J. RADIAT. RES., 44, 319–328 (2003) http://jrr.jstage.jst.go.jp

Resuspension: Decadal Monitoring Time Series of the Anthropogenic Radioactivity Deposition in Japan

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Atmospheric deposition/Resuspension/Half-residence time/^{137}Cs/^{90}Sr activity ratio/Soil dust

Monthly atmospheric depositions of ^{90}Sr and ^{137}Cs have been observed at the Meteorological Research Institute (MRI), Tsukuba, Japan. This study reports temporal trends and levels of ^{90}Sr and ^{137}Cs depositions in the 1990s. Although the current ^{90}Sr and ^{137}Cs concentrations declined dramatically, they have been found continuously in the deposition samples throughout the 1990s. During this period, the annual ^{90}Sr (^{137}Cs) deposits at MRI ranged from 70–180 (140–350) mBq/m^2/year. With a sufficiently long time series, the decreasing trend of the deposition evidently differs from the past stratospheric fallout; it is far slower. Thus, reservoirs other than the stratosphere provide small amounts of ^{90}Sr and ^{137}Cs to the atmosphere. A simple calculation clearly refutes the significance of the ocean as a potential source of airborne anthropogenic radioactivity. We will demonstrate that these radionuclides in the deposited materials originate from resuspension processes (soil dust suspension processes). The temporal trends of the time series monitoring reveal differences from those in the UNSCEAR Report 2000, which were predicted by a model that disregarded resuspension. The specific activity of ^{90}Sr (^{137}Cs) in the annual depositions exhibited a 10-year (20-year) half-life. Those data were comparable with values reported in the literature for the half-residence time (HRT) of ^{90}Sr and ^{137}Cs in Japanese surface soils. They were also comparable to those calculated from nationwide data of ^{90}Sr and ^{137}Cs concentrations in the surface soil (0–10 cm) obtained from the Ministry of Education, Culture, Sports, Science and Technology Environmental Radiation Database (the MEXT Database). Regarding the activity ratio of ^{137}Cs/^{90}Sr, the Japanese nationwide surface soil data collected during the 1990s in the MEXT Database (median: 5.3, n = 584) did not accord with that in the deposition samples (average: 2.1, n = 82) at MRI. This supports our previous hypothesis that Asian dust may transport a large fraction of anthropogenic radioactivity into the Japanese atmosphere. We need to study the fate of long-lived anthropogenic radioactivity dispersed in the environment over greater spatial and temporal scales.

INTRODUCTION

Investigating the current concentration level of ambient anthropogenic radionuclides is essential for evaluating the long-term effects on humans. At Meteorological Research Institute (MRI), Japan Meteorological Agency, we have conducted monthly monitoring of ^{90}Sr and ^{137}Cs depositions since 1957.[1–3] After the Chernobyl accident, no atmospheric nuclear tests were conducted, and no severe nuclear accident has occurred; there has thus been no additional serious atmospheric contamination. Accordingly, the present global atmospheric ^{90}Sr and ^{137}Cs concentration levels plummeted. Nevertheless, these radionuclides have still been detected in atmospheric deposition samples at the MRI.[4–8] We are endeavoring to derive numeric data using monthly monitoring information, so as to research the transport processes in the atmosphere. Such injection of anthropogenic radioactivity allows meteorologists to better understand the atmospheric transport processes (e.g. Refs. 9 and 10). However, the activities of many research institutes that contributed to the international monitoring networks for global fallout have waned. As far as we know, there are only a few Russian and German publications documenting continuous radioactivity deposition through the 1990s.[11,12] Our data contributes to that small group of publications. Even the United Nations Scientific Committee

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on the Effects of Atomic Radiation (UNSCEAR) Report 2000[13] does not account for the resuspension processes.

Because the atmospheric deposition material is no longer radioactive, we use the term "radioactivity deposition." As pointed out in our previous paper,[4] the decrease of radioactivity deposition at the MRI does not exhibit the stratospheric half-residence time of aerosols; it is far slower. The deposition through the stratosphere, which had been an important pathway of global radioactive fallout,[2] has diminished significantly. Hence, it becomes more and more obvious that the radioactivity is supplied from reservoirs other than the stratosphere. The deposition is, therefore, controlled by a process called resusupension,[4,5] which functions as follows. Radioactive materials deposited on the ground are suspended again in the air along with soil particles by storm activity, as described below. This paper describes monitoring data on radioactivity deposition during the 1990s and data analysis of the temporal trends in relation to resuspension.

Effluents exhausted from nuclear facilities into the air do not cause the current radioactivity deposition. In general, nuclear facilities in Japan do not enhance the radioactivity concentration level in our ambient air, with only a few exceptions [^{3}H, ^{14}C, ^{131}I, noble gases, etc. (see Tables 31–40, Annex C of Ref. 13)]. Even in the recent accident in Japan, the released radioactivity did not severely contaminate the surrounding environment. A former PNC fire and explosion at a peripheral facility of the Tokai fuel reprocessing plant occurred in 1997 and emitted GBq order or less of the anthropogenic radioactivity.[14] The JCO criticality accident in 1999 introduced little extended contamination into the environment.[15] East Asian nuclear facilities do not release a significant amount of radioactivity either (see Tables 31–40, Annex C of Ref. 13). However, there still remains substantial anthropogenic radioactivity in the environment from previous nuclear detonations (Annex C of Ref. 13).

