Tellus (2002), 54B, 127–133
Printed in UK. All rights reserved

TELLUS
ISSN 0280–6509

Latitudinal distribution of radon-222 flux from continents

By F. CONEN* and L. B. ROBERTSON, *Institute of Ecology and Resource Management, University of Edinburgh, Mayfield Road, Edinburgh EH9 3JU, UK*

(Manuscript received 13 August 2001; in final form 6 November 2001)

ABSTRACT

Global atmospheric transport models are frequently tested by using ^{222}Rn as a tracer. Generally this tracer is assumed to be emitted at a uniform rate (1 atom $cm^{-2} s^{-1}$) from all ice-free land surfaces. The analysis of published data suggests a strong decrease from 30°N northwards to 0.2 atom $cm^{-2} s^{-1}$ at 70°N. This could be a result of increasing water tables and proportions of organic soils as indicated by larger proportions of wetlands in northern latitudes.

1. Introduction

The natural radioactive noble gas ^{222}Rn is produced by the decay of ^{226}Ra, a trace element present in all mineral soils. Its emission from ice-free land surfaces is assumed to be relatively uniform. Ice cover inhibits its emission. Oceans, where ^{226}Ra is in solution, emit about two orders of magnitude less ^{222}Rn than continents. This source distribution and its short half-life ($t_{1/2} =$ 3.82 d) make ^{222}Rn a useful tracer in atmospheric transport studies, an application first proposed by Israël (1951). It is often used in validating global atmospheric transport models (Genthon and Armengaud, 1995; Li and Chang, 1996; Jacob et al., 1997; Stockwell et al., 1998; Dentener et al., 1999; Stockwell and Chipperfield, 1999; Rasch et al., 2000). For this purpose, its large-scale emission rate needs to be known.

By inverse modelling of longitudinal variations in ^{210}Pb flux in the northern (15–55°N) and southern (15–55°S) hemispheres, Turekian et al. (1977) estimated ^{222}Rn flux from continents to be 1.2 atom $cm^{-2} s^{-1}$. Later, Lambert et al. (1982) used a global inventory of ^{222}Rn and its daughter products based on 20 yr of measurements and derived an average ^{222}Rn flux of 0.72 atom $cm^{-2} s^{-1}$.

In model validations, ^{222}Rn flux is generally assumed to be spatially uniform and around 1 atom $cm^{-2} s^{-1}$. However, Lee and Feichter (1995) concluded that a non-uniform distribution of the emission rate of ^{222}Rn over land would improve predictions of global transport and deposition of ^{210}Pb. They demonstrated this by comparing calculations using a constant emission rate with calculations using a simple distribution of latitudinal changes in the emission rates. A similar assumption was made in a comparison of scavenging and deposition processes in global models at the WCRP Cambridge Workshop 1995 (Rasch et al., 2000).

We use this empirical indication of ^{222}Rn flux changing with latitude as a hypothesis in re-assessing published data and in evaluating possible implications of own flux measurements on different soils in northern Britain.

2. Radon-222 flux measurements and estimates

2.1. Direct ^{222}Rn flux measurements

Radon-222 flux can be directly measured by accumulator methods. In principle, they involve

* Corresponding author.
e-mail: franz.conen@ed.ac.uk

the interception of gas flux between soil and the atmosphere by an inverted box (chamber) placed onto the soil surface. In a closed, or static system, this results in a usually linear increase in gas concentrations within the chamber. Flux is calculated from the observed concentration change over time, the average chamber height and the gas density. In an open, or dynamic system, outside air is continuously flushed through the chamber and concentration differences between incoming and outgoing air are measured. Flux is then a function of this concentration difference between in-flowing and out-flowing air, flow rate, chamber area, and gas density. Reviews of direct measurements of ^{222}Rn flux can be found in Wilkening et al. (1975) and Turekian et al. (1977). They generally show a large variability among different sites and between seasons. Apart from genuine differences in flux, differences in measurement techniques and apparatus also need to be considered. Hutter and Knutson (1998) found a coefficient of variation of 34% for ^{222}Rn flux measured at the same location by nine different groups.

