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Model study of atmospheric transport using carbon 14 and strontium 90 as inert tracers

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Abstract. The observed excess carbon 14 in the atmosphere from 1963 to 1970 provides unique, but limited, data up to an altitude of about 35 km for testing the air motions calculated by 11 multidimensional atmospheric models. Strontium 90 measurements in the atmosphere from 1964 to mid-1967 provide data that have more latitude coverage than those of carbon 14 and are useful for testing combined models of air motions and aerosol settling. Model calculations for carbon 14 begin at October 1963, 9 months after the conclusion of the nuclear bomb tests; the initial conditions for the calculations are derived by three methods, each of which agrees fairly well with measured carbon 14 in October 1963 and each of which has widely different values in regions of the stratosphere where there were no carbon 14 measurements. The model results are compared to the stratospheric measurements, not as if the observed data were absolute standards, but in an effort to obtain new insight about the models and about the atmosphere. The measured carbon 14 vertical profiles at 31°N are qualitatively different from all of the models; the measured vertical profiles show a maximum mixing ratio in the altitude range of 20 to 25 km from October 1963 through July 1966, but all modeled profiles show mixing ratio maxima that increase in altitude from 20 km in October 1963 to greater than 40 km by April 1966. Both carbon 14 and strontium 90 data indicate that the models differ substantially among themselves with respect to stratosphere-troposphere exchange rate, but the modeled carbon 14 stratospheric residence times indicate that differences among the models are small with respect to transport rate between the middle stratosphere and the lower stratosphere. Strontium 90 data indicate that aerosol settling is important up to at least 35 km altitude. Relative to the measurements, about three quarters of the models transport carbon 14 from the lower stratosphere to the troposphere too rapidly, and all models with all three sets of initial conditions appear to sweep carbon 14 out of the midstratosphere (above 28 km) much more slowly than was observed from 1963 to 1970. Atmospheric dynamicists should seriously consider what the carbon 14 measurements imply with respect to advection in the midstratosphere.

1. Introduction

1.1. Background

The Atomic Energy Commission (AEC), Department of Defense, and National Oceanographic and Atmospheric Administration carried out extensive measurements of radioactive products of atmospheric nuclear bomb tests as a high-priority, high-technology activity. These tracers were produced by the above-ground nuclear bomb tests of 1952–1958 and 1961–1962. From 1953 to July 1969, aircraft collected whole air samples at many latitudes and up to pressure altitudes of 20 km, and balloons collected whole air samples from 20 km up to about 35 km. The whole air samples were brought back to laboratories for analysis of carbon 14 and several other radioactive species: zirconium, strontium, cesium, etc. Strontium 90 is produced only by fission reactions, and carbon 14 is formed by nuclear reaction of atmospheric nitrogen with neutrons, which are produced by fission and fusion.

Balloons collected whole air samples for carbon 14 analysis at several latitudes from 1953 to 1959 but only at 31°N from 1959 to July 1966 [*Hagemann et al.*, 1965, 1966; *Telegadas*, 1967, 1971]. After termination of the high-altitude collection of carbon 14 in whole air samples in 1966, AEC developed a molecular sieve collector specifically for carbon 14 in carbon dioxide. This detector was tested in the AEC high-altitude test chamber under realistic stratospheric

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Table 1. Bomb Cloud Stabilization Heights of Large Nuclear Bombs for September 1961 to December 1962

Country	TNT Equivalent, Mt*	Firing Dates	Latitude, °N	Bomb Rise Altitude Range, km
USSR	98	Aug. 2 to Sept. 4, 1961	75	8–33
United States	23	May 2 to July 11, 1962	2	15–24
USSR	188	Aug. 5 to Dec. 25, 1962	75	8–30
United States	13	Sept. 18 and Sept. 30, 1962	17	15–30

*One megaton equals 10^9 kg of TNT.

conditions and tested in the stratosphere with a series of balloon launches at 31°N latitude in 1968 and 1969. Molecular sieve instruments gave carbon 14 values in agreement with values found by the method of whole air sampling. AEC conducted an atmospheric research program during 1970–1971 with 84 balloon launches from six latitudes: 65°N, 42°N, 31°N, 9°N, 23°S, and 34°S [*Telegadas*, 1971; *Telegadas et al.*, 1972].

Carbon 14 is reported in units of 10^5 atoms of carbon 14 per gram of dry air, which is proportional to mixing ratio and is referred to as carbon 14 "mixing ratio unit"; multiply by 4.82×10^{-18} to obtain carbon 14 mixing ratios by volume. The natural background of stratospheric carbon 14 was derived from prenuclear age standards, and the value is 74 units. In the AEC reports the natural background value is subtracted from the observed values, and the difference is referred to as "excess carbon 14." In the rest of this article, the "carbon 14 mixing ratio" is understood to be "excess carbon 14 mixing ratio units."

Strontium 90 is reported in terms of in units disintegration per minute in 1000 cubic feet (28 m^3) of dry air, which is proportional to mixing ratio; multiply by 0.013 to obtain picocuries per kilogram of air. Balloons collected whole air samples for carbon 14 analysis at several latitudes until mid-1969.

Large nuclear bombs were tested in the atmosphere during the periods 1952–1958 and 1961–1962 [*Bauer*, 1979]. In this context, "large" means the explosions were energetic enough to push a major fraction of the bomb cloud into the stratosphere. During the test series of September 1961 to December 1962, the rough vertical profiles of the stabilized bomb clouds are given in Table 1, derived from *Bauer* [1979, Tables B2 and B12]. The atmospheric nuclear test moratorium between the United States and USSR ended aboveground nuclear tests in December 1962. After January 1, 1963, the distribution of these tracers became dependent only on the transport processes in the atmosphere.

The French and Chinese performed a small number of atmospheric nuclear detonations after 1967 in which the bomb rise height was primarily below 20 km [*Telegadas and List*, 1969]. *Telegadas et al.* [1972] discussed whether the Chinese and French atmospheric nuclear bomb tests of 1967–1970 are significantly included in the 1970 observations. During 1961–1962 the bombs included 15 greater than 10 Mt, 7 greater than 20 Mt, and 1 of about 60 Mt. The bombs having energy equivalents greater than 10 Mt rose well into the stratosphere. After December 1962, atmospheric bomb tests were made as follows: by the Chinese (at 40°N), 3 Mt in 1967, 3 Mt in 1968, 3 Mt in 1969, and 3 Mt in October 1970; by the French (at 21°N), 3.5 Mt in 1968, and 2 Mt in summer 1970. For 3-Mt bombs detonated at these latitudes, the top of the initial bomb cloud was about 22 km. In 1970–1971 the maximum mixing ratio of carbon 14 was observed at altitudes of 24–28 km, and the maximum mixing ratio of zirconium 95 and cesium 137 was between 14 and 18 km in the northern hemisphere and between 15 and 19 km in the southern hemisphere. *Telegadas et al.* [1972] concluded that almost all excess carbon 14 observed above about 21 km in 1970–1971 was from pre-1963 nuclear bomb tests.

1.2. Goal of This Study

The goal of the study reported here is to compare the dynamical transport predictions of models against the observed carbon 14 and strontium 90 distributions as tests that are independent of chemistry and photolysis rates. This study is divided into two parts: (1) modeling carbon 14, which is in the form of CO_2 and acts as a passive gaseous tracer, and (2) modeling strontium 90, which sticks to stratospheric aerosols and thus requires the inclusion of a settling velocity in the model.

