*Journal of Atmospheric Chemistry* **17**: 61–71, 1993.



# Wet Deposition of Radionuclides Derived from the Chernobyl Accident

KATSUMI HIROSE
*Geochemical Laboratory, Meteorological Research Institute, Nagamine 1-1, Tsukuba, Ibaraki 305 Japan*

SUKEYOSHI TAKATANI
*Koube Marine Observatory, Nakayamatedouri 7-14-1, Chuou-ku, Koube, Hyougo 650 Japan*

and

MICHIO AOYAMA
*Japan Marine Science and Technology Center, Natsushima 2-15, Yokosuka 237, Japan*

(Received: 13 March 1992; in final form: 14 January 1993)

**Abstract.** Chernobyl radioactivity in precipitation was measured at Tsukuba, Japan, as were both surface-air concentrations and particle-size distributions of Chernobyl-released radionuclides. To understand the wet removal processes of the Chernobyl radionuclides, i.e. $^{137}Cs$, $^{103}Ru$, and $^{90}Sr$, wet deposition velocities were calculated. The wet deposition velocities of the Chernobyl radioactivity for individual rainfall events varied largely. The wet deposition velocity is given as the product of washout ratio and rainfall rate. Typically, it was found that the washout ratios of $^{90}Sr$ are systematically larger than those of $^{137}Cs$. In order to explain this fact, we examined the relationship between the washout ratios and particle sizes of radionuclide-bearing aerosols. A positive correlation between corrected washout ratios and particle size was found with a particle diameter range from 0.35 to 1.2 μm. The result strongly suggests that the factors controlling the wet removal of the Chernobyl radioactivity for individual rainfall events are surface air concentration, particle size, and rainfall rate, rather than precipitation amount, which is in agreement with previous understandings. This suggests that high contamination areas of radioactivity are formed during heavy rainfall events with high rainfall rates in the case of tropospheric injection such as the Chernobyl accident.

**Key words.** Wet deposition, Chernobyl radioactivity, precipitation scavenging, washout ratio.

## 1. Introduction

Wet deposition is the most important pathway for the removal of radioactivity from the atmosphere. The theoretical and experimental studies of wet removal processes for particles and gases have been carried out and reviewed by many researchers (Junge, 1963; Pruppacher and Klett, 1978). However, despite these studies, many uncertainties remain in understanding the wet removal processes because wet removal is a highly complex process, which depends upon meteorological conditions such as temperature, the microphysical structure of clouds, the dynamic structure of clouds, both determining the rainfall rate, and so on, and the characteristics of the cloud chemistry involving the physical and chemical properties of airborne particles.

The theoretical models which contain microphysics cloud processes were constructed for the wet removal (Flossmann *et al.*, 1985; 1987; Flossmann and Pruppacher, 1988; Flossman, 1991). Considerable uncertainty exists in current theoretical predictions and experimental determinations of wet deposition in the natural environment. There is also a lack of field measurements to make reliable model prediction tests. Especially, field verification of the major factors controlling the wet removal processes during an individual rainfall in the natural environment is required.

To provide model field observations, we studied the Chernobyl accident of 26 April 1986 when real accident radionuclides were released into the lower atmosphere. This release presented a unique opportunity to study the behavior of radionuclides in the environment. In fact, pronounced high-level radioactivity has been observed in the surface air and in rain waters in Japan since 3 May 1986 (Aoyama *et al.*, 1986). As a result of systematic studies of the environmental effect of the Chernobyl fallout, we obtained information on the transport of the radioactivity to the stratosphere (Aoyama, 1988; Aoyama *et al.*, 1991) and the dry deposition process (Aoyama *et al.*, 1992). The Chernobyl radioactivity also provided a good chance for environmental experiments of the wet removal processes.

In this paper, we describe the Chernobyl-derived long-lived radionuclides in precipitation as well as observations on the surface-air concentrations and the particle-size distributions of the Chernobyl radioactivity, and discuss the factors which control the wet deposition in individual rainfalls.

## 2. Sampling and Measurements

A high volume sampler that was installed into a precipitation shelter, was used to collect samples of airborne particulate materials on glass fiber filters (GB-100R, with a nominal collection efficiency of 99.9% for larger than 0.3 μm particles) at a flow rate of 1000 l $min^{-1}$. The sampling site was the observation field of the Meteorological Research Institute at Tsukuba, Japan. To separate the coarse and fine fractions from each other, a 10 μm cut-off filter was used on the sampler. The filter was changed every morning and the backup filter (less than 10 μm of particle sizes) was usually analyzed daily.

