

DISTRIBUTION OF ^{222}Rn OVER THE NORTH PACIFIC: IMPLICATIONS FOR CONTINENTAL INFLUENCES.

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ABSTRACT. The atmospheric distribution of ^{222}Rn over the north Pacific is simulated with a three-dimensional chemical tracer model using meteorological input from the NASA-GISS general circulation model ($4^\circ \times 5^\circ$ resolution). Radon-222 (half-life 3.8 days) is a tracer of continental air. Model results are in good agreement with measurements from ships and aircraft. Strong Asian influence is found throughout the tropospheric column over the north Pacific in spring, reflecting a combination of frequent convection over the continent, strong westerly winds at altitude, and subsidence over the ocean. In summer, the upper troposphere over the north Pacific is heavily affected by deep convection over China; however, Asian influences at the surface are then at their yearly minimum. In winter, strong Asian influence is found near the surface but not at high altitudes. Transport of American air over the Pacific is important only at low latitudes. American sources account for 11% of total ^{222}Rn in the model at Midway, 30% at Mauna Loa and 59% at Oahu. Results for Hawaii indicate two seasonal peaks of American influence, one in summer and one in winter. The tropical western Pacific is particularly remote from continental influences year round.

Key words: North Pacific, global atmospheric chemistry, modeling, radon.

1. Introduction

Measurements of atmospheric composition over the north Pacific indicate a wide range of continental influences (Duce et al., 1983; Merrill et al., 1985; Prospero et al., 1985; Prospero and Savoie, 1989; Durkee et al., 1991). Understanding the origin and extent of these continental influences is important in the context of several environmental issues. Recent observations suggest that the primary productivity of the north Pacific is limited by the availability of iron transported as aerosol from the continents (Martin and Fitzwater, 1988; Young et al., 1991). Continental aerosols over the oceans provide a supply of cloud condensation nuclei, which could influence weather and increase cloud albedo (Twomey et al., 1984; Charlson et al., 1987; Durkee, 1990; Penner et al., 1991). Long-range transport of anthropogenic CO, nitrogen oxides, and ozone from the polluted continents may affect significantly the photochemistry of the atmosphere over vast expanses of the north Pacific (Chameides et al., 1987, 1989; Fishman et al., 1987).

The natural radioisotope ^{222}Rn is a particularly sensitive tracer of continental influences in marine air. Radon-222 is emitted ubiquitously from land surfaces, at a rate 2 to 3 orders of magnitude higher than from the oceans (Broecker et al., 1967; Wilkening et al., 1975). It is removed exclusively by radioactive decay (half-life 3.8 days). Concentrations of ^{222}Rn in continental boundary layers are typically 50-250 pCi/SCM, while background concentrations in marine air are less than 1 pCi/SCM (1 pCi/SCM = 6.6×10^{-22} atoms of ^{222}Rn per molecule of air). Measurements of high ^{222}Rn concentrations over the oceans thus provide an unambiguous signal of recent continental history.

We present in this paper a three-dimensional climatological simulation of the atmospheric distribution of ^{222}Rn over the north Pacific, using the Harvard/GISS chemical tracer model (CTM). Model results are evaluated with ^{222}Rn concentration data from ships and aircraft. Section 2 briefly describes the computational structure of the CTM and the simulation of ^{222}Rn emission; complete descriptions are given by Prather et al. (1987) and Jacob and Prather (1990). Comparisons of model results with data are presented in Section 3. Spatial and seasonal variations of continental influences over the north Pacific are discussed in section 4 in light of model results. Concluding remarks are in section 5.

2. Description of the Model

The CTM simulates the atmospheric distribution of ^{222}Rn by solving the three-dimensional mass conservation equation. Meteorological input is provided by a 1-year record of winds and other variables from a general circulation model (GCM) developed at the Goddard Institute of Space Studies (GISS). We use the GISS GCM II version with $4^\circ \times 5^\circ$ resolution and full diurnal cycle (Hansen et al., 1983). The CTM grid replicates that of the GCM, i.e., $4^\circ \times 5^\circ$ in the horizontal with 9 layers in the vertical extending from the surface to 10 mb along a sigma (terrain-following) coordinate. The 3 lowest layers extend approximately to 450, 1200, and 2600 m above the surface. The time step is 4 hours; meteorological quantities are updated at the end of each time step. Advection is computed with an upstream method that conserves the first- and

second-order moments of tracer concentrations (Prather, 1986). The concentrations within a grid-box are thus described by a second-order polynomial in 3 dimensions, affording some subgrid resolution. Conservation of moments has the principal advantage of reducing numerical diffusion during advection (Prather, 1986).

The treatment of dry and wet convection follows the scheme used in the GISS GCM II to transport momentum and heat (Hansen et al., 1983). When an air column is unstable with respect to the dry adiabat (dry convection), the air within that column is assumed to mix uniformly over the 4-hour time step. When an air column is unstable with respect to the moist adiabat (wet convection), 50% of the air in the lowest layer is moved directly to the highest layer in the column, with no entrainment, followed immediately by compensating subsidence in the intermediate layers. The model can in this manner simulate the rapid vertical transport of air from the lower troposphere to the upper troposphere during deep convection.

Emission of ^{222}Rn from land surfaces is simulated using the formulation of Jacob and Prather (1990). A standard emission flux of $1.0 \text{ atoms cm}^{-2} \text{ s}^{-1}$ is assumed from all unfrozen land, intermediate between two global average estimates of $0.75 \text{ atoms cm}^{-2} \text{ s}^{-1}$ (Wilkening et al., 1975) and $1.2 \text{ atoms cm}^{-2} \text{ s}^{-1}$ (Turekian et al., 1977). This standard flux is reduced by a factor 3 under freezing conditions, based on data from George and Breslin (1979) and George (1980, 1981). It is further modified by changes in surface pressure following Clements and Wilkening (1974) and Edward and Bates (1980). The pressure dependence of ^{222}Rn emission induces some day-to-day variability in the simulated boundary layer concentrations over the continents; it has negligible effect over the oceans.

Emission of ^{222}Rn from oceans and ice packs is ignored. Oceanic emission could possibly account for a background concentration of 0.5-1.0 pCi/SCM in marine surface air (Lambert et al., 1982), but our interest here is in examining the elevations over this background resulting from continental influences. A test simulation was conducted including a uniform oceanic emission flux of $5 \times 10^{-3} \text{ atoms cm}^{-2} \text{ s}^{-1}$, which can be considered an upper limit (Wilkening and Clements, 1975; Lambert et al., 1982). This simulation produced marine background concentrations of 0.9-1.8 pCi/SCM at the surface, decreasing rapidly with altitude. The continental enhancements discussed below are usually larger.

The distribution of ^{222}Rn over the north Pacific reflects mainly influences from Asia and America, owing to the short lifetime of the gas. Figure 1 shows the model domain used in our simulations; fixed background concentrations of 0.15 pCi/SCM are assumed as boundary conditions at the edges. The evolution of ^{222}Rn concentrations is computed for 1 model year, following 1 month of initialization. The choice of model domain was tested by comparison with a 2-month simulation using the global CTM grid. We found no significant differences in the simulated concentrations over the north Pacific.