Previous studies have used carbon 14 [*Johnston et al.*, 1976; *Kinnison*, 1989; *Shia et al.*, 1989, 1993; *Jackman et al.*, 1991] and strontium 90 [*Kinnison*, 1989] as tracers in one- and two-dimensional models. The injected radioactive species in the lower stratosphere are transported somewhat similar to the exhaust gases from proposed high-speed civil transports, and the history of these radionuclides provides a suitable test of the models used to study stratospheric aircraft emissions. The first version of this paper is by *Kinnison* [1989], and the present version is a reduced manuscript from section I (volume 3) of the Report of the 1992 Model and Measurements Workshop of the High Speed Research Program/Atmospheric Effects of Stratospheric Aircraft [*Prather and Remsberg*, 1993]. The research reported within is a combination of work performed by the three authors of this article and the combined efforts of the atmospheric modeling community (see acknowledgments). Model descriptions are summarized by *Prather and Remsberg* [1993]. For the LLNL model, two different dynamical representations are shown and labeled "LLNL" and "LLNLND" (ND is short for new dynamics). Detailed descriptions of both LLNL dynamical formalisms are given by *Prather and Remsberg* [1993] (models are identified in the footnote to Table 2).

2. Carbon 14

2.1. Carbon 14 Initial Conditions for the Models as Found by Three Methods

The carbon 14 measurements provide a unique record of globally distributed inert gaseous tracer. The advantage of these data is that they are free from dependence on chemis-

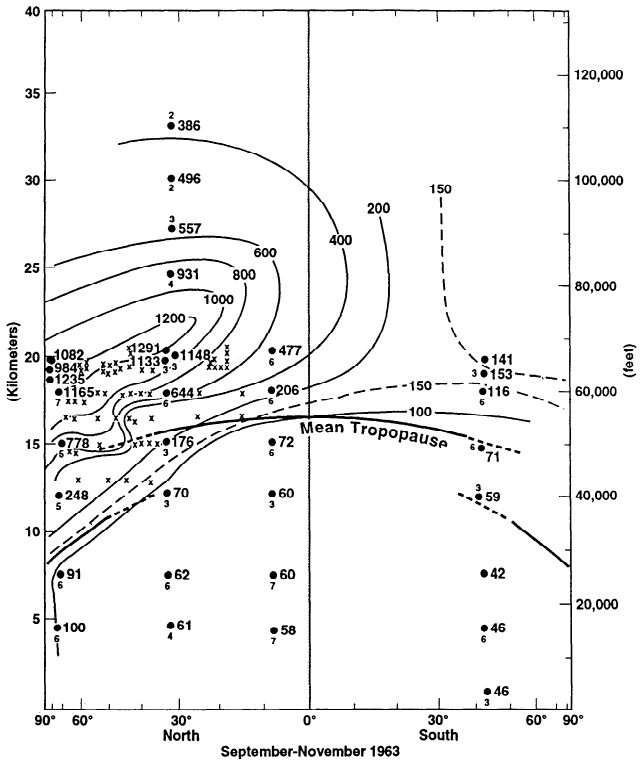

Figure 1a. Measured carbon 14 altitude-latitude distribution for October 1963 [*Telegadas*, 1971], which is an average of September, October, and November data.

try and photolysis rates. The disadvantage of this record is the sparseness of the atmospheric sampling. This sparseness is especially critical with respect to providing the initial conditions for the model calculations, and these initial conditions are highly uncertain. The initial conditions are based on measurements for October 1963, where these are available, and the initial conditions are extended to other regions by three methods of extrapolation, which produce widely different values in regions where no measurements were made.

2.1.1. Method 1. *Telegadas* [1971] reported the measured, atmospheric, excess carbon 14 data from March 1955 through July 1969 at four latitudes (70°N, 31°N, 9°N, and

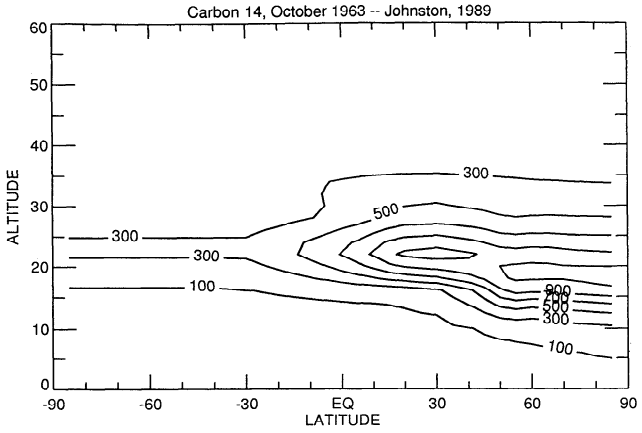

Figure 1b. Method 1 carbon 14 initial conditions for October 1963 used in this study [*Johnston*, 1989].

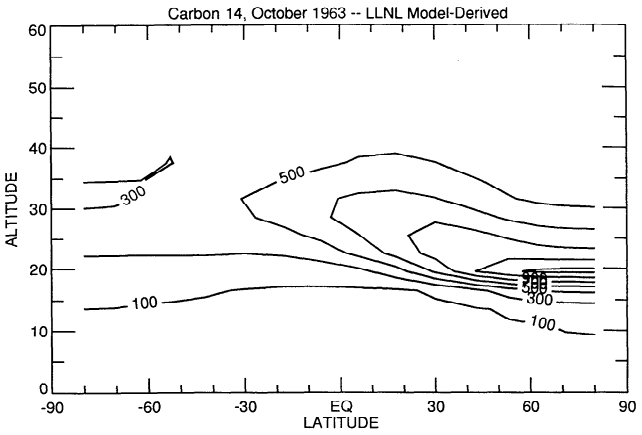

Figure 1c. Method 2 model-derived carbon 14 initial conditions for October 1963 [*Kinnison*, 1989].

42°S), but only at 31°N were measurements made at altitudes greater than 20 km. Using these data, his experience with other radioactive isotopes (Sr, Cs, Zr, etc.), and his aircraft-based observations of early-stage bomb clouds, Telegadas constructed contour plots of carbon 14 mixing ratios for each quarter year over the 15 years. Each plot represents an average of all aircraft and balloon data for a 3-month period (e.g., October is an average over September, October, and November). Figure 1a of this paper shows the altitude-latitude distribution of carbon 14 for October 1963, including numerical values of measurements at four latitudes; crosses where measurements were made at other latitudes and contour lines drawn by Telegadas are also shown. *Johnston* [1989] concluded that October 1963 provided good initial conditions for testing the dynamical representation of two-dimensional models and used Telegadas' data points and contour lines for this purpose. October 1963 was 9 months after the conclusion of the nuclear bomb test series and was the first time at 31°N that the local carbon 14 mixing ratios at all measured altitudes passed through their maximum value. Since the nuclear tests were in the northern hemisphere above the Arctic circle and in the tropics, there were large

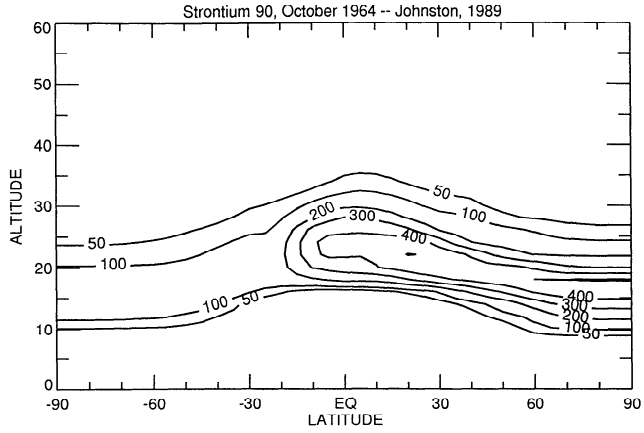

Figure 1d. Strontium 90 initial conditions for October 1964 [*Telegadas*, 1967; *Johnston*, 1989]. Contours of strontium 90 are in units of disintegrations per minute per 1000 cubic feet (28 m^3) of dry air, which are proportional to mixing ratio by volume.