MATERIALS AND METHODS

Monthly precipitation and dust deposits (total deposition) were collected using a 4 m^2 plastic open surface collector installed on a small building roof in the observation field of the MRI in Tsukuba (36°03′N, 140°08′E), 60 km northeast of Tokyo. The MRI is located in a planned city, Tsukuba, which is a typical Japanese suburb, surrounded by a combination of rice paddies, cultivated fields, residential areas, small woods and coppices. The total deposition sample was evaporated to dryness using a porcelain evaporation dish or an electronic vacuum-rotary evaporator (Eyela NE-12, Tokyo) with a 10 liter glass flask. The dried residue was packed into a plastic vessel and examined for ^{137}Cs by γ-ray spectrometry using intrinsic Ge detectors. Next, ^{90}Sr was determined by low-background γ-ray counting coupled with conventional radiochemical separation. The detection limits of the radionuclides were as low as 5 mBq gross. Although the determined values were all statistically significant, 11 of the ^{90}Sr data values (14%) and 14 of the ^{137}Cs data (17%) included over 10% and 25% counting errors, respectively. Data precision was confirmed through intercomparison runs.[16] Details of experimental procedures are described elsewhere.[4,16] All results were decay-corrected as of the end of the sampled month.

RESULTS

Table 1 summarizes data of the monthly ^{90}Sr and ^{137}Cs depositions observed at the MRI from 1993 to 1999, along with other relevant information, such as the monthly precipitation, etc. The annual ^{90}Sr (^{137}Cs) depositions ranged from 70 to 180 (140 to 350) mBq/m^2/y during the aforementioned period. Fig. 1 depicts a time series of those data since 1957. A few unexplained Sr anomalies[6] are omitted from the table and figure. As described in our previous papers, though the MRI moved from Tokyo to Tsukuba in 1980, the temporal record of radioactivity deposition is regarded as being continuous. As shown in the figure, the current monthly deposits of ^{90}Sr and ^{137}Cs are low, up to only a few tens of mBq/m^2/month, which is 10^{-4} to 10^{-5} lower than the maximum number recorded in the early 1960s.

DISCUSSION

Figure 2 displays the annual ^{137}Cs deposition temporal trends. Calculating the annual deposition is similar to averaging data in order to remove seasonal variations in a monthly time series. This calculation can demonstrate a temporal decreasing trend over a few years. As is apparent in the figure, the decrease of the annual radioactivity deposition observed at the MRI in the 1990s, especially after 1993, no longer exhibits the stratospheric half-residence time of aerosols, and is far slower. In the figure, a curve displays the typical stratospheric half-residence time of aerosols of about 1 year.[1,2,17,18] This indicates that the decrease in ^{137}Cs deposition is very slow, and that ^{137}Cs was supplied to the lower troposphere, from reservoirs other than the stratosphere. We can anticipate that other long-lived radionuclides, such as introduced into the environment from nuclear weapons tests and severe nuclear accident, would behave similar to ^{137}Cs due to the global transport and dispersion processes in the atmosphere. Strontium-90 and ^{137}Cs, having almost the same 30-year half lives, remained in the Earth's surface. From 1945 to 1997, Aoyama[19] estimated cumulative depositions of ^{137}Cs as 3.4 kBq/m^2 at the mid latitude in the Northern Hemisphere. The UNSCEAR Report states that, as of 2000, 250 PBq of ^{90}Sr and 1.5 times more of ^{137}Cs still exist (Annex C of Ref. 13). Regarding this, Fig. 1 reveals the absorption of ^{90}Sr and ^{137}Cs into the land near Tokyo from 1954 to the present. Consequently, contamination of ^{90}Sr and ^{137}Cs in the surface soil and waters is detectable in global samples.

Hence, there are only two major candidates as reservoirs of

Table 1. Monthly ^{90}Sr and ^{137}Cs depositions observed at the MRI, Tsukuba, Japan during 1993 to 1999 together with related informatiom

