Aerosol residence times and iodine gas/particle conversion over the North Pacific as determined from Chernobyl radioactivity

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Radioactivity from the Chernobyl accident present on aerosols was measured at several island sites and on a ship in the North Pacific. The radioactivity arrived at the various sites located from 11°–64°N between 6–14 May 1986. Isentropic trajectory analyses and measurement of the natural radioactive species ⁷Be and ²²²Rn indicate that the continental air containing Chernobyl fission products (e.g., ¹³¹I, ¹³⁷Cs) was transported to the central North Pacific through the middle troposphere. We estimate the residence time and gas-to-particle conversion times by combining the radionuclide data with surface concentration estimates in Japan. The atmospheric residence time over the North Pacific for the aerosols containing the fission products was 3–5 days. The calculated mean gas-to-particle conversion time for radioactive ¹³¹I was 2–3 weeks, with a minimum of ~12 days. The results indicate that the radioactive submicrometer aerosol and the iodine gas from the Chernobyl explosion have residence and conversion times similar to those of naturally occurring aerosols and gases in the atmosphere.

INTRODUCTION

An explosion occurred at the Chernobyl nuclear power complex near Kiev, USSR (51°N, 30°E), on 26 April 1986 (GMT). The resulting plume of radioactivity was measured at many locations over Europe during the next several days (Devell *et al.*, 1986; Fry *et al.*, 1986; Kaupinen *et al.*, 1986; Thomas *et al.*, 1986) and over Japan one week later (Aoyama *et al.*, 1986).

When the Chernobyl accident was reported, we began special air filter sampling for radioactive materials at the island stations in the Sea/Air Exchange Program (SEAREX) Air Sampling Network in the North Pacific (see Fig. 1) and on the R/V *Moana Wave* which had put

to sea on 28 April 1986 from Honolulu for an intensive atmospheric/ocean sampling experiment as part of SEAREX.

We report here about long-range transport of the fission products from Chernobyl to the North Pacific and estimates of aerosol residence times and iodine gas-to-particle conversion rate during the transport using natural and artificial radioactive materials.

EXPERIMENTAL

The 24 hour aerosol samples for the radionuclides were collected daily at the island sites and on the ship by using high-volume air-sampling systems (~1.1 m³min⁻¹; Uematsu *et*

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Fig. 1. Sampling stations in the North Pacific and part of the track of R/V Moana Wave. The line (—) is the track between 12 and 18 May; data from that period are used for the model calculations (see text). In addition, an isentropic trajectory back in time from the position of R/V Moana Wave early on 12 May, 1986 ($\sim 45^\circ\text{N}$, 152°W) at 295 K is given (---). The dots are 1 day apart. The white dots show the locations and the altitude of the air mass on 30 April, 5 and 10 May, respectively. The air mass crossed the central northern North Pacific toward the east at ~ 4 km altitude, then descended as it moved to lower latitudes and finally was advected from east to west in the marine boundary layer.

al., 1983). The filters used were Whatman 41 filters (20×25 cm) at the island sites and Gelman glass fibers filters (20×25 cm) on the ship. The samples were analyzed for fission products with gamma-ray spectrometry systems at the Rhode Island Nuclear Science Center. There was no apparent difference of the trapping efficiency between the two different kinds of filters examined on the ship, although the mean diameter of particles bearing the fission products were reported to be $0.3\text{--}0.9 \mu\text{m}$ (Bondietti and Brantley, 1986; Kauppinen *et al.*, 1986). Radon-222 was determined by counting the radioactivity of Rn daughters (^{214}Pb and ^{214}Bi) on aerosols using techniques described by Larson and Bressan (1978). Hour long samples for ^{222}Rn were taken twice daily on the ship.

RESULTS AND DISCUSSION

Transport of fission products to the North Pacific

From the samples collected in early May, apparent peaks of ^{131}I (half-life 8.0d), ^{140}La (half-life 1.7d), ^{103}Ru (half-life 39d), ^{140}Ba (half-life 13d), ^{134}Cs (half-life 2.1y) and ^{137}Cs (half-life 30y) were identified by the gamma ray spectra, while there was little or no peak due to the radioactivities of these fission products in earlier samples.

