

Use of radon for evaluation of atmospheric transport models: sensitivity to emissions

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ABSTRACT

We present comparative analyses of atmospheric radon (Rn) distributions simulated using different emission scenarios and the observations. Results indicate that the model generally reproduces observed distributions of Rn but there are some biases in the model related to differences in large-scale and convective transport. Simulations presented here use an off-line three-dimensional chemical transport model driven by assimilated winds and two scenarios of Rn fluxes (atom cm⁻² s⁻¹) from ice-free land surfaces: (A) globally uniform flux of 1.0 within ±60° and 0.5 within 60°N–70°N and (B) uniform flux of 1.0 between 60°S and 30°N followed by a sharp linear decrease to 0.2 at 70°N. We considered an additional scenario (C) where Rn emissions for case A were uniformly reduced by 28%. Results show that case A overpredicts observed Rn distributions in both hemispheres. Simulated Northern Hemisphere Rn distributions from cases B and C compare better with the observations, but are not discernible from each other. In the Southern Hemisphere, surface Rn distributions from case C compare better with the observations. We performed a synoptic-scale source–receptor analysis for surface Rn to locate regions with ratios B/A and B/C less than 0.5. Considering the maximum uncertainty in regional Rn emissions of a factor of 2, our analysis indicates that additional measurements of surface Rn, particularly during April–October and north of 50°N over the Pacific as well as Atlantic regions, would make it possible to determine if the proposed latitude gradient in Rn emissions is superior to a uniform flux scenario.

1. Introduction

Evaluation of the representation of large-scale and subscale transport processes in an atmospheric model is a pre-requisite for its application to the study of chemistry–climate interactions. Long-lived tracers such as chlorofluorocarbons (CFCs) and SF₆ are generally used to evaluate large-scale atmospheric transport features such as interhemispheric exchange (Prather et al. 1987; Denning et al. 1999; Gupta et al. 2001). Radon (²²²Rn, or simply Rn), characterized by its short radioactive decay time constant ($\tau_{1/2} = 3.825$ d) and primarily land-based emissions, has been extensively used as a tracer for the study of subscale convective and synoptic transport processes in the troposphere (Liu et al. 1984; Dentener et al. 1999; Chevillard et al. 2002). Radon has been used for model intercomparison studies (Jacob et al. 1997; Rasch et al. 2000), assessment of convective parametrizations (Mahowald et al. 1995; Allen et al. 1996) and evaluation of model transport of continental air to remote oceanic locations (Heimann et al. 1990; Balkanski and Jacob 1990).

Emissions of Rn from radioactive decay of radium in soils are known to be highly variable. Factors controlling Rn emissions include the ²²⁶Ra concentration and the physical characteristics of the soil affecting its escape into the atmosphere (Turekian et al. 1977; Nazaroff 1992). Jacob and Prather (1990) and Genthon and Armengaud (1995) have derived algorithms to account for some of the variations in Rn emissions. Observations used to develop these algorithms are limited and do not uniformly represent the entire globe, therefore a simple emission rate of 1.0 atom cm⁻² s⁻¹ over the ice-free land surfaces between 60°S and 60°N has been widely used (Jacob et al. 1997; Rasch et al. 2000). This emission rate is considered to be accurate to within 25% globally and within a factor of 2 regionally (see Jacob et al. 1997, and references therein). Also, this Rn flux falls between two other estimates of 0.72 atom cm⁻² s⁻¹ and 1.2 atom cm⁻² s⁻¹, suggested by Lambert et al. (1982) and Turekian et al. (1977). Oceanic contributions to the Rn flux are considered negligibly small (0.005 atom cm⁻² s⁻¹).

Earlier, Lee and Feichter (1995) concluded that the use of latitudinally varying Rn emission rates in their modelling of transport and deposition of ²¹⁰Pb improved the model results against observations, particularly at high Northern Hemisphere latitudes. In this study, they used uniform Rn emission rates of

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1.0 atom cm⁻² s⁻¹ and 0.005 atom cm⁻² s⁻¹ for land and oceanic surfaces respectively between 60°S and 60°N. Between 60° and 70°, in both hemispheres, they decreased the ice-free land-based emission rate to 0.005 atom cm⁻² s⁻¹. Jacob et al. (1997) also used this emission rate between 60°N and 70°N. However, in the intercomparison study of Rasch et al. (2000), a Rn emission rate of 0.5 atom cm⁻² s⁻¹ was used. Various models have simulated atmospheric distributions of Rn using emission rates of Rasch et al. (2000) and assimilated meteorology and compared them against observed surface concentrations and vertical profiles (e.g. Dentener et al. 1999; Taguchi et al. 2002).