Since the last review of which we are aware [Turekian et al. (1977)], many more ^{222}Rn flux measurements from natural soils have been reported (Table 1). Some of this work specifically focussed on the determination of ^{222}Rn flux from larger areas (e.g. Schery et al., 1989; Whittlestone et al., 1998; Nielson et al., 1996). In some of the other studies, ^{222}Rn flux measurements were limited to a few locations, or part of an experiment with a different focus. These directly measured fluxes show a similar average flux and variability, as previous flux measurements reported. Given the large variability of the data, there is no clear indication of a trend in ^{222}Rn flux across latitudes.

2.2. *Indirect ^{222}Rn flux estimates*

Indirect flux estimates from measurement of atmospheric ^{222}Rn profiles and the assumption of steady-state conditions between flux and decay have the advantage of integrating over large areas. Unlike direct measurements, they are not subject to small-scale variations in soil properties. However, for a near equilibrium between flux and decay to establish, air masses have to be in contact with a land surface for over a week. Indirect flux estimates of this kind have been made in the mid-latitudes of the northern hemisphere in France (Servant, 1964, cited in Wilkening et al., 1975), the former USSR (Kirichenko, 1970) and in the USA (Anderson and Larson, 1974; Wilkening et al., 1975). When plotted against latitude, they exhibit a decreasing trend with increasing latitude (Fig. 1). The same data plotted against longitude did not exhibit any trend.

Indirect estimation of ^{222}Rn flux is also possible from measurements of ^{210}Pb flux. Decay of ^{222}Rn in the atmosphere results in the formation of ^{210}Pb ($t_{1/2} = 22.3$ yr) which attaches to submicron-sized aerosols. The mean residence time of these particles in the atmosphere is around 6.5 d (Lambert et al., 1982) before they are scavenged by precipitation or deposited dry. While emission of the parent element ^{222}Rn is almost exclusively from continents, the daughter product ^{210}Pb is deposited on continents and oceans likewise. In order to estimate the average ^{222}Rn flux from continents in a certain latitudinal band, we might therefore divide the average ^{210}Pb deposition flux by the proportion of ice free continental area within this band.

Such an estimate is based on several assumptions. Firstly, that the number of ^{210}Pb atoms deposited is equal to the number of ^{222}Rn atoms emitted. This is very likely to be true because of the principal of continuity (Turekian et al., 1977) and the absence of any significant other sources of ^{210}Pb (Lambert et al., 1982). Secondly, that within a certain latitudinal band there is no bias in the allocation of ^{210}Pb measurement sites to areas with particularly low or high ^{210}Pb deposition fluxes. This seems correct for the latitudes north of 30°N, where ^{210}Pb flux has been measured at a large number of sites in America, Europe and Japan, including continental, coastal and maritime sites. However, south of 30°N, ^{210}Pb flux measurements are more sparse and are concentrated in a few regions, rendering this assumption less likely. The third assumption is that transfer of air masses from one latitudinal band into another is negligible. This is supported by the prevailing atmospheric circulation being either from west to east (30–60°N), or from east to west (10–30°S). Nevertheless, some loss or entrainment of ^{210}Pb from one latitudinal band into another is possible and might lead to an under- or over-estimation of ^{222}Rn flux.

To estimate ^{222}Rn flux based on this approach we can use the average values of ^{210}Pb flux for

Table 1. *Reported ^{222}Rn flux from natural soils since review by Turekian et al. (1977)*

Country	Latitude	^{222}Rn flux (atom cm^{-2} s^{-1})	No. of samples	No. of different sites sampled	Temporal scale	Reference
Canada	54°N	0.54	8	2	Jul + Aug	Ussler et al. (1994)
Canada	50°N	0.33	51	51	Aug + Sept	Kuhlmann et al. (1998)
Germany	50°N	0.08–0.19	11	6	[a]	Keller and Schütz (1988)
Germany	49°N	0.88	[a]	[a]	Annual average	Dörr et al. (1983)
Germany	49°N	0.75	>1000	5	8 year average	Schüßler (1996); cited in Levin et al. (1999)
Romania	46°N	0.95	9	3	Oct, Nov + Mar	Cosma et al. (1996)
Spain	37°N	0.60	235	4	Jan, Feb, Jun + Oct	Dueñas et al. (1997)
Japan	36°N	0.48	9	2	Jul–Oct	Uchida et al. (1997)
Japan	35°N	0.48	[a]	13	Annual average	Tojo (1989); cited in Moriizumi et al. (1996)
Japan	35°N	0.41	[b]	1	Oct, Dec + Jan	Koarashi et al. (2000)
USA	26–31°N	0.70	882	882	Apr + Jun	Nielson et al. (1996)
India	15°N	1.43	13	13	[a]	Somashekarappa et al. (1996)
Brazil	3°S	0.38	20	8	April + May	Trumbore et al. (1990)
Australia	11–39°S	1.05	–	–	Seasonally adjusted annual average	Schery et al. (1989)
		1.20	78	78	Jun + Jul	
Tasmania	41–44°S	1.10	74	37	Feb + Jan	Whittlestone et al. (1998)

