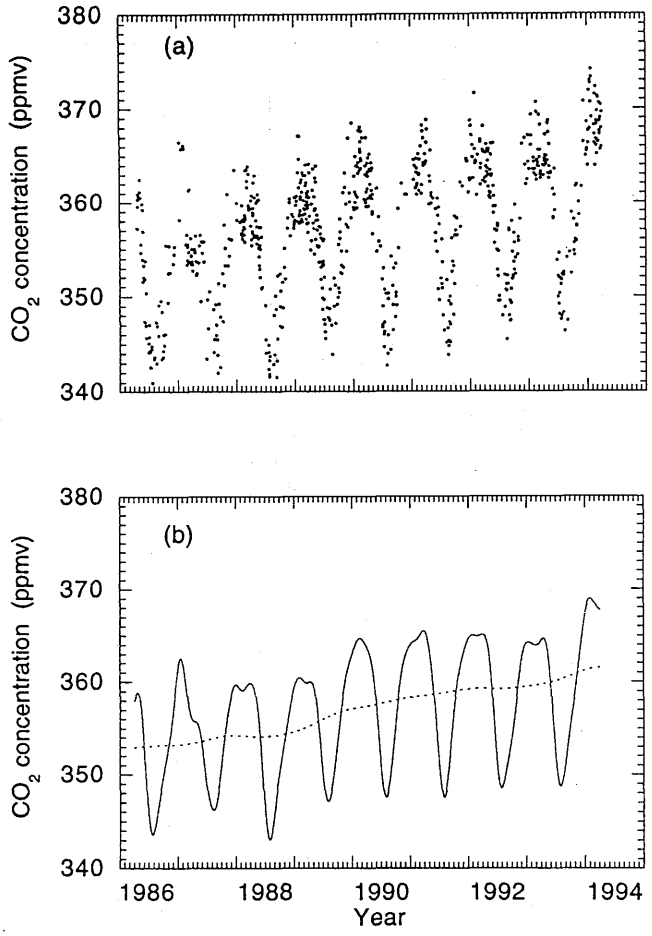

Fig. 95-6 Temporal variations in daily average when hour-to-hour changes are less than 1 ppmv (Cp) between 11:00 JST and 16:00 JST. (a) displays all raw data from April 13, 1986, to March 31, 1994. The solid curve in (b) is for a low-pass filter with cutoff at 100 days and the dashed curve is for cutoff at 667 days.
Reprinted from *J. Atmospher. Chem.*, **23** (1996), 137-161, Variations in atmospheric CO_2 at the Meteorological Research Institute, Tsukuba, Japan, Inoue and Matsueda, fig. 7 (©1996 Kluwer Academic Publishers. Printed in the Netherlands.) with kind permission from Kluwer Academic Publishers.

RADIOACTIVE NUCLIDES (^{90}Sr, ^{137}Cs, and Pu Isotopes)

3. Radioactive Materials in Air, Fallout, Rainwater, and Seawater

Global radioactive contamination derived from the fallout of nuclear weapon tests, waste disposal, or accidents at nuclear facilities has brought about scientific and social consequences. To evaluate the feedback effect on society and to trace the fate in the global environment, meteorological and oceanographic studies on the environmental radioactivity have been conducted for the last 40 years.

3.1 Geochemical studies on Chernobyl radioactivity in environmental samples

Hirose (1995a)

The Chernobyl accident on April 26, 1986, contaminated a wide area of the Northern Hemisphere. GRD scientists have comprehensively studied the environmental effect of Chernobyl fallout. Pronounced, high-level radioactivity has been observed in surface air and rainwater in Japan since May 3, 1986 and higher concentrations of Chernobyl radionuclides in surface air continued until early July 1986 (Aoyama *et al.*, 1986). Chernobyl fallout caused partly increasing ^{137}Cs concentrations in surface water and riverwater (Hirose *et al.*, 1990). They obtained some new evidence about the transport of Chernobyl radioactivity to the stratosphere, dry deposition, and wet deposition (Aoyama *et al.*, 1987; Aoyama *et al.*, 1991; 1992; Hirose *et al.*, 1993).

