Estimation of the Global ^{222}Rn Flux Density from the Earth's Surface

Shigekazu HIRAO,*¹ Hiromi YAMAZAWA*¹ and Jun MORIIZUMI*¹

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Global ^{222}Rn flux density distributions with a spatial resolution of 1° in latitude and longitude were estimated for each month in the period of 1979–2007 using an ^{222}Rn exhalation model based on a porous media diffusion theory. This exhalation model calculated the ^{222}Rn flux density as a function of the ^{226}Ra content, soil water saturation, and soil temperature. The influence of the topographical ruggedness of the ground surface on the flux density was also considered in the model. The global average of the flux density during the 29-year period was estimated to be $21.3 \text{ mBq m}^{-2} \text{ s}^{-1}$. The annual variation in the globally averaged flux density showed a slight but steady increase since 1999. The seasonal variation in the globally averaged flux density was estimated to range from a minimum of $18.8 \text{ mBq m}^{-2} \text{ s}^{-1}$ in March to a maximum of $23.6 \text{ mBq m}^{-2} \text{ s}^{-1}$ in September. The regional averages for the annual flux density varied from $8.3 \text{ mBq m}^{-2} \text{ s}^{-1}$ for the northern part of North America to $33.5 \text{ mBq m}^{-2} \text{ s}^{-1}$ for Australia. The latitudinal distribution of zonally averaged flux density was found to be in good agreement with the latitudinal decline at high latitudes in the Northern Hemisphere. The spatial and temporal variations were caused mainly by variations in the soil water content and soil ^{226}Ra content, but to a lesser extent by the soil temperature. The topographical ruggedness was found to increase the flux density by only 2% globally but by more than 10% in mountainous regions. Atmospheric transport simulations using the estimated flux density as input data showed that the simulated atmospheric ^{222}Rn concentration was substantially lower than that measured at Hachijo Island, implying that the flux density of the present study with topographical ruggedness is still underestimated in the East Asian region.

KEY WORDS: radon exhalation, flux density, soil water saturation, topographical ruggedness, validation, atmospheric transport model.

1 INTRODUCTION

Radon-222 has been widely studied with two purposes: 1) to evaluate the public exposure to ^{222}Rn and its decay products and 2) to be used as a tracer for atmospheric transport. It is commonly known that the inhalation of ^{222}Rn and its short-lived decay products accounts on average for about one-half of the effective dose from all natural sources of ionizing radiation, and the induction of lung cancer by the inhalation of the decay products is one of the most important problems for public radiation protection.¹⁾ Therefore, information on the regional and continental distribution of the ^{222}Rn flux density from the earth's surface is considered to be useful for identifying areas with a risk of high ^{222}Rn exposure to the public.

On the other hand, ^{222}Rn has been observed to be a tracer for atmospheric transport because it is an inert gas and has a half-life of 3.82 d, which is desirable for such studies.^{2–6)} Observations of the concentrations of ^{222}Rn and its decay products in the atmosphere are very useful in evaluating chemical transport models, which simulate horizontal and

vertical atmospheric transport, along with the removal processes of aerosols.^{7–10)} For this purpose, a uniform ^{222}Rn flux density of $21.0 \text{ mBq m}^{-2} \text{ s}^{-1}$ ($1 \text{ atom cm}^{-2} \text{ s}^{-1}$) from ice-free land surfaces has commonly been used. The ^{222}Rn flux density is often assumed to be constant with space and time because of the lack of sufficient observations across continents. However, it is well known that the flux density varies in space and time depending on the ^{226}Ra content and soil conditions. Current high-resolution atmospheric transport models also require a more accurate ^{222}Rn source term for more quantitative validation of the models. Therefore, a more reliable flux map covering the earth's surface is desirable for the effective use of ^{222}Rn as a tracer.¹¹⁾

Previous studies have provided useful information for estimating a ^{222}Rn flux map.^{12–14)} SCHERY and WASIOLEK¹²⁾ proposed a global ^{222}Rn flux map based on a porous media transport theory and calibrated it with experimental ^{222}Rn flux density data from Australia and Hawaii. The map proposed by them has a large uncertainty because of the lack of sufficient global data on soil conditions such as the ^{226}Ra content and soil moisture; however, HIRAO et al.¹⁵⁾ provided evidence that the flux map could be used to improve the performance of an atmospheric transport model for ^{222}Rn and ^{210}Pb , as compared with the conventional source term of $21.0 \text{ mBq m}^{-2} \text{ s}^{-1}$ ($1 \text{ atom cm}^{-2} \text{ s}^{-1}$).

