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Monte Carlo Calculation of Global ^{222}Rn Transport at Middle Latitude Using a Simple One-dimensional Model

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A simple Monte Carlo computational technique was developed to simulate general circulation of ^{222}Rn *in the troposphere between 30° and 60° North. Fitting the calculated results to previously published observed profiles, averaged vertical eddy diffusion coefficients were derived, along with exhalation rate values of 1.5 and 1.0 atoms/cm²·s for summer (June-August) and winter (December-February), respectively.*

To assess the validity of the method provided by the present report, the calculated results were compared with a number of experimental measurements of other researchers.

Contribution of ^{222}Rn *originated in the Eurasian continent to the Japan Islands was calculated as an example of applications.*

Key Words : radon-222, global transport, Monte Carlo calculation, middle latitude, eddy diffusion coefficient, exhalation rate

1. Introduction

Global air pollution is becoming more and more a matter of public concern recently. It must be of significance under such circumstances to develop a simple and convenient computational technique capable of making rough estimation speedily even using a personal computer so that many persons concerned can apply it easily.

The pollutant concentration may be governed by vertical mixing and horizontal transport or 'advection'. In other words, the profiles of eddy diffusion coefficient (K) and wind speed (U) would be needed in the calculations. While global U profiles have been reported here and there, there are still very few accurate data of K profile available at present for computing global pollutant transport. Radon-222 (Rn) and its short-lived daughters have frequently been used as tracers for studying local and/or regional vertical mixing in the atmosphere. In addition, the lifetime and source characteristics (*i.e.*, emitted at the surface) of Rn are similar in several ways to many air pollutants such as NO_x, SO_x and other moderately reactive hydrocarbons.

In this study, we try to make a Monte Carlo model based on the general circulation of Rn at middle latitude in the troposphere. The longitudinally averaged vertical distribution of eddy diffusion coefficient will be determined by comparing observed profiles with the results calculated by the present model for various K profiles. Although Liu et al.[1] have estimated the K profiles for the North American continent under relatively simplified assumptions, we will evaluate the profile using a more realistic model. Some authors[2),3)] have proposed a global one-dimensional model or the transport of Rn and its long-lived daughters. Those were a one- or two-layer model using vertically averaged wind speed, since their purpose was to obtain a mean residence time of aerosols in the troposphere. This paper deals with the altitude distribution of Rn as a function of longitude taking into account globally averaged U and K profiles.

2. Method of Computation

We treat a one-dimensional flow along the horizontal axis. The transport equation can be expressed as

$$\frac{\partial C}{\partial t}+U\frac{\partial C}{\partial x}=\frac{\partial}{\partial z}\left(K\frac{\partial C}{\partial z}\right)-\lambda C, \quad (1)$$

where, C, Rn concentration, t, time, U, wind speed, x and z, horizontal and vertical directions, respectively, K, eddy diffusion coefficient, and λ, decay constant of Rn. Here, diffusion along the x axis, compared to the transport by advection (the second term of the left-hand side of the equation), is assumed to be negligible; the vertical wind speed is taken to be zero.

The boundary condition is represented by

$$\int_0^\infty\int_0^\infty \lambda C \mathrm{d}x\mathrm{d}z=E, \quad (2)$$

where, E is the exhalation rate.

Equation (1) can be rewritten as

$$\frac{\partial C}{\partial t}+U\frac{\partial C}{\partial x}=K\frac{\partial^2 C}{\partial z^2}+\frac{\partial K}{\partial z}\frac{\partial C}{\partial z}-\lambda C. \quad (3)$$

A segment model is used in this calculation. A Rn trajectory is represented in this model as a set of straight-line segments for a small time interval, and the directions are allowed to take place only at the end of each segment. Although these approximations introduce a systematic error, it can often be reduced by making the segments sufficiently small.

Several points in altitude distributions of U and K are stored as input data in advance. The values at an altitude of interest are calculated from linear interpolation in case of need.

We get one Rn atom to exhale from the ground at $t=0$; $x=0$; $z=0$. A life of each Rn is randomly sampled just before tracing every trajectory from an exponential distribution according to the last term of the right-hand side of equation (3), *i.e.*,

$$\tau=-\frac{1}{\lambda}\ln\xi, \quad (4)$$

where, ξ is a uniform random number between zero and one.