To determine the particle-size distribution of the Chernobyl radioactivity in the surface air, we used a high-volume cascade impactor (Five stages: I > 7 μm, II 3.3–7.0 μm, III 2.0–3.3 μm, IV 1.1–2.0 μm, V < 1.1 μm) and an Andersen sampler (Seven stages: I > 7 μm, II 3.3–7.0 μm, III 2.0–3.3 μm, IV 1.1–2.0 μm, V 0.65–1.1 μm, VI 0.43–0.65 μm, VII < 0.43 μm), of which the flow rates were 600 and 16 l $min^{-1}$, respectively. Airborne particles in each size were collected on a glass fiber filter. The air samples were collected during a period of between 4 to 7 days.

Daily precipitation samples of the Chernobyl radioactivity were collected in a plastic sampler with a surface area of 1 $m^2$, which was installed at 1 m height above ground level and had a smooth surface. The effect of dry deposition is negligible for

daily precipitation samples. Precipitation samples were concentrated to about 30 ml on a hot plate and transferred in a package with a volume of 50 ml.

The radioactivity of $\gamma$-spectrometer. The $^{90}Sr$ counting was carried out by using a low background $\beta$ counter after the separation and purification of strontium using the radiochemical method as described in detail elsewhere (Aoyama *et al.*, 1991).

## 3. Results

### 3.1. *Surface Air Concentrations of the Chernobyl Radioactivity*

The nuclear reactor accident occurred at Chernobyl on 26 April 1986 (Devell *et al.*, 1986). The Chernobyl radioactivity contaminated most of the Northern Hemisphere (Larsen *et al.*, 1986; Juzdan *et al.*, 1986). Figure 1 shows temporal variations of surface air concentrations of $^{137}Cs$ observed at Tsukuba in Japan. On 3 May, abrupt high concentrations of volatile radionuclides, containing $^{131}I$, $^{103}Ru$, $^{106}Ru$, $^{134}Cs$, $^{137}Cs$, $^{136}Cs$, and so on, were initially detected in the surface air at Tsukuba, of which $^{131}I$, $^{103}Ru$, and $^{137}Cs$ are remarkable (Aoyama *et al.*, 1986). The $^{131}I$, which is a main nuclide of the Chernobyl radioactivity, showed a maximum concentration in airborne particles (494 mBq $m^{-3}$) on 5 May 1986. A maximum $^{103}Ru$ concentration in the surface air occurred on 9 May (161 mBq $m^{-3}$). The $^{137}Cs$ concentration in the surface air increased by about four orders of magnitude

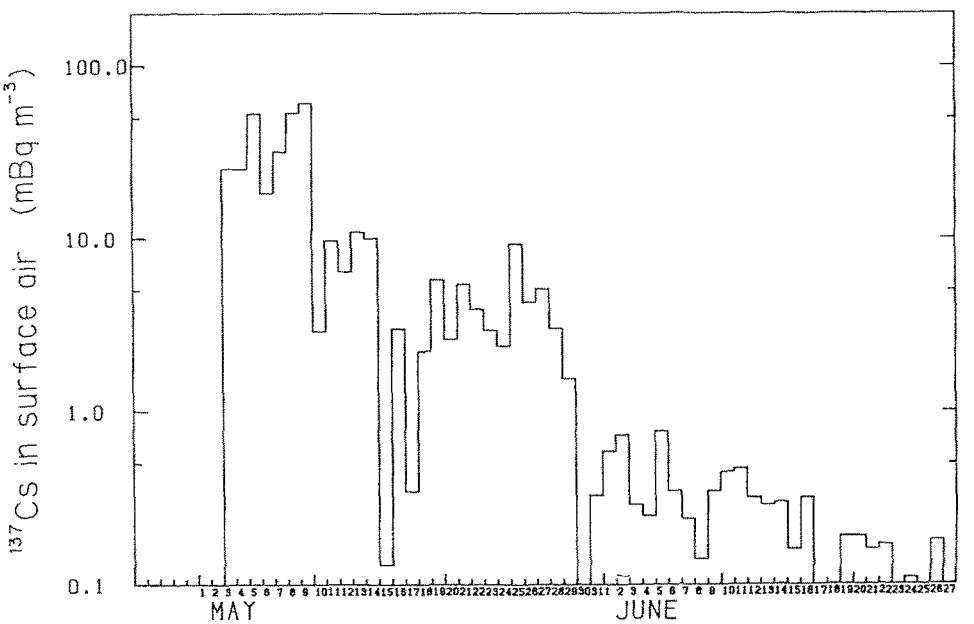


Fig. 1. The surface air concentration of $^{137}Cs$ observed at Tsukuba, Japan, in May–June 1986.

higher than the previous month and reached a maximum of 61.2 mBq $m^{-3}$ on 9 May.