Year	Month	Res. wt. (g/m^2)	Depo. (mBq/m^2)	^{90}Sr error (mBq/m^2)	Specific act. (mBq/g)	Depo. (mBq/m^2)	^{137}Cs error (mBq/m^2)	Specific act. (mBq/g)	$^{137}Cs/^{90}Sr$	Monthly ppt. (mm)
1993	Jan.	6.80	9.9	0.7	1.5	14.6	2.9	2.2	1.48	91.0
	Feb.	2.87	10.5	0.3	3.7	19.4	3.5	6.8	1.84	64.0
	Mar.	6.09	25.2	0.6	4.1	55.6	5.9	9.1	2.21	58.5
	Apr.	4.77	23.0	1.4	4.8	54.2	4.0	11.4	2.36	35.5
	May	7.65	21.5	0.4	2.8	79.0	5.4	10.3	3.68	103.0
	Jun.	4.36	15.9	0.6	3.6	69.0	6.2	15.8	4.34	166.0
	Jul.	2.88	11.1	0.4	3.9	10.9	2.8	3.8	0.98	224.0
	Aug.	2.86	12.1	0.2	4.2	8.8	2.5	3.1	0.73	241.5
	Sep.	2.88	8.0	0.4	2.8	9.8	3.0	3.4	1.23	92.0
	Oct.	2.88	5.3	0.2	1.9	8.6	2.1	3.0	1.61	124.5
	Nov.	2.16	7.5	0.0	3.5	8.7	2.5	4.0	1.17	119.0
	Dec.	1.82	3.9	0.7	2.1	7.2	2.2	4.0	1.85	61.5
	Sum	48.0	153.9	3.2	345.9	6.4	1.96	1380.5		
1994	Jan.	3.17	20.1	0.7	6.3	82.7	4.1	26.1	4.12	45.5
	Feb.	5.31	15.9	0.5	3.0	45.0	7.1	8.5	2.83	71.5
	Mar.	4.44	11.6	0.4	2.6	22.0	3.6	5.0	1.90	116.0
	Apr.	5.86	20.0	0.6	3.4	39.9	4.7	6.8	2.00	39.0
	May	5.93	26.1	0.9	4.4	40.3	5.7	6.8	1.54	101.5
	Jun.	4.41	15.8	0.5	3.6	18.8	3.1	4.3	1.19	72.0
	Jul.	6.06	22.9	0.7	3.8	30.8	5.2	5.1	1.35	54.5
	Aug.	3.40	8.6	0.1	2.5	13.6	2.6	4.0	1.57	77.5
	Sep.	4.77	7.4	0.2	1.6	10.5	3.5	2.2	1.42	348.0
	Oct.	3.50	15.6	0.5	4.4	21.6	7.2	6.2	1.39	54.0
	Nov.	2.80	7.0	0.5	2.5	8.8	2.4	3.2	1.27	49.0
	Dec.	1.50	4.4	0.6	2.9	8.5	4.1	5.7	1.94	22.0
	Sum	51.1	175.3	3.4	342.6	7.0	1.9	1050.5		
1995	Jan.	1.23	4.5	0.1	3.7	11.5	3.0	9.3	2.56	27.5
	Feb.	2.05	8.5	0.3	4.1	13.4	2.6	6.5	1.59	15.5
	Mar.	3.73	23.9	0.2	6.4	19.9	3.4	5.3	0.84	142.0
	Apr.	4.00	19.1	0.3	4.8	26.7	3.6	6.7	1.39	110.5
	May	3.59	8.7	0.3	2.4	25.9	4.1	7.2	2.97	188.5
	Jun.	1.73	4.5	0.2	2.6	8.6	2.0	5.0	1.92	180.0
	Jul.	2.94	20.9	0.7	7.1	21.4	3.6	7.3	1.02	155.5
	Aug.	3.31	11.0	0.4	3.3	10.1	2.3	3.1	0.92	69.5
	Sep.	2.76	*	*	*	11.6	2.3	4.2	*	171.5
	Oct.	2.51	*	*	*	7.1	1.9	2.8	*	60.5
	Nov.	2.99	18.3	0.5	6.1	19.9	3.9	6.7	1.09	38.5
	Dec.	0.78	3.5	0.3	4.6	4.8	1.3	6.2	1.37	0.0
	Sum	31.6	122.9	4.5	181.0	5.9	1.6	1159.5		
1996	Jan.	2.35	19.7	0.6	8.4	16.0	3.3	6.8	0.81	17.0
	Feb.	2.52	12.3	0.6	4.9	27.1	4.1	10.8	2.20	32.0
	Mar.	3.55	11.4	0.4	3.2	34.6	4.5	9.8	3.04	95.0
	Apr.	3.73	14.5	0.3	3.9	33.9	4.0	9.1	2.34	66.0
	May	3.22	9.9	0.3	3.1	22.4	3.6	7.0	2.26	103.0
	Jun.	4.59	8.8	0.4	1.9	18.6	3.6	4.1	2.10	42.0
	Jul.	2.74	8.7	0.7	3.2	17.5	2.7	6.4	2.02	191.5
	Aug.	2.63	5.6	0.4	2.1	12.9	2.1	4.9	2.31	36.0
	Sep.	1.47	3.7	0.2	2.5	6.9	1.5	4.7	1.89	306.5
	Oct.	1.36	2.6	0.1	1.9	6.3	1.4	4.6	2.39	63.0
	Nov.	2.14	4.5	0.2	2.1	7.0	1.4	3.3	1.55	94.0
	Dec.	1.93	4.5	0.5	2.3	14.7	1.4	7.6	3.25	36.5
	Sum	32.2	106.3	3.3	218.0	6.6	2.2	1082.5		

Table 1. (continued) Monthly ^{90}Sr and ^{137}Cs depositions observed at the MRI, Tsukuba, Japan during 1993 to 1999 together with related information