A vertical displacement after a time interval Δt can be derived from the first (diffusion) and the second (quasi-advection) terms of the right-hand side of the equation as follows. One component (due to the quasi-advection term) is expressed by

$$\Delta z_a=\frac{\partial K(z)}{\partial z}\Delta t. \quad (5)$$

Namely, for the i-th position z_i, the tentative coordinate is approximately derived as

$$z'=z_i+\frac{\Delta K(z_i)}{\Delta z}\Delta t. \quad (6)$$

Another one (the diffusion term) is determined by random sampling from a Gaussian distribution, *i.e.*,

$$\Delta z_d=\sqrt{2K(z_m)\Delta t}\sqrt{-2\ln\xi_1}\sin 2\pi\xi_2, \quad (7)$$

where, ξ_1 and ξ_2 are the random number and

$$z_m=\frac{z_i+z'}{2}. \quad (8)$$

The new coordinate is then given by

$$z_{i+1}=z'+\Delta z_d. \quad (9)$$

A small displacement in the x direction can be calculated from the second (advection) term of the left-side hand of equation (3) by

$$\Delta x=U\Delta t. \quad (10)$$

To put it in the concrete, it is approximated as

$$x_{i+1}=x_i+\frac{U(z_i)+U(z_{i+1})}{2}\Delta t. \quad (11)$$

Thus, the new position (x_{i+1}, z_{i+1}) can be successively computed through equations (4)-(11) until the Rn decays at $t=\tau$.

When the Rn atom gets to ground- or sea-level due to diffusion, we make it reflect at $z=0$, since it is an inert gas. The steady state Rn concentrations can be obtained from the density of segments normalized by equation (2) as a function of x and z.

To test our computational method, we attempt to make a sample calculation for a more simplified condition and compare it with other data. Jacobi and Andre[4] have approximately solved the steady state diffusion equation

$$\frac{\mathrm{d}}{\mathrm{d}z}\left(K\frac{\mathrm{d}C}{\mathrm{d}z}\right)-\lambda C=0. \quad (12)$$

Figure 1 is the two profiles among five idealized K profiles presented by Jacobi and Andre. Figure 2 shows the comparison between their numerical calculations and our results. The Monte Carlo calculations were performed for $\Delta t=1$ hour and the sample size were 10 000 histories. The same time interval and sample size will be chosen for all the calculations in this paper. The agreement

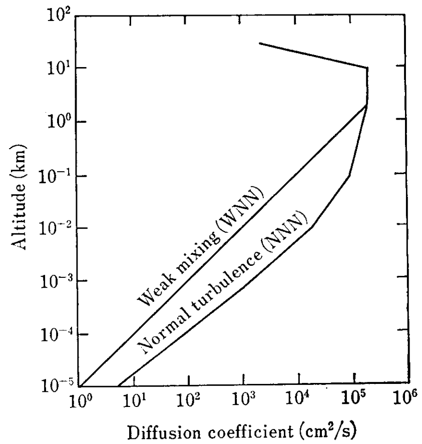

Fig. 1 Eddy diffusion coefficient profiles adapted by Jacobi and Andre.

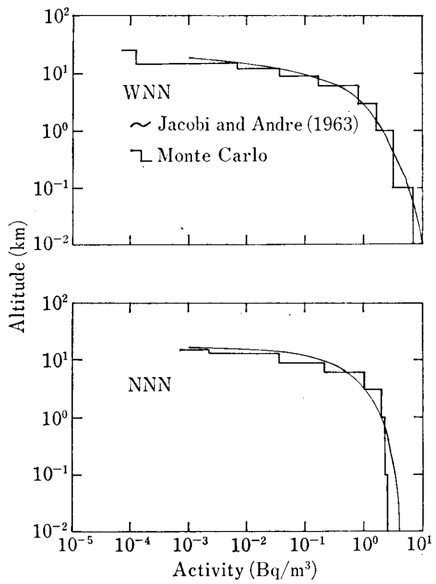

Fig. 2 Comparison of the Monte Carlo calculated vertical distributions of Rn with numerically obtained results by Jacobi and Andre for K profiles shown in Fig. 1.

between them is fairly well. Jacobi and Andre neglected the quasi-advection term (corresponding to the second term of the right-hand side of equation (3)) implicitly included in equation (12). Differences at very low and very high altitudes seen in Fig. 2 may be attributable to the above neglect.

3. Basic Data

According to equations (1) and (2), U and K profiles and the distribution of E are required in the calculations. We use longitudinally averaged profiles in this study.

Figure 3 is the profiles of wind speed parallel to longitude averaged over latitudes between 30° and 60° N deduced from ref. 5), where positive component means the westerly.