The monthly mean $^{90}Sr$ concentration in the surface air at Tsukuba in April 1986 was 0.41 μBq $m^{-3}$. On 3–6 May, as shown in Figure 2, the $^{90}Sr$ concentration increased to 32 μBq $m^{-3}$, which is about two orders of magnitude higher than the previous month. A typical feature of the Chernobyl $^{90}Sr$ observed in Japan is that its increase was lower than that expected from total released amounts. After the Chernobyl fallout, therefore, $^{90}Sr/^{137}Cs$ activity ratios changed from 0.7 of nuclear-bomb derived radionuclides to 0.005.

### 3.2. *Particle-Size Distributions of the Chernobyl Radioactivity*

A typical example for the particle size distributions of $^{131}I$, $^{103}Ru$, and $^{137}Cs$ in the airborne particles collected on 6–10 May 1986, which corresponds to the period observed maximum concentrations of the Chernobyl radioactivity, is shown in Figure 3. A marked feature is that the Chernobyl radioactivity, such as $^{131}I$, $^{103}Ru$ and $^{137}Cs$, was mostly sub-micrometer in size, which is consistent with the results observed by Bondietti and Brantley (1986), Jost *et al.* (1986), Knuth and Sanderson (1986) and Ooe *et al.* (1987). On the other hand, $^{90}Sr$ and plutonium isotopes released from Chernobyl, were found in larger particle size (Aoyama *et al.*, 1992).

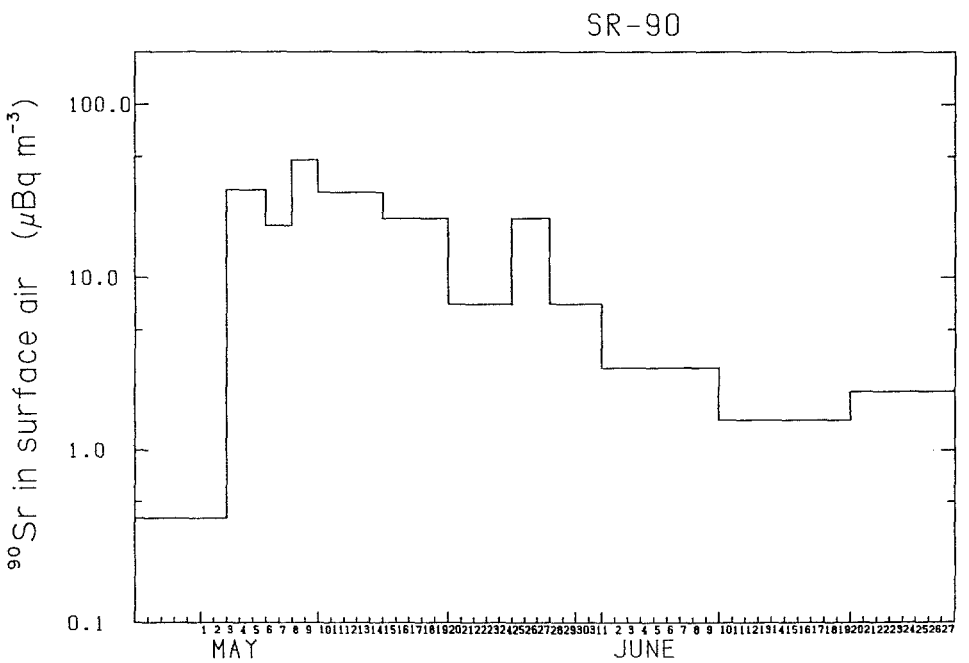


Fig. 2. The surface air concentration of $^{90}Sr$ observed at Tsukuba, Japan, in May–June 1986.