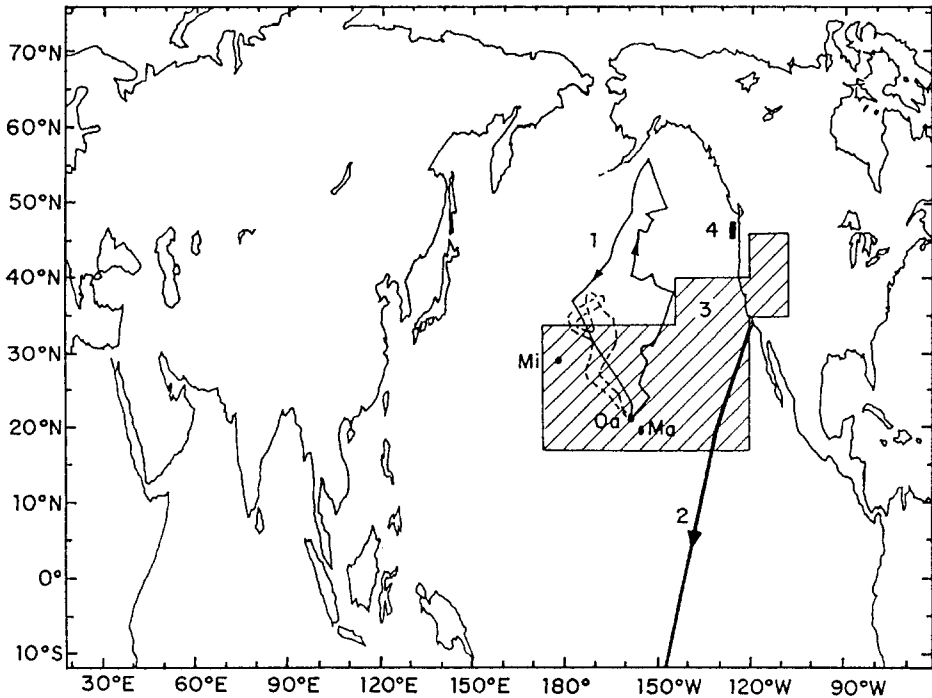


Figure 1. Model domain used in the simulation of ^{222}Rn over the north Pacific. The edges of the figure represent the boundaries of the domain, where fixed background concentrations (0.15 pCi/SCM) are assumed. The locations of the ^{222}Rn measurements discussed in the text are identified by numbers. (1) Two ship cruises north of Hawaii in May 1986 (solid line) and in June-July 1986 (dashed line). (2) Ship cruise from California to Antarctica in November 1972 (thick line). (3) Aircraft flights at 9.4-12.5 km between California and Hawaii in July-August of 1983 and 1984 (hatched area). (4) Aircraft flights at 0.1-3.6 km off the northwest coast of the United States in May 1985 (thick line). References for the data are: (1) Atlas (1988); (2) Wilk-niss et al. (1974); (3) Kritz et al. (1990); (4) Andreae et al. (1988). The locations of Midway (Mi), Oahu (Oa), and Mauna Loa (Ma) are shown.

3. Comparison of Model Results with Observations

3.1. CRUISES BETWEEN HAWAII AND ALASKA

Concentrations of ^{222}Rn were measured aboard the University of Hawaii research vessel Moana Wave cruising between Hawaii and Alaska in May-July 1986. The measurements were made by R. Arimoto and colleagues from the University of Rhode Island, and have been reported by Atlas (1988). The aerosol ^{222}Rn daughters were collected on glass fiber filters, and the β -activity of the filters was counted (Larson and Bressan, 1978). The detection limit was 0.2 pCi/SCM and the uncertainty was 15-20%. Samples were collected twice a day (06 and 18 local time) at the bow of the ship, 7 m above deck.

Figure 2 compares the simulated and observed time series of concentrations. The simulated time series was constructed by sampling model output twice a day along the path of the cruise, on the days of observations. A prominent feature of the observations is the occurrence of episodes during which concentrations are elevated sharply relative to background. The model captures well the occurrence of these episodes, at least in a qualitative way. The general decrease of concentrations from May to July, both in the model and in the observations, reflects a seasonal trend in continental influence over the north Pacific. This trend is discussed further in Section 4.

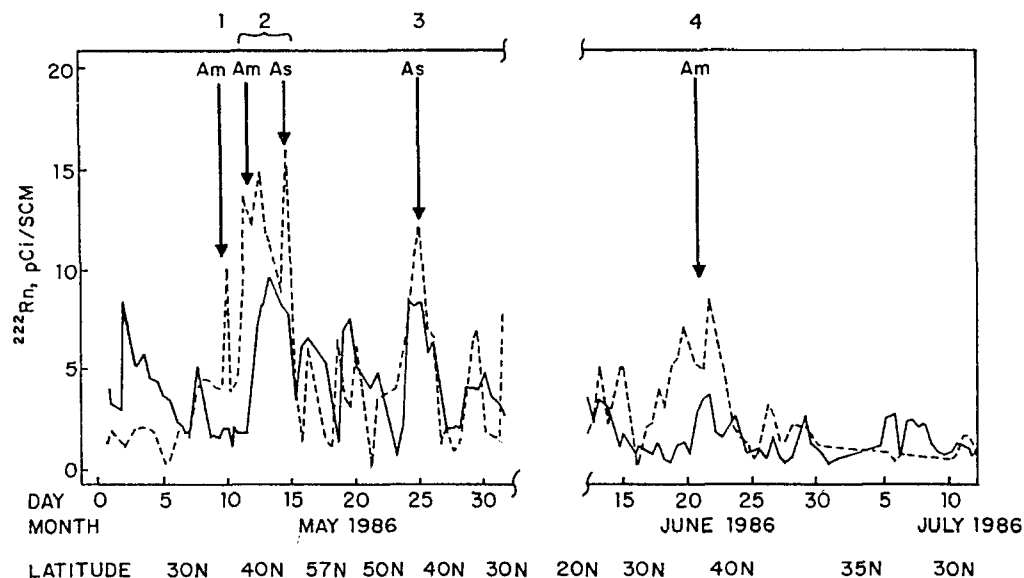


Figure 2. Concentrations of ^{222}Rn along two cruise tracks north of Hawaii in May-July 1986 (see Figure 1 for location of cruise tracks). The abscissa gives dates and latitudes. Model results (dashed line) are compared to observations (solid line). The measurements were made every 12 hours. The model time series was constructed by sampling output along the path of the cruise every 12 hours, for the days of observations. Arrows identify the 5 model episodes (see text). The labels "As" or "Am" for each episode indicate a dominant Asian or American origin.

The GISS GCM simulates a typical meteorological year, not necessarily 1986; therefore the timing of high- ^{222}Rn episodes in the model should not be expected to match precisely the observations. We can however evaluate model results in a quantitative way by examining some statistical features of the time series. Figure 3 shows the cumulative probability distributions of the observed and simulated concentrations. The slope of the ^{222}Rn distribution increases sharply above the median, both in the model and in the observations, reflecting the bimodal character of the distributions. The lower mode represents background conditions and the upper mode represents the high- ^{222}Rn episodes. We find that the model reproduces well most of the observed distribution; the high tail is overpredicted, largely because of the strong model episode on 11-14 May.