A different estimation of the ^{222}Rn source term was

*¹ Department of Energy Engineering and Science, Graduate School of Engineering, Nagoya University; Furo-cho, Chikusa-ku, Nagoya 464–8603, Japan.
Corresponding author, E-mail address: s-hirao@ees.nagoya-u.ac.jp

proposed by CONEN and ROBERTSON,¹³⁾ based on atmospheric profile measurements by integrating over larger areas. This indicated a latitudinal decline in the ^{222}Rn flux density from an ice-free land surface from $21.0 \text{ mBq m}^{-2} \text{ s}^{-1}$ ($1 \text{ atom cm}^{-2} \text{ s}^{-1}$) at 30°N to $4.2 \text{ mBq m}^{-2} \text{ s}^{-1}$ ($0.2 \text{ atom cm}^{-2} \text{ s}^{-1}$) at 70°N . This source term was found to improve the simulation results of the atmospheric ^{222}Rn transport model but it was speculated that the ^{222}Rn flux density might begin to decline from a latitude between 30°N and 50°N .¹⁶⁾

More recently, SZEGVARY et al.¹⁴⁾ developed a new method based on an empirical correlation between the ^{222}Rn flux density and the terrestrial gamma dose rate and calibrated it in Switzerland, Germany, Finland, and Hungary to create a ^{222}Rn flux map for Europe. The basic concepts of their method involved the two assumptions of a constant contribution of the ^{238}U series to the total gamma dose rate and a proportionality of the terrestrial gamma dose rate to the ^{222}Rn flux density. It was also pointed out that a consideration of additional physical soil parameters might further improve the prediction of the fluxes on the small scale.

Although physical soil parameters are considered to be important for estimating a more detailed ^{222}Rn flux map, sufficient global data on physical soil parameters are unavailable, such as the global distributions of ^{226}Ra content, soil water content, and soil temperature. In recent year, global meteorological reanalysis datasets have been produced with data assimilation systems and worldwide observational data inclusive satellite data. Of these global meteorological reanalysis datasets, the 25-year reanalysis data (JRA-25) provided by the Japan Meteorological Agency (JMA) and the Central Research Institute of Electric Power Industry (CRIEPI) have a high quality.¹⁷⁾ The purpose of the present study is to estimate the global ^{222}Rn flux density distribution with latitudinal and longitudinal resolutions of 1° for each month from 1979 to 2007. The ^{222}Rn flux density is calculated with a ^{222}Rn exhalation model based on a porous media transport theory.¹⁸⁾ The global physical parameters of the water saturation and temperature of the soil from JRA-25 are used to investigate the temporal and spatial variations in the flux density. In addition, the influence of the topographical ruggedness at the earth's ground surface on the flux density is also considered. The uncertainties in the estimated flux density distribution are discussed from the viewpoint of the exhalation model and the input physical parameters. We test the estimated flux density distribution by comparing the concentration measured at a solitary island in Japan to atmospheric ^{222}Rn concentrations simulated by inputting the estimated flux map into the atmospheric ^{222}Rn transport model.

II MODELS OF ^{222}Rn EXHALATION AND ATMOSPHERIC TRANSPORT

2.1 Radon-222 exhalation model

The exhalation of ^{222}Rn from the earth's surface involves two processes, emanation and transport. Emanation is the process whereby a ^{222}Rn atom escapes from ^{226}Ra -bearing grains to the pore space in soil. The transport of ^{222}Rn in the pore space involves two processes, diffusion and advection.

Diffusion, which is the main process of ^{222}Rn transport, is caused by the ^{222}Rn concentration gradient. Advection is caused by a pressure difference between the air in the pore space and the ground surface.

In this study, only diffusion is considered because the influence of most meteorological parameters on the advection can be assumed to be negligible when it is averaged over one month. The ^{222}Rn exhalation model used in this study basically follows that proposed by a previous study.¹⁸⁾ Assuming a semi-infinite and homogeneous soil, the ^{222}Rn flux density from the earth's surface is derived by solving a one-dimensional diffusion equation.¹⁸⁾ The influence of the water content and temperature of the soil on the flux density is taken into account by expressing the emanation and diffusion processes as a function of soil water saturation and soil temperature. The flux density from a rugged ground surface is considered to be larger than that from a flat surface by a factor that is the ratio of the surface area of the rugged ground surface to that of the flat area projected onto a horizontal plane. The ^{222}Rn flux density F from the ground surface is described as:

$$F = mR\epsilon\rho\sqrt{\lambda D}, \quad (1)$$

where R is the ^{226}Ra content in the soil (Bq kg^{-1}), ϵ the ^{222}Rn emanation coefficient, ρ the dry soil bulk density (kg m^{-3}), λ the ^{222}Rn decay coefficient (s^{-1}), and the ruggedness factor. The effective diffusion coefficient D ($\text{m}^2 \text{ s}^{-1}$) is:

$$D = D_0 p \left(\frac{T}{273} \right)^{1.5} \exp(-6Sp - 6S^{14p}), \quad (2)$$

to consider an increase in the kinetic diffusion in pore air due to a higher temperature, and a decrease in diffusion due to water in the soil.¹⁹⁾ In Eq. (2), D_0 is the ^{222}Rn diffusion coefficient in open air, p the total porosity of the soil, T the soil temperature (K), and S the water saturation, which is the ratio of the volumetric water content to p . The emanation coefficient ϵ has a dependence on water saturation²⁰⁾ and on soil temperature,²¹⁾ as follows:

$$\epsilon = \epsilon_0 [1 + 1.73(1 - \exp(-19S))] \cdot [1 + 0.01(T - 298)], \quad (3)$$

where ϵ_0 is the ^{222}Rn emanation coefficient for dry soil at 25°C . The emanation coefficient ϵ is assumed to be constant between soil textures.