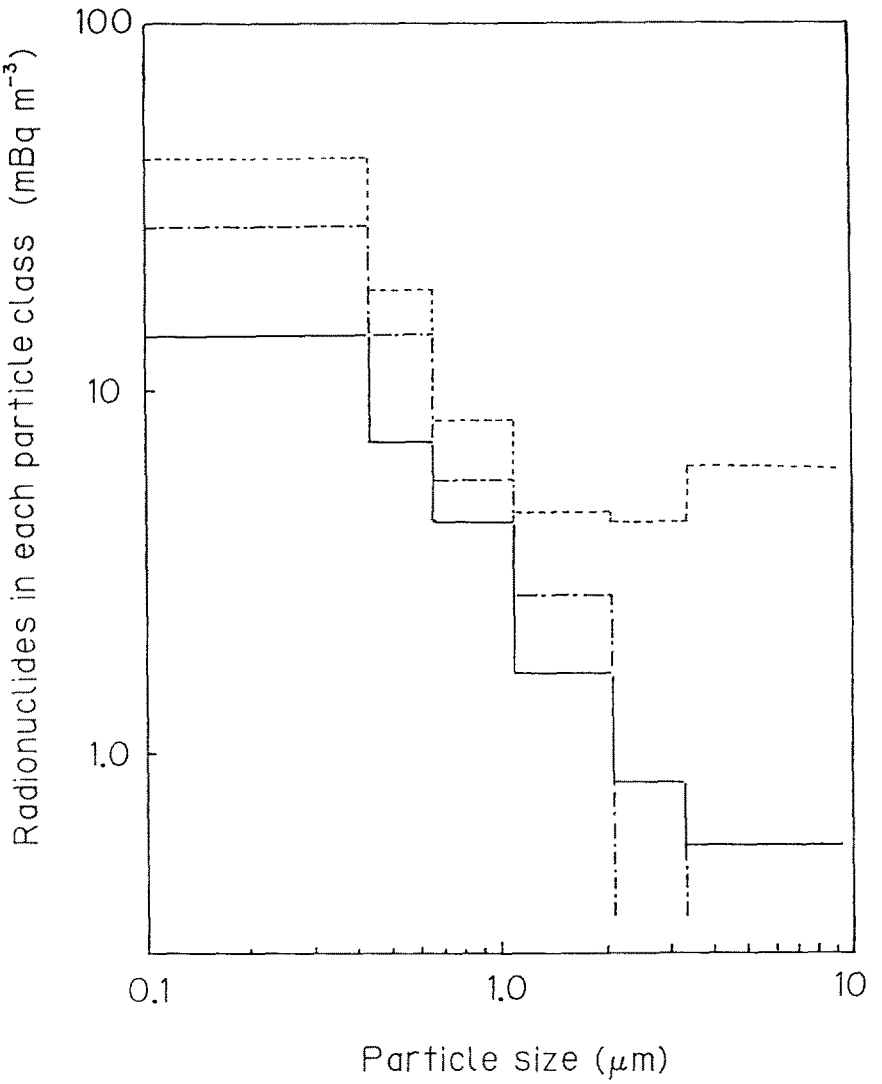


Fig. 3. Particle size distributions of $^{131}$I (-----), $^{103}$Ru (– · –) and $^{137}$Cs (——) observed at Tsukuba, Japan, during the period of 6–10 May 1986.

Another feature is that the size distribution pattern of $^{131}$I differed significantly from others and showed no lognominal distribution.

The activity median aerodynamic diameters (AMAD) of the radionuclides, which are determined from the size distributions of four sampling periods of 6–10, 13–17, 17–24, and 24–31 May 1986, are summarized in Table I. The AMAD values of the Chernobyl radionuclides varied in each sampling period, whose tendency is consistent with the results of Bondietti and Brantley (1986) and Ooe *et al.* (1987). The temporal variation of the AMAD for the Chernobyl radioactivity may be due to a difference in the release process at Chernobyl and/or the fractionation in the transport process of the Chernobyl radioactivity.

Table I. The activity median aerodynamic diameters (AMAD) of the Chernobyl radioactivity

| Sampling period | AMAD (μm) | | |
|---|---|---|---|
| | $^{137}Cs$ | $^{103}Ru$ | $^{90}Sr$ |
| 6–10 May 1986 | 0.38 | 0.35 | – |
| 13–17 May | 0.51 | 0.54 | 1.2 |
| 17–24 May | 0.71 | 0.65 | – |
| 24–31 May | 0.45 | 0.38 | 1.0 |

### 3.3. *Wet Deposition of the Chernobyl Radioactivity*

In May 1986, marked high radioactive deposition occurred in Japan as well as high air concentrations (Aoyama *et al.*, 1987; 1991). The monthly $^{137}Cs$ deposition, which is the sum of wet and dry deposition, increased to 131 Bq $m^{-2}$, which was the same level as that of the highest deposition period in 1963 after the 1961–62 large-scale atmospheric nuclear tests. This pronounced peak of deposition was clearly attributable to the Chernobyl accident because of the observation of short-lived radionuclides and $^{134}Cs$ (Cambray *et al.*, 1987; Aoyama *et al.*, 1987). In the same period, the monthly $^{90}Sr$ deposition increased to 1.37 Bq $m^{-2}$, which is about 60 times larger than for the previous month.

Wet deposition of $^{137}Cs$, $^{103}Ru$, and $^{90}Sr$ due to individual rainfall events, observed at Tsukuba in May 1986, are summarized in Table II.

Maximum wet deposition of the Chernobyl radioactivity occurred with a relative heavy rainfall on 14–15 May, which was not consistent with the period of maximum surface air concentrations of the Chernobyl radioactivity. Another typical feature is that higher deposition, whose $^{103}Ru$ was a maximum within the sampling period, was observed in a thunderstorm on 16–17 May, although the precipitation amount of the storm was lower than that on 14–15 May.