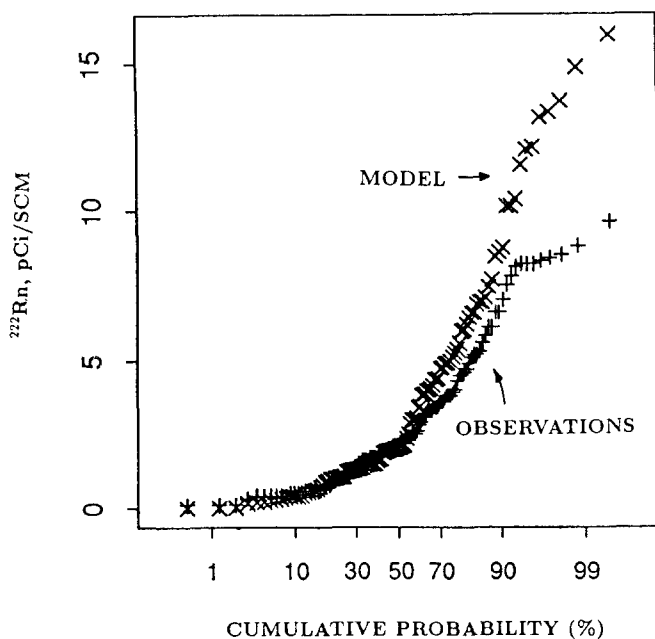


Figure 3. Cumulative probability distributions of ^{222}Rn concentrations in the simulated and observed time series of Figure 2. The abscissa in this Figure is scaled such that a normal distribution plots as a straight line (probability axis). Both time series contain 118 points.

We can also compare the numbers and durations of episodes in the two time series. Here we define an episode as the continuous occurrence of ^{222}Rn concentrations at least twice higher than the mean of the time series. Both the observed and simulated time series include 4 such episodes, lasting up to 2 days in the observations and up to 4 days in the model. The model episodes are identified with numbers in Figure 2, along with their origins (Asian or American); these origins

were established in a sensitivity simulation with the American source shut off. We find that Asian and American influences do not overlap. The 11-14 May episode actually represents a rapid succession of two episodes, one American (11-13 May) and one Asian (14 May).

The model thus simulates 5 distinct episodes along the path of the cruise: 2 are of Asian origin and 3 are of American origin. Both Asian episodes are caused by air transported from northern Asia in the middle troposphere and subsiding over the path of the cruise. This transport is illustrated in Figure 4, which shows surface concentrations and areas of subsidence on May 14 (for the sensitivity simulation with the American source shut off). The 3 American episodes in the model are all due to direct boundary layer transport of air from Alaska in transient weather systems, as illustrated in Figure 5 for the 19 June episode. We find that significant Alaskan influence in the boundary layer over the Pacific can extend as far south as 30°N during the summer months.

3.2. CRUISE FROM CALIFORNIA TO ANTARCTICA

Concentrations of ^{222}Rn in surface air were measured by Wilkniss et al. (1974) during a cruise from California to Antarctica in November-December 1972. The measurement procedure was that described in Section 3.1. Figure 6 compares model results with observations. Excellent agreement is found; the model captures well the sharp concentration decrease as the ship moves south from 20°N to 10°N , and the low concentrations south of 10°N . Figure 7 shows the mean surface concentrations simulated over the north Pacific in November. We see that the decrease in concentration from 20°N to 10°N reflects the movement of the ship out of the American plume.

3.3. AIRCRAFT MEASUREMENTS BETWEEN HAWAII AND ALASKA (9.4-12.5 KM ALTITUDE)

Kritz et al. (1990) measured ^{222}Rn concentrations from an aircraft flying at 9.4-12.5 km altitude (300-200 mb) between Hawaii and California. Measurements were made on 13 different days in July-August 1983 and 1984. The aerosol ^{222}Rn daughters were collected on paper filters, using an aerodynamic inlet and isokinetic sampling, and the α -radioactivity of the filters was counted. The detection limit was 1 pCi/SCM and the measurement uncertainty was 20%. The observed concentrations were bimodally distributed, with one mode centered below 1 pCi/SCM and the other centered at 11 pCi/SCM; the median concentration was 4 pCi/SCM, remarkably high for that altitude. Concentrations measured only 150 to 300 km apart during a given flight often differed dramatically, indicating that the continental influences were concentrated in narrow plumes.

Figure 8 shows the means and standard deviations of ^{222}Rn concentrations simulated by the model at 200 mb in July-August, as a function of latitude and for the region sampled by Kritz et al. (1990). Also shown in Figure 8 are the individual data of Kritz et al. (1990), and separate averages of the 1983 and 1984 data over the 4° latitude bands of the CTM. Concentrations in the model average 4 pCi/SCM north of 25°N , reasonably consistent with the observations and indicating substantial continental influence in the upper troposphere. The small standard deviations of concentrations in the model (≈ 1 pCi/SCM) indicate excessive averaging of continental influences; the episodic structure of the observations is not simulated.

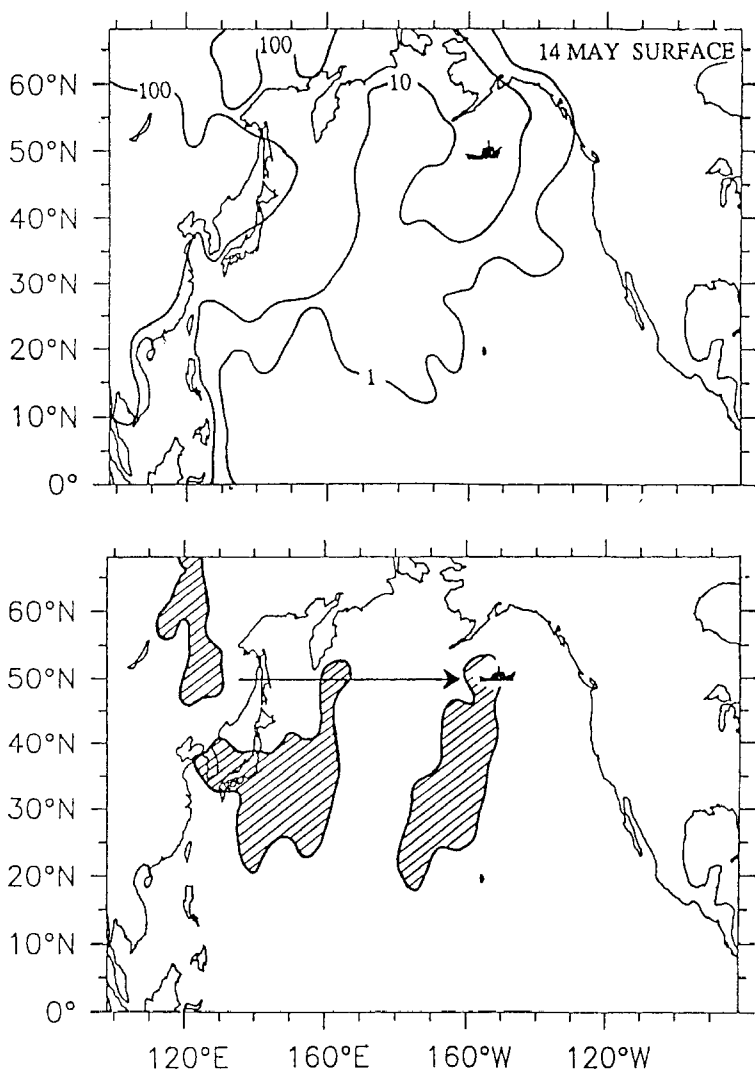


Figure 4. Model results on 14 May for the simulation with the American source shut off. The top panel shows the 24-hour average surface concentrations of ^{222}Rn (pCi/SCM). The location of the cruise ship is also shown. The shaded areas in the bottom panel indicate regions of strong ^{222}Rn subsidence (24-hour average downward flux $> 1 \times 10^{-2} \text{ atoms cm}^{-2} \text{ s}^{-1}$ at 720 mb). The arrow in the bottom panel indicates the origin of the ^{222}Rn subsiding over the location of the ship.