Figure 4 is the K profiles used in the calculations. How to estimate the profiles will be stated later.

Figure 5 is the contour maps of Rn concentration calculated for winter (December-February) and summer (June-August) seasons for a point source on the x axis. We notice in the figure that there are depressed parts at an altitude of about 2 to 3 km, especially in winter. We consider as one of the reasons that this is caused by reflection of Rn from

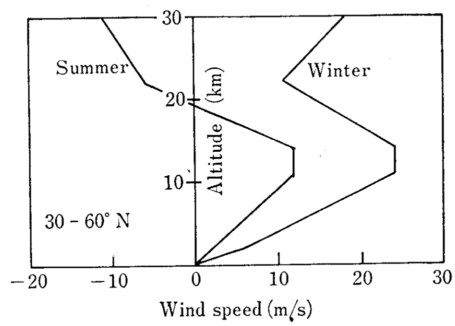

Fig. 3 Wind speed profiles used in the calculations.

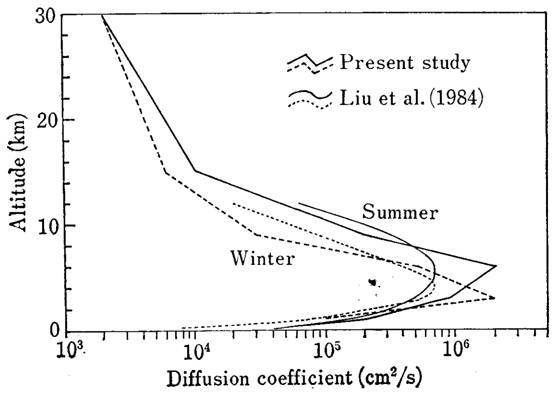

Fig. 4 Eddy diffusion coefficient profiles used in the calculations.

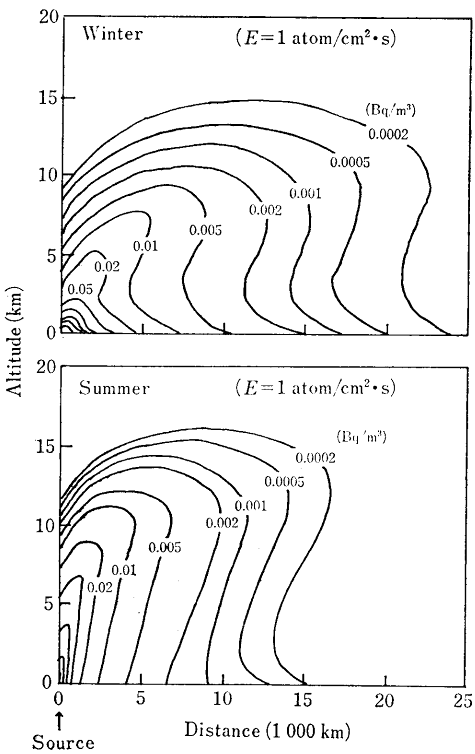

Fig. 5 Monte Carlo calculated results for point Rn sources for U and K profiles shown in Figs. 3 and 4.
The concentrations for curves without numerals drawn near the original point increase with the same rate as that for lower ones.

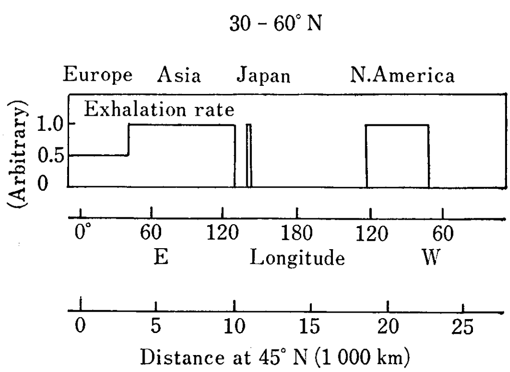

Fig. 6 The model of global exhalation rates of Rn used for the calculations.

the ocean and the existence of a peak at this altitude in K profile. This might also be affected materially by the time interval of 1 hour, since the variation in K is too large there.

We will be able to obtain the global Rn profiles by integrating point source data (Fig. 5) over the exhalation rate distribution on the lands. We assume a simple relative distribution of exhalation rate at middle latitude as is seen in Fig. 6. The rate in Europe is assumed to be half the rate in other places, since around one second in area is occupied by the Mediterranean Sea, the Bay of Biscay, the North Sea, the Baltic Sea and the Black Sea at latitudes between 30° and 60° in Europe.

