SHORT-LIVED DECAY PRODUCTS OF RADON-222 AND RADON-220
IN AIR AT HACHIJO-JIMA, CHICHI-JIMA AND NAGOYA :
APPLICATION FOR DETERMINING AEROSOL
RESIDENCE TIME

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Abstract. Concentrations of radon-222 short-lived decay products were measured in surface air on the windward sides of Hachijo-jima and Chichi-jima and on the campus of Nagoya University. Concentrations of radon-222 gases and thoron short-lived decay products were also obtained simultaneously on the same site at Hachijo-jima. On the basis of the radon concentration data and meteorological trajectories, decay time constants for tropospheric aerosols due to dilution and mean residence times for atmospheric aerosol particles over ocean areas are estimated to be 0.48~2.5 days and 0.13~0.67 days, respectively.

1. Introduction

Radon-222(Rn), a naturally occurring chemically inert radioactive gas, is produced by the decay of Ra-226 in soils. The Rn exhalation rate over continents is approximately 100 times that over oceans. The only significant mechanism for removing Rn from atmosphere is by radioactive decay(3.82 days half life). These facts make Rn an ideal tracer for large scale transport of air masses across oceans.

In recent years, the decay time constant for lower atmospheric aerosols due to dilution has been estimated from the concentrations of short-lived decay products of radon(Rn-Dts) which has been assumed to be in equilibrium state with Rn (Misaki et al., 1975, Mochizuki and Tanji, 1981). Misaki et al. presented to be about 2.5 days for the decay time constant for aerosol particles due to dilution and Mochizuki and Tanji indicated the values ranging from 1.3 to 2.7 days.

The aerosol residence time in the lower atmospheric air has been estimated from the rate of the concentration of long-lived radioactive nuclides (e.g. Poet et al., 1972, Moore et al., 1979, and others), and from the concentration of aerosol particles (Morita et al., 1973). A number of independent estimations based on Po-210/Pb-210 ratio indicated fairly long tropospheric aerosol residence time of almost one month. However, Poet et al. represented an apparent aerosol residence time averaging only a few days or less. Tsunogai(1980) presented to be 10~14 days for aerosol residence times on the basis of Bi-210/Pb-210 and Po-210/Pb-210 ratios. On the other hand, from the results of observations of aerosol particles, Misaki et al. estimated apparent aerosol residence times to be 0.35 days (for particle radius 0.5 μm) and 5 days (for particle radius 0.03 μm), and Morita et al. obtained those ranging from 0.15 to 0.63 days.

The purpose of the present study is to estimate the decay time constant for lower atmospheric aerosols due to dilution(τ_d) and the

aerosol residence time(τ_r) on the basis of the data observed — (1) concentration of Rn-Dts in surface air carried from the Japan Islands to Hachijo-jima and Chichi-jima, (2) the radioactive equilibrium state between Rn and Rn-Dts in surface air on Hachijo-jima, and (3) Tn-Dts concentration in surface air on the two islands.

2. Measurements

The Rn-Dts were collected on a 1.2 μ m pore size filter (Millipore Filter RA) ; this filter was nearly 100 percent efficient and also had very little self absorption during counting. Sampling times were 60 min, 30 min, and 15 min at almost 10 l/min, 50 l/min, and 20 l/min, at Hachijo-jima, Chichi-jima, and Nagoya, respectively. The activities on the sampled filter counted with zinc-sulfide alpha scintillation counting system. The instruments used at Chichi-jima and Nagoya were operated automatically except once exchange of filter paper a day.

The Rn concentration was measured by the use of a continuous flow-type ionization chamber. A sample air, passed through a filter to eliminate Rn- and Tn-Dts, was admitted to the ionization chamber at almost 1.0 l/min. Ionization current was measured by a vibrating-reed-electrometer ; the electric signals caused by alpha particles were selected from those by beta-, gamma, and cosmic rays using an advanced pulse technique. This technique will be described in detail in the near future.

The measurement of Tn-Dts was carried out with manual operation using the filter-pack technique applied for Rn-Dts measurement. The sampling time was about one day and the counting time to obtain a decay curve was several hours.

The measurements of Rn- and Tn-Dts concentrations were carried out from January 14 to 20, 1981, on the two islands (Hachijo-jima and Chichi-jima) and the measurement of Rn-Dts concentration at the campus of Nagoya University. In addition to these measurements, Rn concentration was also obtained at Hachijo-jima.

Fig. 1. Diurnal variations of Rn and Rn-Dts concentration.