For snow covered soils, the decay of ^{222}Rn in a snow layer of depth z_s (m) is taken into consideration as:

$$F = F_0 \exp\left(-\sqrt{\frac{\lambda}{D_s}} z_s\right), \quad (4)$$

where F_0 is the ^{222}Rn flux density at the ground-snow interface. The effective diffusion coefficient in the snow layer D_s is calculated by substituting $S = 0$ into Eq. (2).

2.2 Influence of topographical ruggedness

Ruggedness and cracks in the earth's surface are important topographic features when considering ^{222}Rn flux density. The rate of the ^{222}Rn exhaled from the ground surface is proportional to the surface area, which varies according to topographic features. Therefore, the rate of ^{222}Rn exhalation from a rugged ground surface will be larger than that from

a flat surface. The diffusion length for the ^{222}Rn in soil is typically from a few tens of centimeters to a meter. In this study, the horizontal scale of ground surface ruggedness affecting the flux density is assumed to be 1 m. The ruggedness at a horizontal scale of 1 m is newly introduced to account for the effect of terrain features on the flux density, as designated by ruggedness factor m in Eq. (1). Since the horizontal resolution of the available elevation data is generally lower than 1 m, the relationship between m and the horizontal resolution of the elevation data was examined as follows.

We used four digital elevation model (DEM) data sets: 1) the 1 arc-second DEM derived by the Advanced Spaceborne Thermal Emission and Reflection Radiometer (ASTER), 2) the finished Shuttle Radar Topography Mission Data at 3 arc-seconds (SRTM3), 3) the finished Shuttle Radar Topography Mission Data at 30 arc-seconds (SRTM30), and 4) the 0.4 arc-second DEM provided by the Japan Geographical Survey Institute (GSI). Detailed descriptions of these DEMs have already been published for SRTM²²⁾ and for ASTER.²³⁾ The GSI DEMs represent the actual ground surface, whereas the ASTER and SRTM elevation data refer to the canopy top.

To calculate the ruggedness factor m as illustrated in **Fig. 1**, we calculated the area S_{rugg} of the tilted triangular plane covered by the DEM and the S_{flat} area of the horizontal base plane, and evaluated the ratio of the rugged surface area to the flat one, $m = \Sigma S_{\text{rugg}} / \Sigma S_{\text{flat}}$, as a ruggedness factor for each 1° grid tile. To calculate an actual length on the spherical earth's surface, Hubeny's law²⁴⁾ was used because equirectangular projection was used for the DEMs in this study. For simplicity in discussing the horizontal resolution of the elevation, we assumed that the DEM resolution δ was the grid size at an equatorial position, for example, 90 m for the SRTM3 DEM and 10 m for GSI DEM.

Figure 2 shows the calculated ruggedness factor for each DEM averaged over Japan and the entire globe. There are linear relationships between the logarithm of m and the logarithm of the inverse of the horizontal resolution of the DEM. In the Japan case, m increases with increasing $\log_{10} \delta^{-1}$ in the range of about -3 to -1 . Therefore, we assume that m is linearly related to $\log_{10} \delta^{-1}$. The ruggedness factor m at a horizontal resolution of 1 m can be estimated using an extrapolation of the linear relation, as shown in **Fig. 2**. Finally, the global distribution of extrapolated m at a resolution of 1 m is estimated for every 1° by 1° grid tile.

2.3 Input soil condition data

The ^{222}Rn exhalation model requires the ^{226}Ra contents and physical soil parameters. The data for the ^{226}Ra contents used in this study are the country-by-country soil ^{226}Ra content data (for details, see Table 5 in the UNSCEAR 2000 report²⁵⁾). For China, the distribution of the ^{226}Ra content is from the published data.²⁶⁾ The ^{226}Ra content in the Korean peninsula is the value assigned at the neighboring area geologically corresponding to the above-mentioned Chinese map. For other countries, a global median value of 35 Bq kg^{-1} is assumed.

The global data for physical soil parameters from 1979 to 2007 are obtained from the JRA-25/JCDAS reanalysis datasets, which are provided by JMA/CRIEPI and the JMA

Fig. 1 Schematic illustration of calculating the surface area from digital elevation model data (DEM) given at corners (solid circles). The variable S_{rugg} represents the area of the tilted triangular plane and S_{flat} is the area of the horizontal base plane.

