



Estimation of ^{222}Rn Concentration in the Lower Troposphere during Precipitation Using Wet Scavenging Model for its Decay Products

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ABSTRACT

The gaseous ^{222}Rn concentration at the level of clouds was estimated by using the wet scavenging model of its decay products with the observed data of environmental radiation at the ground. And the origin of the ^{222}Rn was also discussed. The estimation was done for a precipitation event on Dec. 26-27, 2003, when a large increase of the radiation was observed in Tokai-mura in Ibaraki, Japan. From a backward trajectory analysis, the origin of ^{222}Rn atoms for that event was traced back to the north-eastern part of China, and it was expected that the large amount of ^{222}Rn emitted in the northeastern part of China was transported to Tokai-mura by the Eurasian continental air mass.

Key words: ^{222}Rn , Atmosphere, Precipitation, Decay products, Scavenging model, Back trajectory analysis

1. INTRODUCTION

Radon (^{222}Rn), which has a half-life of 3.82 days, is the gaseous radionuclide that may diffuse into the atmosphere after being produced by the decay of ^{226}Ra in soil. The ^{222}Rn in the atmosphere disperses with advection and diffusion, and decays to radionuclides including ^{218}Po (half-life: 3.10 min), ^{214}Pb (half-life: 26.8 min), ^{214}Bi (half-life: 19.9 min) and etc., according to the decay scheme. The ^{222}Rn and its decay products are useful tracers for investigating the dynamics of harmful substances and aerosols in the atmosphere, since their half-life is in the same time scale as that of the residence time of the atmospheric substances.

The ^{222}Rn decay products in the atmosphere are transported to the ground by the scavenging process

of precipitation, which causes an increase in environmental gamma radiation at the ground surface. Ikebe *et al.* (1985) and Takeyasu *et al.* (2006) proposed scavenging models of the decay products in the atmosphere by precipitation, and these models have been validated by comparing with the observed ^{214}Pb and ^{214}Bi concentrations in rainwater and their dependency on the precipitation. In these models, the gaseous ^{222}Rn concentration at the level of clouds is one of the parameters for determining the concentrations of ^{214}Pb and ^{214}Bi in rainwater. Few studies, however, have investigated the gaseous ^{222}Rn concentration at the level of clouds.

In this study, an attempt was done to estimate the gaseous ^{222}Rn concentration at the level of clouds by the wet scavenging model of the decay products proposed by Takeyasu *et al.* (2006) with the observed data of the increase of the environmental gamma radiation. This paper also discussed the origin of the ^{222}Rn when a large increase of the radiation at the ground was observed.

2. MATERIALS AND METHODS

2.1 Wet Scavenging Model of the Atmospheric ^{222}Rn Decay Products

In general, the two scavenging mechanisms of radionuclides in the atmosphere by precipitation, namely, rainout and washout, are important (Engelmann, 1968). It is known that ^{222}Rn decay products contained in rainwater are mainly removed by rainout (Minato, 1983; Takeuchi and Katase, 1982). In this study, a rainout model was designed, the outline of which was shown in Fig. 1. The conditions of rain clouds were set as follows:

- Gaseous ^{222}Rn is distributed uniformly in the atmosphere at the height of rain clouds.