To evaluate the percentage of the radioactive deposition due to precipitation

Table II. Wet deposition of long-lived radionuclides observed at Tsukuba

| Sampling period | Precipitation (mm) | $^{137}Cs$ (Bq $m^{-2}$) | $^{103}Ru$ (Bq $m^{-2}$) | $^{90}Sr$ (Bq $m^{-2}$) |
|---|---|---|---|---|
| 5–6 May | 2.2 | 11.6 ± 1.5 | 29.6 ± 3.5 | 0.045 ± 0.006 |
| 6–7 May | 23.5 | 16.8 ± 2.5 | 25.5 ± 3.3 | 0.045 ± 0.006 |
| 14–15 May | 68.0 | 30.3 ± 7.9 | 22.9 ± 6.6 | 0.62 ± 0.07 |
| 16–17 May | 16.0 | 29.2 ± 6.6 | 68.5 ± 11.5 | 0.43 ± 0.05 |
| 19–20 May | 2.6 | 3.8 ± 0.7 | 6.0 ± 0.9 | 0.060 ± 0.010 |
| 20–21 May | 11.7 | 2.7 ± 0.6 | 5.6 ± 0.9 | 0.038 ± 0.008 |
| 29–30 May | 70.0 | 7.2 ± 1.1 | 27.7 ± 3.3 | 0.092 ± 0.016 |

scavenging, we calculated the total wet deposition in May 1986, which is a sum of radioactive deposits from individual rainfalls. The percentages of the total wet deposition to the monthly deposition in May 1986 were 88% for $^{137}$Cs, 87% for $^{103}$Ru, and 96% for $^{90}$Sr. The result implies that the precipitation scavenging is the major pathway to transport the Chernobyl radioactivity from the atmosphere to the ground. In this connection, Ballestra *et al.* (1987) indicated that the major part of the radioactive deposits observed at Monaco was through rain, rather than dry deposition. The dominance of wet deposition for the Chernobyl radioactivity is also evident from the fallout date of Milan (Queeirazza *et al.*, 1987) and Saluggia (Spezzano and Giacomelli, 1990).

## 4. Discussion

The major pathway to the deposition of the Chernobyl radioactivity is by precipitation scavenging. This finding suggests that the distribution of the Chernobyl fallout on the land area depends on the presence of rainfall events when the radioactive cloud passes through it.

To understand the wet removal process of the Chernobyl radioactivity, we introduce wet deposition velocities of the radionuclides. The wet deposition velocity ($V_{\text{wet},R}$) is written as follows:

$$V_{\text{wet},R} = D_R / C_{a,R} \tag{1}$$

where $D_R$ and $C_{a,R}$ are the wet deposition flux (Bq m$^{-2}$ s$^{-1}$) and surface-air concentration (Bq m$^{-3}$) of the Chernobyl radioactivity, respectively. The wet deposition velocities of $^{137}$Cs, $^{103}$Ru, and $^{90}$Sr are summarized in Table III, together with the rainfall rates. The wet deposition velocities varied considerably. This apparently depends on the radionuclides and sampling periods. The wet deposition velocity is represented as

$$V_{\text{wet},R} = WI_R \tag{2}$$

where $I_R$ is rainfall rate (mm h$^{-1}$) and $W$ the washout ratio (scavenging ratio). The washout ratios are usually used to describe the wet removal processes of atmospheric pollutants (Junge, 1963; Engelmann, 1971; Barrie, 1985). We calculated the washout ratios of the Chernobyl radioactivity for individual rain fall events (Table III). The washout ratios of $^{137}$Cs, $^{103}$Ru and $^{90}$Sr derived from the Chernobyl accident, which ranged from $0.19 \times 10^6$ to $0.53 \times 10^6$, from $0.16 \times 10^6$ to $0.37 \times 10^6$, and from $0.50 \times 10^6$ to $1.4 \times 10^6$, respectively, are the same order of magnitude as those of $^7$Be ($9.2 \times 10^5$) and artificial radionuclides ($1 \times 10^6$) derived from atmospheric nuclear tests (Slinn, 1978; Harvey and Matthews, 1989).

