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Notes

A Simple Rainout Model for Radon Daughters

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This paper presents a simplified time-independent model which calculates radioactivities of short-lived radon daughters in rainwater as a function of rainfall rate for unit concentration of radon in cloud. The model incorporates processes of impact collection for cloud droplets by raindrops, removal of raindrops from cloud and transport time of raindrops from the cloud base to the ground as forms of semi-empirical formulas. Radon concentrations in rain clouds are estimated from comparisons between model calculations and ground-level observations made over a few years.

1. Introduction

Rn-222 is of considerable interest in studies of global circulation and the long range transport of pollutants. However, the measurements in higher altitudes take much expense. It would be quite valuable if ^{222}Rn concentrations in cloud air could be estimated from radon daughters contained in rainwater observed at ground level.

Several papers have so far been published describing in-cloud scavenging of radon daughters, which were briefly reviewed by Fujinami et al.[1] and Horng and Jiang.[2] Among those, only the following two papers deal with ^{222}Rn and its daughters in three phases, i.e. cloud air, cloud droplets and raindrops, and take into account microphysical processes occurring in clouds. One is the work of Minato[3] and the other is that of Ikebe et al.[4] However, the former model does not include the removal process of raindrops from cloud, and the latter assumes a constant removal rate of cloud droplets for any type of rainfall event. These treatments may be considered to be somewhat far from reality. The purpose of this paper is to build a more reasonable model by improving the models described in the above two papers.

2. The model

Let us consider here a rain cloud under a steady state condition, where Rn (^{222}Rn) and its daughters are uniformly distributed.

2.1. Cloud droplets. Since the half-life of RaA (^{218}Po) is quite short compared to Rn, we can assume radioactive equilibrium between them in cloud air. Since the produced RaA atoms attach to the cloud droplets immediately,[4] the radon daughters in cloud droplets per unit volume can be expressed as follows.

$$a_{\mathrm{Rn}} = \lambda_{\mathrm{A}} n_{\mathrm{A}} + \psi_{\mathrm{c}} n_{\mathrm{A}}\,, \tag{1}$$

$$\lambda_{\mathrm{A}} n_{\mathrm{A}} = \lambda_{\mathrm{B}} n_{\mathrm{B}} + \psi_{\mathrm{c}} n_{\mathrm{B}}\,, \tag{2}$$

$$\lambda_{\mathrm{B}} n_{\mathrm{B}} = \lambda_{\mathrm{C}} n_{\mathrm{C}} + \psi_{\mathrm{c}} n_{\mathrm{C}}\,. \tag{3}$$

Here, a_{Rn} is the radioactivity of Rn, n_{A}, n_{B}, and n_{C} are the number of cloud droplets with RaA, RaB (^{214}Pb), and RaC (^{214}Bi), respectively, λ_{A}, λ_{B}, and λ_{C} are the decay constants of RaA, RaB, and RaC, and ψ_{c} is the removal rate of cloud droplets by raindrops.

Then, the activities of RaA, RaB, and RaC on cloud droplets per unit volume in cloud take the forms,

$$a_{\mathrm{A}} = \lambda_{\mathrm{A}} n_{\mathrm{A}} = \frac{\lambda_{\mathrm{A}}}{\lambda_{\mathrm{A}} + \psi_{\mathrm{c}}}\, a_{\mathrm{Rn}}\,, \tag{4}$$

$$a_{\mathrm{B}} = \lambda_{\mathrm{B}} n_{\mathrm{B}} = \frac{\lambda_{\mathrm{B}} \lambda_{\mathrm{A}} n_{\mathrm{A}}}{\lambda_{\mathrm{B}} + \psi_{\mathrm{c}}} = \frac{\lambda_{\mathrm{B}}}{\lambda_{\mathrm{B}} + \psi_{\mathrm{c}}}\, a_{\mathrm{A}}\,, \tag{5}$$

$$a_{\mathrm{C}} = \lambda_{\mathrm{C}} n_{\mathrm{C}} = \frac{\lambda_{\mathrm{C}} \lambda_{\mathrm{B}} n_{\mathrm{B}}}{\lambda_{\mathrm{C}} + \psi_{\mathrm{c}}} = \frac{\lambda_{\mathrm{C}}}{\lambda_{\mathrm{C}} + \psi_{\mathrm{c}}}\, a_{\mathrm{B}}\,, \tag{6}$$