Hirose (1995a) described the deposition of ^{137}Cs, ^{90}Sr, and plutonium isotopes at Tsukuba in 1986 and summarized geochemical studies of Chernobyl fallout in Japan (Table 95-3). Artificial radionuclides in deposition and airborne dust samples in 1986 were measured at Tsukuba and 11 stations in Japan. Significant amounts of ^{238}Pu and ^{241}Pu were detected in deposition samples in May 1986 at Tsukuba. $^{238}Pu/^{239,240}Pu$ and $^{241}Pu/^{238}Pu$ ratios in monthly deposition show that meaningful amounts of Chernobyl-derived plutonium isotopes were transported to Japan, about 8,000 km distant from Chernobyl. Hirose found that the $^{241}Pu/^{239,240}Pu$ activity ratio was useful as a geochemical marker because its isotopic ratios changed significantly for different sources of plutonium.

The Chernobyl ^{90}Sr and plutonium isotopes, especially Pu, were preferentially scavenged from the atmosphere by wet and dry deposition, compared to volatile radionuclides such as ^{131}I and ^{137}Cs. This is due to the particle size difference between radionuclide-bearing aerosols; the order of particle size was Pu isotopes > ^{90}Sr > ^{137}Cs (Fig. 95-7). These findings suggest that large amounts of actinides were deposited near the accident site. This will require an assessment of the environmental effects of actinides because of their high toxicity and long radioactive life.

Table 95-3 Monthly Deposition of ^{137}Cs, ^{90}Sr, and Plutonium Isotopes at Tsukuba in 1986

Month	^{137}Cs	^{90}Sr	$^{239,240}Pu$	^{238}Pu
	(Bq m^{-2})		(mBq m^{-2})	
January	0.036±0.010	0.021±0.003	0.065±0.013	-
February	0.083±0.016	0.027±0.004	0.343±0.060	-
March	0.080±0.016	0.094±0.011	1.02±0.11	0.061±0.020
April	0.097±0.013	0.024±0.002	0.155±0.023	-
May	131±13	1.24±0.14	0.650±0.045	0.107±0.020
June	2.51±0.27	0.164±0.019	0.075±0.014	0.021±0.010
July	0.783±0.084	0.015±0.002	0.069±0.012	0.020±0.010
August	0.369±0.045	0.018±0.002	0.133±0.019	0.010±0.007
September	0.087±0.014	0.011±0.002	0.148±0.021	0.017±0.009
October	0.106±0.020	0.016±0.002	0.266±0.027	0.030±0.010
November	0.071±0.012	0.019±0.002	0.129±0.019	0.025±0.010
December	0.078±0.016	0.010±0.001	0.150±0.020	0.025±0.009

The data of monthly ^{137}Cs and ^{90}Sr were cited from the results of Aoyama, *et al.*, 1991.

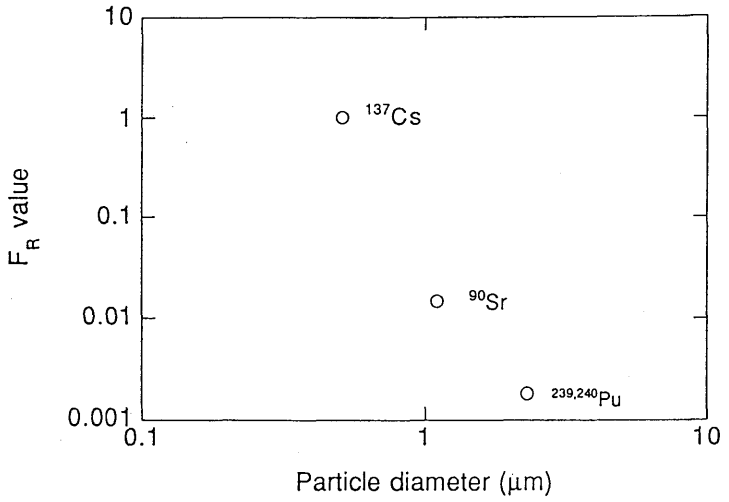

Fig. 95-7 F_R and particle size of radionuclide-bearing particles.