Actually, the K profile was determined as follows. First, for a given trial K profile, the calculation is carried out for E=1 by the method described above. Next, the calculated result is compared with the observed altitude distribution of Rn (Fig. 8) compiled by Liu et al.[1]. If the shape of them is quite different from each other, the procedure just mentioned is repeated. If the shape of calculated altitude distribution is judged to be acceptable, the exhalation rate is evaluated by fitting that to the observed curve. The exhalation rates were found through the above procedure to be 1.0 and 1.5 atoms/cm²·s for winter and summer seasons, respectively. Liu, et al. have already estimated the K profiles based on the observed data. However, as they neglected the transport of Rn above the ocean to the continent at that time, the estimated values are somewhat different from our profiles as shown in Fig. 4.

4. Results and Comparison with Observations

The global distribution of Rn at middle latitude calculated by the present method are shown in Fig. 7 as a parameter of altitude.

Figure 8 gives the altitude distribution of Rn. The observed data were taken from the work of Liu et al.[1]. They have compiled 38 previously published data measured at the Eurasian and North American continents and illustrated the average values with one standard deviation. The Monte Carlo calculation was performed so that the observed data may coincide with the calculations for a cen-

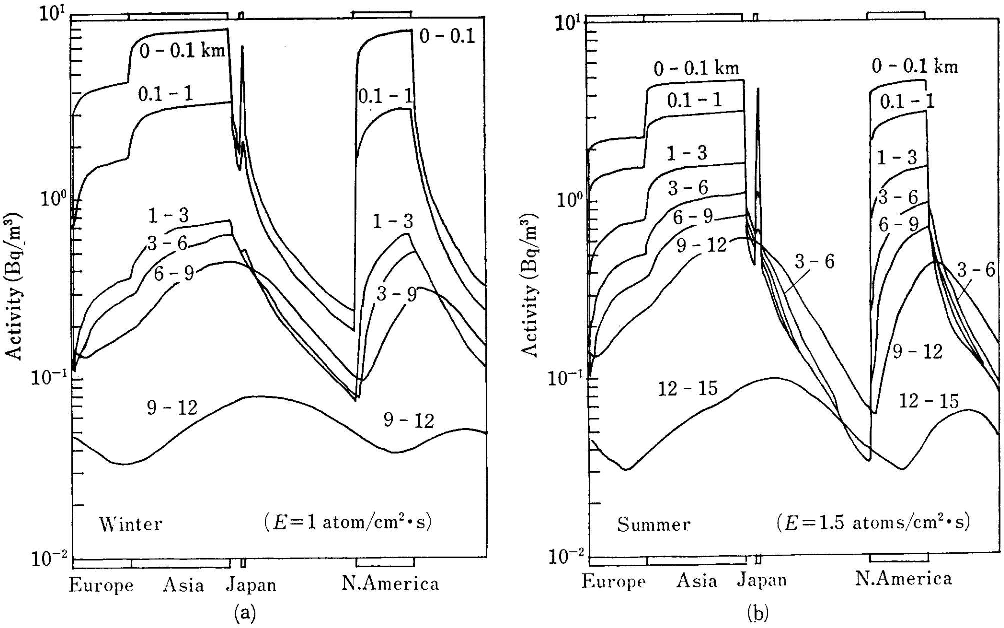

Fig. 7 Monte Carlo calculated distributions of Rn at various altitudes for (a) winter (December-February) and (b) summer (June-August).

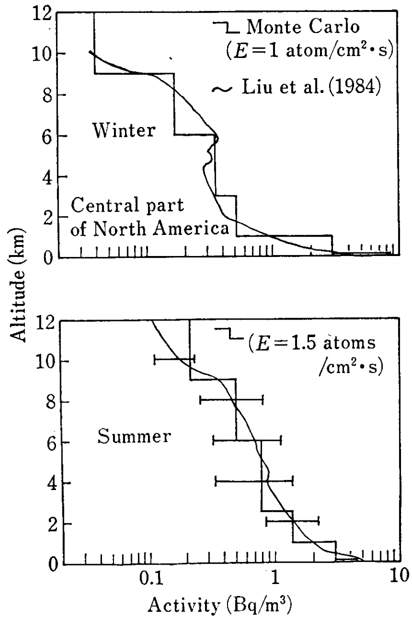

Fig. 8 Comparison of the Monte Carlo calculated Rn profiles with observed data compiled by Liu et al.

tral position of North America by trial and error method as was stated in the preceding section.