Year	Month	Res. wt. (g/m^2)	Depo. (mBq/m^2)	^{90}Sr error (mBq/m^2)	Specific act. (mBq/g)	Depo. (mBq/m^2)	^{137}Cs error (mBq/m^2)	Specific act. (mBq/g)	$^{137}Cs/^{90}Sr$	Monthly ppt. (mm)
1997	Jan.	2.06	6.0	0.8	2.9	14.9	1.6	7.2	2.49	28.0
	Feb.	8.70	15.3	0.7	1.8	92.5	4.0	10.6	6.05	30.5
	Mar.	3.21	10.7	0.2	3.3	22.5	2.1	7.0	2.10	100.5
	Apr.	3.36	12.0	1.1	3.6	24.8	2.5	7.4	2.08	79.0
	May	4.67	12.9	0.3	2.8	34.8	3.4	7.5	2.71	154.5
	Jun.	3.21	7.2	0.2	2.2	17.3	3.1	5.4	2.42	161.0
	Jul.	2.98	11.6	0.7	3.9	9.8	1.8	3.3	0.84	95.5
	Aug.	2.41	6.1	0.3	2.5	8.7	1.7	3.6	1.43	35.5
	Sep.	5.49	*	*	*	15.7	2.9	2.9	*	148.0
	Oct.	4.35	4.9	0.7	1.1	17.3	3.2	4.0	3.57	21.5
	Nov.	2.19	5.8	0.5	2.7	6.5	1.6	3.0	1.12	102.0
	Dec.	1.42	3.9	0.3	2.8	4.5	1.3	3.2	1.15	46.5
	Sum	44.0	96.3	2.7	269.5	5.4	2.4	1002.5		
1998	Jan.	1.75	3.3	0.2	1.9	5.7	1.0	3.3	1.71	78.5
	Feb.	2.06	5.2	0.2	2.5	11.5	2.0	5.6	2.22	62.5
	Mar.	4.28	12.1	0.3	2.8	26.8	2.4	6.3	2.21	80.5
	Apr.	3.27	10.9	0.5	3.3	25.4	2.5	7.8	2.33	205.5
	May	2.25	5.2	0.4	2.3	10.7	1.8	4.7	2.04	148.0
	Jun.	2.48	4.5	0.3	1.8	6.4	1.1	2.6	1.42	155.5
	Jul.	2.14	5.5	0.3	2.6	9.7	1.4	4.6	1.76	144.0
	Aug.	2.35	5.0	0.6	2.1	5.3	0.7	2.3	1.06	199.5
	Sep.	3.47	6.5	0.2	1.9	7.9	1.3	2.3	1.21	275.5
	Oct.	2.39	4.8	0.5	2.0	4.5	0.6	1.9	0.94	128.0
	Nov.	1.71	3.9	0.1	2.3	9.0	0.9	5.2	2.30	3.0
	Dec.	2.42	7.9	0.2	3.3	18.8	1.4	7.7	2.38	42.5
	Sum	30.6	74.9	2.4	141.6	4.5	1.8	1523.0		
1999	Jan.	0.99	3.1	0.3	3.1	9.2	1.2	9.3	2.98	9.5
	Feb.	3.37	8.0	0.3	2.4	29.9	3.5	8.9	3.73	43.0
	Mar.	2.54	9.7	0.4	3.8	24.8	3.2	9.8	2.54	112.5
	Apr.	3.41	8.2	0.1	2.4	20.6	1.3	6.0	2.52	194.5
	May	2.58	6.5	0.3	2.5	16.8	2.6	6.5	2.58	101.0
	Jun.	5.25	6.4	0.3	1.2	15.5	1.2	2.9	2.42	149.0
	Jul.	7.01	21.4	0.3	3.1	66.0	4.9	9.4	3.08	196.0
	Aug.	2.87	10.0	0.8	3.5	12.5	2.0	4.4	1.24	129.5
	Sep.	6.47	*	*	*	14.3	1.2	2.2	*	50.0
	Oct.	4.57	*	*	*	13.9	2.6	3.0	*	89.5
	Nov.	1.56	5.2	0.4	3.3	4.2	0.5	2.7	0.81	77.5
	Dec.	1.69	3.7	0.5	2.2	6.9	1.4	4.1	1.86	12.5
	Sum	42.3	82.3	2.8	234.5	5.8	2.4	1164.5		

*A few anomalous ^{90}Sr data are not included.

environmental anthropogenic radionuclides: land and ocean. Because Japan is surrounded by the sea, sea salt could carry anthropogenic radioactivity. We determined that the ocean's contribution to radioactivity deposition is negligible, based on the sea-salt deposition at the MRI and contents of the radionuclides in the surface seawater. The Current concentration levels of ^{90}Sr and ^{137}Cs in the surface seawater are reported to be 2 mBq/L[20] and 3 mBq/L.[21] The annual deposition of Na at the MRI is about 1 g/m^2/year (our unpublished data). The weight composition of Na in sea salt is 30%.[22] Thus, the annual deposition of bulk sea salt is 3 g/m^2/year, which corresponds to 0.1 liter seawater annually poured onto 1 m^2 of land surface of Tsukuba. Therefore, the annual ocean deposition of ^{90}Sr and ^{137}Cs would be less than 0.2 and 0.3 mBq/m^2. These correspond to less than 0.3% and 0.2% of the annual ^{90}Sr and ^{137}Cs depositions at the MRI in Tsukuba, and are therefore negligible.