3.2 ^{137}Cs concentration temporal and spatial variation in western North Pacific and marginal seas from 1979 to 1988

Aoyama and Hirose (1995)

After the Chernobyl accident in May 1986, intensified observation was conducted to study the geographical distribution of Chernobyl radioactivity. Aoyama and Hirose (1995) studied temporal and spatial variation of ^{137}Cs concentration in the western North Pacific and its marginal seas during the period from 1979 to 1988. ^{137}Cs concentrations in surface water along the 137°E transect from 1979 to 1988 were 5.0–10.6 mBql^{-1} between 30°N and 24°N, 3.4–7.8 mBql^{-1} between 24°N and 7°N, and 2.1–6.9 mBql^{-1} between 7°N and the Equator (Fig. 95-8). ^{137}Cs concentrations at stations between 30°N and 7°N did not show clear temporal variation, while those at stations south of 7°N increased from 1986 to 1988.

The ^{137}Cs concentration south of 7°N became the same magnitude as those in two latitude bands between 30°N and 7°N in 1987 and 1988. Chernobyl-derived ^{134}Cs was detected in surface water of marginal seas around Japan north of about 30°N, which reflects the meridional distribution of Chernobyl-derived ^{134}Cs in surface air over the western North Pacific (Fig. 95-9). The particulate cesium isotope concentration ranged from 0.1 to 1.0 % of the total cesium isotope concentration in 1986 and 1987.