Recently, Conen and Robertson (2002) reported that land-based Rn fluxes decrease linearly from 1.0 atom cm⁻² s⁻¹ at 30°N to 0.2 atom cm⁻² s⁻¹ at 70°N. Their conclusion was based on indirect estimates of Rn emissions. The authors speculated that the northward decrease in Rn emissions could be due to more organic soils with decreasing radium content and shallow water tables. They emphasized the potential importance of this finding in the validation of atmospheric transport models. This revised estimate modifies Rn emission from 59% of the Northern Hemisphere land-cover between 31°N and 71°N.

Here, we investigate the impact of different emission estimates of Rn on simulated atmospheric distributions. We compare simulated Rn distributions against corresponding observations. This comparative analysis will also evaluate treatment of physical processes and meteorological inputs to the model that are responsible for long-range as well as vertical transport of Rn in the atmosphere.

2. Model simulations

All the simulations presented here were performed using the Goddard Space Flight Center (GSFC) off-line three-dimensional parametrized chemistry and transport model (PCTM) (Nielsen and Douglass 2001). Meteorological fields (surface pressure, temperature and winds), and convective and diffusive fluxes input to PCTM were taken from a reanalysis run of the Goddard Earth Observation System version 4 (GEOS-4) data assimilation system given at a resolution of 2.5° × 2° with 55 hybrid sigma-pressure levels between the surface and 80 km. GEOS-4 is an updated version of the finite-volume data assimilation system (FVDAS) used by Douglass et al. (2003). The time-dependent tracer mass continuity equation was solved at 15 min time intervals using linearly interpolated wind fields drawn from 6 h archives of GEOS-4 meteorology. Advection was calculated using a flux-form semi-Lagrangian scheme (Lin and Rood 1996) on the GEOS-4 grid. A pressure fixer, applied following Prather et al. (1987), adjusts the horizontal winds slightly to remove the mass increase that results from inconsistency between the forecast and observed surface pressures used in the assimilation process. A semi-implicit numerical scheme was applied for

convective tracer transport based on the three-dimensional cloud mass flux produced by the FVDAS.

Atmospheric Rn simulations were performed using analysed meteorology for 1993 and 1994. For both years, two emission scenarios were used: (A) 1.0 atom cm⁻² s⁻¹ between 60°N–60°S and 0.5 atom cm⁻² s⁻¹ between 60°N–70°N (Rasch et al. 2000), and (B) latitude-dependent emissions suggested by Conen and Robertson (2002). For these two cases, Rn emission rates differ for the latitude region of 30°N–70°N. Fractional land-based emissions were uniformly applied in coastal and island-containing model grids using a high-resolution (1/6° × 1/6°) surface topography. No alterations in land-based Rn fluxes in these model grids were introduced, even though there is evidence of lower Rn emanation rates at the island of Hawaii due to water saturation (Whittlestone et al. 1996). Also, surface Rn emissions were zero for model grids with surface temperature below freezing point. We lack knowledge of the temporal variations in land-based Rn sources; therefore, constant emissions were applied at each time step. For both cases, the model was allowed to spin-up for 3 months before comparing the results with observations. We also considered another Rn emission scenario as proposed by Lambert et al. (1982) by scaling down the simulated Rn concentrations for case A by a factor of 0.72. This model scenario is termed C. For cases A, B and C, annual global Rn emissions were 62.1, 55.3 and 45.0 mol respectively.

To investigate the impact of latitudinal gradient in Rn emissions, we concentrate on its Northern Hemisphere distribution. Validation was performed by comparison with observations from the same period as the analysed wind (1993–1994). Measured Rn vertical profiles over Moffett Field during 1994 (Kritz et al. 1998) and from the North Atlantic Regional Experiment (NARE) Intensive-1993 (Zaucker et al. 1996) were compared with model profiles for all three emission scenarios. Surface measurements of Rn over Bermuda (64.45°W, 32.2°N, 28 m above sea level (asl); BER) for 1993 reported by the Environmental Measurement Laboratory (EML) (Hutter et al. 1995) were also used. EML reports data for BER at a 60 min time interval. Even though Rn data for other Northern Hemisphere stations, e.g. an island site (Mauna Loa) and continental sites (Freiburg and Schauinsland, Germany) are also available, we considered Bermuda as a representative site for model-data comparison because of its remote location, low surface elevation and negligible local emissions (surface land area = 53 km²). For the purpose of model evaluation to examine the influence of uniform reduction in surface emissions we also include comparison between simulated and observed surface Rn for 1993 at two Southern Hemisphere stations: Amsterdam Island (77.34°E, 37.5°S, 50 m asl; AMS) and Cape Grim (144.41°E, 40.41°S, 94 m asl; CGA) following Dentener et al. (1999). CGA is located on the coastal site of Australia and AMS is remotely located in the Indian Ocean with a land surface area of 61 km².