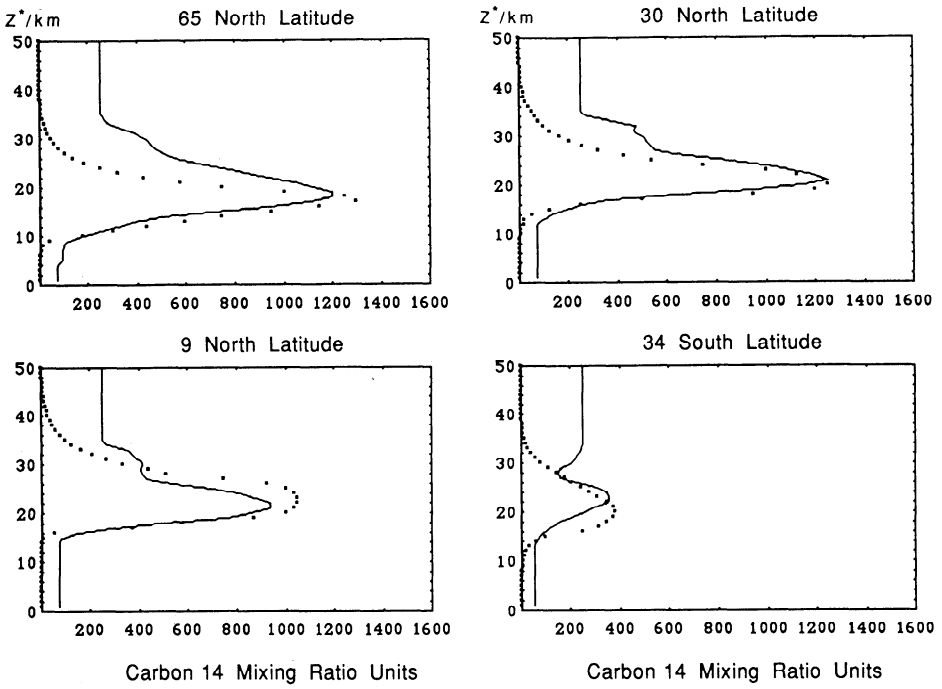

Figure 1e. Comparisons at four latitudes of method 1 (solid curve) and method 3 (dots) used to find carbon 14 initial conditions for October 1963. Contours of carbon 14 are in units of 10^5 atoms of carbon 14 per gram of dry air as obtained from a total air sampler. These units are proportional to mixing ratio; multiply by 4.82×10^{-18} to obtain carbon 14 mixing ratios by volume.

spatial gradients of carbon 14 at that time. Values outside the range of Telegadas' contour lines for October 1963 were extrapolated by *Johnston* [1989], largely on the basis of observed trends during the period 1958–1961, to give the initial conditions shown in Figure 1b. A more detailed description of this procedure is given by *Johnston* [1989, supplementary appendix] and by *Prather and Remsberg* [1993, pp. I-1 to I-3].

2.1.2. Method 2. Starting with the initial conditions for July 1960 (compare Figure 3 of *Johnston* [1989, p. 18,488]), *Kinnison* [1989] ran the LLNL two-dimensional model as a function of time through the explosion of each bomb at its appropriate latitude during the 1961–1962 test series; the calculations were continued to October 1963 to provide the second set of initial conditions. Kinnison assigned each cloud its altitude range according to two bomb rise models: (1) *Seitz et al.*'s [1968] formula, which was calibrated from tests in the tropics and from tests carried out in Arctic regions, and (2) *Peterson*'s [1970] formula, which was calibrated only in the tropics and thus overestimates the cloud rise heights for the USSR Arctic tests, especially during the winter. The lower boundary condition is based on observed carbon 14 near the ground. This calculation gives two estimates of the October 1963 initial conditions for a given two-dimensional model, an example of which is given by Figure 1c. Initial conditions found by *Kinnison* [1989] have carbon 14 mixing-ratio units that are distinctly larger than method 1 initial conditions in the global upper stratosphere and tropical stratosphere, regions where there are no measurements.

2.1.3. Method 3. The strontium 90 initial conditions are based on four measured vertical profiles at 64°N, 31°N, 9°N, and 34°S (see Figure 4 of *Johnston* [1989]) in October 1964, and these observations span the profiles from essentially zero values at 35 km down to essentially zero values at 4.5 km altitude. Compared to carbon 14, there is less uncertainty in extrapolating strontium 90 data to the top of the stratosphere and to the poles, and the interpolated, extrapolated, two-dimensional distribution of strontium 90 for October 1964 is given by Figure 1d. With a scaling factor of 2.5 applied to the strontium 90 mixing ratio units relative to carbon 14 mixing ratio units, the initial conditions for strontium 90 in October 1964 happen to be in fairly good agreement with the carbon 14 initial conditions of October 1963 between 15 and 28 km, as illustrated in Figure 1e at four latitudes, but the amount of inert tracer above 35 km, where there were no measurements, is drastically lower than that in method 1. In Figure 1e the solid curves give carbon 14 initial conditions as found by method 1, the dots give carbon 14 initial conditions as found by method 3, and the panels are successively at 65°N, 30°N, 9°N, and 34°S. Starting in October 1964, the strontium 90 model calculations by *Prather and Remsberg* [1993] were carried out twice, once assuming that strontium 90 acted as a gas and once assuming that the strontium 90 particles were attached to the sulfate aerosol and settled out with it. The model calculations carried out for strontium 90 without aerosol settling, that is, as if strontium 90 were an inert gaseous tracer, can be interpreted as modeling carbon 14 after October 1963 with a different set of initial conditions. Method 3 initial carbon 14 mixing-ratio units agree fairly well with method 1 between 15 and 28 km, are lower than method 1 below 15 km, and are distinctly lower than method 1 initial conditions in the global upper stratosphere.

2.2. Results of Model Calculations

For method 1 initial conditions, the time-dependent lower boundary values specified by the models for the northern and southern hemispheres were obtained from data by *Johnston* [1989] and are given by the following functions:

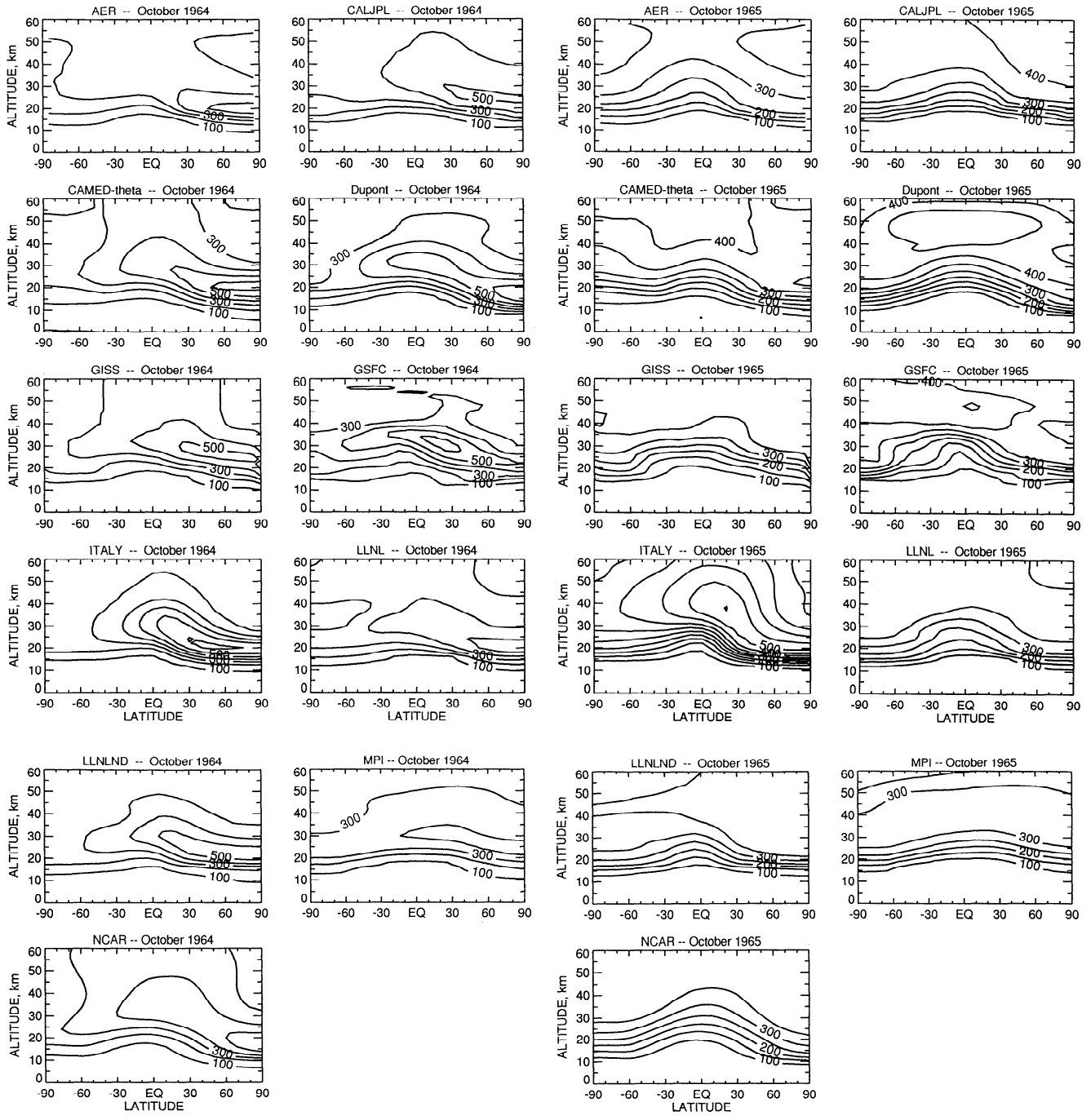

Figure 2. Model-derived carbon 14 altitude-latitude contour plots for (left) October 1964 and (right) October 1965. Initial conditions are those given in Figure 1b. Contours are in units of 10^5 atoms of carbon 14 per gram of dry air as obtained from a total air sampler. These units are proportional to mixing ratio.

To 1971

$$^{14}C\ (\text{N-Hem.}) = 73.0 - 0.27823t - 3.45648 \times 10^{-3}t^2 + 4.21159 \times 10^{-5}t^3$$

To 1968

$$^{14}C\ (\text{S-Hem.}) = 44.5 + 1.02535t - 2.13565 \times 10^{-2}t^2 + 8.61853 \times 10^{-5}t^3$$

where t is the time in months after October 15, 1963, and the southern hemisphere lower boundary values are the same as those of the northern hemisphere after 1968. These functions are based on observed data in the lowest troposphere during this time period. The upper boundary conditions were specified as zero flux.

The models are used to derive the distribution of carbon 14 between October 1963 and January 1971. The three-dimensional GISS model is integrated only from October 1963 to October 1968. In Figure 2, altitude-latitude contour plots of carbon 14 are shown for each model 1 year after and 2 years after the initial condition (that is, for October 1964 and October 1965).

The models obtain results at all latitudes and at all

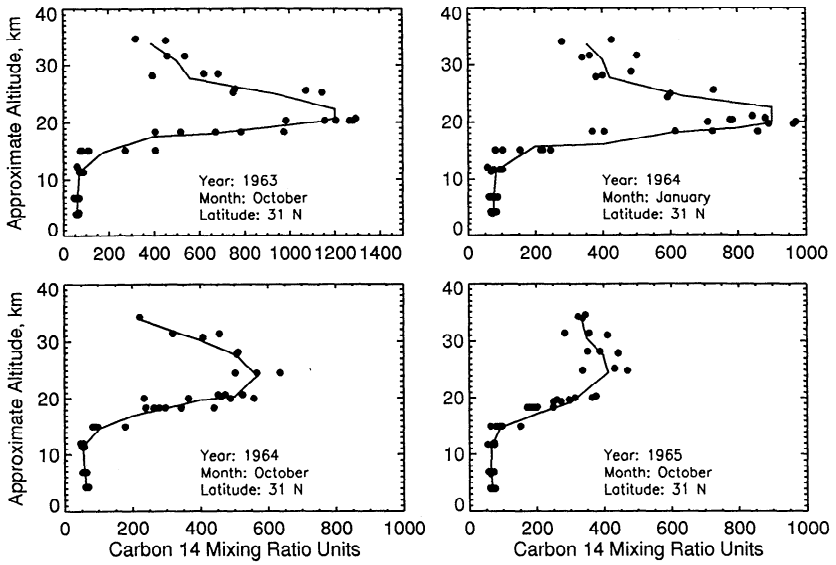

Figure 3. Individual measurements at 31°N of carbon 14 over four separate 3-month periods centered on October 1963, January 1964, October 1964, and October 1965. The solid curve for each time period is the profile derived by *Johnston* [1989] from work by *Telegadas* [1971].

altitudes from 0 to 60 km. The measurements were made at a limited number of altitudes and latitudes, and comparisons between models and measurements are possible only where the two overlap. Observed carbon 14 profiles up to 20 km are available for three latitudes (70°N, 9°N, and 42°S) and profiles up to 32 ± 2 km (31°N) are available every 3 months through July 1966 [*Telegadas*, 1971; *Johnston*, 1989]. In this study, model results are compared with the average measured vertical profiles. As a qualitative estimate of the minimum uncertainty in these profiles, we compare locally measured data and seasonally averaged profiles at 31°N for four times between October 15, 1963, and July 15, 1966 (Figure 3). Among the individually measured data there is low variability in the troposphere but significant variability in the stratosphere. Within this variability there is reasonably good agreement between locally measured data and the profiles derived from averaged data. There are additional profiles between July 1966 and July 1969, which have no data above 20 km altitude and are not discussed in this study.

Figure 4 compares measured and modeled vertical profiles of carbon 14 mixing ratio units at 31°N for eight times between January 1964 and April 1966. The measured profiles are labeled by crosses, and each of the 11 models in this study is labeled by a distinctive symbol. Figure 5 compares measured and modeled vertical profiles of carbon 14 mixing ratio units at 70°N, 9°N, and 42°S for January 1965, 15 months after initial conditions. Figure 6 compares measured and modeled vertical profiles of carbon 14 mixing ratio units at four latitudes (65°N, 30°N, 9°N, and 34°S) for November 1970, 7 years after initial conditions.

2.3. Intermodel Comparisons

2.3.1. Global stratospheric residence times. The global, model stratospheric (16–60 km) inventory of excess carbon 14 molecules was calculated for each 3-month period for each of 11 models, and the residence times were found by a least squares fit of ln (inventory) versus time; the values are given in Table 2. In Table 3, for each time interval of Table 2, the models are grouped according to their relative stratospheric residence time: short, medium, and long. Between October 1963 and October 1964 (Table 3, column 1), there are seven models in the short stratospheric residence time section, and the range of residence times among all the models is from 1.3 to 3.2 years. When the stratospheric residence times are compared between January 1965 and July 1966, three models have a relatively short stratospheric residence time (2.9–3.2 years), six models have medium residence times (3.4–3.9 years), and two have long residence times (4.4–4.9 years). Table 3 shows the period between October 1963 and July 1966; the range in stratospheric residence times is from 2.3 years (GISS) to 4.1 years (Italy). Table 3 also gives the full time, October 1963 through January 1971, and the corresponding residence times, which range from 3.8 to 4.9 years. For the time interval January 1965 through January 1971, the range of calculated residence times is 4.3 to 5.4 years. Italy and DuPont have the longest stratospheric lifetimes, and GSFC and GISS have the shortest residence times. During the first year after initial conditions, the range in carbon 14 stratospheric residence times varied by a factor of 2.5 among the 11 models. Whatever the residence time is during the first year, each model shows a considerably longer carbon 14 lifetime at later times. The explanation of this deceleration of stratospheric sweep-out rate with passage of time appears to be that the models relatively rapidly remove carbon 14 from the lower stratosphere but the model transfer of carbon 14 from the middle stratosphere to the lower stratosphere is slower than the rate of removal from the lower stratosphere.