2.2. Raindrops. The radon daughters in raindrops per unit volume in the cloud are represented by

$$\psi_{\mathrm{c}} n_{\mathrm{A}} = \lambda_{\mathrm{A}} N_{\mathrm{A}} + \psi_{\mathrm{r}} N_{\mathrm{A}}\,, \tag{7}$$

$$\psi_{\mathrm{c}} n_{\mathrm{B}} + \lambda_{\mathrm{A}} N_{\mathrm{A}} = \lambda_{\mathrm{B}} N_{\mathrm{B}} + \psi_{\mathrm{r}} N_{\mathrm{B}}\,, \tag{8}$$

$$\psi_{\mathrm{c}} n_{\mathrm{C}} + \lambda_{\mathrm{B}} N_{\mathrm{B}} = \lambda_{\mathrm{C}} N_{\mathrm{C}} + \psi_{\mathrm{r}} N_{\mathrm{C}}\,, \tag{9}$$

where N_{A}, N_{B}, and N_{C} are the number of raindrops with RaA, RaB, and RaC, respectively, and ψ_{r} is the removal rate of raindrops from the cloud. From these equations, the activities of RaA, RaB, and RaC in raindrops per unit volume can be rewritten as

$$A_{\mathrm{A}} = \lambda_{\mathrm{A}} N_{\mathrm{A}} = \frac{\psi_{\mathrm{c}} \lambda_{\mathrm{A}} n_{\mathrm{A}}}{\lambda_{\mathrm{A}} + \psi_{\mathrm{r}}} = \frac{\psi_{\mathrm{c}}}{\lambda_{\mathrm{A}} + \psi_{\mathrm{r}}}\, a_{\mathrm{A}}\,, \tag{10}$$

$$A_{\mathrm{B}} = \lambda_{\mathrm{B}} N_{\mathrm{B}} = \frac{\psi_{\mathrm{c}} \lambda_{\mathrm{B}} n_{\mathrm{B}} + \lambda_{\mathrm{B}} \lambda_{\mathrm{A}} N_{\mathrm{A}}}{\lambda_{\mathrm{B}} + \psi_{\mathrm{r}}} = \frac{\psi_{\mathrm{c}}}{\lambda_{\mathrm{B}} + \psi_{\mathrm{r}}}\, a_{\mathrm{B}} + \frac{\lambda_{\mathrm{B}}}{\lambda_{\mathrm{B}} + \psi_{\mathrm{r}}} A_{\mathrm{A}}\,, \tag{11}$$

$$A_{\mathrm{C}} = \lambda_{\mathrm{C}} N_{\mathrm{C}} = \frac{\psi_{\mathrm{c}} \lambda_{\mathrm{C}} n_{\mathrm{C}} + \lambda_{\mathrm{C}} \lambda_{\mathrm{B}} N_{\mathrm{B}}}{\lambda_{\mathrm{C}} + \psi_{\mathrm{r}}} = \frac{\psi_{\mathrm{c}}}{\lambda_{\mathrm{C}} + \psi_{\mathrm{r}}}\, a_{\mathrm{C}} + \frac{\lambda_{\mathrm{C}}}{\lambda_{\mathrm{C}} + \psi_{\mathrm{r}}} A_{\mathrm{B}}\,. \tag{12}$$

2.3. Ground level. The radon daughters under the cloud base are calculated from the following equations.

$$\frac{dA_{\mathrm{A}}}{dt} = -\lambda_{\mathrm{A}} A_{\mathrm{A}}\,, \tag{13}$$

$$\frac{dA_{\mathrm{B}}}{dt} = \lambda_{\mathrm{B}}(A_{\mathrm{A}} - A_{\mathrm{B}})\,, \tag{14}$$

$$\frac{dA_{C}}{dt} = \lambda_{\mathrm{C}}(A_{\mathrm{B}} - A_{\mathrm{C}})\,. \tag{15}$$

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The activities at ground level can be obtained by substituting the transport time from the cloud base to the ground into the solutions of eqs 13–15.