Fig. 2 Dependence of the ruggedness factor on the horizontal resolution of the DEM δ in meters. Solid circles are m averaged over only tiles in Japan, while open circles are m averaged over all tiles in the globe.

Climate Data Assimilation System (JCDAS). Monthly averages of soil water saturation, soil temperature, and snow depth with a horizontal resolution of 1.125° are used. The soil water saturation and temperature values are averages over about 0.0–0.1 m and 0–1 m soil depth, respectively. These data are interpolated to a horizontal resolution of 1° in the exhalation model. The total porosity of the soil has the same horizontal resolution but is constant with time. The dry bulk density p , the ^{222}Rn diffusion coefficient in air D_0 , and the emanation power of dry soil ϵ_0 used in the exhalation model are $1,600 \text{ kg m}^{-3}$, $1.1 \times 10^{-5} \text{ m}^2 \text{ s}^{-1}$, and 0.1, respectively.

2.4 Atmospheric ^{222}Rn concentration

In this study, the measured ^{222}Rn concentration in the surface air at Hachijo Island^{5, 6)} is used to test the estimated ^{222}Rn flux density distribution. Hachijo Island is located in the Pacific Ocean about 200 km away from Japan's main

island. The local ^{222}Rn level on this island has been measured to be less than $1 \text{ mBq m}^{-2} \text{ s}^{-1}$, because its surface is covered by basalt scoria due to relatively recent volcanic activities. Therefore, the atmospheric concentration measured on this island can be considered to be mainly contributed from long-range transported ^{222}Rn from the continent and the main island of Japan. The data used in this study were measured with an electrostatic radon monitor, which had a minimum detection limit of 0.3 Bq m^{-3} for hourly averaged concentrations.²⁾ The measured concentration at Hachijo Island in 2006 averaged about 1.5 Bq m^{-3} , with a variation from the detection limit to values as high as about 5.0 Bq m^{-3} .

2.5 Atmospheric ^{222}Rn transport model

The atmospheric transport is simulated in this study using a three-dimensional atmospheric ^{222}Rn transport model to test the flux density distribution. This model consists of a meteorological model MM5,²⁷⁾ and a Eulerian transport model HIRAT¹⁵⁾ for ^{222}Rn and its decay products. These models are off-line connected through a data set of three-dimensional wind, turbulence, and precipitation. The models are described in detail by HIRAO et al.²⁸⁾ The simulation domain is $9,792 \text{ km (W-E)} \times 7,776 \text{ km (S-N)} \times 10 \text{ km}$, divided by a 72 km -square horizontal grid. The 6-hourly global analysis data from the Japan Meteorological Agency (GANAL) and the monthly SST data from the National Centers for Environmental Prediction (NCEP) are used for the meteorological data and sea surface temperature (SST) data, respectively.

III RESULTS

3.1 Topographical ruggedness on the earth's surface

The global distribution of the ruggedness factor at a horizontal scale of 1 m is shown in **Fig. 3**. The land area weighted global average for the ruggedness factor is 1.03. The maximum for the ruggedness factor is 1.51 in the Himalayan range. For East Asia, the average value is 1.09. Large values are obtained in high altitude regions, such as the Rocky

Mountains, Alaska Range, Andes Mountains, and Alps. The ruggedness factors for island countries such as Japan and New Zealand are high. However, the ruggedness factors are almost unity in most of the plane surface and in parts of high altitude regions. The topographical ruggedness increases the land area-weighted global average ^{222}Rn flux density by 2%. As listed in **Table 1**, the topographical ruggedness in East Asia results in a 9% increase in the ^{222}Rn flux density.

3.2 Spatial distribution of ^{222}Rn flux density

Figure 4(a) depicts the global distribution of the estimated ^{222}Rn flux density with the influence of the topographical ruggedness, as averaged over the period of 1979 to 2007. The global average of the flux density weighted by land area is $21.3 \text{ mBq m}^{-2} \text{ s}^{-1}$ during the 29-year period. Low fluxes are found in the high latitudes of the northern hemisphere, the equatorial band, Europe, the central part of China, the eastern and northern parts of North America, and the southern part of South America. The flux density in the Amazonian area is relatively large in the equatorial band. In contrast, the fluxes for Australia, Africa, the western part of the United States of America, and the northern as well as southern parts of China are high. The regional averages for the annual flux densities by region are summarized in **Table 1**. The regional average flux densities range between the lowest value of $8.3 \text{ mBq m}^{-2} \text{ s}^{-1}$ for the northern part of North America and the highest value of $33.5 \text{ mBq m}^{-2} \text{ s}^{-1}$ for Australia. This variation in the regional averages is larger than the annual and seasonal variations, as will be shown in the next subsection.