3. Results and discussion

3.1. Comparison for Rn vertical profiles

3.1.1. Vertical profiles during the NARE-1993 campaign. Simulated and observed Rn for the NARE Intensive period 16–31 August 1993 are compared in Fig. 1. Sixty-six Rn samples were collected between the boundary layer and 5.5 km over the North Atlantic Ocean and the continent in the vicinity of Nova Scotia and New Brunswick, Canada (Zaucker et al. 1996). To facilitate the comparison, observed Rn concentrations and corresponding modelled values were divided into three subdivisions: 0–2 km for the marine boundary layer (MBL); 0–2 km for the continental boundary layer (BL); and free troposphere (FT). The total numbers of samples in the MBL, BL and FT subdivisions are 28, 13 and 25 respectively. Average concentrations and $\pm 1\sigma$ deviation for each division were computed for analysis.

Case A predicts larger than observed Rn concentrations for all three regions. Modelled Rn concentrations from cases B and C closely correspond to observed values, particularly for MBL and FT. For case B the agreement is within $\pm 5\%$, and for case C it is within -3 and 16% . For the BL, cases B and C overpredict observed average Rn by 27% and 38% respectively. This stated discrepancy could be due to uncertainty in Rn emissions and insufficient vertical transport of continental boundary layer air mass to the free troposphere. The latitudinal band covered during the NARE-1993 Intensive is 42°N – 46°N where the Rn flux was reduced by 24–32% in case B. The mean value of this reduction roughly corresponds to the decrease in global Rn surface emissions for case C as compared with that for case A. This comparison indicates that although the ratio (B/C) for integrated Northern Hemisphere Rn source strength is 1.25, the simulated Rn distributions from both cases for the NARE geographical area are not clearly distinct from each other.

3.1.2. Vertical profiles over Moffett Field during 1994. A comparison between modelled and observed Rn concentrations over Moffett Field (37.4°N , 122.0°W) is shown in Fig. 2. Observations are adopted from Kritz et al. (1998), who report Rn profiles from the surface to 11.5 km obtained from 127 samples

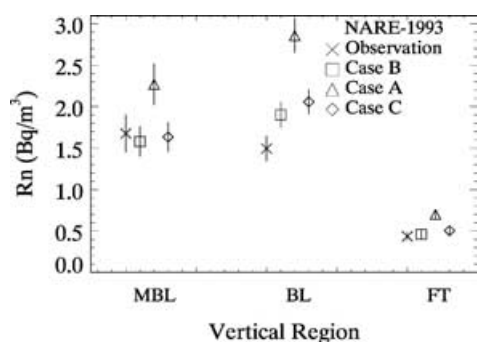


Fig. 1. Comparison between simulated and observed Rn profiles during NARE-1993. Error bars represent $\pm 1\sigma$ deviation from the mean.

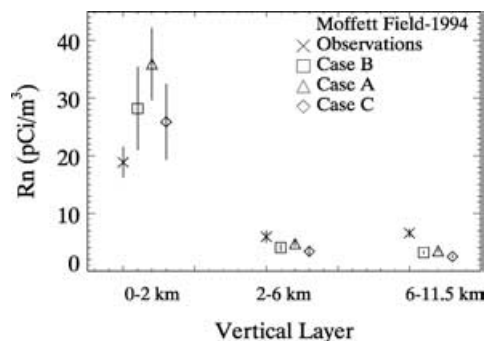


Fig. 2. Comparison between simulated and observed Rn vertical profiles over Moffett Field for the period 3 June to 16 August 1994. Error bars represent $\pm 1\sigma$ deviation from the mean.

on 11 flights during 3 June to 16 August 1994. We averaged Rn concentrations corresponding to three segments of vertical profiles: (1) 0–2 km; (2) 2–6 km and (3) 6–11.5 km. All three emission scenarios overestimate observed average Rn in the lowest 2 km of the atmosphere; however, this comparison is the worst for case A. In the model, Moffett Field is located in the coastal grid (with 64% of land cover) which, as stated earlier, may have a lower Rn emanation rate than what is considered here. At the latitude of Moffett Field, reductions in Rn emission for cases B and C are 15% and 28% respectively relative to case A. For the layer between 2 and 6 km, Rn distributions from all cases closely follow each other within $\pm 1\sigma$ deviation around the mean. Besides atmospheric mixing, a combination of two factors, (1) higher magnitude of Northern Hemisphere emissions and (2) a relatively smaller reduction in local surface flux at Moffett field for case B, resulted in its distributions that are no different from those of case C.

Because of simulated excessive Rn near the surface, irrespective of emission scenarios, the model underpredicts observed Rn at high altitudes. This discrepancy may be due to insufficient long-range transport and possible existence of high Rn emission from the East Asian region (Dentener et al. 1999) which is not considered here. Doubling of convective fluxes in model simulation improved the results by up to 15% at high altitudes. Model simulations for case B with regionally tagged emissions for Indo-China and North American regions show that, above 6 km, more than 50% of the Rn over Moffett Field originates in the Indo-China region (Fig. 3). This result is qualitatively consistent with the conclusions of Kritz et al. (1990) and Stockwell et al. (1998). Together these regional sources account for more than 80% of total atmospheric Rn simulated over the Moffett Field.