In the 1990s, median of the ^{90}Sr and ^{137}Cs concentrations in

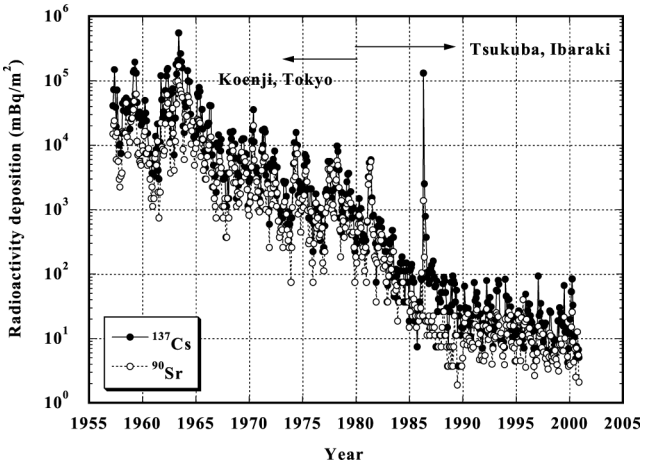

Fig. 1. Temporal variation in monthly ^{90}Sr and ^{137}Cs depositions observed at the Meteorological Research Institute since 1957. Some data have only one effective digit in an obsolete Ci unit, which cannot be converted to Bq unit that are more effective. A few anomalous ^{90}Sr data are not plotted. These are minor limitations; the temporal trends in radioactivity deposition are well documented in the monitoring record.

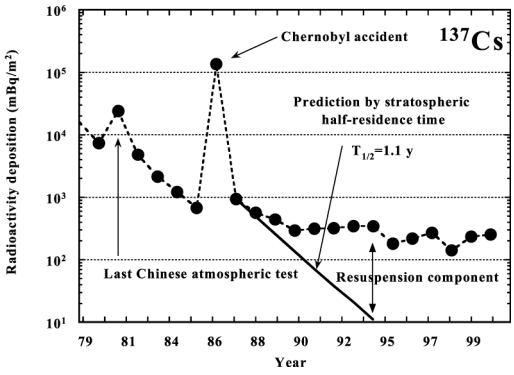

Fig. 2. Temporal trends in the annual ^{137}Cs deposition observed at the MRI, Tsukuba, Japan, during the 1980s to the 1990s. These trends show persistent radioactivity deposition at low levels in the 1990s, due to resuspension.

Japanese surface soils were 2 and 13 mBq/g (see later discussion and figures). Because the soil dust contained in the atmospheric deposition becomes visible, such soil dust increases the anthropogenic radioactivity in the sample. Consequently, we reasonably assume that those anthropogenic radionuclides were resuspended with surface soil dust, and that resuspension is the major factor in radioactivity deposition. In other words, suspended soil dust (surface soil particles) is the source of anthropogenic radionuclides in ambient air.

The above-mentioned temporal trends in radioactivity deposition seem to be quite different from prediction by a model calculation described in Fig. V, Annex C of Ref. 13, and its supporting paper with equivalent figures.[23] The calculation has been based on the monitoring data up to 1985, which has not yet included resuspension phenomena. The resuspension processes are thus not included in the calculation. When considering future long-term consequences and effects of anthropogenic radioactivity on humans, we need to incorporate the resuspension processes into the model analysis.

Temporal features of our monitoring record will be generalizable to other global sites. Gritchenko *et al.*[11] reported about ^{137}Cs deposition at Zelenogorsk (50 km northwest of St. Petersburg, Russia). The temporal trends in monthly ^{137}Cs deposition decreased just after the Chernobyl spike until 1992 or 1993. In following years, however, the decreasing trend slowed. Compared with the activity level, the present Japanese data seem to be at least one order of magnitude lower. No Russian data from the 1990s are as low as a few tens of mBq/m^2/month, most likely due to the effect of the local resuspension of the Chernobyl ^{137}Cs contamination. A similar report was made by Rosner and Winkler[12] on a site near Munich, Germany. They have continued observing activity concentrations of ^{90}Sr, ^{137}Cs and Pu isotopes in aerosol samples and atmospheric depositions since 1970. Because their data are depicted in figures, decay-corrected to May 1986, and account for the effect of Chernobyl-derived radioactivity, a direct comparison with our data is problematic. Annual depositions of ^{90}Sr, ^{137}Cs and ^{239,240}Pu decreased slowly after the primary effects of the Chernobyl accident had waned and were reported as 280, 3800 and 2.1 mBq/m^2/year in 1998. Annual ^{137}Cs depositions are one order of magnitude higher for the Russian data than for our Japanese data.

Atmospheric deposition is generally composed of dust, soot, and sea salt. It always contains soil dust. The land surface has been contaminated by anthropogenic radioactivity by the past global radioactive fallout. Hence, resuspension (surface soil dust suspension) must be a universal process injecting detectable amounts of anthropogenic radioactivity into the ambient air. The surface air contamination, even though extremely low, would be prolonged for several decades after cessation of the direct input of anthropogenic radioactivity into the atmosphere. In addition, the health effects to the individual member of the public of exposure to such resuspension are insignificant at the current level found at the MRI in Japan. Nevertheless, knowing the current atmospheric background level is essential to prepare for any nuclear emergency.