[a] Data not given in reference
[b] Continuous measurement.
Only the arithmetic mean is indicated for locations where more than one value was given in the same reference (i.e. measurements at different times or different soils).

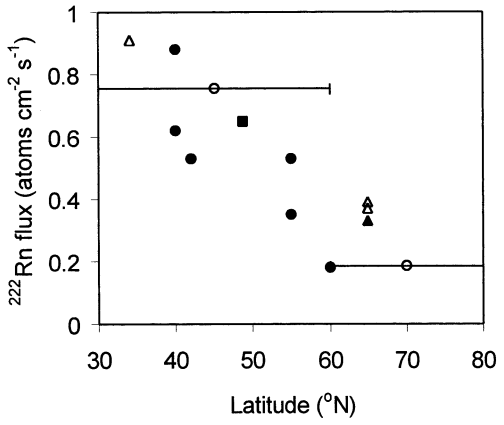

Fig. 1. Indirect measurements of ^{222}Rn flux in the mid-latitudes of the northern hemisphere. The decreasing trend with increasing latitude is supported by estimates derived from ^{210}Pb deposition flux (horizontal bars indicate the latitudes over which this flux is integrated). ■, Servant (1964); ●, Kirichenko (1970); ▲, Anderson and Larson (1974); △, Wilkening et al. (1975); ○, derived from ^{210}Pb fluxes in Preiss et al. (1996).

different latitudinal bands as reported in Preiss et al. (1996). This gives us ^{222}Rn flux values of 0.75 atom cm^{-2} s^{-1} for the latitudinal band 30–60°N, and 0.18 atom cm^{-2} s^{-1} for 60–80°N. These values will probably only have a small error because the discussed assumptions are most likely for these two latitudinal bands. They support the trend indicated by the atmospheric ^{222}Rn profiles in the same latitudes (Fig. 1). Consequently, the ^{222}Rn flux distribution based on indirect estimates suggests a linear decrease from 1 atom cm^{-2} s^{-1} at 30°N to 0.2 atom cm^{-2} s^{-1} at 70°N.

Radon-222 flux derived from ^{210}Pb deposition flux in the 10–30°N band gives a value of 1.8 atom cm^{-2} s^{-1}. This is certainly an overestimate resulting from the concentration of ^{210}Pb flux measurements in monsoon-influenced regions, where fluxes are expected to be much larger than in drier regions, such as the Sahara or Arabia (Rasch et al., 2000), from where there are no reported ^{210}Pb flux measurements. Lack of ^{210}Pb flux data from the equatorial regions precludes a

similar estimate between 10°N and 10°S. The ^{210}Pb-based estimate between 10 and 30°S (0.93 atom $cm^{-2}\,s^{-1}$) is very similar to the average of 1.05 atom $cm^{-2}\,s^{-1}$ for the Australian continent (11–39°S) measured directly by Schery et al. (1989). However, lack of ^{210}Pb flux values from southern Africa and South America reduces the confidence, we might put into the ^{210}Pb-based estimate for these latitudes. For the more southern latitudes (30–50°S), the ^{210}Pb-derived ^{222}Rn flux seems again unreasonably high (2.8 atom $cm^{-2}\,s^{-1}$). Entrainment of small quantities of ^{210}Pb into these latitudes from further north could result in a large over-estimate because of the small proportion of ice free continental area (0.06) in these latitudes.