The latitudinal distribution of the zonally averaged ^{222}Rn flux density is shown in **Fig. 5(a)**. The latitudinal distribution has a minimum at the equator and two maxima at mid-latitudes, and the flux density decreases linearly with increasing latitude from the maxima. The zonal average flux density reaches a maximum in the northern hemisphere of $31.9 \text{ mBq m}^{-2} \text{ s}^{-1}$. In the southern hemisphere, the maximum zonal average flux density is $31.0 \text{ mBq m}^{-2} \text{ s}^{-1}$. This latitudinal

Table 1 Regional averages of ^{222}Rn flux density.

Region	^{222}Rn flux density ($\text{mBq m}^{-2} \text{ s}^{-1}$)		R/F
	Flat [†] (F)	Rugged ^{††} (R)	
Northern part of North America	8.0	8.3	1.03
Southern part of North America	19.5	20.1	1.03
South America	23.3	24.1	1.03
North Asia	9.0	9.2	1.02
East Asia [†]	17.3	18.5	1.07
Japan	10.0	10.8	1.09
Central Asia	24.7	25.4	1.03
South Asia	15.2	15.9	1.05
Europe	17.8	18.3	1.03
Africa	29.1	29.4	1.01
Australia	33.4	33.5	1.00
New Zealand	11.5	12.4	1.08
global average	20.8	21.3	1.02

[†] : except for Japan

^{††} : ^{222}Rn flux density calculated by assuming flat surfaces

^{†††} : ^{222}Rn flux density calculated by assuming rugged surfaces

Fig. 3 Global distribution of the ruggedness factor m at a horizontal scale of 1 m.

decline corresponds to the results of CONEN and ROBERTSON,¹³⁾ in which the flux linearly decreased from $21.0 \text{ mBq m}^{-2} \text{ s}^{-1}$ ($1 \text{ atom cm}^{-2} \text{ s}^{-1}$) at 30°N to $4.2 \text{ mBq m}^{-2} \text{ s}^{-1}$ ($0.2 \text{ atom cm}^{-2} \text{ s}^{-1}$) at 70°N . Although the latitudinal variations estimated in this study are similar to that of the ^{222}Rn flux map by SCHERY and WASIOLEK,¹²⁾ the absolute values of the flux density are substantially different by about $10 \text{ mBq m}^{-2} \text{ s}^{-1}$ in the tropical to subtropical zone and by about $20 \text{ mBq m}^{-2} \text{ s}^{-1}$ or more in the northern mid to high latitudinal zones. **Fig. 5(b)** shows the latitudinal distributions of zonally averaged soil water saturation and soil temperature. The latitudinal distribution of the water saturation has an inversely proportional relationship to that of the flux density. The soil temperature distribution has a maximum at the equator. The zonal averages of the water saturation at the two minima in the northern and southern hemispheres are 0.33 and 0.27, respectively. The latitudinal distribution of the flux density corresponds mainly to that of the water saturation and partially to the soil temperature and ^{226}Ra content.

3.3 Temporal variation of ^{222}Rn flux density

As shown by **Fig. 6(a)**, the globally averaged ^{222}Rn flux density shows annual variations around an average of $21.1 \text{ mBq m}^{-2} \text{ s}^{-1}$ from 1979 to 1999 and a slight increase from $21.1 \text{ mBq m}^{-2} \text{ s}^{-1}$ in 1999 to $22.1 \text{ mBq m}^{-2} \text{ s}^{-1}$ in 2007. There is a remarkable decrease in 1984. This decrease corresponds to the maximum annually averaged soil water saturation of 0.47. The seasonal variation in the flux density averaged over the period of 1979 to 2007 ranges from a minimum of $18.8 \text{ mBq m}^{-2} \text{ s}^{-1}$ in March to a maximum of $23.6 \text{ mBq m}^{-2} \text{ s}^{-1}$ in September (**Fig. 6(b)**), with a peak-to-peak amplitude of about $5 \text{ mBq m}^{-2} \text{ s}^{-1}$. This seasonal variation is larger than the annual variation, which indicates that each estimated annual average flux density has a large variation in the amplitude of the seasonal variation.

The spatial distribution also shows the seasonal variation in a more enhanced manner, as shown in **Figs. 4(b)** and **(c)**. For North Asia, the ^{222}Rn flux density averaged over the

period varies from a minimum of $3.6 \text{ mBq m}^{-2} \text{ s}^{-1}$ in April to a maximum of $14.2 \text{ mBq m}^{-2} \text{ s}^{-1}$ in September. The averaged soil water saturation and soil temperature in this region also vary from 0.80 and -9.1°C in April to 0.65 and 5.2°C in September, respectively. For East Asia, including the Korean peninsula and China, the flux density varies from a minimum of $16.1 \text{ mBq m}^{-2} \text{ s}^{-1}$ in April to a maximum of $20.6 \text{ mBq m}^{-2} \text{ s}^{-1}$ in November. The water saturation and soil temperature are 0.49 and 3.8°C in April, and 0.50 and 12.3°C in November, respectively. For South Asia, the flux density varies from a minimum of $13.2 \text{ mBq m}^{-2} \text{ s}^{-1}$ in October to a maximum of $20.7 \text{ mBq m}^{-2} \text{ s}^{-1}$ in February. The water saturation and soil temperature are 0.76 and 24.7°C in October, and 0.61 and 23.9°C in February, respectively. For North Asia, the amplitude of the seasonal variation in the regional average flux density is found to be $10.7 \text{ mBq m}^{-2} \text{ s}^{-1}$. These temporal variations in the regional average flux density correspond to those of the water content and temperature, as discussed in the next section.