Daily atmospheric concentrations of ^{131}I (half-life 8.0d) observed at the stations are shown in Fig. 2. Radioactive materials from Chernobyl were detected at Midway (28°N , 177°W) between 6 and 7 May; at St. Lawrence (64°N , 172°W) between 6 and 8 May; and at Shemya (53°N , 174°E) between 6 and 8 May. The radioactivity arrived at all three sites at approximately the same time. The leading edge of the air mass containing radioactive materials arrived at Oahu (21°N , 158°W) on 10 May but did not reach the Moana Wave (which was then located at 45°N , 152°W) until 12 May. Weekly samples collected at Enewetak (11°N , 162°E) in-

Fig. 2. Daily atmospheric concentrations of ^{131}I (fCi/SCM: fCi per standard cubic meter) observed at the stations in the North Pacific during the period from April 25 to June 4. After Uematsu and Duce (1986).

dicate that the fission products reached there sometime between 7 and 14 May.

The highest concentration of ^{131}I was observed at Midway, where $\sim 1800 \text{ fCi m}^{-3}$ were measured during the 7–9 May period (see Fig. 2). While the concentrations measured north of Midway were lower, the decrease was not as dramatic as it was to the south: the highest concentrations of ^{131}I measured at Oahu never exceeded 53 fCi m^{-3} , while concentrations north of Midway reached several hundred fCi m^{-3} at all stations. The concentrations of fission products at the North Pacific sites had decreased to near the detection limits by mid-June. No Chernobyl radioactivity was observed in any samples collected at South Pacific stations (e.g., Nauru, American Samoa, Norfolk) by the end of 1986.

Air trajectory and meteorological analyses

In Fig. 1, we show a sketch of the isentropic trajectories (Merrill *et al.*, 1985) at the 295 K (the layer which enters the surface boundary layer near 30°N) for air reaching the ship's location early on 12 May and for air reaching Japan on 2 May. On other days during this period the trajectories for air reaching Japan, Oahu and the

vessel follow a similar pattern indicating a common origin for the air containing the radioactive debris. These trajectories indicate clearly that the slow flow of air around the semipermanent anticyclone northeast of Hawaii accounts for the relatively late appearance of the fission products east of 160°W . The trajectory-based transit times from the Asian coast to Oahu and the ship sites are ~ 8 and ~ 10 days, respectively. The transport speed during the period 6–10 May ($\sim 8 \text{ m s}^{-1}$) is much slower than that during the period 1–3 May ($\sim 20 \text{ m s}^{-1}$). Furthermore, the rapid movement on 3–5 May of air near 45°N , 180° towards the southeast is associated with the intrusion into the high of a cold front associated with a strong low centered in the Bering Sea.

Relationship between artificial and natural radionuclides on the ship

In Fig. 3, the concentrations of ^{222}Rn , ^7Be , ^{131}I and ^{137}Cs during the SEAREX cruise are plotted versus time. Each of these radionuclides has a different source: ^{222}Rn (half-life 3.8d) is emitted from continental soil; ^7Be (half-life 53d) is produced by cosmic rays in the upper troposphere and lower stratosphere; ^{131}I and ^{137}Cs were

Fig. 3. The atmospheric concentrations of ^{222}Rn , ^7Be , ^{131}I and ^{137}Cs observed on the Moana Wave in 1986, decay corrected to the midpoint of the sampling period.

primarily attributable to the Chernobyl accident. The variations of the concentrations with time, however, were similar to each other, especially during the early period of the sampling. As an example, the peak concentrations for each of these substances occur on 13–16 May.

The transport pattern indicated in Fig. 1 would account for the coincident peaks, as ^7Be would be added to the air mass carrying the fission products in the upper troposphere and ^{222}Rn would be injected into the air mass in the middle troposphere along the path over Asia. This pattern is similar to the transport path of mineral aerosol to the central Pacific (Uematsu *et al.*, 1983).

Transit time of fission products from over Japan to the ship

The time series of radioactive materials observed at Tsukuba, Japan (Aoyama *et al.*, 1986)

showed that the first major pulse of Chernobyl radioactivity occurred there during the period 3–8 May. This peak evidently corresponds to the event observed on the ship during the period 13–18 May. If so, the transit time from near the coast of the Asian continent to the ship site was ~10 days, in good agreement with the transit time estimated from the trajectory analysis. Additional trajectory analyses (not shown) indicate that the radioactive materials moved eastward from 90°E to 120°E at $\sim 55^\circ\text{N}$ and that the flow split so that some of this material reached Japan on 3–8 May, while some continued to the east. Some of this material subsided to the surface east of 160°W where the ship was located. Subsiding anticyclonic flow which would bring upper tropospheric material to the North Pacific marine boundary layer has been documented previously (Merrill *et al.*, 1985; Uematsu *et al.*, 1983).