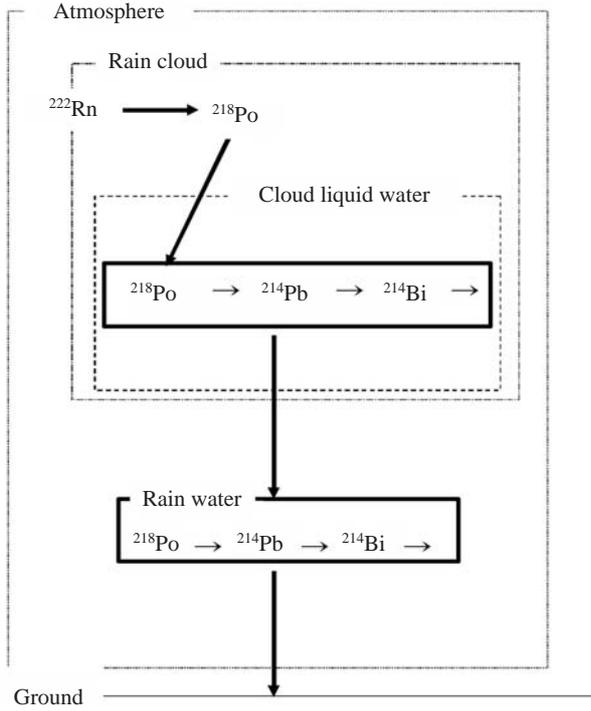


Fig. 1. Outline of the proposed scavenging model by precipitation for ^{222}Rn decay products in the atmosphere.

- (b) Gaseous ^{222}Rn in the atmosphere decays to ^{218}Po , ^{214}Pb , ^{214}Bi , etc., according to the decay scheme.
- (c) The rain cloud is a homogeneous liquid water column.
- (d) A radioactive equilibrium is established between the ^{222}Rn decay products contained in the cloud liquid water.
- (e) All the cloud liquid water removed from rain clouds is transported to the ground as rainwater.
- (f) The transport time of the liquid water from the cloud base to the ground was set in relation to the precipitation.

Under these conditions, the concentrations of ^{222}Rn decay products in the cloud liquid water or the rainwater are given by the following.

2.1.1 In the Rain Clouds

The concentrations of ^{222}Rn decay products absorbed into the cloud liquid water vary with time according to the following equations:

$$\frac{dN_{Po}^W}{dT} = \Lambda N_{Po}^a - (\lambda_{Po} + \Psi) N_{Po}^W \quad (1)$$

$$\frac{dN_{Pb}^W}{dT} = \lambda_{Po} N_{Po}^W - (\lambda_{Pb} + \Psi) N_{Pb}^W \quad (2)$$

$$\frac{dN_{Bi}^W}{dT} = \lambda_{Pb} N_{Pb}^W - (\lambda_{Bi} + \Psi) N_{Bi}^W \quad (3)$$

where N_{Po}^W , N_{Pb}^W and N_{Bi}^W are the number of atoms of ^{218}Po , ^{214}Pb and ^{214}Bi , respectively, in the cloud liquid water (m^{-3} of the cloud air), Λ the transport coefficient of ^{218}Po between the air and the liquid water in the cloud (s^{-1}), N_{Po}^a the number of atoms of atmospheric ^{218}Po (m^{-3}), λ_i the decay constant of the ^{222}Rn decay product i (s^{-1}), and Ψ the removal rate of the cloud liquid water from rain clouds (s^{-1}). Wilkening (1990) showed that ^{218}Po that was produced by the decay of ^{222}Rn in the atmosphere attached to cloud particles rather than to atmospheric aerosols. Therefore,

$$\Lambda N_{Po}^a = \lambda_{Rn} N_{Rn}^a \quad (4)$$

where λ_{Rn} is the decay constant of ^{222}Rn (s^{-1}), N_{Rn}^a the number of atoms of gaseous ^{222}Rn (m^{-3}), which is constant according to the condition (a). The removal rate, Ψ , of the liquid water from various types of clouds was found to correlate well with the precipitation, according to the radar observation carried out by Takeda and Natsuki (1982).

According to the assumption (d), the Eqs. (1)-(3) are solved under the equilibrium condition, that is, $(dN_{Po}^W/dT) = (dN_{Pb}^W/dT) = (dN_{Bi}^W/dT) = 0$, and the number of atoms of ^{222}Rn decay products in the cloud liquid water, $N_i^W(\infty)$ ($i = \text{Po}, \text{Pb}, \text{Bi}$) (m^{-3} of the cloud air), is obtained.

