ON THE ORIGIN AND TRANSPORT OF Rn-222 IN THE ATMOSPHERE

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Abstract. We propose a new analytical treatment about the variation of ^{222}Rn concentration with an aim to clarify origin, transport, and the cause of seasonal variation of ^{222}Rn in the atmosphere. It was concluded that about a half of ^{222}Rn concentration observed at Nagoya originates near the measuring site and shows diurnal changes, and the another half originates far from Nagoya and shows seasonal changes.

1. Introduction

Many researchers have studied the diurnal variations and the vertical profiles of ^{222}Rn mainly based on the stability of the air, eddy diffusivity, and the diffusion equation. However, not so many observations and analyses concerning the seasonal variation of ^{222}Rn have been made, e. g. (Lockhart, 1964), (Shimo and Ikebe, 1979), and until now the knowledge about the seasonal variation is very limited (Wilkening, 1959) (Rangarajan et al, 1974).

In this report, we propose a new analytical treatment about the variation of ^{222}Rn concentration measured at Nagoya with an aim to clarify origin, transport, and the cause of seasonal variation of ^{222}Rn in the atmospheric air at Nagoya.

2. Analysis

Figure 1 shows an example of the variation of ^{222}Rn concentration measured at Nagoya University by a filter-pack method. Until now, one have usually assumed implicitly that all ^{222}Rn atoms measured at 1 meter above the ground surface had originated from the ground near the measuring site when the measurements were made on land. But in this work we assumed that ^{222}Rn concentration can be separated to the following two components :

(i) Rn atoms originated

Fig.1. An example of the variation of ^{222}Rn concentration.

near the measuring site, which is denoted hereafter by "diurnal variation component." For this component, the time variation may be controlled by factors such as atmospheric stability, eddy diffusivity, and Rn exhalation rate near the site. The ordinary diffusion equation may be applied for this component.

(ii) Rn atoms originated far from the measuring site, which is denoted hereafter by "background component." This component is assumed to be distributed uniformly in the atmospheric boundary layer since these Rn atoms have been mixed enough in the boundary layer until they arrive at the measuring site from their origin. The seasonal variation may be explained by this component if the values of diurnal variation component are fairly constant throughout the year.

In Fig.1, a broken line represents the background component. The dotted curve was drawn about 16 pCi/m^3 below the minimum concentration on each day. The value of 16 pCi/m^3 was chosen as the sum of a minimum contribution of the diurnal variation component (15 pCi/m^3)* and a global background of the air above sea surface (1 pCi/m^3) (Wilkening and Clements 1975).

The background component obtained at the campus during summer season (July - Sept., 1977, 1978, 1981) and during winter season (Dec. 1980 - March 1981) was analysed. An air mass trajectory was drawn on each day by using 850 mb weather chart. According to the history of the air arrived at Nagoya, the observed air mass was classified into two groups — maritime air mass and continental air mass.

Figure 2 shows the relation between the background component of Rn concentration and the pass-over-island time in the case of maritime air mass. The pass-over-island time on Japan Island was determined from the trajectories described above.

3. Transport model of background component of Rn

According to the model (ii) described in section 2, the concentration of background component Rn, n , is given by

Fig.2. The relation between the background component and the pass-over-island time.

* According to the model of Jacobi and Andre (1963), minimum Rn concentration corresponding to K-profile for SSN is about 15 % of normal Rn concentration for NWN and NNN. In the work, about 15 % of the mean Rn concentration at Nagoya ($\sim 100 \text{ pCi/m}^3$) is adopted for the minimum Rn concentration on each day.

$$\frac{dn}{dt} = -(\lambda_d + \lambda_r)n + S \quad (1)$$

where, t : the pass-over-islands time (d)

λ_r : removal constant from the atmospheric boundary layer to the upper free atmosphere (d^{-1})

λ_d : radioactive decay constant (d^{-1})

S : rate of Rn supply ($pCi \cdot m^{-3} \cdot d^{-1}$).

S is given by

$$S = E/H \quad (2)$$

where, E : Rn exhalation rate ($pCi \cdot m^{-2} \cdot d^{-1}$)

H : height of the atmospheric boundary layer (m).