Estimates of air mass mixing, removal of particle and gas-to-particle conversion

Based on the transit time and the concentration of radionuclides measured over Japan and on the ship (see Table 1), we can calculate the first order rate constants for mixing (or dilution) of the air mass as it moves across the Pacific, for removal of the aerosol, and for gas to particle conversion for iodine. The decreasing concentration of substance x with time can be expressed as a first order relationship

$$N = N_0 \exp(-\lambda_x t) \quad (1)$$

where N is the concentration measured on the ship, N_0 is the concentration measured in Japan or estimated concentration over the continent, λ_x is an apparent first order process rate constant for x , and t is the transit time between the ship site and Japan or the continent.

For ^{222}Rn , the concentration decreases only by radioactive decay and by mixing or dilution of the air mass with time. Therefore λ_{Rn} , the total apparent rate constant for concentration decrease, can be expressed as

$$\lambda_{\text{Rn}} = \lambda_m + \lambda_{d1} \quad (2)$$

where λ_m is the mixing constant for the air mass and λ_{d1} is the radioactive decay constant for ^{222}Rn , 0.18 d^{-1} . Here λ_{Rn} , 0.30 d^{-1} , is obtained from (1) by assuming that the ^{222}Rn concentration at ground level over Japan, which is the eastern end of the source regions that supplies ^{222}Rn to the North Pacific. By this assumption, λ_m obtained is the maximal value. However, no data for ^{222}Rn in Japan are available during this period in 1986. A value of 125 pCi m^{-3} ($90\text{--}200 \text{ pCi m}^{-3}$) as measured at Nagoya, Japan (Shimo *et al.*, 1983) in 1981 was used. If ^{222}Rn in the air mass containing the fission products was injected only from the Chernobyl region and not from any other places through the transport, λ_{Rn} is 0.19 d^{-1} which is minimal value. According to Lambert *et al.* (1982), a typical concentration of ^{222}Rn is $\sim 125 \text{ pCi m}^{-3}$ at ground level over continents. From (2), λ_m is then $\leq 0.01\text{--}0.12 \text{ d}^{-1}$ and a mean mixing time of $\sim 8\text{--}\geq 100$ days is obtained. This same mean mixing time is assumed

Table 1. The mean and standard deviation of concentrations of radioactivity observed during the first Chernobyl plume at Japan and over the eastern North Pacific

Nuclide	Japan* (fCi m ⁻³)		Ship** (fCi m ⁻³)	
	Mean	s.d.	Mean	s.d.
^{222}Rn	125000†	—	6400	2300
^{137}Cs	890	400	31	15
^{131}I	4100	1500	120	80
$^{131}\text{I}/^{137}\text{Cs}^{\dagger\dagger}$	4.9	1.4	3.3	1.2

*Tsukuba, May 3–8, 1986 (Aoyama *et al.*, 1986).

**45°–57°N, 153°–160°W, May 13–18, 1986.

†Shimo *et al.* (1983).

††The mean of the ratios for each sample.

to apply to all radionuclides in subsequent calculations.

The concentration of ^{137}Cs decreases by mixing and by removal of particles from the air during transport. Radioactive decay for ^{137}Cs can be neglected in this case. Thus

$$\lambda_{\text{Cs}} = \lambda_r + \lambda_m \quad (3)$$

where λ_r is the aerosol removal rate constant. λ_{Cs} is 0.34 d^{-1} , obtained from (1). From (3) λ_r is then $0.22\text{--}0.33 \text{ d}^{-1}$, and the aerosol residence time is $3.0\text{--}4.5$ days.

The concentration of particulate ^{131}I decreases as a result of radioactive decay, air mass mixing and removal of particles. In addition, particulate ^{131}I is formed by gas to particle exchange processes from gaseous ^{131}I . More than 60 percent of the total ^{131}I from the Chernobyl release was present in the gas phase (Aoyama *et al.*, 1986; Bondietti and Brantley, 1986). If we assume that ^{131}I in the air is removed only in particulate form and that the removal and mixing rates of particulate ^{131}I are similar to ^{137}Cs , then

$$\lambda_I = \lambda_{d2} + \lambda_m + \lambda_r - \lambda_g \quad (4)$$

where λ_{d2} is the radioactive decay constant for ^{131}I , 0.086 d^{-1} and λ_g is the rate constant for converting gaseous to particulate iodine (i.e., the conversion is a source for atmospheric particulate ^{131}I). λ_I is 0.35 d^{-1} from (1). λ_g is then 0.08 d^{-1} , equivalent to a mean gas-to-particle

conversion time of ~ 13 days.