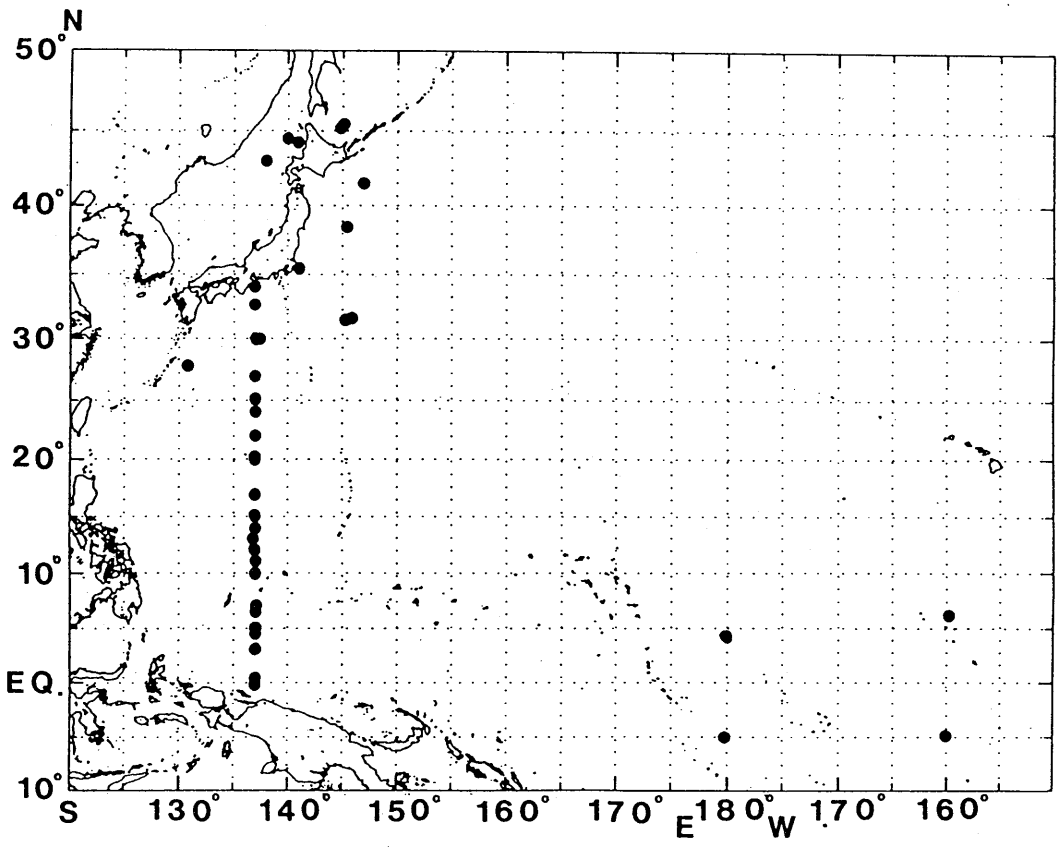

Fig. 95-8 Sampling sites.
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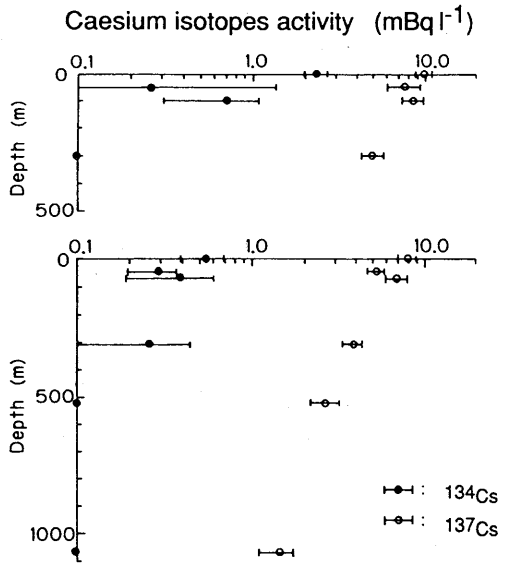

Fig. 95-9 Vertical distribution of ^{137}Cs and ^{134}Cs on August 22, 1986, at 44°30′N, 140°00′E (upper panel) and on August 26, 1987, at 44°15′N, 140°58′E (lower panel). Reprinted from *J. Environ. Radioactivity*, **29**, Aoyama and Hirose, The temporal and spatial variation of ^{137}Cs concentration in the western North Pacific and its marginal seas during the period from 1979 to 1988, 57-74, Copyright (1995), with permission from Elsevier Science.

3.3 Recent ^{90}Sr and ^{137}Cs deposition observed in Tsukuba

Igarashi, Otsuji-Hatori, and Hirose (1996)

Igarashi *et al.*(1996) studied recent deposition of ^{90}Sr and ^{137}Cs observed in Tsukuba. Referring to the trend in annual radioactivity deposition observed at the MRI since 1957, they showed the annual deposition of ^{90}Sr in 1990s to be as low as around 0.15 Bqm^{-2} and that of ^{137}Cs to be 0.3 Bqm^{-2} (Fig. 95-10). This is because no atmospheric nuclear weapons tests have been conducted since 1981. Although the Chernobyl accident brought a significant amount of ^{137}Cs to Japan, no long-term effect seems to have continued in radioactivity deposition in Japan (Fig. 95-11). The present activity level is lower than that in 1985 when minimum annual radioactivity deposition was recorded.

Although a spring peak was found in deposition, it is difficult to explain this seasonal pattern only by stratospheric fallout. The variation in ratios of the radioactivity to corresponding stable elements did not show