3.4 Verification with atmospheric ^{222}Rn concentration

An example of the simulation results are shown in **Fig. 7**, in which the ^{222}Rn concentrations at Hachijo Island in February 2006 simulated by the atmospheric transport model are compared with the measured surface air ^{222}Rn concentration. Two simulations were conducted using the ^{222}Rn flux map estimated in this study and that of SCHERY and WASIOLEK¹²⁾ (hereafter, referred to as the S-W map). The concentrations shown in **Fig. 7** are of the lowest model layer with a 50 depth.

The measured ^{222}Rn concentration in February averages 1.8 Bq m^{-3} . The measured concentration shows several-day cycle variations caused by the passage of ^{222}Rn -bearing air masses originating in East Asia with low-pressure systems. The simulated concentrations in February average 0.8 Bq m^{-3} for the present map and 1.8 Bq m^{-3} for the S-W map. The two simulated concentrations show several-day variations that are similar to the measured concentration in shape but clearly different in the absolute concentration values. The differences between the two simulated concentrations and

Fig. 4 Global distribution of ^{222}Rn flux density averaged over the period of 1979 to 2007 with latitudinal and longitudinal resolutions of 1° , (a) annual average, (b) composite of distribution in April, and (c) in September.

the measured concentration vary with time. For example, the concentrations simulated from the present map from February 3 to 6 are underestimated, whereas those from the S-W map agree with the measured concentrations. From February 16 to 22, the simulated concentrations from the present map agree with the measured ones, whereas those from the S-W map are overestimated. For the entire year of 2006, the biases,

which indicate the under- or over-estimation of the simulated concentration, are -0.96 Bq m^{-3} for the present map and 0.12 Bq m^{-3} for the S-W map, respectively, indicating that the ^{222}Rn concentration simulated from the ^{222}Rn flux density of the present study is underestimated. However, the amplitude of the several-day cycle variations in the simulated concentration in the case with the present map shows better agreement with

Fig. 5 (a) Latitudinal distribution of zonal mean ^{222}Rn flux density averaged over the period of 1979 to 2007, compared to the global flux map by SCHERY and WASIOLEK (1998)¹²⁾ and the linearly decreasing flux map by CONEN and ROBERTSON (2002).¹³⁾ (b) Latitudinal distributions of zonal mean soil water saturation and soil temperature derived from JRA-25/JCDAS for the same period.

Fig. 7 Comparison of the surface air ^{222}Rn concentration values from simulations and measurements at Hachijo Island in February 2006. The simulations were conducted by using the ^{222}Rn flux map from this study and that of SCHERY and WASIOLEK (1998).¹²⁾

the measured concentration than that in the case with the S-W map.

IV DISCUSSIONS

The global average of the ^{222}Rn flux density distribution estimated in this study for the period of 1979 to 2007 is $21.3 \text{ mBq m}^{-2} \text{ s}^{-1}$. Although the estimated global average flux density is consistent with the conventional flux density of $21.0 \text{ mBq m}^{-2} \text{ s}^{-1}$ ($1 \text{ atom cm}^{-2} \text{ s}^{-1}$), the global flux density distributions have significant spatial and temporal variations. In this section, the discussion will focus on the causes of the

Fig. 6 Temporal variation in the global average ^{222}Rn flux density for the period of 1979 to 2007: (a) annual variation and (b) seasonal variation.

variation in the estimated ^{222}Rn flux density distribution and the uncertainties in the flux distribution.

4.1 Temporal and spatial variations

From the viewpoint of the annual and large-scale variations, the change in the global average ^{222}Rn flux density is affected by climate change. **Figs. 8(a)** and **(b)** show the annual and seasonal variations in the global averages for the soil water saturation and soil temperature, respectively, which were used in the exhalation model. The soil temperature shows a slight but steady increase from 14.5°C in 1999 to 15.1°C in 2007. The soil water saturation inversely shows a decrease from 0.47 in 1999 to 0.45 in 2007. These variations resulting from the climate change were taken into consideration in the exhalation model. Consequently, as shown in **Fig. 6(a)**, the decrease in the flux density in 1984 was caused by the increase in the soil water saturation. The increasing trend in the flux density since 1999 is the result of a decrease in water content and an increase in soil temperature. As shown in **Fig. 8(b)**, the seasonal variation in the soil temperature averaged over the period of 1979 to 2007 shows a minimum value in March and a maximum value in September, while the seasonal variation in the soil water saturation shows a minimum value from July to September and a maximum value in April. These seasonal variations are larger than the annual variations. The large seasonal variation in the flux density is also related to those of the soil water content and soil temperature.