3.2. Comparison for surface radon

3.2.1. Northern Hemisphere. We compare modelled and observed Rn surface concentrations for Northern Hemisphere station BER. Bermuda is downwind of North America and routinely experiences a strong influence of its Rn sources due to

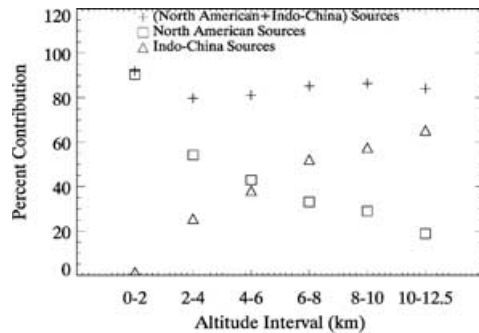


Fig 3. Percent contribution of regional sources of Rn from North America and Indo-China to its vertical profile over Moffett Field during 3 June to 16 August 1994. Note that the combined contribution from both regional sources is more than 80% of the total Rn at all vertical levels.

westerly flow. As should be expected due to the use of assimilated meteorological fields, the model often captures observed synoptic-scale variability in surface Rn as shown for June in Fig. 4. In this figure, we used model results for case B, but it should be emphasized that synoptic variability in simulated Rn is independent of our choice of emission scenario. However, due to differences in emission intensities, the simulated extreme Rn concentrations during these synoptic events differ significantly. The model failed to simulate a prolonged event of high Rn concentration observed around Julian days 169 and 171. It also predicts excessive Rn around Julian days 160–163, but generally corresponds with the data to within a factor of 2. Radon simulations performed using assimilated winds from an earlier version (GEOS-1) exhibited synoptic-scale variability of a similar nature at this location (Allen et al. 1996).

For comparison of modelled and observation data for BER over a year, surface Rn was averaged each month. The model produces the general features of the observed monthly variation in surface Rn at BER (Fig. 5). The standard error of the mean for observation is calculated to be very small. Lower Rn concentrations during late summer and autumn are due to the formation of

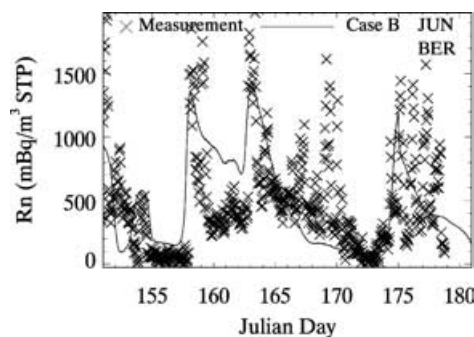


Fig 4. Comparison between synoptic-scale variation in simulated and observed surface Rn at Bermuda for June 1993.

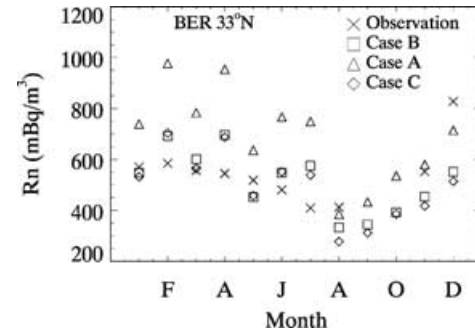


Fig 5. Comparison between simulated and observed monthly averaged surface Rn at Bermuda during 1993. Observational data were missing for September and October.

the so-called ‘Bermuda High’ which prevents outside air from entering into the high-pressure region.

During the first half of the year, case A consistently overpredicts observed monthly averaged surface Rn at BER. Simulated surface Rn concentrations from cases B and C closely follow each other and are in close agreement with the observed values (with correlation coefficients of 0.60 and 0.55 respectively) despite a reduction of only 6% in surface flux at 33°N for case B as compared with 28% for case C. It should be noted that these reductions in Rn emissions are within the range of uncertainties in regional and global sources. However, in the absolute sense, this comparison reinforces previous conclusions that despite a difference in Northern Hemisphere source strength in Rn emissions for cases B and C, their simulated Rn distributions do not significantly differ from each other due to mixing of air masses from other latitudes.

3.2.2. Southern Hemisphere. In this section, we present comparison between simulated and observed surface Rn for the two Southern Hemisphere stations, AMS and CGA. Figure 6 shows a comparison of synoptic changes in observed and simulated surface Rn from case B at (a) CGA for March and (b) AMS for July. Similar to that for station BER, at both sites the model captures most events of observed high and low surface Rn activities. In the case of AMS, the model successfully simulates the baseline Rn surface concentrations and events of radonic storms. In the absence of local emissions due to the very small surface land area, the good comparison of these characteristic features of surface Rn at AMS establishes a confidence in the model’s ability to accurately simulate the transport of the continental air mass to the remote sites.