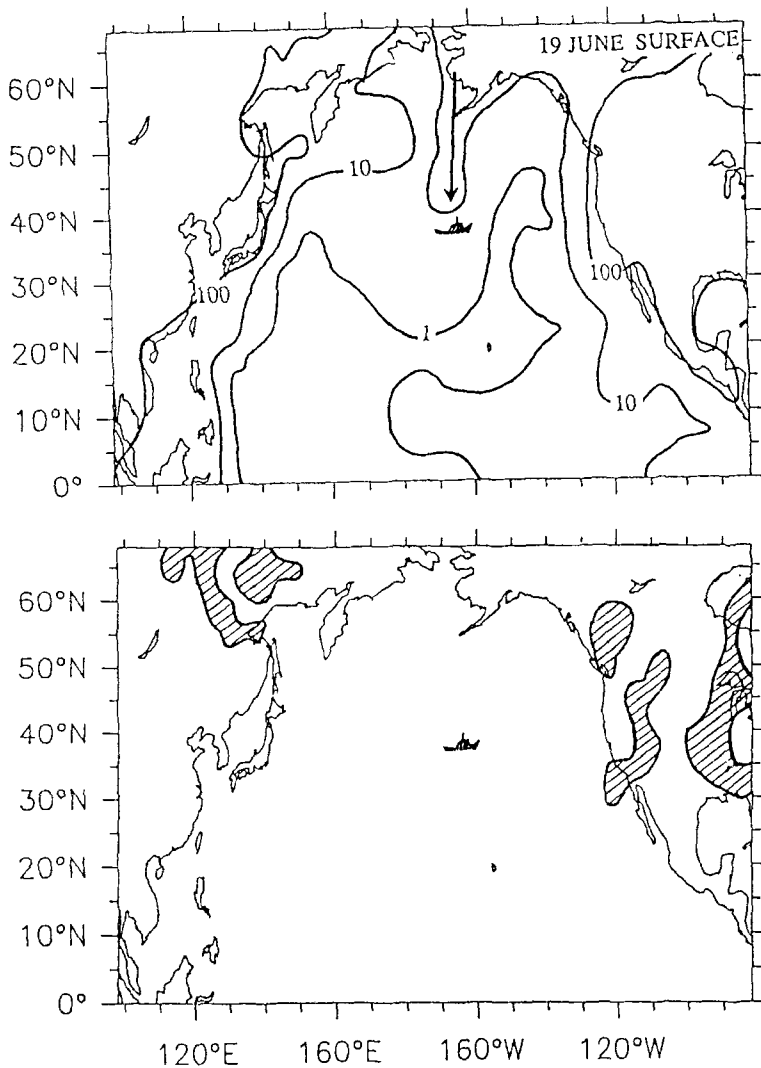


Figure 5. Model results on 19 June. The top panel shows the 24-hour average surface concentrations of ^{222}Rn (pCi/SCM). The location of the cruise ship is also shown. The shaded areas in the bottom panel indicate regions of strong ^{222}Rn subsidence (24-hour average downward flux $> 1 \times 10^{-2}$ atoms $\text{cm}^{-2} \text{s}^{-1}$ at 720 mb). We see that the high concentration of ^{222}Rn at the location of the ship is not due to subsidence of continental air, but rather to direct boundary layer transport of air from Alaska (arrow in the top panel).

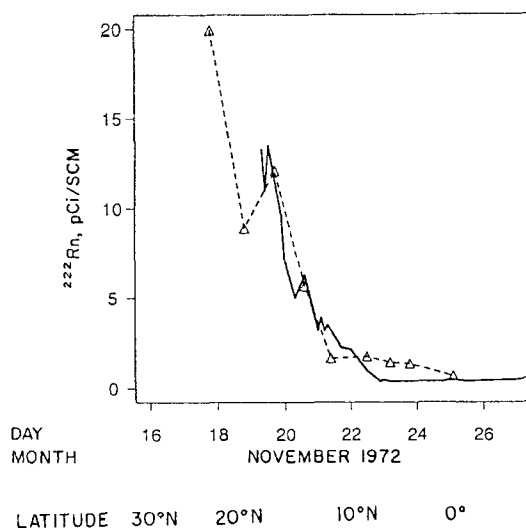


Figure 6. Concentrations of ^{222}Rn along a cruise track from California to Antarctica in November 1972 (see Figure 1 for location of cruise track). The abscissa gives both dates and latitudes. Model results (dashed line) are compared to observations (solid line). The model results are monthly means for gridboxes along the path of the cruise.

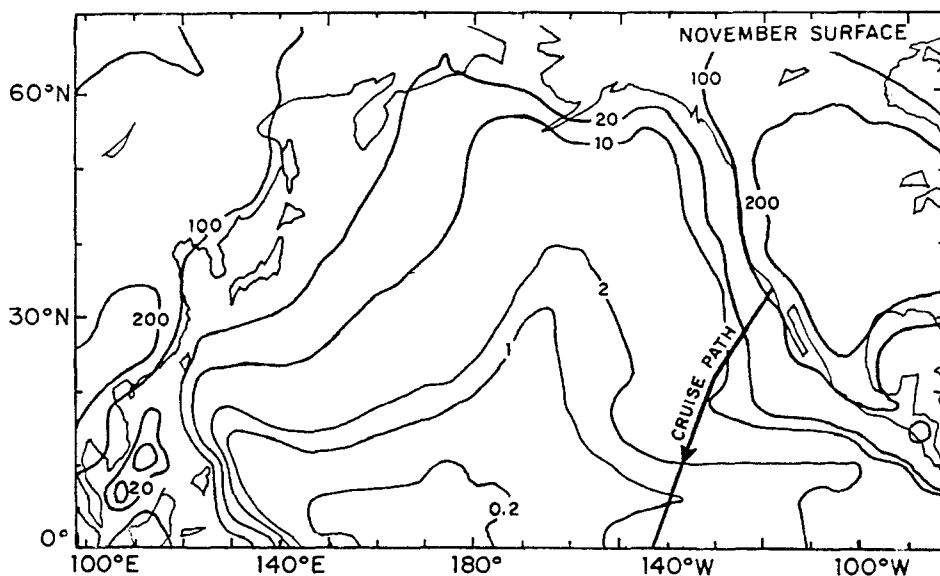


Figure 7. Surface concentrations of ^{222}Rn (pCi/SCM) simulated by the model for November. Values are monthly means. The Wilkniss et al. [1974] cruise track is shown.