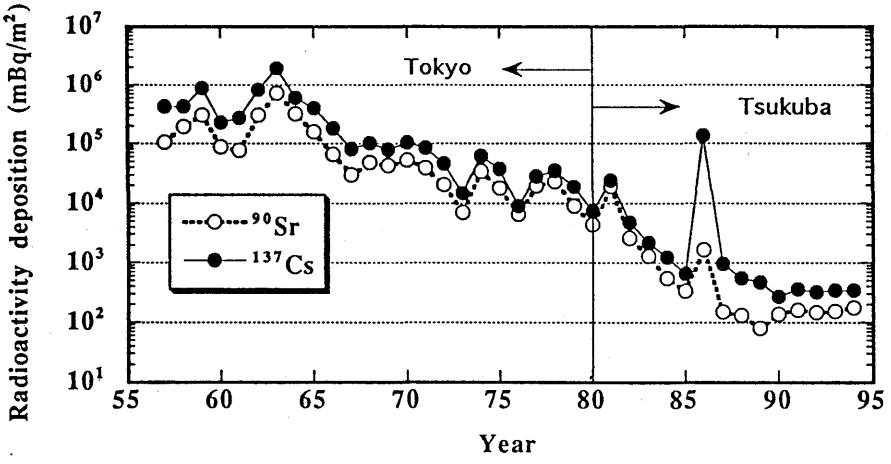

Fig. 95-10 Temporal variation in annual radioactivity deposition observed at MRI.
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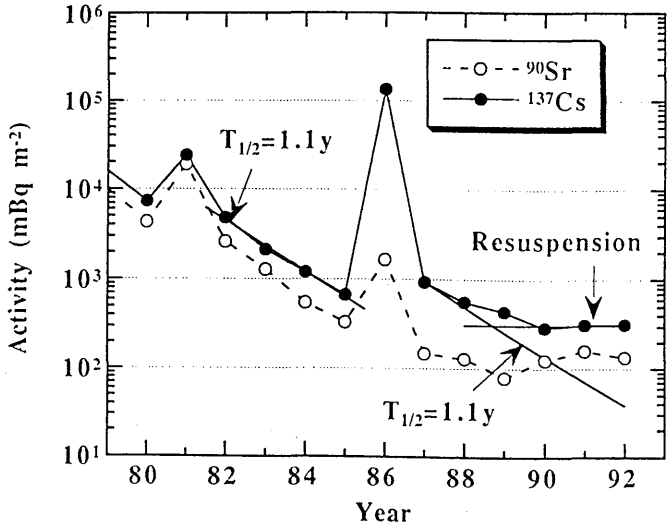

Fig. 95-11 Influence of the stratospheric component on the annual deposition of ^{137}Cs and ^{90}Sr.
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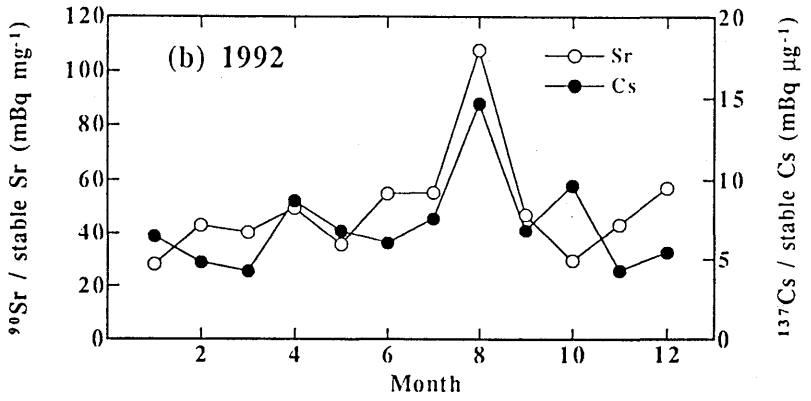

Fig. 95-12 Seasonal r/s ratio variation.
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the maximum in deposition peak months in spring (Fig. 95-12). It was concluded that most ^{90}Sr and ^{137}Cs deposits are of resuspended soil origin. The activity ratio (^{137}Cs/^{90}Sr) suggests that plural sources are present for resuspended radioactivity in Japan.

3.4 Reference fallout material preparation for activity measurements

Otsuji-Hatori, Igarashi, and Hirose (1996)

Since the present environmental radioactivity level is becoming extremely low, quality control in determining fallout has become more important, despite the dearth of reference material for fallout activity determination. Otsuji-Hatori *et al.*(1996) attempted to prepare reference fallout material for activity measurements by using deposition samples collected at 14 stations over Japan in 1963-1979, preserved after gamma-activity measurement (Fig. 95-13). With this reference material, ^{90}Sr, ^{137}Cs, and Pu isotopes were determined by several independent institutions. Results show good accord among individual institutions, meaning that the sample can be used as a reference for the measurement of fallout (Table 95-4). At present, this reference is being effectively used in quality control of radiochemical analysis.