Figure 3 depicts the annual ^{137}Cs deposition, weight of the deposition residue and specific activity (^{137}Cs activity over total weight of the residue) with a fitting curve for specific activities from 1992 to 1999. The deposition rate was little affected by the stratospheric ^{137}Cs during this period. A 22-year half-life was obtained for ^{137}Cs deposition. Similar calculations for ^{90}Sr deposition resulted in a half-life of 10 years (Fig. 4). The temporal decrease may reflect a loss of radionuclides in the resuspension reservoir. Therefore, the half-residence time (HRT) of ^{90}Sr and ^{137}Cs in the surface soil could be

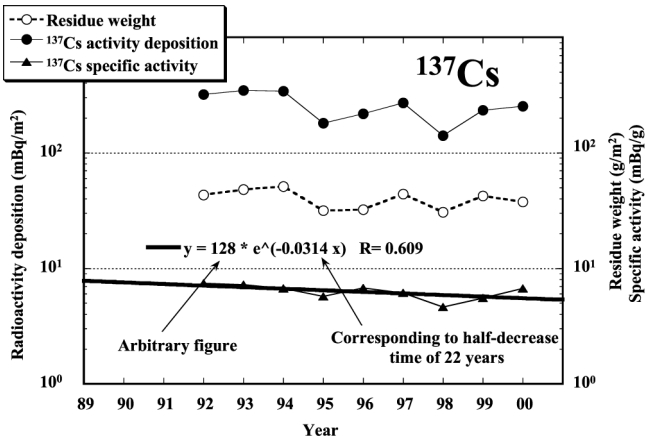

Fig. 3. Annual ^{137}Cs radioactivity deposition, annual deposition material weight, specific activity and estimation of apparent half-decrease time for specific activity (^{137}Cs activity/total weight of the deposition).

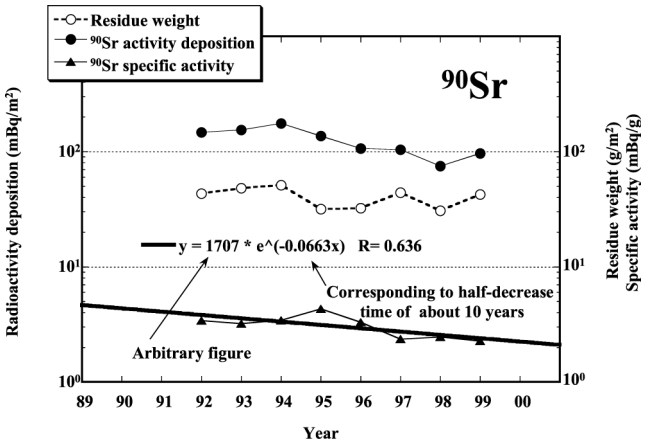

Fig. 4. Annual ^{90}Sr radioactivity deposition, annual deposition material weight, specific activity and estimation of apparent half-decrease time for specific activity (^{90}Sr activity/total weight of the deposition). The anomalous monthly data were evaluated by averaging the data obtained in that month and summed to the corresponding annual data.

Table 2. Comparison of the estimated half- residence time of total ^{90}Sr and ^{137}Cs deposits in Japan and those in Japanese surface soils.

Radionuclides and samples	Sampling site	Annual ppt. (mm)	Apparent half-residence time (y)	Literature
^{90}Sr				
Deposition sample	MRI, Tsukuba	1,236*	10	Present work
^{90}Sr				
Various surface soils (0–10 cm)	MEXT Database, nationwide	1,650**	11.1	Present work
Field surface soil (0–10 cm)	MEXT Database, nationwide		10.7	
^{90}Sr (exchangeable)	Sea of Japan side	–	7.4	Kobayashi *et al.*
Rice paddy (Plow layer)	4 sites		(5.8–10.3)***	(1984)
	Pacific Ocean side	–	6.6	Ref. 25
	9 sites		(4.6–10.5)***	
	Nationwide	1,650**	6.7	
^{90}Sr (exchangeable)				Kobayashi *et al.*
Upland fields (Plow layer)	Nationwide 11 sites	1,650**	7.3	(1984)
			(4.2–15.8)***	Ref. 25
^{137}Cs				
Deposition sample	MRI, Tsukuba	1,236*	22	Present work
^{137}Cs				
Various surface soils (0–10 cm)	MEXT Database, nationwide	1,650**	14.4	Present work
Field surface soil (0–10 cm)	MEXT Database, nationwide	1,650**	16.2	
^{137}Cs	Sea of Japan side	2,350	14.6	Komamura *et al.*
Rice paddy soil (0–20 cm)	6 sites		(10.6–26.6)***	(1999)
	Pacific Ocean side	1,350	21.3	Ref. 24
	8 sites		(10.3–27.4)***	
	Nationwide	1,650**	16.5	

*30 years average at Tateno (Aerological Observatory). **Median of annual precipitation over 30 years from 157 weather observation sites in Japan. ***Range (minimum to maximum).