We obtain the solution of eq. (1) from the initial condition : $t = 0$: $n = 0$,

$$n = \frac{S}{\lambda} (1 - e^{-\lambda T}) \quad (3)$$

where $\lambda = \lambda_r + \lambda_d$ and T is the pass-over-islands time from landing point in Japan Islands to Nagoya. The solid line shown in Fig.2 is drawn using the values of removal time constant

$$\tau_r = \frac{1}{\lambda_r} = 2 \text{ days and } \frac{S}{\lambda} = 100 \text{ pCi} \cdot m^{-3}.$$

The saturation concentration of $n(t \rightarrow \infty) = S/\lambda = 100 \text{ pCi} \cdot m^{-3}$ was determined from the similar relationship (n vs t) in the case of continental air masses. The observed data agree fairly well with this curve. The value of $\tau_r = 2$ days seems to be consistent with the values obtained by Misaki et al (1975) and Mochizuki and Tanji (1981).

4. Discussions and conclusions

In the treatment described above, we can obtain the value $S = 68 \text{ pCi} \cdot m^{-3} \cdot d^{-1}$ from $S/\lambda = 100 \text{ pCi} \cdot m^{-3}$ and $\lambda = 0.68 \text{ d}^{-1}$.

If we adopt the value of $E = 36720 \text{ pCi} \cdot m^{-2} \cdot d^{-1}$ (Wilkening et al 1972) as the mean exhalation rate of Rn over Japan Islands, we get $H = 540 \text{ m}$ from eq.(2), which is consistent with the height of atmospheric boundary layer (0.5 - 1.5 km).

In Fig.3, the monthly mean values of total and background Rn concentrations are shown by open and solid circles, respectively. The former values are about 2 times larger than the latter one on each month. This means that about a half of Rn

Fig.3. Monthly mean values of total and background Rn concentrations.

atoms observed at Nagoya originates near the measuring site, and the remainder (another half) originates far from the measuring site.

Furthermore, we can see a clear seasonal variation as for the background component, but can't see as for the diurnal variation component (the difference between the open and solid circle). This means that a main cause of the seasonal variation at Nagoya is the seasonal change of the concentration of background component. The level of background component may be controlled mainly by the air mass types which are fixed from synoptic weather conditions characteristic for each season. More detailed analyses including the case of continental air masses will be published elsewhere.

References

- Jacobi, W. and K. Andre, the vertical distribution of radon-222, radon-220 and their decay products in the atmosphere, *J. Geophys. Res.*, 68, 3799, 1963.
- Lockhart, L. B. Jr., Radioactivity of the radon-222 and radon-220 series in the air at ground level, "The Natural Radiation Environment", pp. 331, 1964 (Univ. Chicago Press).
- Misaki, M., M. Ikegami, and I. Kanasawa, Deformation of the size distribution of aerosol particles dispersing from land to ocean, *J. Met. Soc. Japan* 53, 111-120, 1975.
- Mochizuki, S. and T. Tanji, Radioactive aerosols in the atmosphere over ocean near Japan Islands (in Japanese), *Res. Lett. Atmosph. Electr.*, 1, 9-14, 1981.
- Rangarajan, C., Smt. S. Gopalakrishnan, and C. D. Eapan, The diurnal and seasonal changes in short-lived radon-thoron daughters' concentrations in the coastal and inland regions of India and their possible relation to regional climatology, *Pure Appl. Geophys.*, 112, 375, 1974.
- Shimo, M. and Y. Ikebe, ^{222}Rn , ^{220}Rn and their short lived daughter products concentrations in the atmosphere, *Hoken-Butsuri*, 14, 251, 1979 (in Japanese).
- Wilkening, M. H., Daily and annual courses of natural atmospheric radioactivity, *J. Geophys. Res.*, 64, 521, 1959.
- Wilkening, M. H., W. E. Clements, and D. Stanley, Radon-222 flux measurements in widely separated regions, *The Nat. Radiat. Env. II*, ed. J. A. S. Adams, 2:717-30, USERDA CONF-720805, 1972.
- Wilkening, M. H. and W. E. Clements, Radon 222 from the ocean surface, *J. Geophys. Res.*, 80, 3828, 1975.

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