The water content shows significant seasonal variations and complex spatial distributions. As shown in **Figs. 4** and **6(b)**, for high latitude zones in the northern hemisphere, the small

Fig. 8 Temporal variations in the global average soil temperature and soil water saturation for the period of 1979 to 2007: (a) annual variation and (b) seasonal variation.

values of ^{222}Rn flux density are caused by a seasonal change in the soil conditions, i.e., the snow coverage of the land surface and surface soils that are frozen in winter and saturated with water from melting snow in spring. For South Asia, the high water content of the soil during the rainy season from the end of spring to the beginning of winter leads to the minimum flux density in October. The low water content during the dry season leads to the maximum flux density in February. The change in water content is found to have the largest influence on the variation in the ^{222}Rn flux density.

The ^{226}Ra content of the soil also has a great influence on the flux density, next to the water content. As shown in **Fig. 4**, the complex spatial distribution of the flux in China is partially caused by the detailed information on the ^{226}Ra content. This result indicates that the influence of the ^{226}Ra content on the ^{222}Rn flux density is as significant as that of the water content of the soil.

The influences of soil temperature and topographical ruggedness are less important than the soil water content and ^{226}Ra content. The annual and regional variations in the soil temperature are more moderate than those of the soil water content. Although large ruggedness factors are found in high altitude regions, as shown in **Fig. 3**, the ruggedness factor for most of the land surfaces is almost unity. In addition, the flux density in the high altitude regions is low because of a high water content and low soil temperature. Therefore, the influence of the topographical ruggedness, as listed in **Table 1**, is only a few percent despite its large value in high altitude regions.

4.2 Uncertainty

As shown in **Fig. 5**, result of a comparison between the ^{222}Rn flux density distribution of the present study and the

maps by SCHERY and WASIOLEK¹²⁾ and CONEN and ROBERTSON¹³⁾ indicates that the ^{222}Rn flux density estimation in this study is successful in reproducing the northward declining flux density. However, for East Asia, a test of the maps using atmospheric ^{222}Rn concentrations implies that the flux density of the present study is too small, whereas that of the S-W map is too large. Therefore, the true ^{222}Rn flux density in East Asia is probably in the range between these two estimates. It was found in the atmospheric transport simulation that the best atmospheric ^{222}Rn concentration at Hachijo Island was found by using the present ^{222}Rn flux density multiplied by a factor of 1.69, which minimized the mean square difference between the simulation and observation values. This indicates that the flux density estimated in this study needs to be increased by 69% for the East Asian region. A similar analysis was performed for the previous version of our map to show that ship-borne atmospheric ^{222}Rn concentration measurements over the Arctic Ocean and Bearing Sea support the northward decrease in the flux density and very small flux values in the arctic regions, as in the present map.²⁹⁾ For other regions, the evaluation is insufficient because of the lack of observation data.

The uncertainty in the ^{222}Rn flux density estimated in this study can be considered to come from: 1) the ^{222}Rn exhalation model, 2) the data input for the soil condition and ^{226}Ra content of the soil, and 3) topographical ruggedness. First, there is an uncertainty in the exhalation model itself, as given by Eq. (1). ZHUO et al.²⁰⁾ verified the ^{222}Rn exhalation model using direct measurements of the flux density. The calculated ^{222}Rn flux densities were found to be in agreement with the measured ones within $\pm 25\%$ at 10 sites. The exhalation model used in this study assumes a constant soil texture of silt. The UNSCEAR 2000 report shows that the ^{222}Rn emanation coefficient for rocks and soils varies from 0.05 to 0.7. The soil texture-related uncertainties of the exhalation model are considered to come from the use of a constant dependence of the emanation coefficient on the water saturation and the use of the value of 0.1 for the emanation coefficient ε_0 for dry soil at 25°C in Eq. (3). Assuming a soil texture of sand or clay, partly according to ZHUO et al.,²⁰⁾ changes the emanation coefficient by only about -7% or 4% , respectively, if a value of 0.1 is used for ε_0 regardless of the soil texture. It is, therefore, highly likely that the dependence of ε_0 on the soil texture leads to the large variability in the emanation coefficient. Although the dependence on soil texture is still unclear, it can be said that this dependence may cause the uncertainty in the flux density estimated in this study.