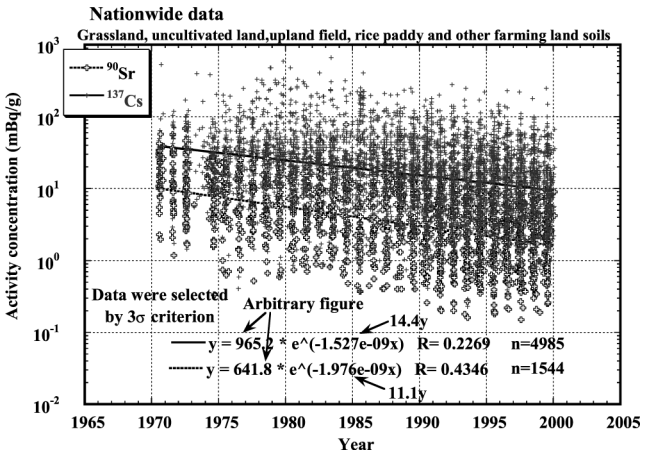

Fig. 5. Temporal trends in activity concentrations of ^{90}Sr and ^{137}Cs in Japanese surface soils (0-10cm thickness) collected during the 1970s to the 1990s (data were obtained from the Ministry of Education, Culture, Sports, Science and Technology Environmental Radiation Database).

compared with the half-life from the same standpoint (hereafter the half-life is also expressed as the HRT). We do not adopt the term "ecological half-life"[12] in the present text. The term "ecological" would be ambiguous, since it expresses biological, chemical and physical processes all together.

Table 2 compares Japanese surface soils from the literature[24,25] and those from this research. The apparent HRT was also evaluated in the present study (Fig. 5) from nationwide data of ^{90}Sr and ^{137}Cs concentrations in the surface soil (0–10 cm) obtained from the Ministry of Education, Culture, Sports, Science and Technology Environmental Radiation Database (the MEXT Database) maintained by the Japan Chemical Analysis Center (http://search.kankyo-hoshano.go.jp/servlet/search.top). The HRTs for the MEXT Database data were calculated by the least-square fitting of exponential functions and parameter conversion in years. The HRTs for ^{90}Sr and the MEXT Database data are in accord, but ^{137}Cs differs slightly. The removal of anthropogenic radioactivity depends on the chemical properties of the radionuclides in question, soil properties, and the annual precipitation rate. It is noteworthy that all HRTs for ^{90}Sr are shorter than those for ^{137}Cs. This detail corresponds with the established fact that ^{90}Sr migrates more rapidly than ^{137}Cs in the soil layer when accompanied by water movement.[18,26,27] Cs-137 is strongly adsorbed in clay and organic matter and is non-exchangeable. This is why ^{137}Cs is used as a tracer for soil erosion and sedimentation.[28] For ^{90}Sr, the exchangeable fraction is considered to be significant for the food chain (e.g. Ref. 25). For this reason, they obtained an exchangeable-fraction ^{90}Sr loss in the plow layer of cultivated soil, which shows a different temporal trend from the total trend.

HRTs for ^{90}Sr and ^{137}Cs depositions are the same order of magnitude as those obtained for Japanese surface soil layers. This is further proof that the ^{90}Sr and ^{137}Cs found in atmospheric depositions are derived from airborne resuspension (dust suspension). However, the HRTs are not helpful in locating the site of resuspension.

There are limitations for the HRT argument. Although there have been several publications regarding HRTs in various surface soils (e.g. Refs. 29–32), it is difficult to define the HRT on the same scale. HRTs are practically different, when considering the quality of surface soil composition and depth of soil or plow layer (0–5 cm or 0–20 cm). The data must thus be normalized. It is also practically difficult to define how much annual erosion would occur in the resuspension processes (soil dust suspension processes). In regions where soil erosion is significant, we need to consider this process. When we evaluate resuspension, we need to know if the erosion is negligible or significant. A direct comparison of these HRTs will not be sufficiently meaningful for identifying the origin of resuspension. There are limitations in utilizing HRTs to locate the site where major resuspension occurs. Employing the ^{137}Cs/^{90}Sr activity ratio is more useful.

In order to investigate the origin of resuspension, Igarashi *et al.*[7] compared the ^{137}Cs/^{90}Sr activity ratios in atmospheric deposition samples with those in various Japanese surface soil samples. They found that in the 1990s, the average activity ratio of ^{137}Cs/^{90}Sr in the deposition samples at the MRI (average: 2.1, n = 82) was far from those of paddies and field soils taken around the MRI (median: 6.8, n = 8), which had been considered the primary source for resuspension. The frequency distribution of the ^{137}Cs/^{90}Sr ratio in the deposition samples was narrow (0.6 to 6.0), while the frequency distribution of the surface soils ranged up to 60. Additionally, no road or roof dust, once considered likely sources, exhibited low activity ratios of 2 to 3.

