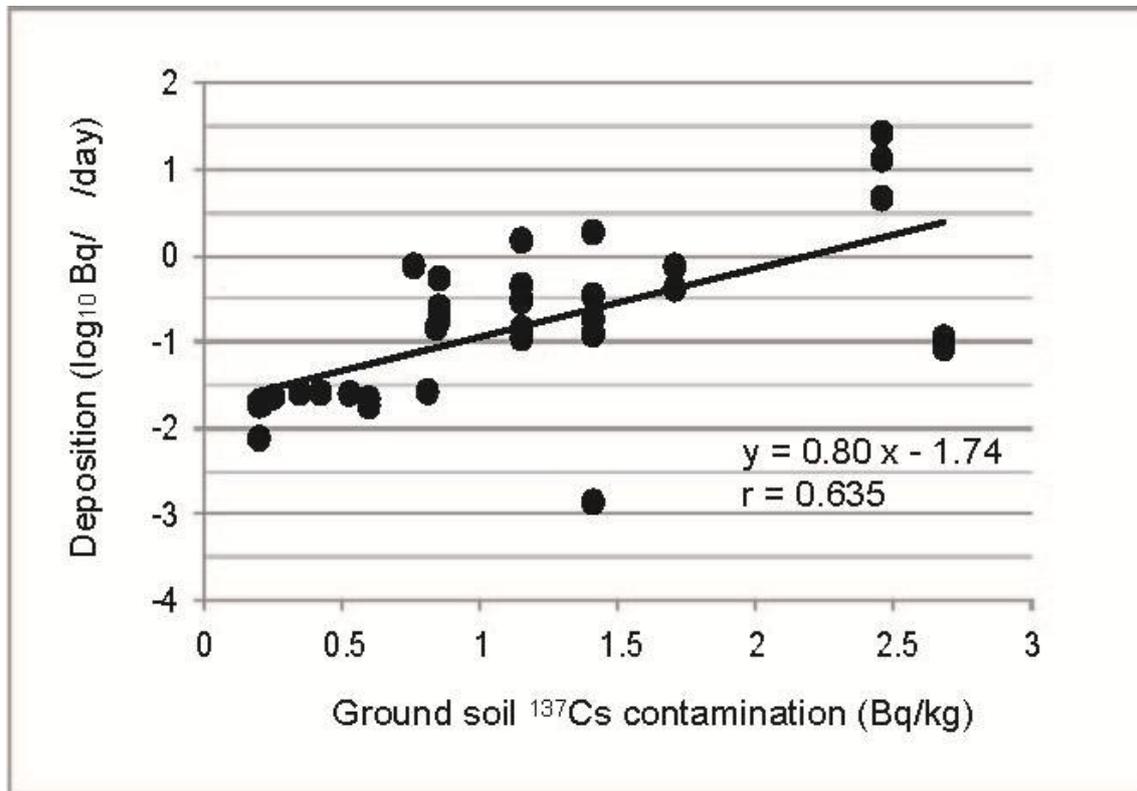


Figure 3. Correlation analysis between depositions into water traps distributed all over Japan, and soil radionuclide contamination measured in July 2011 on the corresponding area (Searched data from Environmental Radioactivity Database of Nuclear Regulation Authority, Japan).

Low radionuclide depositions were recorded at four sites, Yokote (0.02, Akita Pref., 200km), Shibata (0.03, Niigata Pref., 100km), Yamanashi (0.03, Yamanashi Pref., 300km), and Shizuoka (0.03, Shizuoka Pref., 370km), instead of same distance from injured FDNPP than Tokyo and Kanagawa Prefectures. These results were likely due to the fact that these locations, with the exception of Yamagata, were separated from the FDNPP by some large mountain chains.

Another study analyzing fallout in March 2011 (MEXT, March, 2011) showed that ¹³⁴Cs and ¹³⁷Cs levels were at 0.32MBq/km²/month and 0.35MBq/km²/month, respectively, in Sasebo, Nagasaki

Prefecture. This location is 50 km north of Nagasaki and more than 1000km far from FDNPP. In our data from Nagasaki ($0.01\text{Bq}/\text{m}^2/\text{day}$), deposition would be detected owing to the previous fallout.

We measured contaminant levels in an average-sized outdoor swimming pool ($25\text{m}\times 25\text{m}$) and found that radionuclide deposited into the water at a rate of $1.0\text{Bq}/\text{m}^2/\text{day}$ for an estimated $625\text{Bq}/\text{day}/\text{pool}$. The majority of radionuclide bonded with soil particles, sank, and accumulated at the bottom of the pool. Because pools tend to be shallow in Japan (1m depth) because of safer swim practices in school, it is likely that contaminated soil sediments will be stirred with swimming and come into contact with or be swallowed by swimmers. If, the sediments were distributed equally in the pool water by stirring due to students swimming and the pool operation was fixed (the turn-rate (Total circulation volume / pool volume): 4/day; hours used for swimming; six hours/day; daily elimination rate during stirring: 10% (Murotani, 1968)), the resulting radionuclide accumulation was estimated $380,000\text{Bq}/\text{pool}$ at 35 days after refilling. The value exceeds one tenth of the Japanese national guideline of MEXT. In the same manner, $500\text{Bq}/\text{m}^2/\text{day}$ deposition will exceeds the MEXT guideline ($10\text{Bq}/\text{l}$) within 30 days, if elimination from pool or prevention are not performed (Murase, et al., 2013).