Figure 7 shows a comparison between simulated and observed monthly averaged surface Rn at (a) CGA and (b) AMS. Observations show significant variability around the mean, but the corresponding standard error of the mean is calculated to be very small. At CGA, the model reproduces monthly variation in observed surface Rn. For this month both cases A and B overpredict the surface Rn. In general, simulated monthly mean Rn from case C closely follows corresponding observed surface

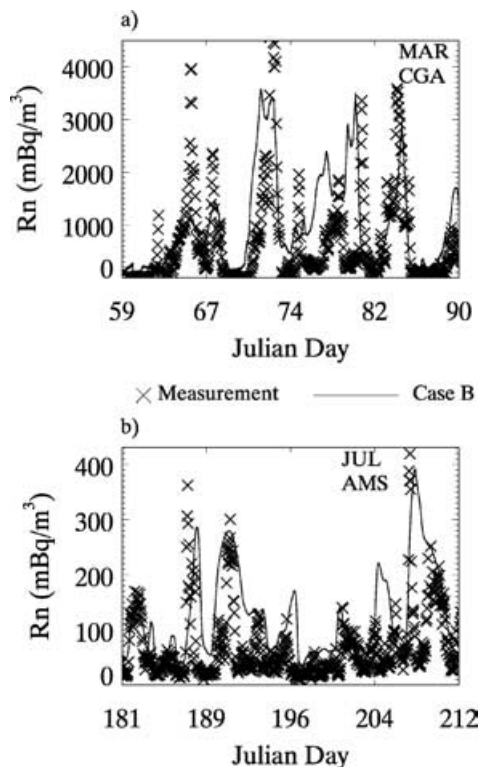


Fig 6. Comparison between synoptic-scale variations in simulated and observed surface Rn at (a) Cape Grim (CGA) for March 1993 and (b) Amsterdam Island (AMS) for July 1993.

values at CGA. For case C, the coefficient of correlation between monthly averaged observed and simulated surface Rn is 0.68. There seems to be a tendency in the model to underpredict the observed mean Rn during May and June when corresponding values from all three cases are significantly lower.

At AMS, all cases reproduce the general features of monthly variation in observed surface Rn but overpredict its magnitude, indicating stronger trapping of the air mass in the boundary layer similar to that for Rn over the Moffett Field. Similar to station CGA, the standard error of the mean for observation data is very small. This overprediction is the least for case C. For this case, the coefficient of correlation between monthly averaged observed and simulated surface Rn is 0.72. For most months, the simulated monthly mean surface Rn from cases A and B significantly differs from the corresponding observed mean values (Fig. 7b). Combined with the inference from the earlier discussion for station CGA, this comparison shows that the emission scenario from case C is better suited for simulation of Southern Hemisphere Rn distribution.

We performed another model simulation identical to case A except that the effect of Rn emissions due to partial land cover in coastal and island grid boxes was neglected. For the AMS site there is no significant effect of neglecting the local emissions due to its small land area, as shown by plus symbols in Fig. 7b. On the

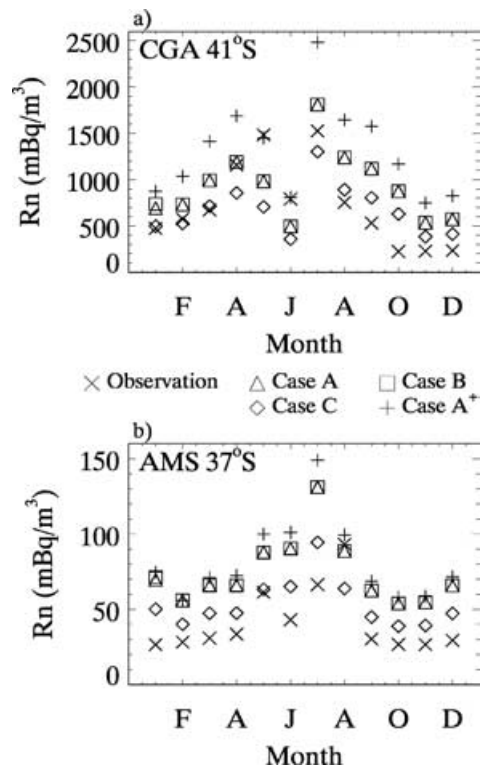


Fig 7. Comparison between simulated and observed monthly averaged surface Rn at (a) Cape Grim (CGA) and (b) Amsterdam Island (AMS) during 1993. Plus symbols correspond to surface Rn distribution from case A when fractional land/ocean covers were neglected.

other hand, improper distribution of Rn emissions in the model grid containing CGA leads to overestimation of its simulated surface concentrations for the months of September to December (Fig. 7a). Similarly, for two other Southern Hemisphere sites, Kerguelen (70.15°E, 49.21°S, 30 m asl) and Crozet (51.52°E, 46.26°S, 150 m asl) (results not shown here) model calculations indicate that local emissions can significantly affect the baseline surface Rn because of their land surface areas (7215 km² and 325 km², respectively). These results indicate that Rn fluxes need to be prescribed carefully in coastal- and island-containing model grids in order to compare with data meaningfully.