2.1.2 Below the Rain Clouds

During the period when the liquid water that was removed from rain clouds is being transported from the cloud base to the ground, the concentrations of ^{222}Rn decay products in the falling liquid water are represented by the following equations:

$$\frac{dN_{Po}^r}{dt} = -\lambda_{Po} N_{Po}^r \quad (5)$$

$$\frac{dN_{Pb}^r}{dt} = \lambda_{Po} N_{Po}^r - \lambda_{Pb} N_{Pb}^r \quad (6)$$

$$\frac{dN_{Bi}^r}{dt} = \lambda_{Pb} N_{Pb}^r - \lambda_{Bi} N_{Bi}^r \quad (7)$$

where N_i^r is the number of atoms of the ^{222}Rn decay product i in the falling liquid water (m^{-3} in the air), and $N_i^r(0) = N_i^W(\infty)$. According to assumption (e), the falling liquid water that was removed from rain clouds becomes rainwater, and the number of atoms of the ^{222}Rn decay product i in the falling liquid water is converted to that in the rainwater, n_i^r (cm^{-3} in the rainwater), using the following equation:

$$\lambda_i n_i^r = \frac{\lambda_i N_i^r}{M} \quad (8)$$

where M is the liquid water content of rain clouds ($\text{cm}^3 \text{m}^{-3}$ (in the cloud air)), which was set to be the mean

value ($0.2 \text{ cm}^3 \text{ m}^{-3}$ (in the cloud air)) (Mizuno *et al.*, 2003).

The time, t (s), is the transport time of the falling liquid water from the cloud base to the ground, and is calculated with the cloud base height and the speed at which the liquid water falls. The cloud base height was set at 3,000 m, which was adopted as the mean height of stratiform clouds by Ikebe *et al.* (1985). Mason (1971) gave the relationship between the mean raindrop diameter and the precipitation and that between the raindrop diameter and the terminal speed of the raindrop. From the cloud base height and Mason's relationships, the falling time of the liquid water can be represented as a function of the precipitation. When the precipitation is 1 mm h^{-1} , the falling time is about 20 min, and when the precipitation is 10 mm h^{-1} , the falling time is about 10 min.

Then, the concentrations of the decay products in rainwater that reaches to the ground surface become functional to the gaseous ^{222}Rn concentration at the level of clouds and the precipitation. This relationship was validated by the comparison with the observation results for the precipitation events in Osaka, Japan in 1992 (Takeyasu *et al.*, 2006).

2.2 Estimation of the Gaseous ^{222}Rn Concentration at the Level of Clouds from the Data of Environmental Radiation during Precipitation

On the basis of the model described in the previous section, the gaseous ^{222}Rn concentration at the level of clouds was estimated from the observed data of the increase of environmental radiation dose rate. In this method, the increase pattern of the dose rate calculated by the model was adjusted to agree with the observed one, and the atmospheric ^{222}Rn concentration, which is one of the model's parameters, was estimated.

First, the gaseous ^{222}Rn concentration at the level of clouds was assumed to be 1 Bq m^{-3} , and the concentrations of its decay products in rainwater were calculated from the precipitation data observed continuously at the Nuclear Fuel Cycle Engineering Laboratories (Cycle Laboratory), JAEA, located in Tokai-mura in Ibaraki, Japan.

The ground deposition amounts of the decay products i ($i=^{214}\text{Pb}$, ^{214}Bi) by the rainfall during the time from t_j-5 to t_j+5 (min), D_{ji} ($\text{atom cm}^{-2} \text{ s}^{-1}$) were obtained by multiplying the concentrations of the decay products in rainwater by rainfall, shown in the equation of (9). The value of j corresponds to the period of rainfall, and t_j the start time of rainfall. For example, if the rainfall period is 60 min, the value of j is from 1 to 6.

$$D_{ji} = \frac{2.8 \times 10^{-5} A_{ji} P_j}{\lambda_i} \quad (9)$$

where A_{ji} is the concentrations of the decay products i in rainwater (Bq cm^{-3}) which fell between the period of t_j-5 to t_j+5 (min), P_j the rainfall (mm h^{-1}) and λ_i the decay constant of the decay products i (s^{-1}).