2.4. Radioactivity of rainwater. Now, in order to compare the model mentioned above with ground-level observations, we have to transform the radioactivities of raindrops per unit volume in cloud air, A_A, A_B, and A_C expressed in units of Bq/m^3 to the activities of rainwater, α_A, α_B and α_C, in units of Bq/mL using the following relation,

$$\alpha_A = \frac{\rho}{L_r}A_A, \quad \alpha_B = \frac{\rho}{L_r}A_B, \quad \alpha_C = \frac{\rho}{L_r}A_C, \qquad (16)$$

where ρ is the density of rainwater in g/mL, and L_r is the water content of raindrops in the cloud in g/m^3, which can be derived from the Marshall-Palmer formula[5] in g/m^3 as a function of rainfall rate P in mm/h,

$$L_r = 0.089P^{0.84}. \qquad (17)$$

The collision rate for a single cloud droplet of radius R by raindrops between R' and R'+dR' is defined as

$$H(R) = \int_{R'} \pi R'^2 E(R', R)[v(R')-v(R)]s(R')dR'. \quad (18)$$

Here, E is the collision efficiency, v the velocities of cloud droplets and raindrops and s the concentration of raindrops.[3] Then, the removal rate can be calculated from the following formula.

$$\psi_c = \frac{\int_C (R)H(R)dR}{\int_C (R)dR}, \qquad (19)$$

where c is the concentration of cloud droplets, which is given in Reference 3 as a function of the mean radius of cloud droplets.

Numerical calculations for the above equations yield the following empirical formula.

$$\psi_c = (0.028-0.036e^{-0.19<R>})P^{0.83}, \qquad (20)$$

where ψ_c is the removal rate of cloud droplets per minute and $<R>$ the mean radius of cloud droplets in μm.

Next is to evaluate ψ_r , i.e. the removal rate of raindrops per minute. The rainfall rate is defined as follows.

$$P = \psi_r L_r D. \qquad (21)$$

Here, D is the cloud depth, i.e. the difference between cloud top and cloud base. Then, we obtain

$$\psi_r = 0.187\frac{P^{0.16}}{D} \qquad (22)$$

where D is in units of km.

Finally, we need an average velocity of raindrops under the cloud $< v >$ to calculate the transport time from the cloud base to the ground, which can be derived from the following relation.

$$P = < v > L_r. \qquad (23)$$

Namely,

$$< v > = 0.187P^{0.16}, \qquad (24)$$

where $<v>$ is in units of km/min.

3. Calculated results and comparisons

Table 1 gives the model calculated results. Figure 1 shows the whole year data of short-time observations made during rainfall events at Nagoya[1] along with a few events data collected in Taiwan[2] and at Kashiwazaki.[6] The two curves in the figure represent fits of the model calculation given in Table 1 to the almost maximum and minimum specific activities. In this way, we can estimate Rn concentrations inside clouds from ground-level observations of rainwater radioactivities using

TABLE 1: Model calculated rainfall rate dependence of specific activity of rainwater at ground level per 1 Bq/m^3 of ^{222}Rn in cloud air