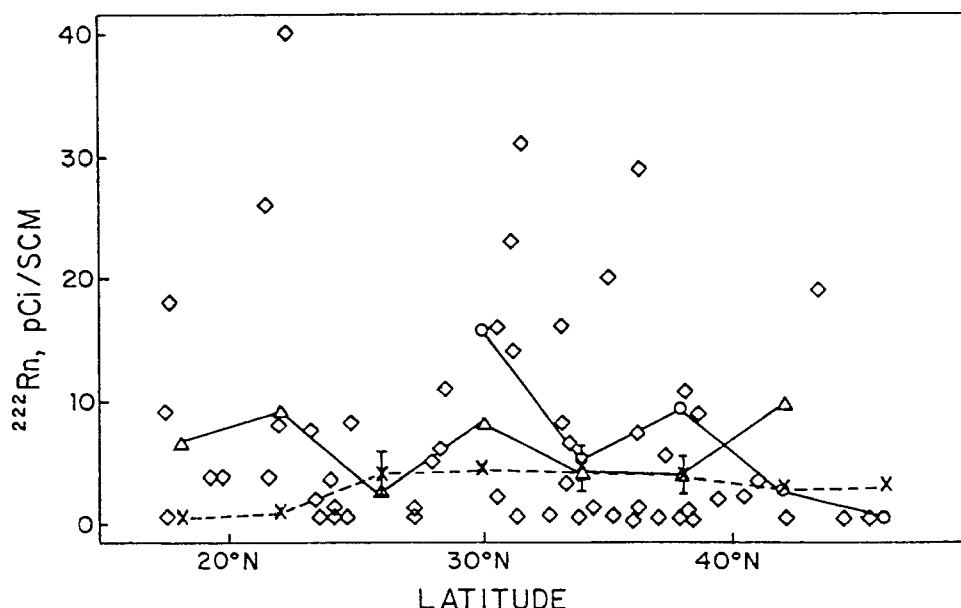


Figure 8. Concentrations of ^{222}Rn at 300-200 mb over the northeast Pacific in July-August, as a function of latitude. The 1983-1984 measurements of Kritz et al. (1990) are shown as diamonds (see Figure 1 for location of flight tracks); the solid lines are average of these observations over the 4° latitudinal bands of the CTM, separately for 1983 (open circles) and 1984 (open triangles). The dashed line shows the mean concentrations simulated by the model in the Kritz et al. (1990) flight region, along with representative standard deviations.

The main origin of the upper troposphere ^{222}Rn in the model is deep wet convection over western and southeastern China, followed by fast transport at high altitudes in the westerly jet. This mechanism is consistent with back-trajectories constructed by Kritz et al. (1990) for two of their observed high- ^{222}Rn episodes. Figure 9 shows the mean distribution of ^{222}Rn at 200 mb in the model in July, along with the corresponding 200 mb geopotential heights in the GCM and in the climatological data. We find that ^{222}Rn concentrations at 200 mb exceed 5 pCi/SCM over much of the north Pacific; the region of maximum concentrations reflects the location of the westerly jet. The strength of the jet in the GCM reproduces well the climatological observations, though the center of the jet is $\approx 5^\circ$ too far south. Further examination of GCM geopotential heights for successive days in July indicates that the location of the GCM jet does not vary significantly from day to day. The meandering of the jet in the observations, which appears to account for much of the variability in the observed ^{222}Rn concentrations (Kritz et al., 1990), is not simulated.

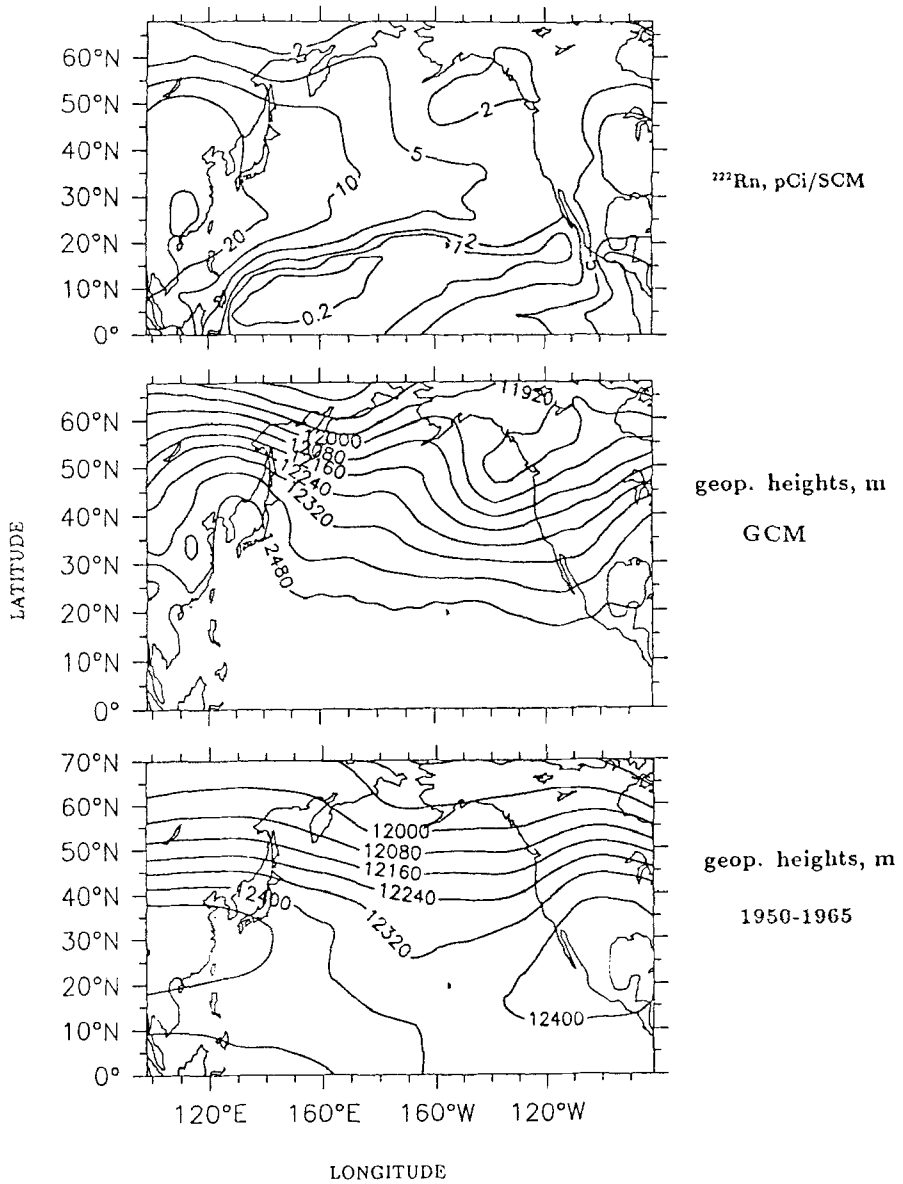


Figure 9. Top panel: simulated concentrations of ^{222}Rn (pCi/SCM) at 200 mb in July. Middle panel: 200 mb geopotential heights (m) in the GCM in July. Bottom panel: observed 200 mb geopotential heights (m) in July (average of 1950-1965 data; Crutcher and Minerve, 1970). All values are monthly means.