The decay products that deposited to the ground were assumed to be not lost by surface runoff. The amount of the decay products i on the ground (N_{ji} ; atom cm^{-2}) which fell by rainfall from the time of t_j-5 to t_j+5 (min) changed according to the following equations;

(1) for the time of t ($t_j-5 < t < t_j+5$)

$$\frac{dN_{jPb}}{dt} = D_{jPb} - \lambda_{Pb} N_{jPb} \quad (10)$$

$$\frac{dN_{jBi}}{dt} = D_{jBi} + \lambda_{Pb} N_{jPb} - \lambda_{Bi} N_{jBi} \quad (11)$$

(2) for the time of t ($t_j+5 < t$)

$$\frac{dN_{jPb}}{dt} = -\lambda_{Pb} N_{jPb} \quad (12)$$

$$\frac{dN_{jBi}}{dt} = \lambda_{Pb} N_{jPb} - \lambda_{Bi} N_{jBi} \quad (13)$$

The amount of the decay product i (S_{ji} ; atom cm^{-2}) at the time of t ($t_j+5 < t$) (min) resulting from the deposition amount by the rainfall until the time of t_j+5 (min) is given by the following equation;

$$S_{ji} = \sum_{k=1}^j N_{ki} \quad (14)$$

The conversion from the amounts of ^{214}Pb and ^{214}Bi on the ground to the dose rate was done using the conversion factor calculated by Minato (Minato, 1980). Minato (1980) calculated the conversion factor from the amount of ^{214}Pb and ^{214}Bi on the ground to the dose rate at the height of 1m above the ground, assuming that ^{214}Pb and ^{214}Bi on the ground formed semi-infinite plane sources. Finally, the ^{222}Rn concentration as a parameter in the model was adjusted for the calculated increase of the dose rate to agree with the observed one at a monitoring post installed in flat ground inside the Cycle Laboratory.

The estimation of the ^{222}Rn concentration was done for a precipitation event on Dec. 26-27, 2003 when a large increase of the dose rate was observed and for another event on Nov. 20, 2003 when no such increase was observed.

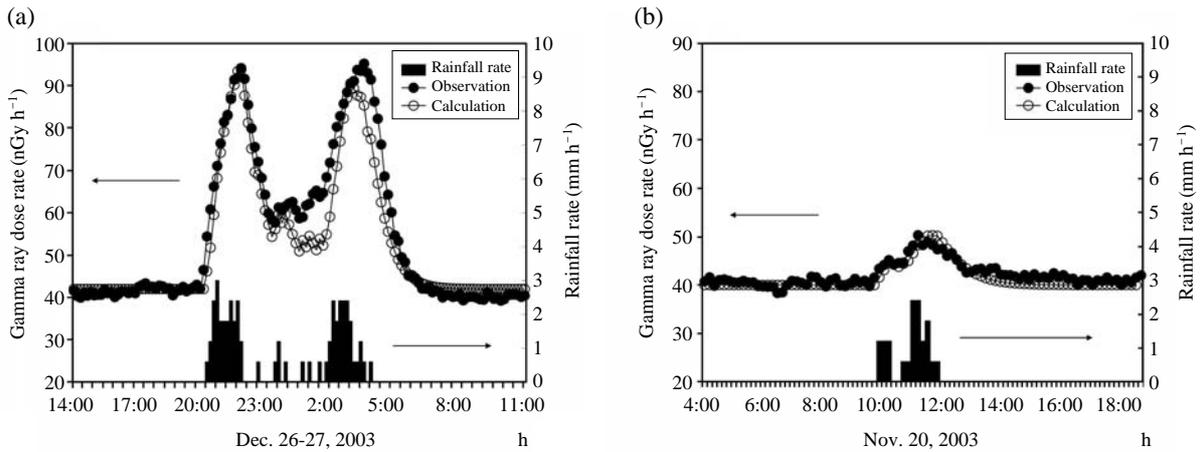


Fig. 2. Results of the adjustment of the increase pattern of the dose rate between the calculation and the observation on (a) Dec. 26-27 and (b) Nov. 20, 2003.