Based on comparisons with the observations presented here, this study clearly shows that the model, in general, is capable of reproducing the observed distributions of atmospheric Rn. Additionally, comparisons show that the use of either the newly proposed Northern Hemisphere latitude-dependent Rn flux scenario (case B; Conen and Robertson 2002) or the uniform global ice-free land-based Rn surface flux of 0.72 atom cm⁻² s⁻¹ (case C; Lambert et al. 1982) in model simulations leads to Northern Hemisphere atmospheric distributions of Rn that are in good agreement with the observations. Because of the limited observational database, Rn distributions from these scenarios are not clearly discernible from each other. Case A (with a continental

Rn surface flux of $1.0 \text{ atom cm}^{-2} \text{ s}^{-1}$) overpredicts observed Rn concentrations in both hemispheres. At Southern Hemisphere sites, Rn distributions from case C compare better with the corresponding observations.

3.3. Comparison between Northern Hemisphere Rn distributions simulated using different emission scenarios

In this section, we concentrate on how the atmospheric Rn distribution for emission scenario B differs from those of A and C in the Northern Hemisphere on various timescales.

3.3.1. Annual averaged distributions. Figure 8 shows the Northern Hemisphere latitude–longitude distribution of the ratio of annually averaged surface Rn mixing ratios between cases B and A. In this figure, we also show locations of Rn measurement sites that are influenced by the differences in both emission distributions. Because of the short lifetime of Rn and the prescribed differences in its Northern Hemisphere emissions, values of B/A lower than 1.0 are mostly confined to the Northern Hemisphere and decrease towards higher latitudes (shown by solid lines). South of 45°N , the ratio B/A, in general, is more than 0.75 which is within the range of global uncertainty in Rn surface emissions. At high latitudes over continental areas this ratio is lower than 0.5. There are two factors that contribute to this pattern of B/A: (1) northward decline in Rn emissions and (2) about 12% fewer total Northern Hemisphere emissions for case B. After uniformly scaling down Northern Hemisphere land-based emissions for case A by 12%, values of B/A are as low as 0.7 at high latitudes (shown by dashed lines) indicating that it is not the emission strength but the latitudinal gradient that has a strong impact on the distribution of B/A. Calculated lower values of B/A persist throughout the troposphere for most of the Northern Hemisphere, as shown in Fig. 9. Note that the

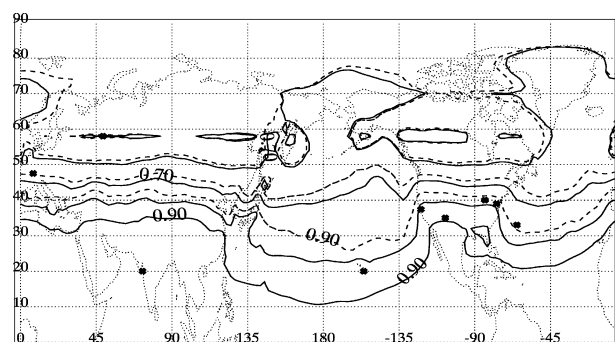


Fig 8. Horizontal cross-section of the ratio B/A (solid lines) for annually averaged surface Rn. The contour levels decrease northwards with an interval of 0.1. Corresponding ratios involving case A in which its Northern Hemisphere Rn surface fluxes were uniformly reduced by 12% are shown by dashed lines. Also shown are locations of some of the Northern Hemisphere Rn measurement sites affected by differences between emissions for cases A and B.

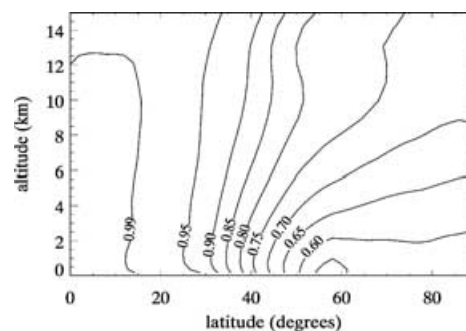


Fig 9. Zonally averaged distribution of ratio B/A for simulated annually averaged Rn.

lowest value of B/A is centred between 55°N and 60°N . At the mid-latitudes, there is a tendency towards an increase in B/A values with altitude indicating a decline in impact of the latitudinal gradient in Rn emissions. At 50°N and 4 km altitude, annually and zonally averaged Rn distributions from simulations B and C would not be discernible as both ratios B/A and C/A are roughly equal to 0.72.