Equations (1), (3), and (4) can also be combined to yield an alternate estimate of λ_g , λ_{g*} , by using the ratios of particulate ^{131}I to ^{137}Cs at the two sites to obtain the ^{131}I gas-to-particle conversion rate.

$$\lambda_{g*} = (\ln(R/R_0)/t) + \lambda_{d2} \quad (5)$$

where R and R_0 are the ratios of particulate ^{131}I to ^{137}Cs on the ship and in Japan, respectively. If the decrease of the R/R_0 ratio followed the radioactive decay of ^{131}I with time, this would indicate no gas-to-particle conversion. If the decrease of the ratio is smaller than that attributable to the radioactive decay of ^{131}I , then we can ascribe the difference to the gas-to-particle conversion of iodine. λ_{g*} is calculated to be 0.05 d^{-1} . The relationship between R/R_0 and the mean gas-to-particle conversion time ($1/\lambda_{g*}$) is plotted in Fig. 4. The mean gas-to-particle conversion time is ~ 20 days (see Fig. 4). In contrast to the other calculations the uncertainty in the transit time, t , does not significantly affect the value of $1/\lambda_{g*}$. However, the variation of R/R_0 (coefficient of variation is 46 percent) results in a large uncertainty in $1/\lambda_{g*}$. It appears from Fig. 4 that the gas-to-particle conversion time for ^{131}I is a minimum of 12 days.

Comparison of the constants with that of naturally occurring aerosols and gases

The results of the various calculations are summarized as follows. A relatively long mean mixing time ($1/\lambda_m$) of >8 days was estimated from ^{222}Rn data. The residence time ($1/\lambda_r$) for aerosol particles in the marine atmosphere containing the radioactive material during this event was 3–5 days, a value that agrees well with the mean tropospheric residence time of continental submicrometer aerosol reported by Graustein and Turekian (1986) using ^{210}Pb and ^{137}Cs measurements. The mean gas-to-particle conversion time ($1/\lambda_g$ or $1/\lambda_{g*}$) for iodine over the Pacific was calculated to be 2–3 weeks, with a minimum of 12 days. This estimate also agrees with the previously reported residence time of 17–19 days for total gaseous iodine (Rahn et al., 1976). By

Fig. 4. The relationship between R/R_0 and the mean gas-to-particle conversion time. t is the transit time from Japan to the ship location. The symbol (●) shows the mean R/R_0 of 0.67 ± 0.31 , and the bar represents ± 1 standard deviation from the mean.

propagating the uncertainties in the mean values of the radionuclide concentrations and the Rn concentration in Japan, as well as the range of estimated travel times we estimate that the uncertainty of the mixing, residence, and conversion times is approximately a factor of two. Despite these uncertainties, this approach provides a means of obtaining an independent estimate of mixing rates, aerosol residence times, and gas/particle conversion rates for iodine. This study shows that the radioactive submicrometer aerosol and the iodine gas from the Chernobyl explosion have residence and conversion times similar to those of naturally occurring aerosols and gases in the atmosphere.

CONCLUSION

Our measurement of atmospheric radionuclides from the Chernobyl accident collected at several island sites and on a ship in the North Pacific lead to the following conclusions:

1. The radioactivity arrived at the various

sites located from 11°–64°N over the North Pacific between 6–14 May 1986. The transit times were about 10–14 days (more than 7000 km from Chernobyl to these sites).

2. Isentropic trajectory analyses and measurement of the natural radionuclides indicate that the continental air containing Chernobyl fission products was transported to the central North Pacific through the middle troposphere.

3. The mean mixing time for boundary layer air parcels with the free troposphere is >8 days. The atmospheric residence time over the North Pacific for the aerosols containing the fission products was 3–5 days. The calculated mean gas-to-particle conversion time for radioactive ¹³¹I was 2–3 weeks, with a minimum of ~12 days.

4. The radioactive submicrometer aerosol and the iodine gas from the Chernobyl explosion have residence and conversion times similar to those of naturally occurring aerosols and gases in the atmosphere.

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