To assess the validity of the technique and results described here, we compare with several other observations.

The calculated Rn concentrations at heights between 0 and 100 m are compared to many observations[6)-22)] in Fig. 9. The calculated results agree, in general, with observed ones within a factor of 2 except for some measurements at eastern sites of North America in summer. If we need a much better agreement between them near the ground, a more detailed model may be required in exhalation rate and K profile there. Furthermore, if we adopt a shorter time interval for calculating small displacements, a more or less complete picture must be given.

Figure 10 shows a comparison with the average vertical profile obtained by Moore et al.[23)]. They took five sets of observed data offshore near San Francisco in every season. The mean of the Monte Carlo calculated results for winter and summer is presented in the figure. While the calculated values are to some extent small compared to the measurements, those coincide, as a whole, with each other within one standard deviation. Giving a more adequate wind speed profile might

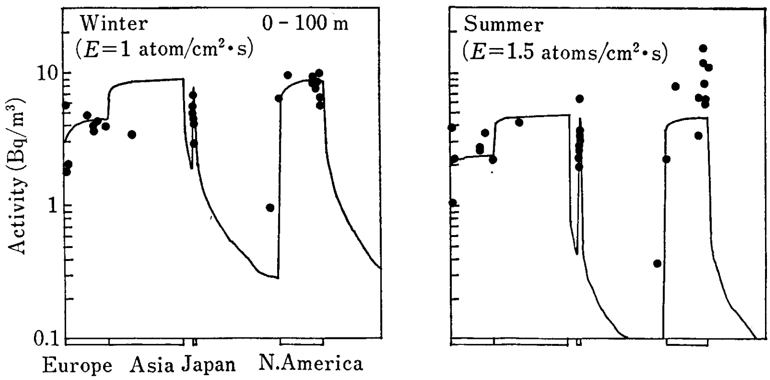

Fig. 9 Comparison of the Monte Carlo calculated concentrations with measured values averaged over the periods December-February and June-August.

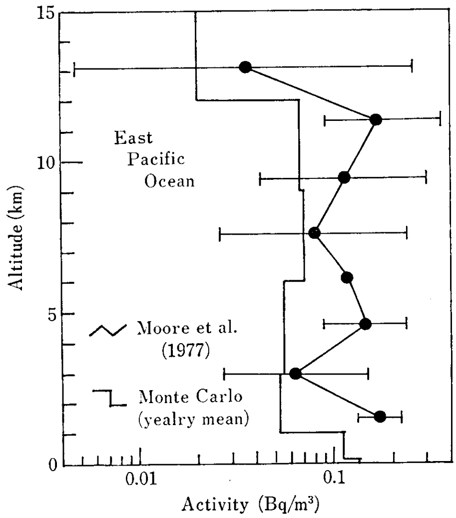

Fig. 10 Comparison of the Monte Carlo calculated concentrations with observed average one for Rn offshore near California.

result in a much better agreement, taking into account the horizontal diffusion effect from North America.

Figure 11 shows a comparison with measurements made on the sea[24] and at Nagoya[14] and islands in the Pacific Ocean[25),26]. An error bar of Nagoya data represents one standard deviation and the others maximum and minimum values. The horizontal axis in the figure gives the distance from the nearest neighbour land parallel to longitude. The trend that the Rn concentration decreases with distance from lands is well illustrated.

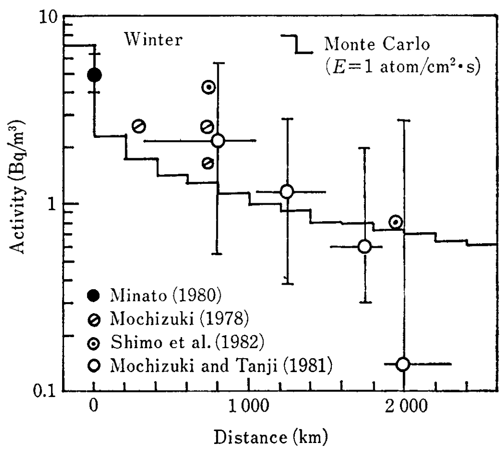

Fig. 11 Comparison of the Monte Carlo calculated results for the heights up to 100 m with the data taken on the sea.