3.4. AIRCRAFT PROFILES OVER THE NORTHEAST PACIFIC (0.1-3.6 KM ALTITUDE)

Andreae et al. (1988) measured 3 vertical profiles of ^{222}Rn concentrations at 0.1-3.6 km altitude (1000-650 mb) off the northwest Pacific coast of the United States in May 1985. The measurements were made 50 km from the coast at approximately 47°N , 125°W (Figure 1). The flight days were chosen to minimize local continental influences, and to maximize influences from the open ocean to the west. The measurement technique was that described in section 3.3. The observed concentrations ranged from 4 to 10 pCi/SCM, substantially above marine background in the entire column up to 650 mb (Figure 10, right panel). The authors argued on the basis of back-trajectory analyses that the ^{222}Rn had originated from Asia, with transit times of 4-8 days in the middle troposphere.

Figure 10 (top left panel) shows the mean simulated ^{222}Rn concentrations at 46°N in May, as a function of longitude and altitude. The concentrations at the longitude of the flights exceed 10 pCi/SCM in the lower troposphere, substantially higher than observed; these high values reflect at least in part the coarse model resolution. The model grid square corresponding to the observations is partly land (Washington and Oregon), and thus receives emissions which mix horizontally over the scale of the gridbox. In addition, numerical diffusion during advection can affect significantly the ^{222}Rn concentrations as far as two gridboxes (10°) away from the coast (Prather, 1986; Balkanski and Jacob, 1990). The model cannot therefore resolve North American influences at the location of the Andreae et al. (1988) flights.

We can however examine the extent of Asian influence by considering results from the sensitivity simulation with the American source shut off. These results are shown in the bottom panel of Figure 10. We find that Asian sources contribute on average less than 2 pCi/SCM to the column at 125°W in May, much lower than the concentrations measured by Andreae et al. (1988). Further inspection of model results indicates that Asian sources never contribute more than 3 pCi/SCM to the column on any 4-hour period during May. It is possible that the model does not resolve episodes of particularly high Asian influence in the region, as previously discussed in our analysis of the Kritz et al. (1990) data. It is however also possible that the ^{222}Rn concentrations measured by Andreae et al. (1988) were due in part to American sources not identified in the back-trajectory analyses.

4. Discussion

The results of Section 3 show that the model provides in general a good simulation of observed ^{222}Rn concentrations over the north Pacific. Unfortunately, there are no long-term time series of observed ^{222}Rn concentrations that may be used to test the simulated seasonal trends. Such time series are however available for continental aerosols at island sites. We compare in Figure 11 the simulated seasonal trends of ^{222}Rn concentrations at Midway, Oahu, and Mauna Loa (see Figure 1 for locations) to observations for nitrate and ^{210}Pb (Feely et al., 1988; Larsen and Sanderson, 1990; Savoie et al., 1990; Turekian et al., 1990). Nitrate over the north Pacific is dominantly of combustion origin (Levy and Moxim, 1989). We find that the model simulates successfully the springtime maximum of Asian influence documented by the aerosol data (Uematsu et al., 1983; Merrill, 1990). The model further indicates minimum Asian influence in summer, consistent with the low nitrate and ^{210}Pb concentrations observed during that season at

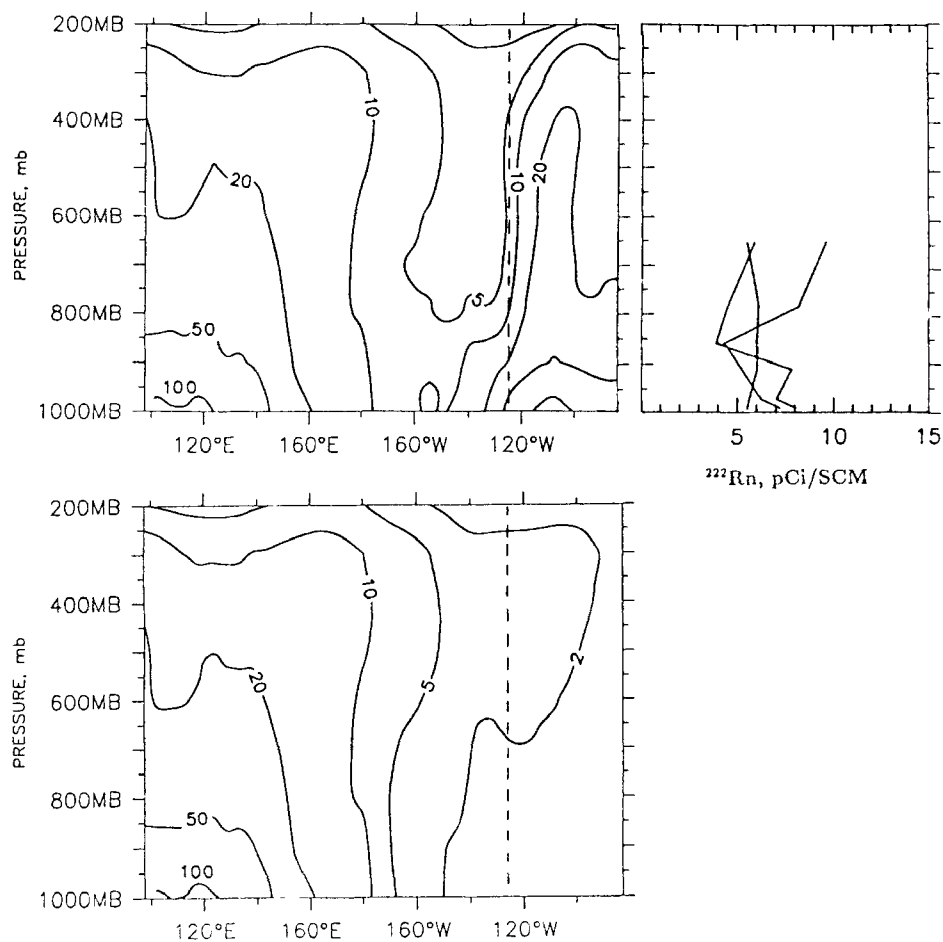


Figure 10. Vertical profiles of ^{222}Rn concentrations over the north Pacific in May. The right panel shows data from 3 flights at 47°N , 125°W extending up to 650 mb (Andreae et al., 1988). The left panels show monthly mean longitude-altitude cross-sections at 46°N in the standard model simulation (top) and in the sensitivity simulation with the American source shut off (bottom). The dashed lines in the left panels indicate the longitude of the Andreae et al. (1988) flights.