The washout ratio depends on meteorological, physical, and chemical factors. According to the theoretical consideration (Mason, 1971; Slinn, 1978; Harvey and Matthews, 1989), the following weak dependence of washout ratio on the rainfall rate is predicted:

Table III. Washout ratios and wet deposition velocities of long-lived radionuclides observed at Tsukuba

| Sampling period | Rainfall rate (mm $h^{-1}$) | Wet deposition velocity (mm $s^{1-2}$) | | | Washout ratio ($\times 10^6$) | | |
|---|---|---|---|---|---|---|---|
| | | $^{137}Cs$ | $^{103}Ru$ | $^{90}Sr$ | $^{137}Cs$ | $^{103}Ru$ | $^{90}Sr$ |
| 5–6 May | 0.49 | 2.6 | 3.8 | 16 | 0.19 | 0.23 | 1.2 |
| 6–7 May | 1.7 | 10 | 7.6 | 27 | 0.22 | 0.16 | 1.4 |
| 14–15 May | 7.6 | 70 | 35 | 230 | 0.32 | 0.19 | 1.1 |
| 16–17 May | 16 | 110 | 220 | 180 | 0.25 | 0.41 | 0.50 |
| 19–20 May | 0.8 | 7.4 | 6.3 | 31 | 0.34 | 0.28 | 1.4 |
| 20–21 May | 1.5 | 12 | 8.2 | 28 | 0.29 | 0.20 | 1.4 |
| 29–30 May | 3.6 | 53 | 15 | 39 | 0.53 | 0.37 | 1.4 |

$$W = \alpha I_R^{-\beta}, \tag{3}$$

where $\alpha$ and $\beta$ are constant. For $^{137}Cs$ and $^{103}Ru$ in the Chernobyl radioactivity, the washout ratio was nearly independent on the rainfall rate. The constancy of the washout ratio is in agreement with the result observed to $^{7}Be$ (Harvey and Matthews, 1989). On the other hand, the washout ratio for $^{90}Sr$ seems to be dependent on the rainfall rate with $\beta = 0.22$. In this connection, $\beta$ is expected to be in the range of 1/4 (for frontal storms) to 1/2 (for convective storms).

Gatz *et al.* (1975) indicated that the washout ratio is a function of the mass median diameter (MMD) for a number of trace metals. For the Chernobyl radioactivity, the AMADs of radionuclide-bearing particles varied through the sampling period as shown in Table I. To confirm the dependence of the washout ratios to the particle diameters, we examined the relationship between the washout ratios and AMADs for $^{137}Cs$, $^{103}Ru$ and $^{90}Sr$. Figure 4 shows the logarithmic plot of the corrected washout ratio to the diameter of Chernobyl radionuclide-bearing particles for individual rainfall events. The result reveals that the scavenging behavior of $^{90}Sr$ for the Chernobyl fallout differs from that of $^{137}Cs$ and $^{103}Ru$. Its causes may be attributable to the chemical and physical properties of radionuclide-bearing particles. Unfortunately, we have no information on the chemical properties of aerosols containing each Chernobyl radionuclide. On the other hand, it is known that the scavenging coefficient, which is defined as the reduction in the aerosol particles during a rain event and the parameters related with the washout ratio, is a function of the particle diameter of aerosols (Davenport and Peters, 1978; Schumann *et al.*, 1988; Flossmann, 1990). Then we examined the correlation between the washout ratio and AMADs of the Chernobyl radioactivity. The result indicates a positive correlation (correlation factor: 0.82) within the particle diameter from 0.35 to 1.2 μm. The result regarding the dependence of the corrected washout ratio on the particle diameter qualitatively reflects a 'Greenfield gap' behavior (large decrease of the scavenging coefficient in the diameter in the diameter range 0.1–1.0 μm), which