Rainfall rate	RaB+RaC/Bq mL^{-1}		
/mm h^{-1}	Winter	Summer	Whole
0.1	6.49	11.4	9.45
0.2	5.50	9.93	8.16
0.3	4.83	8.90	7.26
0.5	3.94	7.45	6.01
0.8	3.12	6.04	4.82
1	2.74	5.38	4.28
2	1.73	3.52	2.75
3	1.26	2.63	2.04
5	0.817	1.76	1.35
8	0.533	1.18	0.892
10	0.432	0.967	0.728
20	0.220	0.512	0.379
30	0.146	0.349	0.256
50	0.0871	0.213	0.155
70	0.0617	0.154	0.111

Conditions; average cloud droplet radius=11 μm, cloud base height=2 km, and cloud top height= 10, 5, and 7.5 km for winter, summer, and whole year, respectively.

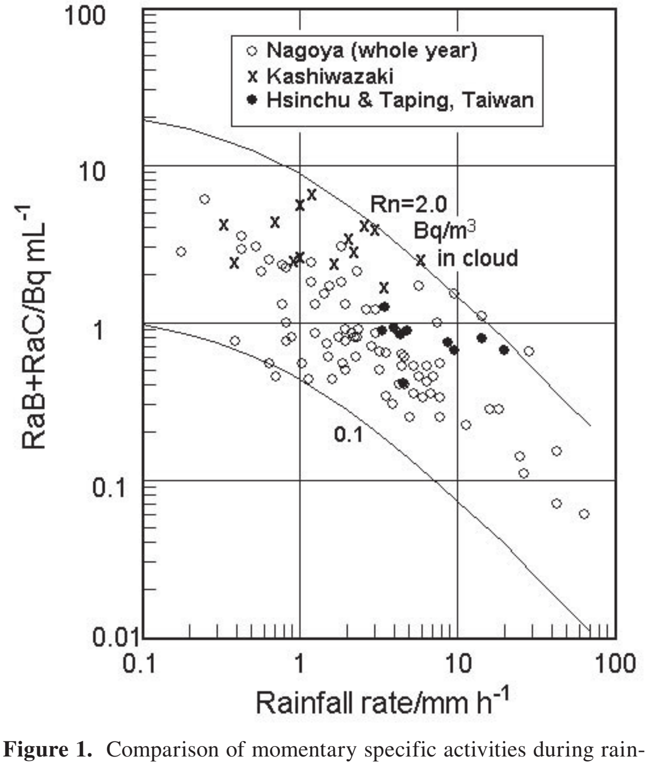

Figure 1. Comparison of momentary specific activities during rainfall observed at ground level with model calculations for the corresponding ^{222}Rn concentrations in cloud air using the parameter for whole year given in Table 1