The collecting method in this paper was not able to detect soluble ^{134}Cs and ^{137}Cs because scavenging (Evangelidou et al., 2009) was not applied on our collections. The present school swimming pool guide by MEXT Japan only provides guidelines on soluble or water radioactivity. Safer outdoor swimming pools must be realized through additional information on deposited radioactivity invasion. Water used to fill swimming pools is subject to current water quality laws; however, the direct deposition or transfer of radioactive contaminants into swimming pools from surrounding soils has not been considered under these laws. Given the potential human health impacts of swimming in radioactive water, an understanding of the quantities of radioactive materials that deposit or are transferred into swimming pools is critical for

maintaining recreational safety standards. Our largest radioactivity deposition (garden: 45.5Bq/m²/day; pool side: 16.9Bq/m²/day) was collected from a contaminated and residential area, these depositions will not exceed MEXT guideline without any other specific pool cleaning. The future restriction removal of contaminated area will be emerged larger radiation deposition or direct invasion than 500Bq/m²/day.

Ground-surface soils and litters surrounding the outdoor swimming pool tested in this study were contaminated following the FDNPP accident in March 2011. It is likely that radioactive materials deposited from the source of release due to wind and rain. The surface soils collected around the pool (Figure 3, ⑥ - ⑪) contained 200 to 2,700Bq/kg of radioactive contaminants, and soil particles collected from paved surfaces (① - ⑤) had levels of 12,000 to 38,000Bq/kg, which is about 60 to 200 times the present concentration. A pool-side trap (1m above the ground-surface) collected 4.60Bq/m²/day of Cesium, and a roof-top trap near the pool (10m above the ground-surface) contained 0.96Bq/m²/day of Cesium. This suggests that dust stirred by the wind is a major source of pool contaminants (Table 3). We recommend that pools be covered with a plastic sheet when not in use and that they be closed on windy days.

Table 3. Radioactivity levels for soils collected around a swimming pool in Tsukuba (Ibaraki Prefecture, Japan).

Location/Sample	$^{134}\text{Cs} \pm 1\sigma$ (Bq/kg)	$^{137}\text{Cs} \pm 1\sigma$ (Bq/kg)	$^{134}\text{Cs} + ^{137}\text{Cs}$ (Bq/kg)
Soil ①	16,300±11	14,500±15	30,800
Soil ②	6,600±22	5,900±29	12,500
Soil ③	9,100±17	9,400±22	18,500
Soil ④	18,400±11	18,400±14	36,800
Soil ⑤	18,100±11	20,000±13	38,100
Soil ⑥	80±220	100±250	200
Soil ⑦	390±110	400±140	800
Soil ⑧	270±130	400±150	700
Soil ⑨	190±150	220±190	400
Soil ⑩	710±70	970±80	1,700
Soil ⑪	1,400±50	1,300±70	2,700
Background	0±74	0±90	0

Location numbers①-⑪are indicated in Figure 1. Soils①-⑤): Sandy residues on pavement came from surrounding soils washed by rains. Soils⑥-⑪): Scraped ground-surface soils with plant litter. Values in this table were adjusted for decay by 1July 2011.

The data in this paper showed that the Japanese citizens have been facing post-emergency exposure situation (IAEA, 2005), today.

The sediment radioactivity of this pool before our study was 439Bq/kg water (equivalent 33,000Bq/m²), which exceeds the limit for safe swimming. We removed the sediments by two vacuum pool cleaners (Type: Compact 400, Mariner 3S AG) and found that the radioactivities were reduced by 41Bq/m² for a 98.88% cleaning efficiency. These vacuum cleaners, designed for daily use in indoor pools, over-flowed and stirred sediments when the filter capacity was exceeded. To compensate for this defect, we recommend that sediments be removed through a thorough, primary cleaning at the start of the recreational season.

Conclusion

The FDNPP failure released a huge radionuclide and contaminated Kanto and Tohoku districts where 30 or more million populations lived in. Magnitude of Gamma radiation in Sv unit, food, and water contaminations in Bq unit have been already pushed forward as they may affect directly health of the people. We do not know how the litter or the soil radioactive contamination affect our daily lives while it is well known that is a determinant factor of the magnitude of terrestrial gamma rays ($\mu\text{Sv}/\text{hour}$). Three Japanese authorities (Ministry of Education, Culture, Sports, Science and Technology, Ministry of Health, Labour and Welfare, and Ministry of the Environment) have their water guidelines for swimming pool or bathing. However, they have not been referred any other direct environmental radionuclide contamination except water supply. This study estimated a direct environmental deposition or transfer into swimming pool as $50\text{Bq}/\text{m}^2/\text{day}$, and showed that it will not exceed the guidelines when no other particular protection or elimination procedures. And, when 10 times more deposition occurred, this study concluded that our guidelines will be broken through within 30 days if any effective procedures are not introduced in the outdoor swimming pools. Today, the restricted area are diminished, schools in the area restarted sequentially. The swimming class participants must be protected from a huge radionuclide deposition.

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