3. Possible relations between latitude and ^{222}Rn flux

While the evidence for decreasing ^{222}Rn flux in northern latitudes is strong, it remains open which

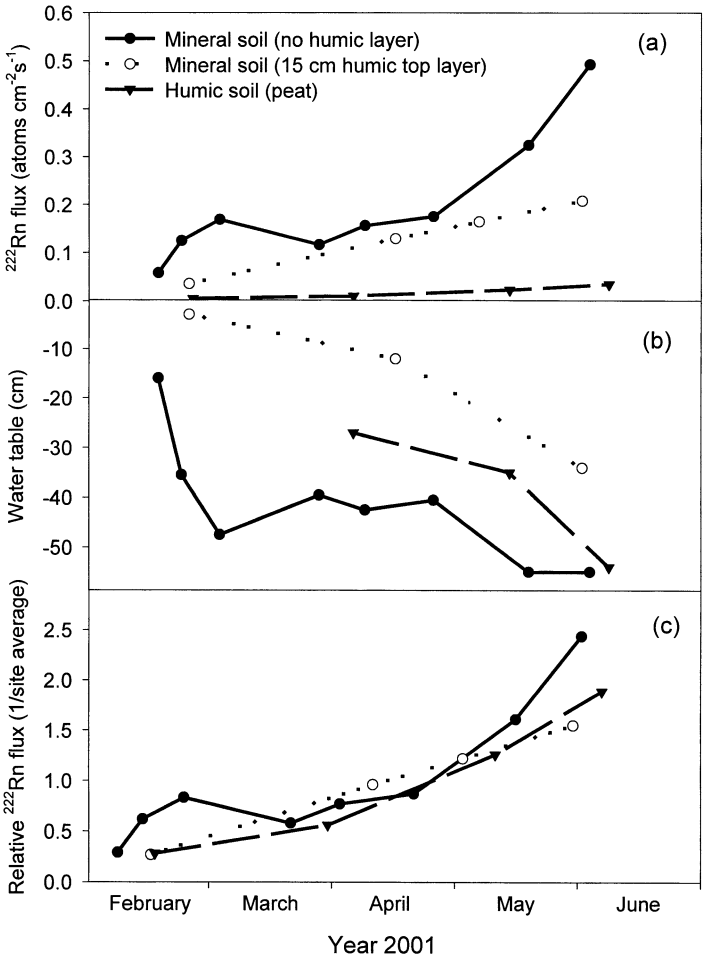

Fig. 2. (a) Radon-222 flux from February to June 2001 at three sites in Britain differing in the proportion of the humic layer in the soil profile. As (b) water tables decrease, (c) fluxes increase relative to respective site average.

factors or processes are causing this. One possibility could be wetter, and more organic, soils and shallower water tables in increasingly northern latitudes. Such a trend is indicated in the increasing proportion of peat bogs and other wetlands per unit land area with increasing latitude in the northern hemisphere (Stillwell-Soller et al., 1995). Compared to mineral soils, organic soils or peat have orders of magnitude lower concentrations of ^{226}Ra. This would result in reduced ^{222}Rn fluxes from humic soils, since ^{222}Rn flux is proportional to soil ^{226}Ra content, if all other factors are equal. Further, shallow water tables in wetlands reduce the thickness of the soil profile from which ^{222}Rn can escape into air spaces and diffuse into the atmosphere, as ^{222}Rn produced below the water table is unlikely to enter air space before its decay. Higher soil moisture contents associated with shallower water tables further reduces soil gas diffusivity and ^{222}Rn flux. This results in increased retention times of ^{222}Rn within the soil pore space and a larger proportion of it decaying before escape into the atmosphere (Dörr and Münnich, 1990).

To investigate these assumptions, we made direct measurements in the north of Britain at sites between 55 and 57°N contrasting in the thickness of their humic layer and at times of different water table depth. Volumetric soil water content was on average around 35% and rarely below 25%. The soil was never frozen when the measurements were made. The underlying material at all sites is of sedimentary origin. Fluxes were measured with four replicate closed chambers and samples (1 litre) were analysed in an ionisation chamber. Radon-222 flux decreased in the order: mineral soil, no humic layer > mineral soil with 15 cm humic top layer > humic soil (peat) (Fig. 2). We also found that fluxes increase with decreasing water table. It is unlikely that temperature had a significant effect on the observed changes in ^{222}Rn flux, since ^{222}Rn emanation and transport processes in soils are only very weakly affected by temperature, if at all (Schery et al., 1989; Nazaroff, 1992). These results support the assumption that an increasing proportion of more organic soils and water tables closer to the soil surface would decrease ^{222}Rn flux in northern latitudes.