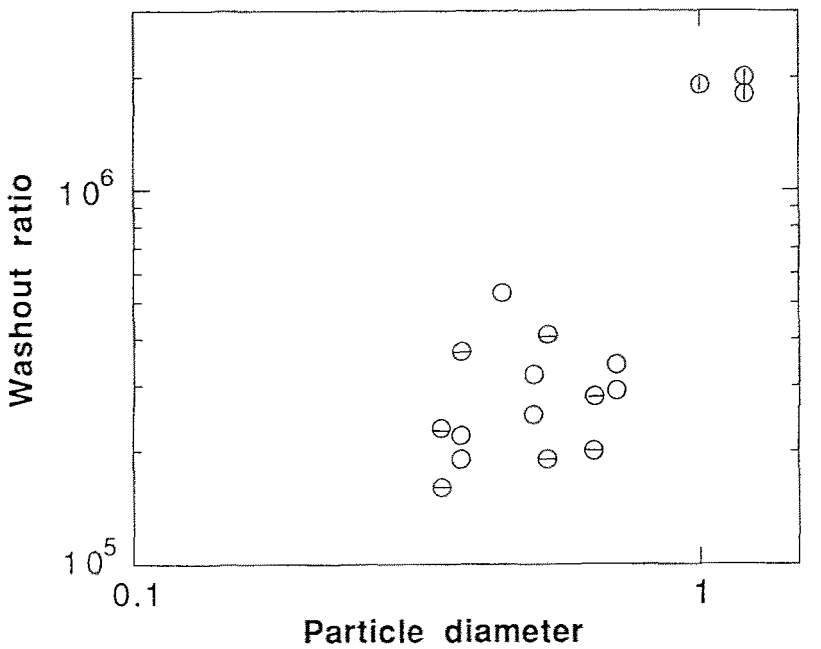


Fig. 4. Relationship between corrected washout ratios and AMAD of the Chernobyl radionuclides. $^{137}$Cs: ○, $^{103}$Ru: ⊖, $^{90}$Sr: ⓪. To eliminate the effect of the rainfall rate to the washout ratio for $^{90}$Sr, the α values, which mean the washout ratio corrected by the rainfall type, were calculated from Equation (3). The correlation factor between the corrected washout ratio and AMAD was 0.82. The washout ratio was represented as a function of the particle diameter (unit: μm): $\alpha = 10^{5.99} r^{1.67}$.

have been observed by the scavenging coefficients of aerosols (Schumann *et al.*, 1988). These findings suggest that the particle diameter is one of the essential factors for controlling the precipitation scavenging of the Chernobyl radioactivity.

The result indicates that the factors for controlling the wet deposition flux of the Chernobyl radioactivity for individual rainfall events are air concentration, particle diameter, and rainfall rate rather than precipitation amount. This suggests that the radioactivity contaminated areas, so-called hot spots, following large-scale nuclear accidents are formed by not only the presence of a rainfall event itself but also in particular a high rainfall rate during this event.

## 5. Conclusions

For the Chernobyl fallout, precpitation scavenging is the most significant process to remove pollutants from the atmosphere; 88, 87, and 96% of the total deposition being scavenged by rainwater for $^{137}$Cs, $^{103}$Ru and $^{90}$Sr derived from the Chernobyl accident, respectively.

The wet removal of the Chernobyl radioactivity for individual rainfall events is controlled by three factors; (1) air concentrations, (2) particle size of the radionuclide-bearing aerosols, and (3) rainfall rate rather than precipitation amount.

This suggests that high contamination areas of the Chernobyl radioactivity are formed at the heavy rainfall events with the high rainfall rate and that radionuclides resident on larger airborne particles are preferentially scavenged by rain.

## Acknowledgements

We thank the staff members of the Geochemical Laboratory, MRI, for help with the sampling and for useful discussion. We are also grateful to the anonymous reviewers for valuable comments.

## References

Aoyama, M., 1988, Evidence of stratospheric fallout of ceasium isotopes from the Chernobyl accident, *Geophys. Res. Lett.* **15**, 327–30.

Aoyama, M., Hirose, K., Suzuki, Y., Inoue, H., and Sugimura, Y., 1986, High level radionuclides in Japan in May, *Nature* **321**, 819–820.

Aoyama, M., Hirose, K., and Sugimura, Y., 1987, Deposition of gamma-emitting nuclides in Japan after the reactor-IV accident at Chernobyl, *J. Radioanal. Nucl. Chem., Articles* **116**, 291–306.

Aoyama, M., Hirose, K., and Sugimura, Y., 1991, The temporal variation of stratospheric fallout derived from the Chernobyl accident, *J. Environ. Radioactivity* **13**, 103–115.

Aoyama, M., Hirose, K., and Takatani, S., 1992, Particle size dependent dry deposition velocity of the Chernobyl radioactivity, in *Precipitation Scavenging and Atmospheric-Surface Exchange Processes: Fifth International Conference*, Hemisphere, Vol. 3, pp. 1581–1593.

Ballestra, S. B., Holm, E., Walton, E., and Whitehead, N. E., 1987, Fallout deposition at Monaco following the Chernobyl accident, *J. Environ. Radioactivity* **5**, 391–400.

Barrie, L. A., 1985, Scavenging ratios, wet deposition, and in-cloud oxidation: an application to the oxides of sulphur and nitrogen, *J. Geophys. Res.* **90**, 5789–5799.

Bondietti, E. A. and Brantley, J. N., 1986, Characteristics of Chernobyl radioactivity in Tennesse, *Nature* **322**, 313–314.

Cambray, R. S., Cawse, P. A., Garland, J. A., Gibson, J. A. B., Johnson, P., Lewis, G. N. J., Newton, D., Salmon, L., and Wade, B. O., 1987, Observations on radioactivity from the Chernobyl accident, *Nuclear Energy* **6**, 77–101.

Davenport, H. M. and Peters, L. K., 1978, Field studies of atmospheric particulate concentration changes during precipitation, *Atmos. Environ.* **12**, 997–1008.

Devell, L., Tovedal, H., Bergstrom, U., Appelgren, A., Chyssler, J., and Andersson, L., 1986, Initial observations of fallout from the reactor accident at Chernobyl, *Nature* **321**, 192–193.