2.3.2. Altitude-latitude contour maps. Figure 1b gives the altitude-latitude contour plot of carbon 14 mixing ratio units used by all models as initial conditions for October 1963. Figure 2 gives altitude-latitude contour plots calculated by 11 models for October 1964 and for October 1965, 1 year after and 2 years after initial conditions. A rough measure of the agreement and disagreement among the 11 models for October 1964 (Figure 2) was carried out by superimposing on tracing paper all curves for contour 200, contour 300, contour 400, and contour 500, each on a separate page. The superimposed curves for contour 200 spread from pole to

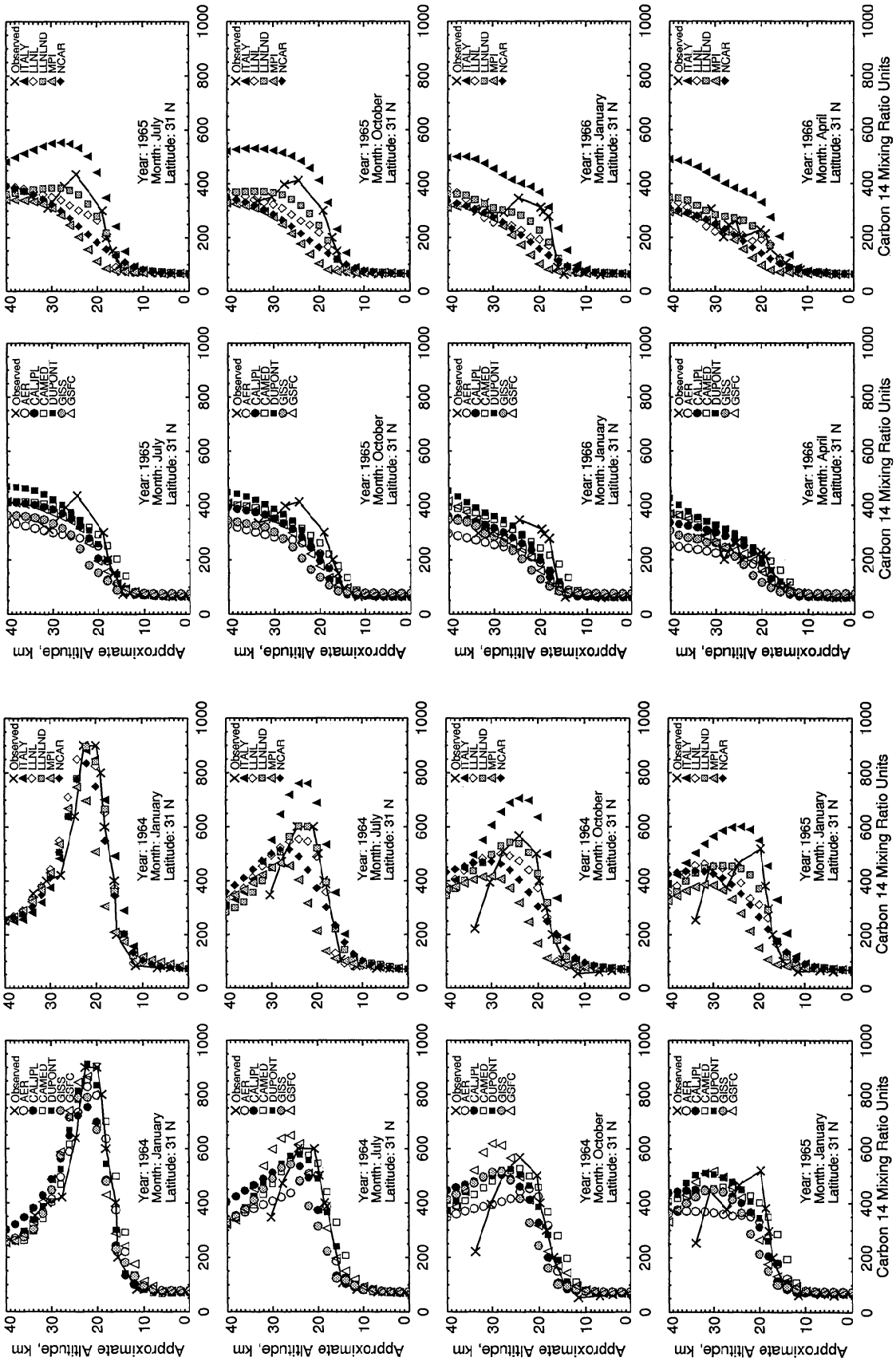

Figure 4. Comparison of model-derived carbon 14 profiles and measured profiles at 31°N for eight times between January 1964 and April 1966. Profiles are in units of 10^5 atoms of carbon 14 per gram of dry air; units are proportional to mixing ratio.

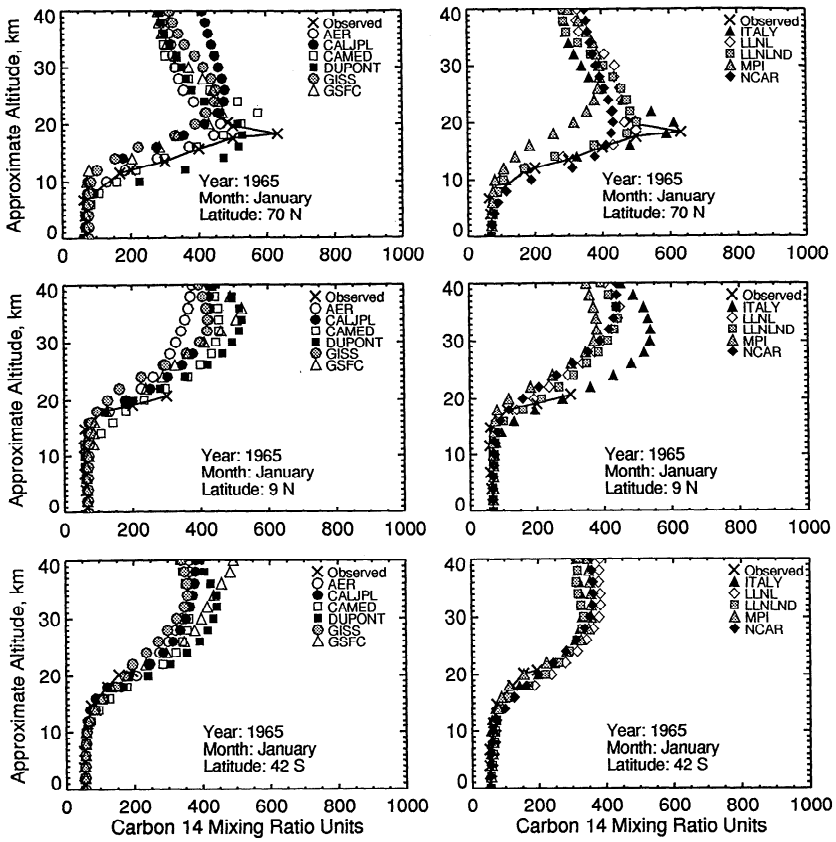

Figure 5. As in Figure 4, except for January 1965 and for latitudes of 70°N, 9°N, and 42°S.

pole with maximum altitude at tropical latitudes, and at each latitude there is a vertical spread, among the 11 models, of about 4 km. The contour 300 curves appear both at low altitudes and at high altitudes. The low-altitude band is similar in shape to the band of contour 200 curves, and its vertical spread among the 11 models is about 5 km. The upper altitude contour 300 curves appear as a random tangle from 30 to 60 km in middle and high latitudes and from 40 to 60 km in tropical latitudes. The superimposed contour 400 curves stretch from the north pole to southern midlatitudes and show at low altitudes a band of about 7 km vertical spread and at upper altitudes a band of about 15 km vertical spread. One model has a 400 contour as high as 50 km, and another has no 400 contour curve above 30 km. In each model, the 500 contour is a closed loop stretching from the north pole to some latitude between 60°N and 25°S. This rough preview of the model calculations indicates large differences among their results.