Another possible cause for decreased ^{222}Rn flux with increasing latitudes could be differences in ^{226}Ra content in mineral soils. Evidence pointing in this direction has been found in China, where concentrations of ^{238}U (a source of ^{226}Ra) in soils have been found to decrease from south to north (Xu et al., 1993). This trend has been attributed to the pattern of geochemical weathering and the effect of leaching on soil development.

4. Proposed ^{222}Rn flux distribution across latitudes

Starting from the assumption of a uniform ^{222}Rn flux of 1 atom cm^{-2} s^{-1} from ice-free continental areas, we might now apply the constraint of linearly decreasing ^{222}Rn flux with increasing latitude from 1 atom cm^{-2} s^{-1} at 30°N to 0.2 atom cm^{-2} s^{-1} at 70°N. From 30°N into the equatorial region, measurements are sparse, but reported direct flux measurements are generally in the same range as at 30°N (Table 1), supporting the assumption of 1 atom cm^{-2} s^{-1} for these latitudes. The area-weighed average for the northern hemisphere based on this distribution would be 0.7 atom cm^{-2} s^{-1}, which is in accordance with the estimate of 0.72 atom cm^{-2} s^{-1} by Lambert et al. (1982). Constraining flux distribution for the southern hemisphere is limited by low data density. Nevertheless, directly measured fluxes in Australia agree well with an indirectly derived ^{222}Rn flux estimate for the 10–30°S latitudinal band and support the general assumption of 1 atom cm^{-2} s^{-1} for these latitudes. The southernmost reported flux measurements are from Tasmania (41–44°S) and are in a similar range

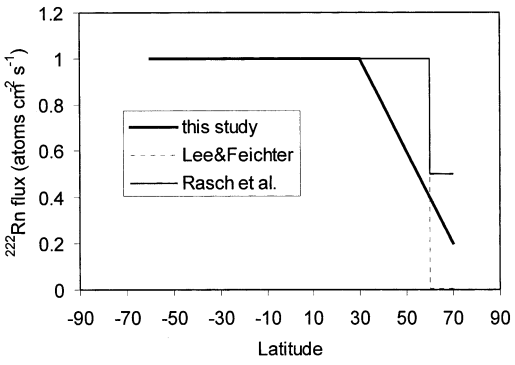

Fig. 3. Distribution of ^{222}Rn flux from ice-free land surfaces across latitudes as used in modelling studies and as proposed in this study.

(Whittlestone et al., 1998). In absence of information on ^{222}Rn flux in lower latitudes, we might extrapolate the value of 1 atom $cm^{-2}\,s^{-1}$ to other ice-free land surfaces further south. As a result of the very low proportion of ice-free land surface in this region, the potential error of this extrapolation is only of minor significance. The global average ^{222}Rn flux for ice-free land surfaces based on this distribution (Fig. 3) would be 0.8 atom $cm^{-2}\,s^{-1}$. Compared to ^{222}Rn flux distributions used in the modelling studies by Lee and Feichter (1995) and Rasch et al. (2000), the distribution proposed here differs mainly in its lower values between 30°N–60°N. This finding could be of importance in model validations, as it concerns the latitudinal band with the highest land/ocean ratio.

REFERENCES

Anderson, R. V. and Larson, R. E. 1974. Atmospheric electricity and radon profiles over a closed basin and the open ocean. *J. Geophys. Res.*, **79**, 3432–3435.

Cosma, C., Ristoiu, D., Poffijn, A. and Meesen, G. 1996. Radon in various environmental samples in the Herculane Spa, Cerna Valley, Romania. *Environ. Int.* **22** (Suppl. 1), S383–S388.

Dentener, F., Feichter, J. and Jeuken, A. 1999. Simulation of the transport of ^{222}Rn using on-line and off-line global models at different horizontal resolutions: a detailed comparison with measurements. *Tellus,* **51B**, 573–602.

Dörr, H., Kromer, B., Levin, I., Münnich, K. O. and Volpp, H.-J. 1983. CO_2 and radon 222 as tracers for atmospheric transport. *J. Geophys. Res.* **88**, 1309–1313.

Dörr, H.and Münnich, K. O. 1990. ^{222}Rn flux and soil air concentration profiles in West Germany. Soil ^{222}Rn as a tracer for gas transport in the unsaturated soil zone. *Tellus* **42B**, 20–28.