Englemann, R. J., 1971, Scavenging prediction using ratios of concentrations in air and precipitation, *J. Appl. Meteorol.* **10**, 493–497.

Flossmann, A. I., 1991, The scavenging of two different types of marine aerosol particles calculated using a two-dimensional detailed cloud model, *Tellus* **43B**, 301–321.

Flossmann, A. I., Hall, W. D., and Pruppacher, H. R., 1985, A theoretical study of the wet removal of atmospheric pollutants. Part I: The redistribution of aerosol particules captured through nucleation and impaction scavenging by growing cloud drops, *J. Atmos. Sci.* **42**, 582–606.

Flossmann, A. I., Pruppacher, H. R., and Topalian (1987), A theoretical study of the wet removal of atmospheric pollutants. Part II: The uptake and redistribution of $(NH_4)_2SO_4$ particles and $SO_2$ gas simultaneously scavenged by growing cloud drops, *J. Atmos. Sci.* **44**, 2912–2923.

Flossmann, A. I. and Pruppacher, H. R., 1988, A theoretical study of the wet removal of atmospheric pollutants. Part III: The uptake, redistribution and deposition of $(NH_4)_2SO_4$ particles by a convective cloud using a two-dimensional cloud dynamics model, *J. Atmos. Sci.* **45**, 1857–1871.

Gatz, D. F., 1975, Scavenging ratio measurement in Metromex, in R. W. Beadle and R. G. Semonin (eds.), *Precipitation Scavenging 1974*, ERDA Symposium Series, Springfield, VA, pp. 71–87.

Hales, J. M., 1982, Mechanistic analysis of precipitation scavenging using a one-dimensional time variant model, *Atmos. Environ.* **16**, 1175–1783.

Harvey, M. J. and Matthews, K. M., 1989, $^{7}$Be deposition in a high-rainfall area of New Zealand, *J. Atmos. Chem.* **8**, 299–306.

Jost, D. T., Gaggeler, H. W., Baltensperger, U., Zinder, B., and Haller, P., 1986, Chernobyl fallout in size fractionated aerosol, *Nature* **324**, 22–23.

Junge, C. E., 1963, *Air Chemistry and Radioactivity*, Academic Press, New York.

Juzdan, Z. R., Helfer, I. K., Miller, K. M., Rivera, W., Sanderson, C. G., and Silvestri, S., 1986, Deposition of radionuclides in the Northern Hemisphere following the Chernobyl accident, in *A Compendium of the Environmental Measurement's Research Projects Related to the Chernobyl Nuclear Accident, EML-460*, U.S. Department of Energy, New York, pp. 105–154.

Knuth, R. H. and Sanderson, C. G., 1986, Particle size distribution of Chernobyl related aerosols in New York City, in *A Compendium of the Environmental Measurement's Research Projects Related to the Chernobyl Nuclear Accident, EML-460*, U.S. Department of Energy, New York, pp. 291–300.

Larsen, R. J., Sanderson, C. G., Rivera, W., and Zamichieli, M., 1986, The characterization of radionuclides in the North American and Hawaiian surface air and deposition following the Chernobyl accident, in *A Compendium of the Environmental Measurement's Research Projects Related to the Chernobyl Nuclear Accident, EML-460*, U.S. Department of Energy, New York, pp. 1–104.

Mason, B. J., 1971, *The Physics of Clouds*, 2nd edn., Clarendon Press, Oxford.

Ooe, H., Seki, R., and Ikeda, N., 1987, Particle-size distribution of fission products in airborne dust collected at Tsukuba from April to June 1986, *J. Environ. Radioactivity* **6**, 219–223.

Pruppacher, H. R. and Klett, J. D., 1978, *Microphysics of Clouds and Precipitation*,. Elsevier, Amsterdam.

Querrirazza, G., Bozzani, A., and Bonforte, G., 1987, Distribuzione temporale e spazziale della radioattivita nelle ricadute umide e secche, *Aqua-Aria* **7**, 807–817.

Schumann, T., Zinder, B., and Waldvogel, A., 1988, Aerosol and hydrometeor concentrations and their chemical composition during winter precipitation along a mountain slope-I. Temporal evolution of the aerosol, microphysical and meteorological conditions, *Atmos. Environ.* **22**, 1443–1459.

Slinn, W. G. N., 1978, Parameterizations for resuspension and for wet and dry deposition of particles and gases for use in radiation dose calculations, *Nuclear Safety* **19**, 205–219.

Spezzano, P. and Giacomelli, R., 1990, Radionuclide concentrations in air and their deposition at Salguggia (Northwest Italy) following the Chernobyl nuclear accident, *J. Environ. Radioactivity* **12**, 79–91.