2.4. Comparisons Between Calculated and Observed Excess Carbon 14

2.4.1. Lower stratosphere. The boundary values at the Earth's surface are the time-dependent observed carbon 14, which implicitly gives a flux into the surface, and there is no significance to the agreement between models and data in the troposphere. The altitude at which carbon 14 begins its sharp increase is a measure of the tropopause height, both for the data and for the models. At 70°N, 9°N, and 42°S, the measurements were made only from aircraft, and the maximum altitude was 20 km. The observed data at 70°N (Figure 5) appear in some cases to reach the maximum carbon 14 mixing ratio and give a good comparison of models and data in the lowest stratosphere, but the tropopause is so high at 9°N and 42°S that the comparison between models and data is of little value at these latitudes. From about April 1964 to April 1965 the altitude of the calculated carbon 14 tropopause is lower than (−), about the same as (≈), or higher (+) than that observed, as follows:

Altitude at 70°N	Altitude at 31°N
NCAR −	Italy −
CAMED −	CAMED −
DuPont ≈	DuPont ≈
Italy ≈	LLNLND ≈
AER ≈	AER ≈
LLNL ≈	LLNL ≈
GSFC +	GSFC +
CALJPL +	CALJPL +
GISS +	GISS +
LLNLND +	NCAR +
MPI +	MPI +

A quantitative comparison of the data and the models is given by Table 4, where the ratio of calculated carbon 14 to observed carbon 14 at 31°N latitude and 20 km altitude (the altitude of the observed maximum carbon 14 mixing ratio) is given from January 1964 through October 1965:

$$\text{ratio} = {}^{14}C \text{ (calculated)}/{}^{14}C \text{ (observed)}$$

In October 1963, all models are set to agree with each other and with the data, and in January 1964, 3 months later, 10 out of 11 models have this ratio slightly below 1, with the range of values being 0.57 to 1.00. During the next 2 years this ratio decreases: to about 0.3 for one model, to 0.5 for three models, to between 0.6 and 0.7 for four models, and to between 0.8 and 1.2 for three models. For all the models

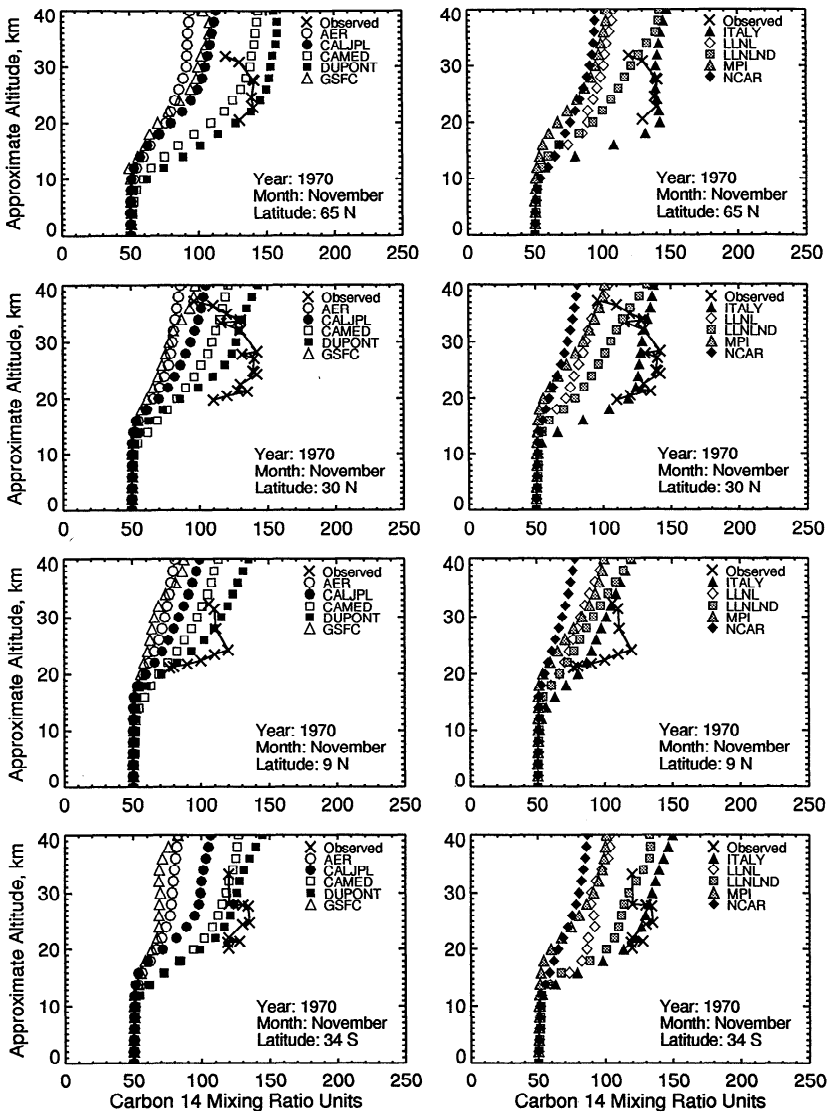

Figure 6. As in Figure 4, except for November 1970 and for latitudes of 65°N, 30°N, 9°N, and 34°S.

after 2 years, the average ratio varies from 0.36 to 1.23, spanning a factor of 3.4. At 20 km, 31°N, the 2-year average model results are higher than the measured data for one model and lower for 10 models. If the ratio ^{14}C (calculated)/^{14}C (observed) is less than 1, it could mean any of several calibration errors or real differences between model and observations; but when this ratio decreases systematically with time, it means that the model is removing carbon 14 from the area in question more rapidly than was observed. Except for three models, the changing ratios of Table 4 indicate that the models reduce carbon 14 at 30°N and a 20-km altitude at a rate somewhat faster than that observed.

A similar comparison is given for 70°N latitude and 16 km altitude (the altitude of the observed maximum carbon 14 mixing ratio at 70°N) in Table 5, but these data show greater irregularities than those at 20 km. In January 1964, 3 months after initial conditions, the initial ratio of 1.00 changes to values between 0.8 and 1.2. After 2 more years this ratio decreases: to about 0.4 for one model, to between 0.5 and 0.7 for four models, to between 0.7 and 0.8 for three models, and to between 0.9 and 1.2 for three models. For all the models the average ratio varies from 0.56 to 1.4. At 16 km, 70°N, the 2-year average model results are higher than the measured data for five models and lower for six models. Except for four models, the changing ratios of Table 5 indicate that the models reduce carbon 14 at 70°N and a 16-km altitude at a rate somewhat faster than that observed.

At the altitude of carbon 14 maximum mixing ratio, at both 31°N and 70°N, results from the 11 models as a group span the range of measured values at all times between January 1964 and July 1966. On average of the set of 11 models, the models reduce carbon 14 in the lower stratosphere at a rate slightly faster than that observed.