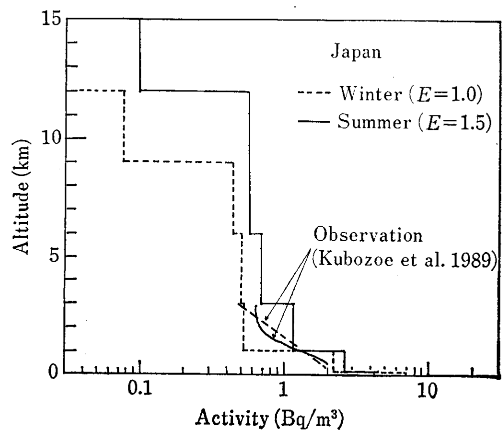

Fig. 12 Monte Carlo calculated and observed altitude distributions of Rn over Japan Islands.

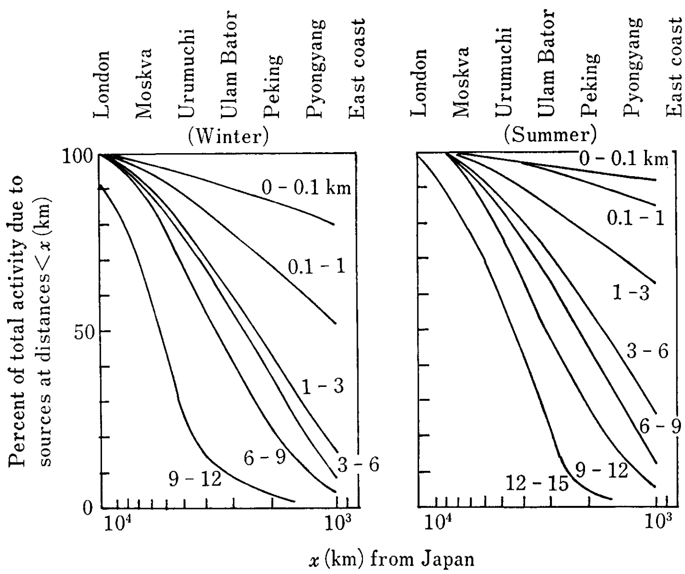

Fig. 13 Monte Carlo calculated relative contribution to the total Rn concentration in Japan from sources as a function of distance.

Vertical distributions of Rn for Japan Islands is shown in Fig. 12 along with the two sets of averaged values of 7 data for two seasons observed by Kubozoe et al.[27]. The agreement between them is fairly well.

We have derived average exhalation rates at middle latitude to be 1.0 and 1.5 atoms/$cm^2 \cdot s$ for winter and summer, respectively. Wilkening et al.[28] have obtained a world-wide mean value to be 0.75 atom/$cm^2 \cdot s$ from 994 measurements. Turekian et al. have found it from a one-layer model to be 1.2 atoms/$cm^2 \cdot s$. These values are not so different from ours. In particular, the value given by Turekian et al.[2] is almost equal to a mean of our values for two seasons. It may be all right to consider that our values are somewhat useful for speculating seasonal variations of Rn concentration.

Finally, we present the calculations of contribution of Rn exhaled from the Eurasian continent to Japan Islands as an application. Figure 13 shows integral Rn concentrations due to sources at various distances, which are normalized to the calculated profile given in Fig. 12. We find from the figure that Rn originated in the continent plays an important part in the concentration in Japan, especially for that in upper atmosphere.

5. Concluding Remarks

We have described how to evaluate global Rn concentrations using quite a simple and easy Monte Carlo technique and model in the present report. From the viewpoint of rough estimation, the results provided in this study is sufficiently acceptable.

The method described here would be applicable to several problems by modifying or extending it in some ways. We would, for instance, be able to calculate seasonal variations in Rn concentration for regional and/or local scale. Moreover, replacing the decay constant of Rn with the removal rate due to sink and washout, we would also be able to deal with global pollutants transport.

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要 旨

簡単な一次元モデルを用いる中緯度での ^{222}Rn 大循環のモンテカルロ計算

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北緯30－60°での対流圏における ^{222}Rn 大循環をシミュレートするための簡単なモンテカルロ計算技法を開発した。既刊の垂直分布の観測値に計算値をフィットさせることで平均的な渦拡散係数の垂直分布を導いた。同時に散逸率として夏（6－8月）に対して1.5，冬（12－2月）に対して1.0原子/cm^2･s を得た。

本方法の妥当性を調べるために他の研究者による多くの測定値と計算結果を比較した。

応用の一例としてユーラシア大陸起源の ^{222}Rn の日本列島への寄与を計算した。