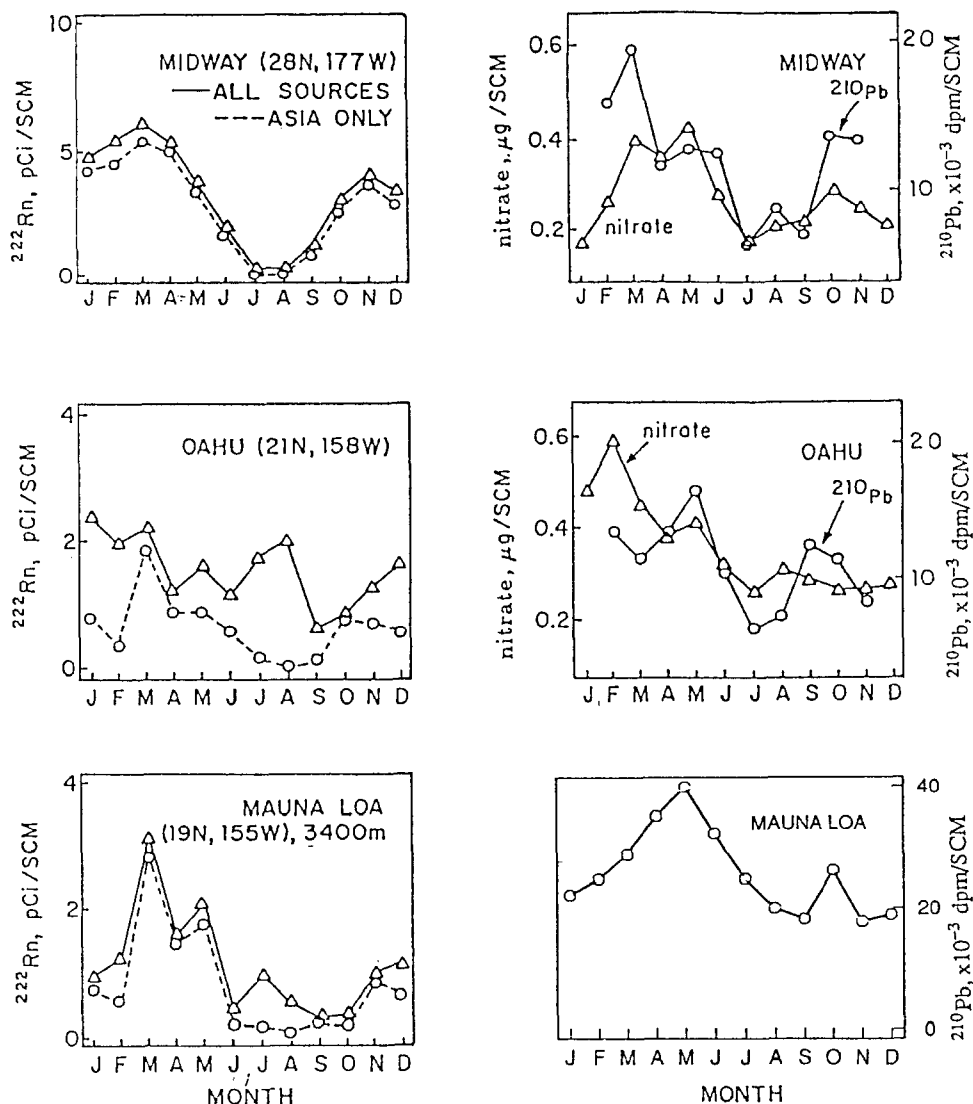


Figure 11. Seasonal variations of continental tracer concentrations at Midway, Oahu, and Mauna Loa. Model results for ^{222}Rn are shown in the left panels, with Asian contributions indicated by the dashed lines. Observations for nitrate and ^{210}Pb are shown in the right panels. Each point represents a monthly mean value. The nitrate observations at Midway and Oahu include 7 years of data from 1981 to 1987 (Savoie et al., 1990). The ^{210}Pb observations at Midway and Oahu include 10 months of data from February to November 1981 (Turekian et al., 1990). The ^{210}Pb observations at Mauna Loa include 6 years of data from 1983 to 1988 (Feely et al., 1988; Larsen and Sanderson, 1990).

Midway. At Oahu and Mauna Loa, the summertime minima of Asian influence are partly offset by efficient transport of American air.

Figure 12 (top panel) shows the longitude-altitude cross-sections of simulated ^{222}Rn concentrations at 34°N , for March and July. We find that Asian influences over the north Pacific in March extend throughout the tropospheric column, due to a combination of frequent convection over Asia and strong westerlies. The continental air advected to the Pacific in the middle troposphere subsides in subtropical high-pressure regions (Figure 12, bottom panel), producing the high concentrations at the surface. Such a mechanism for transport has been previously invoked by J. T. Merrill (Merrill et al., 1985; Merrill, 1990) to explain episodes of high continental aerosol concentrations at north Pacific island sites. A similar mechanism also operates in the model in autumn, but with less intensity.

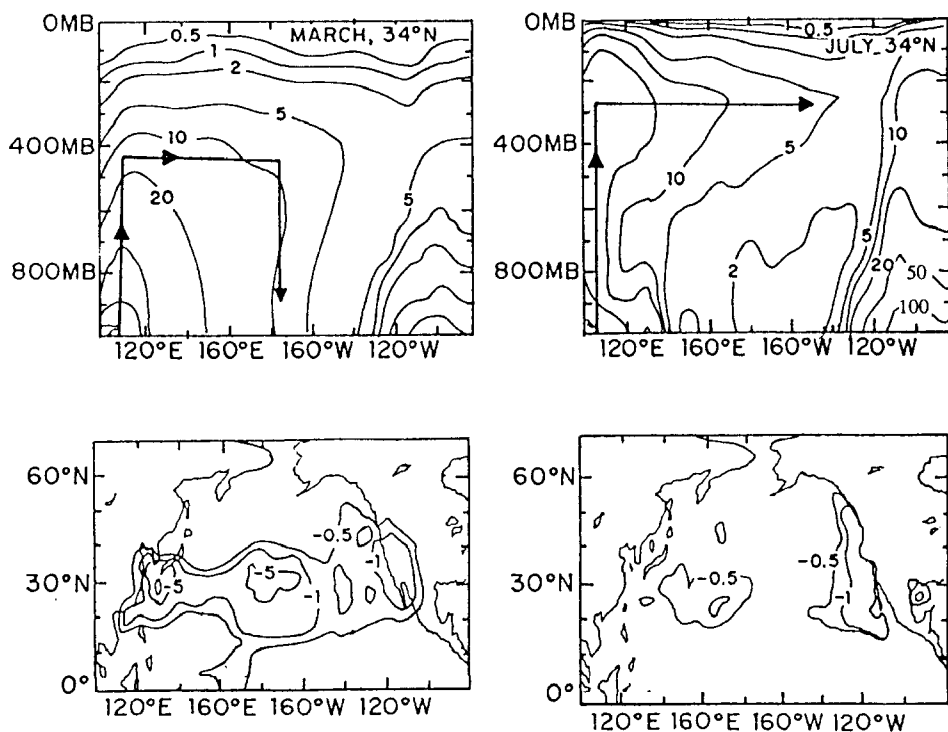


Figure 12. Monthly mean model results for March and July. Top panels: longitude-altitude cross-sections of ^{222}Rn concentrations (pCi/SCM) at 34°N . The arrows illustrate the general direction of the ^{222}Rn flow from Asia. Bottom panels: subsidence fluxes of ^{222}Rn at 720 mb. The flux contours are in units of $1 \times 10^{-2} \text{ atoms cm}^{-2} \text{ s}^{-1}$, and only negative (downward) values are shown.

Asian influences over the north Pacific in summer are largely confined to the upper troposphere above 500 mb; this is illustrated in Figure 12 by the cross-section of ^{222}Rn concentrations at 34°N in July. The high ^{222}Rn concentrations in the upper troposphere are due to deep convection over southern Asia, as discussed above in Section 3.1. Figure 13 shows the mean ^{222}Rn concentrations at 468 mb in July, indicating particularly extensive Asian influence over both the north Pacific and the Arctic. High-altitude transport of Asian air to the Arctic in summer has been documented by aircraft halocarbon measurements during the ABLE-3A expedition to Alaska (Blake et al., 1991). Figure 13 also shows the surface concentrations of ^{222}Rn in July, which average less than 1 pCi/SCM over much of the north Pacific. Such concentrations, 2 orders of magnitude lower than over the continents, identify air masses that have received no continental input for the past several weeks. The lack of Asian influence at the surface of the north Pacific in summer reflects a combination of frequent convection over the continent, ventilating the boundary layer, and weak westerlies in the lower troposphere.