2.4.2. Middle stratosphere. At 31°N latitude the observed carbon 14 data show a bell-shaped vertical profile, skewed toward the vertical, whose altitude of maximum mixing ratio increased from 21 km in January 1964 to 25 km in January 1966 (Figure 3; also, *Johnston* [1989, Figure 5]), and the upper portion of the bell-shaped distribution slowly opens up. The model-calculated vertical profiles show different behavior. The modeled vertical profiles open upwardly over the course of time much faster than the measured vertical profiles (Figure 4). In all models the altitude of maximum carbon 14 mixing ratio increased from about 22

Table 2. Carbon 14 Stratospheric Residence Times in Years

Model*	Oct. 1963 through Oct. 1964	Jan. 1965 through July 1966	Oct. 1963 through July 1966	Oct. 1963 through Jan. 1971	Jan. 1965 through Jan. 1971
AER	1.7	3.5	2.7	3.9	4.5
CALJPL	1.7	3.9	2.8	4.2	4.8
CAMED	2.4	3.4	3.4	4.5	5.0
DuPont	2.7	4.9	3.6	4.9	5.4
GISS	1.3	3.2	2.3	NA	NA
GSFC	1.7	2.9	2.4	3.8	4.3
Italy	3.2	4.4	4.1	4.9	5.1
LLNL	2.7	3.5	3.1	4.1	4.7
LLNLND	2.0	3.6	2.9	4.6	5.3
MPI	1.3	3.5	2.4	4.3	5.3
NCAR	1.9	3.0	2.5	3.8	4.4
			Range		
	1.3–3.2	2.9–4.9	2.3–3.2	3.8–4.9	4.3–5.4

Values are based on linear least squares regression analysis. For each period the model-derived burden was integrated globally between 16 and 60 km. NA, not available.

*AER, Atmospheric and Environmental Research, Inc.; CALJPL, Jet Propulsion Laboratory, California Institute of Technology; CAMED, University of Cambridge and University of Edinburgh; DuPont, E. I. du Pont de Nemours, Inc.; GISS, Goddard Institute for Space Studies; GSFC, Goddard Space Flight Center; Italy, Università degli Studi L'Aquila; LLNL, LLNLND, Lawrence Livermore National Laboratory; MPI, Max-Planck-Institut für Chemie; NCAR, National Center for Atmospheric Research.

km in January 1964 to altitudes greater than 40 km by April 1966 (Table 6). By April 1965 the maximum calculated carbon 14 mixing ratio for AER and NCAR is above 40 km, the upper borders of the panels in Figure 4, but the carbon 14 data profile has its maximum at 23 km. By July 1965, all models but LLNLND and Italy have their mixing ratio maxima above 40 km, and by April 1966 these two models also have their maxima above 40 km.

Another representation of this effect is shown in Table 7. The Italy model most nearly resembles the carbon 14 data with respect to the rise of its altitude of maximum mixing ratio, and the CALJPL model is average of the other models in this respect (Table 6). The average slopes of the carbon 14 mixing ratio with altitude, $(\Delta\mu/\Delta z)$, between 25 and 33 km are presented for the observed data, the Italy model, and the CALJPL model in Table 7 from January 1964 to July 1966:

$$\text{average slope} = [\mu(33\text{ km}) - \mu(25\text{ km})]/(33 - 25)$$

The two models and the observed data have a decrease of 32 mixing ratio units per kilometer in January 1964, and a year later the carbon 14 data have the same vertical slope. During this year, January 1964 to January 1965, the upper arm of the CALJPL mixing ratio profile opened up from a slope of −32 units to −18 to −11 to −2, which is almost straight up. The Italy model gives results intermediate between the observed data and the results of the CALJPL model; it remained strongly negative until October 1964 and then rotated upward, passing the vertical by October 1965. This feature had been noted in the LLNL model [*Kinnison*, 1989]; it is a universal feature of the 11 models studied here.

In part, this disagreement between the observed carbon 14 data and all the models appears to be based on a too-fast transfer to the troposphere by the models, as can be seen from examination of Table 8, which shows actual mixing ratios at 31°N instead of the ratios of Table 4. At 32 km altitude the observed mixing ratios are essentially constant at 350 ± 50 from April 1964 to April 1966, falling to 280 units by July 1966. At 32 km the calculated mixing ratios are larger than the observed values in April 1964, increase slightly (from 450 to 520) between April 1964 and July 1964, and decrease slowly during the next 2 years to 290 units by July 1966. At 20 km the observed carbon 14 mixing ratios fall, with some seasonal irregularity, from 580 units in April 1964 to 280 units in July 1966; over the same period the calculated mixing ratios start with essentially the same value (560 units) and fall to 150 units, a value 54% of that observed. This too-rapid decrease of model carbon 14 at 20 km would contribute to the disagreement between models and measurements shown in Tables 6 and 7 and in Figure 4. (In section 4 we propose that the other component of this effect is that all models sweep carbon 14 too slowly out of the middle stratosphere.)

2.4.3. Long-time observations. At the conclusion of atmospheric carbon 14 monitoring by the AEC, a special investigation was carried out in the fall of 1970, almost 8 years after the conclusion of atmospheric nuclear bomb testing [*Telegadas et al.*, 1972]. The results for the fall of 1970 (September through December) are reproduced in Table 9, the calculated and observed vertical profiles are given in Figure 6, and Telegadas et al.'s contour plot is shown in Figure 7. The observed vertical profiles are of qualitatively different shape from all the calculated profiles (Figure 6; the complete set is given by *Prather and Remsberg* [1993, section I, volume 3]. Each observed vertical profile shows a broad maximum carbon 14 mixing ratio at more or less 25 km and a distinct decrease with altitude

Table 3. Various Time Interval–Model Groupings in Order of Increasing Global Stratospheric Residence Times of Carbon 14

Oct. 1963 through Oct. 1964*	Jan. 1965 through July 1966†	Oct. 1963 through July 1966‡	Oct. 1963 through Jan. 1971§	Jan. 1965 through Jan. 1971‖
		Short		
GISS	GSFC	GISS	GSFC	GSFC
MPI	NCAR	MPI	NCAR	NCAR
AER	GISS	GSFC	AER	AER
CALJPL		NCAR	LLNL	
GSFC		AER	CALJPL	
NCAR		CALJPL		
LLNLND		LLNLND		
		Medium		
CAMED	CAMED	LLNL	MPI	LLNL
LLNL	AER	CAMED	CAMED	CALJPL
DuPont	LLNL	DuPont	LLNLND	CAMED
	MPI			
	LLNLND			
	CALJPL			
		Long		
Italy	Italy	Italy	DuPont	Italy
	DuPont		Italy	LLNLND
				MPI
				DuPont

Times are in years.
*Short, 1.3–2; medium, 2.4–2.7; long, 3.2.
†Short, 2.9–3.2; medium, 3.4–3.9; long, 4–4.9.
‡Short, 2.3–2.9; medium, 3.1–3.6; long, 4.1.
§Short, 3.8–4.2; medium, 4.3–4.6; long, 4.9.
‖Short, 4.3–4.5; medium, 4.7–5; long, 5.1–5.4.