3.3.2. Source–receptor analysis on synoptic timescales. To further identify regions where synoptic-scale variations in surface Rn from case B can be consistently and distinctly separated from those of cases A and C, we concentrate on 184 surface receptors. These receptors were placed between the latitude band of 25°N – 73°N and longitude bands of 10°W – 75°W (Atlantic region), 132.5°E – 180°E and 125°W – 180°W (Pacific region) as shown in Fig. 10. The size of each receptor is $7.5^\circ \times 6.0^\circ$. We sampled the simulated Rn distributions at these receptors from simulations A and B over the entire year at a time resolution of 15 min. We scaled down Rn distributions from case A at each receptor by 0.72 to generate Rn distributions for case C. Considering that regional Rn emissions have an uncertainty of a factor of 2, we selected only those synoptic events at each receptor where ratios B/A and B/C of surface Rn are less than 0.5. We performed three simulations for case B in which Rn emissions

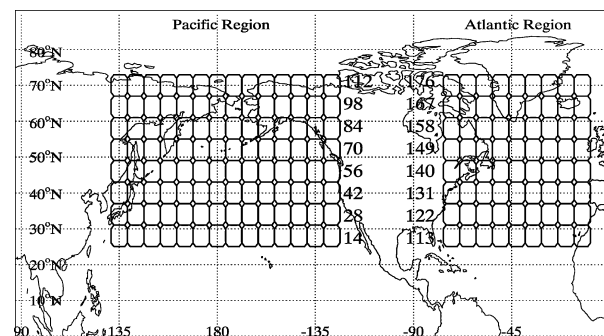


Fig 10. Geographical locations of 184 regional surface receptors placed within Pacific and Atlantic regions between 25°N and 73°N . For each region, receptors are identified by their serial number which increases from left to right and from bottom to top.

were tagged for latitudinal bands: (B1) 29°N–43°N, (B2) 43°N–57°N and (B3) 57°N–71°N. The purpose of these additional simulations is to quantify relative sizes of contributions from these latitude bands during the selected synoptic events at the receptors. Here we concentrate on simulations A, B and B3. Our analysis indicates that during March–September, on average for 44% of the time, each receptor placed between 50°N and 60°N in Pacific region experienced B/A ratios that were less than 0.5. Most of these selected synoptic events occurred when the contribution from region B3 to surface Rn from case B was more than 50%.

Figure 11 shows the percent frequency distribution of selected events each month for the (a) Pacific region and (b) the Atlantic region when the ratio B/A is less than 0.5 and the contribution from region B3 is more than 50%. Figure 12 displays similar plots for percent frequency distribution for the ratio B/C. There are several points that can be noted from these figures. First of all, for receptors numbered below 43 (i.e. below 43°N located in the Pacific region), the occurrence of the events for ratio B/A constrained by both conditions is very limited over the entire year

(Fig. 11a). But, as one moves northwards up to 55°N (i.e. for receptor numbers 43–70) this frequency increases dramatically during the months of April to October. For all these receptors, transport of air mass from higher latitudes played a major role in the displayed frequency pattern in the B/A ratio because differences in local emissions south of 55°N will not solely force this ratio to be less than 0.5. There is negligible occurrence of these events all around the year for receptors numbered below 149 located in the Atlantic region below 50°N (Fig. 11b). Above 55°N, both conditions are met more frequently for most of the year in the Pacific as well as the Atlantic regions. As should be expected due to lower concentrations of surface Rn for case C relative to those for case A, ratio B/C meets both conditions less frequently for receptors placed above 50°N for both regions (Fig. 12). Note that most of the receptors between 50°N and 60°N are placed over the oceanic surface and are free from uncertainties in *in situ* emissions. Based on the implicit consideration of the maximum uncertainty of a factor of 2 in local emissions, this analysis suggests that additional surface measurements of atmospheric Rn between 50°N and 60°N (i.e. for receptors 57–84 and 149–166) over the Pacific and Atlantic regions, mainly over the oceanic sites, during April to October would make it possible to confirm or refute the latitudinal gradient in Rn source distribution (case B). We further stress that these measurements of atmospheric Rn must conform to the same reference of reporting to