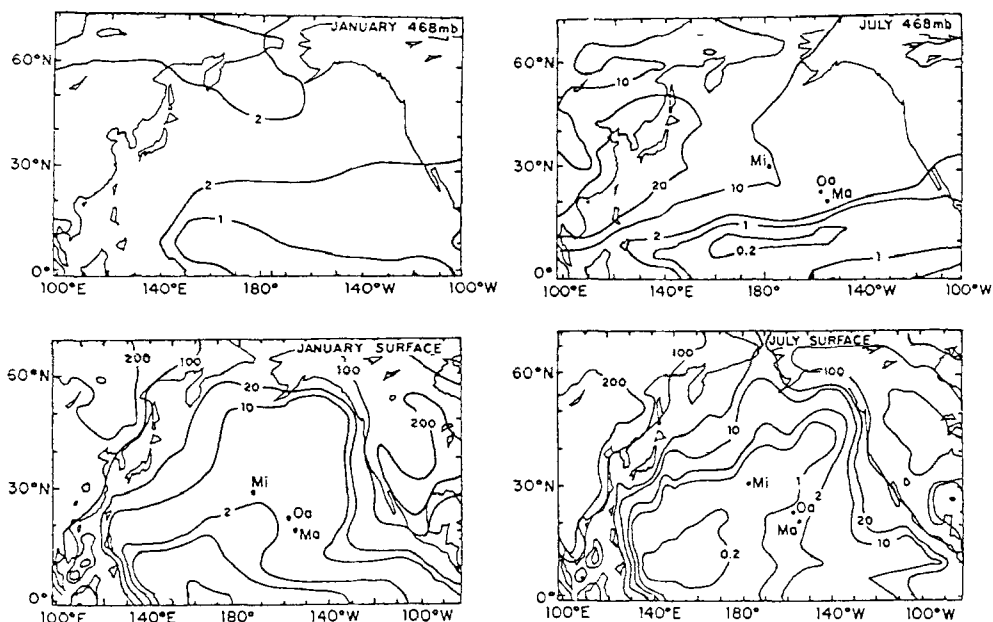


Figure 13. Concentrations of ^{222}Rn (pCi/SCM) simulated by the model in surface air and at 468 mb (6.0 km), for January and July. Values are monthly means. The locations of Midway (Mi), Oahu (Oa) and Mauna Loa (Ma) are indicated.

Model results in January (Figure 13) indicate substantial continental influence at the surface of the north Pacific but not at high altitudes. Convective activity is infrequent during winter, and transport of continental air over the ocean takes place therefore mainly in the boundary layer. We find relatively high wintertime concentrations of ^{222}Rn at Midway, apparently at odds with the observation of low nitrate concentrations (Figure 11). A possible explanation is that continental

aerosols are efficiently scavenged by deposition during boundary layer transport. In spring and autumn, transport from Asia takes place in the middle troposphere where aerosol residence times are much longer than in the boundary layer (Giorgi and Chameides, 1986; Balkanski, 1991).

Transport of American air to the north Pacific in the model is confined mainly to the low latitudes, with the exception of southward incursions of Alaskan air in summer (as discussed in Section 3.1). On a yearly average basis we find that American sources contribute only 11% of ^{222}Rn concentrations at Midway, but 30% at Mauna Loa and 59% at Oahu (Figure 11). American influences at Mauna Loa and Oahu peak in summer and winter, out of phase with Asian influences which peak in spring. The summertime transport of American air to Hawaii is initiated by dry convection over the southwestern U.S. and northern Mexico, pumping continental air to the middle troposphere; this air then subsides around the Pacific High (see subsidence fluxes in Figure 12) and is advected westward in the trade winds. Such a transport mechanism has previously been proposed by Moxim (1990) to explain observed summertime episodes of high nitrate concentrations at Mauna Loa (Galasyn et al., 1987). In winter we find that the transport of American air to Hawaii proceeds directly in the boundary layer, due to particularly strong trade winds off the coast of central America. Climatological observations indeed indicate a winter maximum for trade winds in that region (Hastenrath and Lamb, 1977). Rapid transport of American air to Hawaii could possibly explain the winter maximum of nitrate concentrations observed at Oahu (Figure 11). The stronger American influence at Oahu than at Mauna Loa reflects the dominance of boundary layer transport in supplying American air to the Hawaiian islands.

Figure 13 indicates that the western tropical Pacific is particularly remote from continental influence, at all times of year and throughout the tropospheric column (see also Figure 9). This region is in the trade winds regime but lies far downwind of the American continent. Winds at high altitude are weak (Figure 9). The western tropical Pacific is known to contain extremely low concentrations of tropospheric ozone, < 5 ppb near the surface and 20-30 ppb in the upper troposphere (Newell and Wu, 1985; Fishman et al., 1990; Johnson et al., 1990). Our model results suggest that these low ozone concentrations could possibly reflect a lack of continental influence in the region; such an argument would imply that ozone concentrations elsewhere over the north Pacific are elevated by long-range transport of anthropogenic pollution. Aircraft observations of ozone concentrations over the northeast Pacific in spring and autumn indeed show evidence of significant anthropogenic perturbation (Chameides et al., 1987, 1989; Fishman et al., 1987).

5. Conclusions

The distribution of ^{222}Rn over the north Pacific was simulated with a 3-d model based on meteorological information from the GISS general circulation model. Model results are generally in good agreement with observations. We find that strong Asian influences extend throughout the tropospheric column over the north Pacific in spring, reflecting a combination of frequent convection over the continent, strong westerly winds, and efficient subsidence over the ocean. In summer, the upper troposphere over the north Pacific is heavily affected by deep convection over China followed by fast transport in the westerly jet; however, Asian influences at the surface are then at their yearly minimum. In winter, rapid transport of Asian air to the north Pacific takes place in the boundary layer but not at higher altitudes. Measurements of aerosol concentrations at north Pacific island sites indicate little Asian influence in winter, possibly because aerosols are

efficiently scavenged during boundary layer transport.

American influences over the north Pacific are largely confined to the low latitudes. Results for Hawaii indicate two seasonal peaks of American influence, one in summer and one in winter, reflecting two different transport mechanisms in the model. Transport from America to Hawaii in summer proceeds by dry convection over the continent, followed by subsidence around the Pacific High and transport in the trade winds, as previously suggested by Moxim (1990). Winter-time transport takes place directly in the boundary layer, and is promoted by particularly strong trade winds off the coast of Central America. American influences may thus be responsible for the observed winter maximum of nitrate concentrations at Oahu.

The western tropical Pacific stands out in the model as a region particularly remote from continental influence, year round and throughout the tropospheric column. This region is known to contain extremely low concentrations of tropospheric ozone. Our results suggest that the low ozone concentrations could possibly be explained by a lack of continental influence; this explanation would imply that ozone concentrations elsewhere over the north Pacific are significantly affected by long-range transport of continental pollution.

Acknowledgments

We thank C. M. Spivakovsky and S. Wofsy for useful comments. This work was supported by the National Science Foundation (grants ATM87-19224, ATM89-21119, OCE84-05605 and OCE87-15955, plus computational resources from the National Center for Atmospheric Research).

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