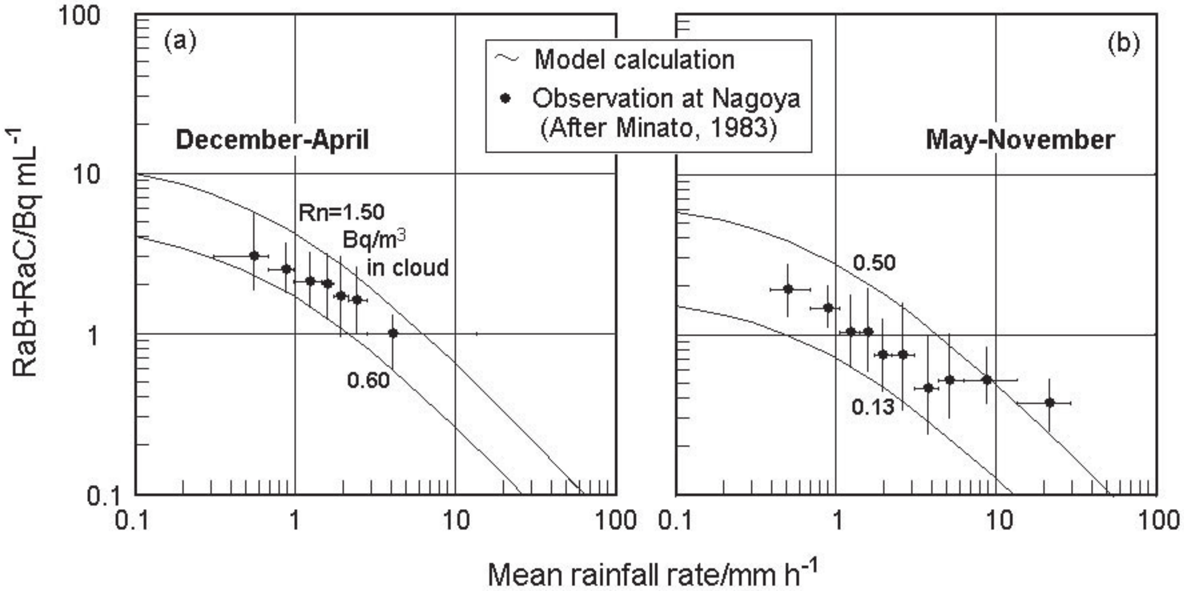

Figure 2. Comparison of specific activities averaged over a single rainfall event observed at ground level[3] with model calculations for the corresponding ^{222}Rn concentrations in cloud air using the parameters in Table 1 for (a) winter and (b) summer. Vertical bars represent one standard deviation.

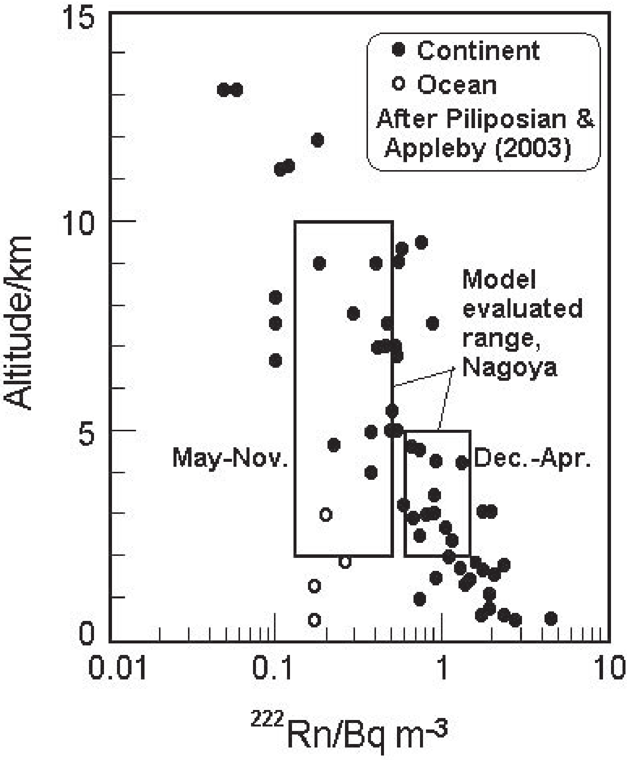

Figure 3. Comparison of observed ^{222}Rn concentrations over continents and a large ocean[7] with model evaluated ranges (±1σ) for those in clouds, based on the data shown in Figure 2.

Table 1.

Figure 2 shows the seasonal data of single rainfall averaged activities observed at Nagoya for a few years.[3] The two curves in the figure correspond nearly to one standard deviation (1σ). Thus, the model evaluated ±1σ ranges of Rn concentrations in clouds amount to 0.60–1.50 Bq/m^3 for a period of December–April and 0.13–0.50 Bq/m^3 for May–November. In order to confirm the validity of the method presented in this paper, let us compare these values with observed data.

Figure 3 shows the data observed over Eurasian and North American continents and over Hawaii, which were summarized by Piliposian and Appleby.[7] Although Nagoya is located at a land-sea interface, it may be all right to use the data taken over the continents for December–April, since continental air masses are dominant in this season. On the other hand, the data of Hawaii would be useful in interpreting the evaluated result for May–November when maritime air masses some times move towards the Japanese Islands.

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