3. RESULTS AND DISCUSSION

Fig. 2 shows the results of the adjustment of the calculated increase of the dose rate to the observed one for the precipitation events on Nov. 20 and Dec. 26-27, 2003. From Fig. 2, the calculated pattern of the increase agreed well with the observed one, and the estimated gaseous ^{222}Rn concentration at the level of clouds for the event on Nov. 20 and on Dec. 26-27 was 1.5 Bq m^{-3} and 6 Bq m^{-3} , respectively. In the model used in this study, the decay products in the atmosphere deposited to the ground by rainout phenomenon, where the cloud base height was set to 3,000 m. If the decay products deposit by washout phenomenon under cloud, it is anticipated that their concentration ratio at the height of the ground changes. However, the calculated ratio of ^{214}Bi to ^{214}Pb by the model agreed relatively well with the observed one (Takeyasu *et al.*, 2006). From this fact, the time from the attachment of the decay products to rainwater to the arrival of rainwater on the ground in this model seem to be correct. However, cloud water content, M , is the mean value in this model. The concentrations of the decay products in rainwater are in inverse proportion to the cloud water content, M . It is anticipated that the cloud water content differs for every rain cloud, and it becomes an error factor for estimating the concentration.

Iida *et al.* (2003) measured vertical profiles of ^{222}Rn concentrations up to 5,000 m in the atmosphere over central Japan during 1999 to 2000 by aircraft. The concentration was $1\text{--}2 \text{ Bq m}^{-3}$ up to 5,000 m over Toyama bay on 23 March 1999. The concentration was about 1 Bq m^{-3} at the height of 3,000 m over Enshunada on 26 April 1999. These concentrations

were the same order as the estimated one at the level of clouds on Nov. 20, 2003.

Fig. 3(a) shows the surface weather chart on 9:00 in Dec. 27, 2003 (JST). From Fig. 3(a), there was an atmospheric depression in the Pacific Ocean, east of the Japanese islands, and the typical winter atmospheric pressure pattern prevailed. A northwest wind was predominant at the meteorological tower of the Cycle Laboratory on Dec. 26-27. Fig. 3(b) shows the chart on 9:00 in Nov. 20, 2003 (JST). From Fig. 3(b), an atmospheric depression located west of the Japanese islands, and a southerly wind was dominant at the meteorological tower. On March 23 and April 26, 1999 when Iida *et al.* (2003) measured the vertical profile of atmospheric ^{222}Rn concentration, the surface weather chart was more similar to that on Nov. 20, 2003. From these facts, it was concluded that the atmospheric condition over the Japanese islands on Dec. 26-27, 2003 was affected by the Eurasian continental air mass, and that the atmosphere on Nov. 20, 2003 and March 23 and April 26, 1999 were not affected by such air mass.

For the precipitation events in Nov. 20 and Dec. 26-27, 2003, the backward trajectory analysis was carried out from the lower troposphere at a height of rain clouds (3,000 m) over Tokai-mura in Ibaraki, Japan as the starting point. The analysis was performed using METEX online system which is offered on the web site of the Center for Global Environmental Research (CGER) in National Institute for Environmental Studies of Japan with meteorological data of the National Centers for Environmental Prediction (NCEP) of USA (National Institute for Environmental Studies of Japan; http://cgermetex.nies.go.jp/metex/index_jp.html). Fig. 4 depicts backward trajectories arrived at Tokai-mura

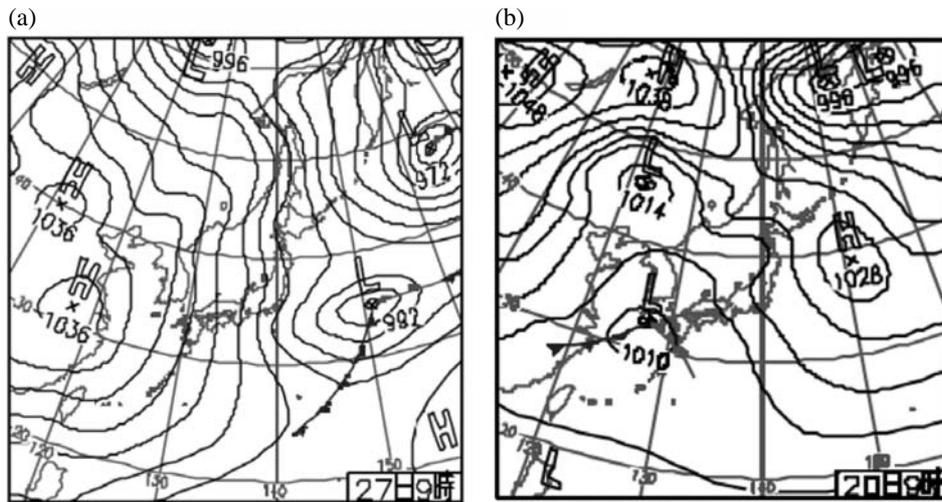


Fig. 3. Surface weather chart on (a) 9:00 in Dec. 27 and (b) 9:00 in Nov. 20, 2003 (JST).