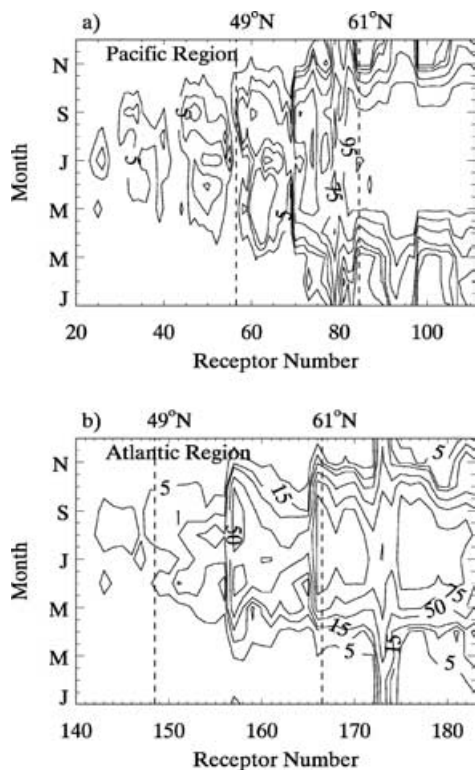


Fig 11. Monthly variations in percent frequency distribution of surface Rn events at receptors placed over (a) Pacific and (b) Atlantic regions. Contour levels on these plots are 5, 15, 25, 50, 75 and 95. These events are characterized by a ratio B/A less than 0.5 and more than 50% contribution from region B3 (57°N–71°N). Vertical dashed lines enclose a latitude band of 49°N–61°N. The latitude correspondence of surface receptors is shown in Fig. 10.

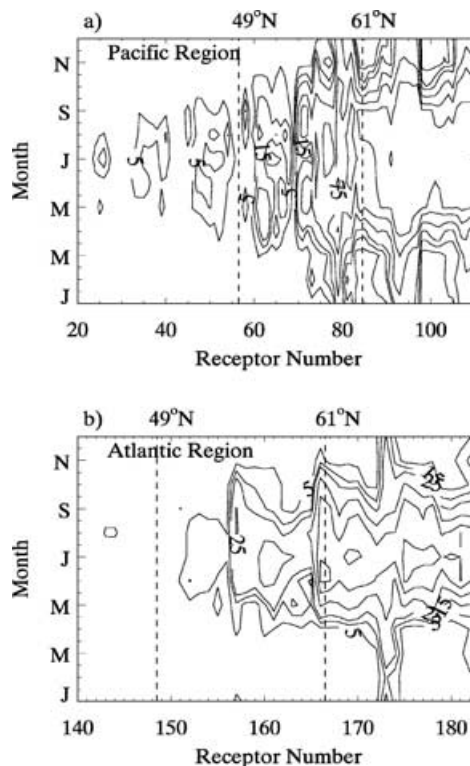


Fig 12. Same as Fig. 11 but for the per cent frequency distribution of occurrence of ratio C/A less than 0.5.

avoid any systematic biases as discussed by Colle et al. (1995) in the intercomparison study of Rn measurements. Four international laboratories participated in this intercomparison study. For ambient air samples from Bermuda, two of these laboratories showed systematic biases of 40% and 65–70% with respect to the common reference. For ambient air containing low Rn, the other two laboratories also showed significant deviations from each other (Colle et al. 1995).

In this study, we simultaneously addressed two interdependent aspects of evaluation study. First, evaluation of the treatment of physical processes and meteorological input to the model using Rn observations and its emissions, and second, use of the model to determine which emission scenario better represents an agreement between observed and simulated atmospheric Rn. For the first case, keeping uncertainties in Rn emissions and its observations in perspective, the discrepancies noted between simulated and observed Rn distributions point to a need for improvement in model processes and input variables. On the other hand, assuming that the model is capable of realistically simulating the atmospheric behaviour of Rn then the discrepancy between the simulated and observed distributions will point towards the need for better representation of emissions. It should be stressed that both of these aspects of the evaluation study guide each other in order to make further progress in model development, flux deductions and observation strategy.

4. Conclusions

We present results from a study in which simulated atmospheric distributions using three scenarios of Rn surface fluxes were compared with each other and with the observations. Comparisons indicate that, in general, the model reproduces observed surface and vertical profiles of atmospheric Rn but that there are some biases in the model related to differences in large-scale and convective transport. We show that a globally uniform land-based Rn surface flux of $1.0 \text{ atom cm}^{-2} \text{ s}^{-1}$ within $\pm 60^\circ$ (Case A) overpredicts atmospheric Rn in both hemispheres. A globally uniform surface flux of $0.72 \text{ atom cm}^{-2} \text{ s}^{-1}$ within $\pm 60^\circ$ (Case C) or a scenario in which emission decreases with latitude north of 30°N (Case B) produces atmospheric distributions of Rn that compare well with the observed Northern Hemisphere distributions but are not discernible from each other. However, case C is a better candidate for Rn emissions in the Southern Hemisphere. Considering the maximum uncertainty in regional Rn emissions of a factor of two and based on the ratios B/A and B/C of surface Rn concentration of less than 0.5 over the Pacific and Atlantic regions north of 25°N , synoptic-scale source–receptor analysis suggests that calibrated measurements of surface Rn between 50°N and 60°N during April to October would be able to validate the suggested latitudinal gradient in surface Rn emissions.

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