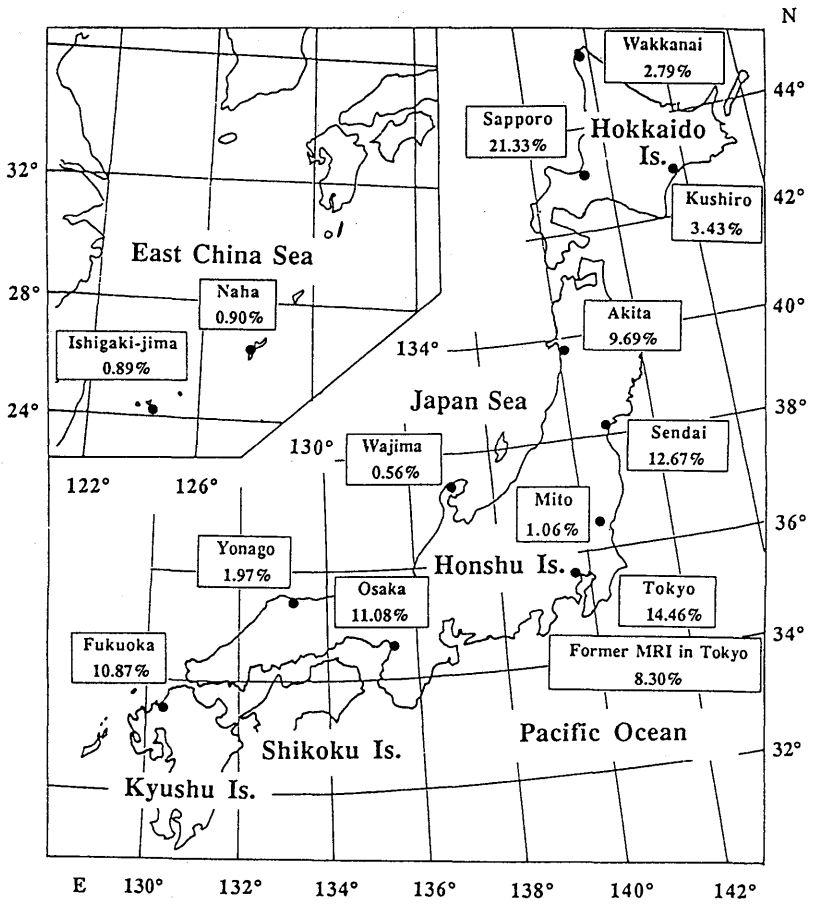

Fig. 95-13 Sampling station site and weight percentage of fallout from each station composing "reference fallout material". Reprinted from *J. Environ. Radioactivity*, **31**, Otsuji-Hatori *et al.*, Preparation of a reference fallout material for activity measurements, 143-155, Copyright (1996), with permission from Elsevier Science.

Table 95-4 Reported Concentrations of Radionuclides in the 'Reference Fallout Material' (mBq g^{-1})

Institution	*Sample* (n)	^{137}Cs	^{90}Sr	$^{239,240}Pu$	^{238}Pu	^{210}Pb	^{226}Ra
A	5	298 ± 3[a]	186 ± 3	6·32 ± 0·10	0·25 ± 0·03		
B	5	304 ± 11					
C	5	328 ± 11					
D	5	314 ± 6					
E	5	342 ± 11	248 ± 7	7·23 ± 0·54	0·40 ± 0·08		
F	5	314 ± 9					
G	5			6·41 ± 0·15	0·27 ± 0·01		
H	5	290 ± 9					
I	4[b]	305 ± 3				661 ± 15	15 ± 2
MRI	5(16)[c]	309 ± 6	198 ± 8	6·49 ± 0·30	0·14 ± 0·08		
Average		312 ± 16	211 ± 33	6·61 ± 0·42	0·27 ± 0·11	661	15
Weighted mean		311	211	6·52	0·28		

[a]Errors indicated in this table show the unbiased SD for each data set.
[b]Although the five bottles were provided, one was broken during transportation. For ^{226}Ra, one sample showed a value below the detection limit.
[c]The figure in parentheses is for ^{137}Cs.

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