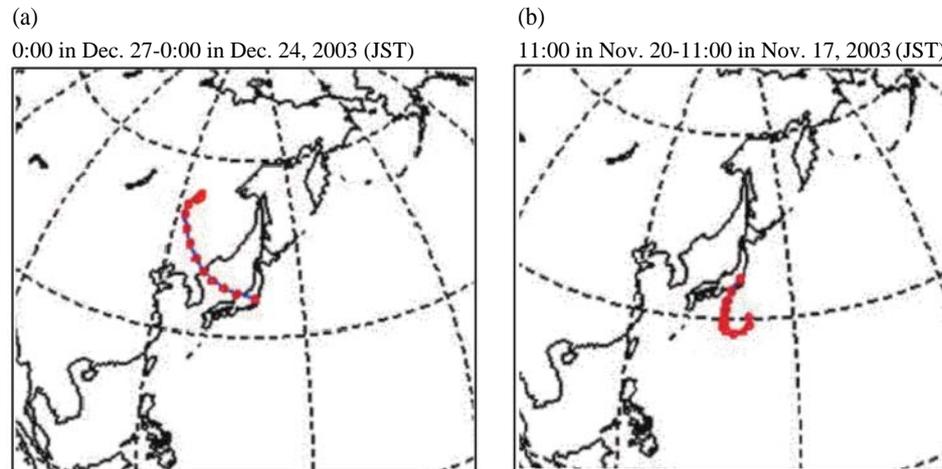


Fig. 4. Backward trajectories for the transport of the ^{222}Rn in the lower troposphere arrived at Tokai-mura on (a) Dec. 27 and (b) Nov. 20, 2003.

for the events. The air mass trajectories for the events on Nov. 20 and Dec. 26-27, 2003 traced back to the Pacific Ocean and the northeastern part of China, respectively.

The backward trajectory analysis was also carried out for March 23 and April 26, 1999 when Iida *et al.* (2003) measured the atmospheric ^{222}Rn concentration using airplane. As the result, the air mass above the Toyama Bay on March 23 came from northern China and the air mass above the Enshunada on April 26 from the Pacific Ocean.

Zhuo *et al.* (2005) measured the atmospheric ^{222}Rn concentration at the ground level in China, and showed that the annual average concentration was 17 Bq m^{-3} in the interior of northeast of China. Jin *et al.* (1985) discussed that the atmospheric ^{222}Rn concentration became high in winter season. They measured and discussed the concentration at the ground level, and

from their discussions, the concentration on the ground level in the winter in northeastern China may become over several tens of Bq m^{-3} . Jacobi and Andre (1963) calculated the vertical profile of ^{222}Rn concentration in the atmosphere, and showed that the ^{222}Rn concentration up to the height of more than 1,000 m from the ground became constant in case of the strong turbulence of the atmosphere. In such meteorological conditions, the atmospheric ^{222}Rn concentration in the lower troposphere in that region of China might become high and the large amount of ^{222}Rn was transported to Tokai-mura by the Eurasian continental air mass on Dec. 26-27, 2003. On the other hand, for March 23, 1999, the atmospheric ^{222}Rn concentration in northern China was not so high because of spring season, and it was suggested that the large amount of ^{222}Rn did not reach to Toyama Bay. In the future, further investigation will be performed in terms of the

transport mechanism of the ^{222}Rn from the continent to Japan and the wet scavenging mechanism of the decay products in the atmosphere.

4. CONCLUSIONS

The gaseous ^{222}Rn concentration at the level of clouds was estimated by using the wet scavenging model of ^{222}Rn decay products with the observed data of environmental radiation at the ground. The origin of the ^{222}Rn was also discussed when a large increase of the radiation at the ground was observed.