Dueñas, C., Fernández, M. C., Carretero, J., Liger, E. and Pérez, M. 1997. Release of ^{222}Rn from some soils. *Ann. Geophys.* **15**, 124–133.

Genthon, C. and Armengaud, A. 1995. Radon-222 as a comparative tracer of transport and mixing in 2 general-circulation models of the atmosphere. *J. Geophys. Res.* **100**, 2849–2866.

Hutter, A. R. and Knutson, E. O. 1998. An international intercomparison of soil gas radon and radon exhalation measurements. *Health Phys.* **74**, 108–114.

Israël, H. 1951 Radioactivity of the atmosphere. In *Compendium of meteorology* (ed. T. F. Malone). Am. Meteorol. Soc., Washington, DC, 155–161.

Jacob, D. J., Prather, M. J., Rasch, P. J., Shia, R. L., Balkanski, Y. J., Beagley, S. R., Bergmann, D. J., Blackshear, W. T., Brown, M., Chiba, M., Chipperfield, M. P., deGrandpre, J., Dignon, J. E., Feichter, J., Genthon, C., Grose, W. L., Kasibhatla, P. S., Kohler, I., Kritz, M. A., Law, K., Penner, J. E., Ramonet, M., Reeves, C. E., Rotman, D. A., Stockwell, D. Z., VanVelthoven, P. F. J., Verver, G., Wild, O., Yang, H. and Zimmermann, P. 1997. Evaluation and intercomparison of global atmospheric transport models using ^{222}Rn and other short-lived tracers. *J. Geophys Res.* **102**, 5953–5970.

Keller, G. and Schütz, M. 1988. Radon exhalation from the soil. *Radiat. Protect. Dosim.* **24**, 43–46.

Kirichenko, L. V. 1970. Radon exhalation from vast areas according to vertical distribution of its short-lived decay products. *J. Geophys. Res.* **75**, 3639–3649.

Koarashi, J., Amano, H., Andoh, M. and Iida, T. 2000. Estimation of ^{222}Rn flux from ground surface based on the variation analysis of ^{222}Rn concentrations in a closed chamber. *Radiat. Protect. Dosim.* **87**, 121–131.

Kuhlmann, A. J., Worthy, D. E. J., Trivett, N. B. A. and Levin, I. 1998. Methane emissions from a wetland region within Hudson Bay Lowland: an atmospheric approach. *J. Geophys. Res.* **103**, 16,009–16,016.

Lambert, G., Polian, G., Sanak, J., Ardouin, B., Buisson, A., Jegou, A. and Le Roulley, J. C. 1982. Cycle du radon et de ses descendants: application à l'étude des échanges troposphère–stratosphère. *Ann. Géophys.* **38**, 497–531.

Lee, H. N. and Feichter, J. 1995. An intercomparison of wet precipitation scavenging schemes and the emission rates of ^{222}Rn for the simulation of global transport and deposition of ^{210}Pb. *J. Geophys. Res.* **100**, 23,252–23,270.

Levin, I., Glatzel-Mattheier, H., Marik, T., Cuntz, M. and Schmidt, M. 1999. Verification of German methane emission inventories and their recent changes based on atmospheric observations. *J. Geophys. Res.* **104**, 3447–3456.

Li, Y. H. and Chang, J. S. 1996. A three-dimensional global episodic tracer transport model. Evaluation of its processes by radon-222 simulations. *J. Geophys. Res.* **101**, 25,931–25,947.

Moriizumi, J., Nagamine, K., Iida, T. and Ikebe, Y. 1996. Estimation of areal flux of atmospheric methane in an urban area of Nagoya, Japan, inferred from atmospheric radon-222 data. *Atmos. Environ.* **30**, 1543–1549.

Nazaroff, 1992. Radon transport from soil to air. *Rev. Geophys.* **30**, 137–160.

Nielson, K. K., Rogers, V. C. and Holt, R. B. 1996. Measurements and calculations of soil radon flux at 325 sites throughout Florida. *Environ. Int.* **22** (Suppl. 1), S471–S476.

Preiss, N., Mélières, M.-A., Pourchet, M. 1996. A compilation of data on lead 210 concentration in surface air and fluxes at the air-surface and water-sediment interfaces. *J. Geophys. Res.* **101**, 28,847–28,862.