3. Results and Discussions

Figure 1 shows diurnal variations of Rn-Dts concentration measured at Nagoya (a) and of Rn concentration on Hachijo-jima (b). On the basis of air masses trajectories, transit times of air masses from the Japan Islands to Hachijo-jima were estimated to be 8~12 hours during observation period. The curve (c) indicates a correction curve ; this is shifted to the left direction on time axis by only transit time from the curve (b). Radioactive decay during transit was neglected since the half life of Rn is 3.82 days. Not only the concentration but also a pattern of variation shown by the curve (c) agreed fairly well with those shown by the curve (a).

Fig. 2. Diurnal variation of Rn-Dts concentration.

Figure 2 shows the result observed on Chichi-jima ; the curve (c) involved correction based on transit times (1~1.5 days) in the same manner as Fig.1(c). In this case, the correction of radioactive decay of Rn during transit was also made. The Rn-Dts concentration of Chichi-jima was almost one-tenth of Nagoya. It was found that the temporary variation of Rn-Dts of Chichi-jima agreed roughly that of Nagoya, although the variation of Chichi-jima was rather blunt.

Figure 3 shows examples of the decay of radioactivities sampled on filters, and indicates that Tn-Dts was able to be detected in surface air on Hachijo-jima but was not detected on Chichi-jima.

Figure 4 shows the good correlation between Rn and Rn-Dts in air on Hachijo-jima and a close agreement of concentration between them suggests the radioactive equilibrium state. Since Rn is in a gaseous state in air, it dissipates from air mass due to radioactive decay and dilution process. On the other hand, Rn-Dts vanish due to the radioactive decay and the scavenging and removal processes as aerosol particles. Nevertheless, the radioactive equilibrium state between Rn and Rn-Dts is presumed in air over oceans since the half life of Rn is considerably long compared with that of Rn-Dts (0.0278 days) and the τ_d is larger than the radioactive half life of Rn-Dts.

Fig. 3. Decay curve of radioactivity.

Figure 5 shows the variation of mean concentrations of Rn and/or Rn-Dts and Tn-Dts as a function of distance from the Japan Islands. The degree of the decrease of Tn-Dts concentration is more acute than that of Rn and/or Rn-Dts ; this is because (1) Tn-Dts does not accompany with its parent nuclide (Tn) whose half life is 54 seconds, (2) the half life of Tn-Dts (ThB : 10.6 hours) is short compared to that of Rn, and (3) Tn-Dts are considered to exist as radioactive aerosol particles in atmospheric air.

Let Q and Q_0 represent radioactivity concentration on the island (Hachijo-jima and/or Chichi-jima) and the Japan Islands, respectively, we have

$$Q = Q_0 \exp(-\lambda t) \quad (1)$$

Fig. 4. Correlation between Rn and Rn-Dts.

Fig. 5. Radioactivity vs. distance.

where Λ is an apparent removal rate for Rn and/or Tn-Dts and t denotes the time length taken after the air mass left the Japan Islands. The apparent removal rate for Rn, Λ^R , is expressed as follows

$$\Lambda^R = \lambda_a^R + \lambda_d \quad (2)$$

where λ_a^R is the radioactive decay constant of Rn (0.182 d^{-1}) and λ_d is the removal rate for atmospheric particles due to dilution process.

In the case of Tn-Dts, the apparent removal rate of Tn-Dts, Λ^T , is represented as follows

$$\Lambda^T = \lambda_a^T + \lambda_d + \lambda_r \quad (3)$$

where λ_a^T is the radioactive decay constant of Tn-Dts (1.57 d^{-1}) and λ_r is the removal rate for atmospheric particles due to the combined action of all particle scavenging and removal processes except dilution process.

The λ_d was estimated to be $0.48 \sim 2.5 \text{ d}^{-1}$ ($\tau_d = 1/\lambda_d = 1.2$ days; averaged individual λ_d) from Eqs. (1) and (2) using the data shown in Fig. 5 and mean wind velocity. On the other hand, the Λ were estimated to be $2.5 \sim 8.5 \text{ d}^{-1}$ ($\tau = 1/\Lambda = 0.12 \sim 0.4$ days) from the data shown in Fig. 5. Hence, the λ_r was found from calculation to be $1.5 \sim 7.5 \text{ d}^{-1}$ ($\tau_r = 1/\lambda_r = 0.67 \sim 0.13$ days) from Λ , λ_d , and λ_a .

The result estimated almost agrees with the value obtained by Morita et al. and roughly agrees with that by Misaki et al. However, there are disagreement between the present work and other researchers (Poet et al. and Tsunogai). These discrepancy shall be discussed after the appointed observation during 1981~1983, taking account of: (1) local anomaly, (2) vertical profile, (3) diurnal variation, and (4) seasonal variation for the concentrations of Rn, its long-lived decay products, and Tn-Dts.

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