The atmospheric ^{222}Rn concentration for the precipitation event on Dec. 26-27, 2003, when a large increase of the radiation was observed in Tokai-mura in Ibaraki, Japan, was estimated to be 6 Bq m^{-3} . On the other hand, the ^{222}Rn concentration for the event on Nov. 20, 2003 when the large increase of the radiation was not observed was estimated to be 1.5 Bq m^{-3} .

The backward trajectory analysis was carried out from the lower troposphere in Tokai-mura as the starting point. From the analysis, the origin of ^{222}Rn atoms for the events on Dec. 26-27, 2003 was traced back to the northeastern part of China. It was expected that the large amount of ^{222}Rn emitted in the northeastern part of China was transported to Tokai-mura by the Eurasian continental air mass.

REFERENCES

- Engelmann, R.J. (1968) The calculation of precipitation scavenging. In *Meteorology and Atomic Energy* (Slade, D.H. Ed.), USAEC, Rep. TID-24190.
- Iida, T., Moriizumi, J., Yamada, K., Ishikawa, T., Nagao, T., Iwasaka, Y. (2003) Vertical profile of radon concentration in the atmosphere. In *Radioecology and Environmental Dosimetry* (Inaba, J., Tsukada, H. and Takeda, A. Eds.), Institute for Environmental Sciences, Japan, pp. 409-415.
- Ikebe, Y., Saijo, K., Iida, T., Nagai, H., Takeda, T. (1985) A model for precipitation scavenging of short-lived ^{222}Rn daughters. *Research Letters of Atmospheric Electricity* 5, 17-21.
- Jacobi, W., Andre, K. (1963) The vertical distribution of Radon-222, Radon-220 and their decay products in the atmosphere. *Journal of Geophysical Research* 68, 3799.
- Jin, Y., Yang, X., Ikebe, Y., Iida, T., Shimo, M., Yamaniishi, H., Guo, Q., Abe, S., Wang, Z., Ren, T., Tian, D., He, Z., Fan, X., Xie, H., Li, S., Lu, S., Zhang, H., Du, K. (1985) A survey of outdoor and indoor ^{222}Rn concentrations by passive method in China. In *Atmospheric radon families and environmental radioactivity III* (Okabe, S. *et al.* Eds.), Japan Atomic Energy Society of Japan, pp. 72-73. (in Japanese)
- Mason, B.J. (1971) *The Physics of Clouds*. Oxford University Press, London.
- Minato, S. (1980) Monte Carlo calculation of gamma radiation field due to precipitation washout of radon daughters from the atmosphere to the ground surface. *Japanese Journal of Health Physics* 15, 19-24.
- Minato, S. (1983) Estimate of radon-222 concentrations in rainclouds from radioactivity of rainwater observed at ground level. *Journal of Radioanalytical Chemistry* 78, 199-207.
- Mizuno, H. (2003) *Kumo to Ame no Kisyogaku*, Asakura Press, Tokyo.
- Takeda, T., Natsuki, S. (1982) Estimation of liquid matter amount in an extended cloud by Nimbus-5 microwaves data. *Journal of Meteorological Society of Japan* 60, 1153-1164.
- Takeuchi, N., Katase, A. (1982) Rainout-washout model for variation of environmental gamma-ray intensity by precipitation. *Journal of Nuclear Science and Technology* 19, 393-409.
- Takeyasu, M., Iida, T., Tsujimoto, T., Yamasaki, K., Ogawa, Y. (2006) Concentrations and their ratio of ^{222}Rn decay products in rainwater measured by gamma-ray spectrometry using a low-background Ge detector. *Journal of Environmental Radioactivity* 88, 74-89.
- Wilkening, M. (1990) *Radon in the Environment*. Elsevier Science Publishers, Amsterdam.
- Zhuo, W., Furukawa, M., Guo, Q., Kim, Y.S. (2005) Soil radon flux and outdoor radon concentration in East Asia. *International Congress Series*, Elsevier Pub., 1276, Amsterdam, 285-286.

(Received 10 October 2009, accepted 13 March 2010)