Rasch, P. J., Feichter, J., Law, K., Mahowald, N.,

Penner, J., Benkovitz, C., Genthon, C., Giannakopoulos, C., Kasibhatla, P., Koch, D., Levy, H., Maki, T., Prather, M., Roberts, D. L., Roelofs, G.-J., Stevenson, D., Stockwell, Z., Taguchi, S., Kritz, M., Chipperfield, M., Baldocchi, D., McMurry, P., Barrie, L., Balkanski, Y., Chatfield, R., Kjellström, E., Lawrence, M., Lee, H. N., Lelieveld, J., Noone, K. J., Seinfeld, J., Stenchikov, G., Schwartz, S., Walcek, C. and Williamson, D. 2000. A comparison of scavenging and deposition processes in global models: results from the WCRP Cambridge Workshop of 1995. *Tellus* **52B**, 1025–1056.

Schery, S. D., Whittlestone, S., Hart, K. P. and Hill, S. E. 1989. The flux of radon and thoron from Australian soils. *J. Geophys. Res.* **94**, 8567–8576.

Schüßler, W. 1996. *Effective Parameter zur Bestimmung des Gasaustauschs zwischen Boden und Atmosphäre*. PhD Thesis, University of Heidelberg.

Servant, J. 1964. *Radon and its short lived daughters in the atmosphere*. Ph.D. Thesis. University of Paris.

Stillwell-Soller, L. M., Klinger, L. F., Pollard, D. and Thompson, S. L. 1995. *The global distribution of wetlands*. NCAR Technical Note, NCAR/TN-416+STR, National Center for Atmospheric Research, Boulder, Colorado.

Somashekarappa, H. M., Narayana, Y., Radhakrishna, A. P., Siddappa, K., Joshi, V. B., Kholekar, R. V. and Bhagwat, A. M. 1996. Atmospheric radon levels and its emanation rate in the environment of Kaiga. *Radiat. Meas.* **26**, 35–41.

Stockwell D. Z., Kritz, M. A., Chipperfield, M. P. and Pyle, J. A. 1998. Validation of an off-line three-dimensional chemical transport model using observed radon profiles. 2. Model results. *J. Geophys. Res.* **103**, 8433–8445.

Stockwell, D. Z. and Chipperfield, M. P. 1999. A tropospheric chemical-transport model: development and validation of the model transport schemes. *Q. J. R. Meteorol. Soc.* **125**, 1747–1783.

Tojo, K. 1989. *Survey of ^{222}Rn exhalation rate from ground and its relationship with atmospheric ^{222}Rn concentration.* Master Thesis, Department of Nuclear Engineering, Nagoya University (in Japanese).

Trumbore, S. E., Keller, M. Wofsy, S. C. and Da Costa, J. M. 1990. Measurements of soil and canopy exchange rates in the Amazon rain rorest using ^{222}Rn. *J. Geophys. Res.* **95**, 16,865–16,873.

Turekian, K. K., Nozaki, Y. and Benninger, L. K. 1977. Geochemistry of atmospheric radon and radon products. *Ann. Rev. Earth Planet Sci.* **5**, 227–255.

Uchida, M., Nojiri, Y.,. Saigusa, N. and Oikawa, T. 1997. Calculation of CO_2 flux from forest soil using ^{222}Rn calibrated method. *Agric. Forest Meteorol.* **87**, 301–311.

Ussler, W., Chanton, J. P., Kelly, C. A. and Martens, C. S. 1994. Radon-222 tracing of soil and forest canopy trace gas exchange in an open canopy forest. *J. Geophys. Res.* **99**, 1953–1963.

Whittlestone, S., Zahorowski, W. and Schery, S. D. 1998. Radon flux variability with season and location in Tasmania, Australia. *J. Radioanal. Nucl. Chem.* **236**, 213–217.

Wilkening, M. H., Clements, W. E. and Stanley, D. 1975. Radon-222 flux measurements in widely separated regions. In *The natural radiation environment II* (ed. J. A. S. Adams). USERDA CONF-720805, 717–730.

Xu, N., Wei, F. S., Ten, E. J. and Chen, L. Q. 1993. Evaluation of indigenous concentrations of uranium and thorium in soils of China. *Commun. Soil Sci. Plant Anal.* **24**, 1795–1803.