This previous finding was further confirmed in the present study. The nationwide Japanese surface soil data collected during the 1990s (from the MEXT Database) had a median ^{137}Cs/^{90}Sr ratio of 5.3 (n = 584). The frequency distribution of the MEXT Database data was exhibited the higher ratio and some data even exceeded 100 (log-normal like distribution). ^{90}Sr and ^{137}Cs deposited and adsorbed in soil particles during the 1960s and 1970s have thus been gradually fractionated through leaching processes in Japan. The original ^{137}Cs/^{90}Sr activity ratio was purported to be 1.5 or 1.6.[13,18] These findings demonstrate that the radioactivity deposition in Tsukuba is not composed of a single local resuspension component, but is a mixture of components and that there must be remote sources responsible for the resuspension.

From the viewpoint of the atmospheric carrier of ^{90}Sr and ^{137}Cs, the resuspension corresponds to soil-dust suspension. It is, therefore, natural to consider large-scale meteorological dust events (e.g. the Kosa phenomena) in Asia[33–35] as remote-source candidates. Igarashi *et al.*[4] were first to assert the following hypothesis regarding common seasonal features in radioactivity deposits during Asian dust events. The Asian

dust transport is active in spring in the East Asia. However, weak events[36] also seem to contribute, even in summer. Igarashi *et al.*[7] performed the activity measurement to obtain $^{137}Cs/^{90}Sr$ ratios in soil samples collected in the arid area of the Asian continent[37,38]. They showed that the activity concentrations of ^{137}Cs in the continental samples were almost the same as those in Japanese surface soils, with higher activity concentration levels of ^{90}Sr. The median $^{137}Cs/^{90}Sr$ activity ratio was 2.0 (n = 4), exhibiting a lower ratio as close to the original value (possibly 1.6 as referred previously) in the arid area. The low precipitation probably prevented fractionation between these two radionuclides. Igarashi *et al.*[7] then evaluated how much radioactivity is elicited by the Asian dust by assuming a simple two-component model of isotope dilution equations and substituting the measured $^{137}Cs/^{90}Sr$ activity ratios into the equation. According to them, the Asian dust may transport about 90% of ^{90}Sr and 70% of ^{137}Cs depositions observed in Tsukuba in the 1990s.[7]

Although precise estimations of the Asian dust contribution and specific source areas are important, these are beyond the scope of the present paper. However, we would like to point out the importance of Asian dust. Precipitation events undoubtedly transport dust from the upper atmosphere to the Earth's surface. Wet deposition may have a greater influence on the composition of total deposits. We need more analysis of the $^{137}Cs/^{90}Sr$ activity ratios in surface soils collected in various regions over the Asian continent to prove this hypothesis.

A general circulation model (GCM) study will also help to better understand the processes (e.g. Ref. 39). In the year 2000, they compared the GCM calculation of monthly total deposited dust (weight) with the measurements at three stations over the Japanese islands, Yonagauni, Nagasaki and the MRI in Tsukuba. Model calculations and measurements were in good accordance with each other with just minor seasonal differences. This is further evidence for the hypothesis that Asian dust may transport a large portion of the anthropogenic radioactivity found in atmospheric deposition. Dust emission within Japan is very low in the GCM calculation. In particular, Yonaguni and Nagasaki are hardly affected by Japanese local dust in the calculation. Hirose *et al.*[40] has further argued that the monthly $^{239,240}Pu$ depositions, obtained from the same samples described here, exhibit a typical seasonal variation of spring maximum (March to April). This seasonal pattern also corresponds to seasonal cycles of soil dust transport originating from the East Asian arid area. Our monitoring records of ^{90}Sr and ^{137}Cs are obtained at only one site, with time trends in radioactivity deposits applying to a relatively large spatial scale.

HRTs of the ^{90}Sr and ^{137}Cs depositions are significantly shorter than their physical half-lives of 30 years. This may suggest that ^{90}Sr and ^{137}Cs migrate deeper into the surface soil layer in the source area of resuspension. This may contradict the previous discussion. The arid and semi-arid areas would produce a modicum of precipitation that could not cause the fractionation between ^{90}Sr and ^{137}Cs in the surface soil. This question remains as a future area of study.

In conclusion, the long-term monitoring of ^{90}Sr and ^{137}Cs depositions in Japan has revealed that the resuspension processes inject anthropogenic radionuclides into air. The half-life for ^{90}Sr and ^{137}Cs depositions were 10 and 20 years, which are comparable to the half-residence time of ^{90}Sr and ^{137}Cs in Japanese surface soils obtained from the MEXT Database data and from the literature. However, there may be a limitation to using the HRTs to locate the source area and to quantify the amount of resuspension components. It was pointed out that the $^{137}Cs/^{90}Sr$ activity ratio seems to be more valuable than the HRTs for studying this. We need to consider larger spatial scales and longer time scales in studying atmospheric anthropogenic radioactivity. Thus, we need to continue the long-term monitoring to asses future environmental impacts and health risks.

ACKNOWLEDGEMENTS

This study was funded in part by the Radioactivity Survey in Japan provided by the former Science and Technology Agency (currently Ministry of Education, Culture, Sports, Science and Technology). Ms. Inukai and Ms. Togashi of KANSO helped in the tedious sample preparation, for which the authors are truly grateful.

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Received on February 17, 2003
1st Revision on September 3, 2003
Accepted on September 3, 2003