Second, the uncertainty in the data input for the soil water saturation and soil temperature affects the temporal and spatial variations of the ^{222}Rn flux density. The flux density decreases significantly by 42% and 80% with an increase in soil water saturation from 0.1 to 0.5, and to 0.8, respectively, with Eq. (1). With the exception of the Amazonian area, the seasonal variations in the water content in the mid-latitudes obtained from JRA25/JCDAS were evaluated to be realistic from a comparison with observations.³⁰⁾ It is, therefore, reasonable to consider that the water saturation values used in this study are reliable for most of the continents, and hence, that the

relative variations in the ^{222}Rn flux density caused by the water saturation variations are proper, except for the Amazonian area. The water saturation in the Amazonian area was pointed out to be unrealistic.¹⁷⁾ This probably resulted in unrealistically large fluxes in the Amazonian area.

An increase in the soil temperature from 0°C and 40°C causes a 10% increase in the flux density with Eq. (3). The soil temperature affects the flux less significantly than the water saturation. We used the country-average ^{226}Ra content of the soil, if available, or the global average of 35 Bq kg⁻¹. Although it is difficult to estimate the uncertainty in the ^{226}Ra content data because of our limited knowledge of the spatial distribution, it can be said that this limitation may cause uncertainty comparable to that caused by the water content of the soil.

Third, the estimated topographical ruggedness has uncertainty depending on the resolution of the DEMs. Topographic ruggedness at a scale smaller than the resolution of a DEM is not captured by the DEM. To overcome this problem, this study extrapolated the linear relationship between the ruggedness factor and the horizontal resolution of a DEM. The results are consistent with a previous report¹²⁾, in which it was pointed out that the total global ^{222}Rn flux density might be enhanced by no more than 10% due to the ruggedness effect. The ASTER and SRTM DEMs refer to the canopy top rather than the ground surface. In addition, it is normal for the actual horizontal resolution of a DEM to be worse than its grid size. The ASTER GDEM Validation Team²³⁾ concluded that the actual horizontal resolution of the ASTER DEM was slightly better than 120 m, in spite of its 30 m grid size. Although the influence of topographic ruggedness may be larger than the present estimate due to this fact, the global average topographic ruggedness will not significantly enhance the flux density globally.

V CONCLUSION

We estimated the global ^{222}Rn flux density distribution with a horizontal resolution of 1° for the period of 1979–2007 by using a ^{222}Rn exhalation model based on a porous media diffusion theory. The global average for the flux density during this 29-year period was estimated to be 21.3 mBq m⁻² s⁻¹, which is consistent with the commonly used flux density of 21.0 mBq m⁻² s⁻¹ (1 atom cm⁻² s⁻¹). The flux density distribution had annual and seasonal variations, along with significant spatial variations. The global average for the ^{222}Rn flux density shows a slight annual increase from 21.1 mBq m⁻² s⁻¹ in 1999 to 22.1 mBq m⁻² s⁻¹ in 2007. In contrast, the seasonal variation averaged over the 29-year period is large, ranging from a minimum of 18.8 mBq m⁻² s⁻¹ in March to a maximum of 23.6 mBq m⁻² s⁻¹ in September. There are significant regional variations, from 8.3 mBq m⁻² s⁻¹ for the northern part of North America to 33.5 mBq m⁻² s⁻¹ for Australia. The latitudinal distribution of the zonal average flux densities in the Northern Hemisphere corresponds well to a previously suggested linear latitudinal decline. The most significant factor causing the variations in the monthly flux density was found to be the soil water content, followed by the

^{226}Ra content, and less importantly the soil temperature. The topographical ruggedness of the earth's ground surface was evaluated to increase the flux density by 2% globally but by more than 10% in mountainous regions. To test the estimated ^{222}Rn flux density in East Asia, we compared surface air ^{222}Rn concentration measurements at Hachijo Island with the values from an atmospheric ^{222}Rn transport simulation that used the present flux map as input. The simulated atmospheric ^{222}Rn concentration indicated that the flux density estimated in the present study in East Asia would result in better agreement with the observed concentration if it was enhanced by 69%. The uncertainties in the flux density were discussed from the viewpoint of the ^{222}Rn exhalation model and the water saturation, soil temperature, ^{226}Ra content, and topographical ruggedness input data. The assumption of the independence of soil textures may have resulted in the underestimation of the flux density. The flux density is more seriously affected by uncertainty in the water saturation than in the soil temperature and topographical ruggedness. The influence of the uncertainty in the ^{226}Ra content seems to be relatively large, although the variability in regional ^{226}Ra content is unclear. When all of these uncertainties are combined, they result in the systematic underestimation of the flux density in East Asia. The estimation of the global ^{222}Rn flux density distribution can be improved by considering the emanation coefficient's dependence on the soil texture.

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Shigekazu HIRAO

Shigekazu HIRAO received the M. E. degree in nuclear engineering from Nagoya University, Japan, in 2005. From 2005 to 2007, he worked in a railway company. He is currently doing the Ph. D. degree in energy engineering and science from Nagoya University. His research interests include transport of radionuclides (specifically radon-222 and its decay products) in the atmosphere, numerical modeling, and field measurements.
E-mail: s-hirao@ees.nagoya-u.ac.jp