Table 4. Ratio of Calculated Carbon 14 to Observed Carbon 14 at 31°N Latitude and an Altitude of 20 km

		Calculated										
Date	Observed	MPI	GISS	NCAR	GSFC	CALJPL	AER	LLNL	DuPont	LLNLND	CAMED	Italy
Jan. 1964	900	0.57	0.78	0.83	0.78	0.78	0.89	0.90	0.92	0.91	1.00	0.98
April 1964	580	0.53	0.81	0.86	0.84	0.95	1.00	1.12	1.14	1.14	1.16	1.29
July 1964	520	0.42	0.65	0.72	0.72	0.73	0.96	0.94	0.94	1.00	1.06	1.33
Oct. 1964	420	0.44	0.51	0.60	0.63	0.65	0.86	0.76	0.90	0.84	0.90	1.28
Jan. 1965	520	0.34	0.46	0.50	0.50	0.60	0.63	0.60	0.67	0.72	0.79	1.06
April 1965	400	0.31	0.48	0.55	0.60	0.70	0.65	0.75	0.78	0.83	0.83	1.20
July 1965	340	0.32	0.49	0.56	0.59	0.60	0.75	0.76	0.76	0.82	1.03	1.32
Oct. 1965	325	0.31	0.46	0.52	0.55	0.62	0.69	0.68	0.68	0.77	0.80	1.26
							Average					
		0.36	0.58	0.64	0.65	0.70	0.80	0.81	0.83	0.88	0.95	1.23

Table 5. The Ratio of Calculated Carbon 14 to Observed Carbon 14 at 70°N Latitude and at an Altitude of 16 km

		Calculated										
Date	Observed	MPI	GISS	CALJPL	GSFC	AER	LLNLND	NCAR	CAMED	LLNL	Italy	DuPont
Jan. 1964	830	0.90	0.82	1.03	0.97	1.04	1.07	1.10	0.83	1.19	0.88	1.20
April 1964	600	0.83	0.83	1.07	0.93	1.32	1.28	1.27	1.17	1.38	1.08	1.48
July 1964	400	0.65	0.98	0.84	0.99	1.33	0.74	1.54	1.40	1.43	1.38	1.88
Oct. 1964	450	0.42	0.57	0.55	0.67	0.73	0.55	1.13	1.09	0.83	1.13	1.33
Jan. 1965	430	0.42	0.53	0.65	0.65	0.77	0.87	0.93	0.80	1.00	1.12	1.21
April 1965	310	0.56	0.79	0.82	0.87	1.16	1.26	1.13	1.26	1.39	1.45	1.45
July 1965	250	0.50	0.72	0.66	0.83	1.00	0.68	1.18	1.32	1.20	1.52	1.52
Oct. 1965	270	0.39	0.56	0.56	0.63	0.74	0.59	0.96	1.07	0.83	1.33	1.19
Jan. 1966	305	0.39	0.49	0.59	0.56	0.66	0.75	0.75	0.79	0.90	1.16	0.98
							Average					
		0.56	0.70	0.75	0.79	0.97	0.87	1.11	1.08	1.13	1.23	1.36

above the maximum (Table 9 and Figure 6); but each calculated profile increases with altitude up to 40 km (Figure 6). At 26 km, 8 out of 10 models calculate carbon 14 to be less than the observed values, the ratios being between 0.49 and 0.73 (Table 10). The excess carbon 14 in the troposphere was 50 units, and a significant mode of comparison of the long-term stratospheric carbon 14 is the difference between the stratospheric value and the tropospheric value, which is also given in Table 10. The model-calculated differences at 26 km range from 18 to 77, compared to the observed difference of 90. The data show greater persistence of carbon 14 in the 25-km range than is shown by any model except Italy and du Pont. Figures 4 (October 1964 and October 1965) and 6 (November 1970, 31°N) taken together indicate that the contradiction between measurements and models in the unfolding of vertical profiles in midlatitude during 1963–1966 (Figure 4) and in the long-term global two-dimensional distribution (Figures 6 and 7) is the same phenomenon.

Figure 7 is a latitude-altitude contour map of carbon 14 mixing ratio units for September through December ("November") 1970 [*Telegadas et al.*, 1972, Figure 4], which is based on the data in Table 9. The data for the southern hemisphere include one vertical profile; the northern hemisphere data include four vertical profiles. In the 23–30 km altitude band between 65°N and 34°S, the data show a minimum with respect to latitude above the tropics. Unlike all the models, the measured carbon 14 mixing ratios decrease about 28 km, and the decrease continues to the topmost observation altitude, 36 km at 31°N.

Table 6. Altitude of Maximum Carbon 14 Mixing Ratio From January 1964 to July 1966

		Calculated										
Date	Observed	AER	NCAR	MPI	DuPont	GSFC	LLNL	CALJPL	GISS	CAMED	LLNLND	Italy
Jan. 1964	21	22	22	24	22	22	22	22	22	20	22	21
April 1964	20	21	24	26	22	24	21	22	24	22	22	22
July 1964	22	21	27	28	24	26	24	25	26	24	23	23
Oct. 1964	24	22	30	30	26	30	28	27	28	24	25	24
Jan. 1965	21	33	32	31	32	30	30	30	30	27	25	24
April 1965	23	>40	>40	34	34	34	34	34	30	30	25	25
July 1965	25	>40	>40	>40	>40	>40	>40	>40	>40	>40	30	25
Oct. 1965	25	>40	>40	>40	>40	>40	>40	>40	>40	>40	35	32
Jan. 1966	25	>40	>40	>40	>40	>40	>40	>40	>40	>40	>40	36
April 1966	>33	>40	>40	>40	>40	>40	>40	>40	>40	>40	>40	>40
July 1966	25	>40	>40	>40	>40	>40	>40	>40	>40	>40	>40	>40

Altitudes are in kilometers.

Table 7. Change of Carbon 14 Mixing Ratio With Altitude Between 25 and 33 km ($\Delta\mu/\Delta z$) at 31°N Latitude

Date	Observed	Calculated	
		Italy	CALJPL
Jan. 1964	−32	−32	−32
April 1964	−40	−44	−18
July 1964	−40	−44	−11
Oct. 1964	−32	−22	−2
Jan. 1965	−24	−12	+2
April 1965	−11	−5	+4
July 1965	−12	−6	+6
Oct. 1965	−10	+4	+7
Jan. 1966	−6	+8	+6
April 1966	+5	+9	+5
July 1966	−2	+9	+5

Expressed in carbon 14 mixing ratio units per kilometer.

2.5. Results From Three Different Initial Conditions

In section 2.1 we described three initial 2-dimensional global distributions of carbon 14 for October 1963. Method 1 is the one used for all model calculations given above. Method 2 models the nuclear bomb explosions during the 1961–1962 test series up to October 1963, and this procedure is carried out using two different bomb rise models, which results in two significantly different distributions for October 1963, each having much larger mixing ratios of carbon 14 in the tropics and in the stratosphere above 35 km than those of method 1 [*Kinnison*, 1989]. Method 3, which agrees fairly well with method 1 between 15 and 28 km at 31°N, 9°N, and 34°S, has nearly zero carbon 14 above 35 km.

2.5.1. Measurements and model calculations using method 2 initial conditions. *Kinnison* [1989] found the initial conditions for October 1963 for two bomb rise models and continued the calculations for every 3 months through December 1970. This calculation for distributions of carbon 14 after October 1963 may be interpreted as being the same sort of model results as those discussed above, but with different initial conditions. *Kinnison* [1989, pp. 178–184] gives comparisons between a method 1 case and two method 2 cases, which is given in Table 11. The large differences in initial conditions in the middle and upper stratosphere are largely eliminated by November 1970, 7 years after the starting point. *Kinnison* [1989] showed the three initial carbon 14 vertical profiles of October 1963 to have similar bell-shaped distributions, which are similar in shape to the measured vertical profile, and the absolute values in the 15–25 km range agree within about 25%. As time advances from October 1963 to October 1964, all three model vertical profiles open upward much faster than the measured vertical profiles at 31°N, and the relative rate of unfolding for the three cases is thesis case B < F < E. With advancing time, this process continues, and the three profiles approach each other in shape and magnitude. The measured and modeled carbon 14 vertical profiles in November 1970 [*Kinnison*, 1989, p. 184] include 65°N, 42°N, 30°N, 9°N, and 34°S, and

Table 8. Actual Mixing Ratios at 31°N

	Date									
	April 1964	July 1964	Oct. 1964	Jan. 1965	April 1965	July 1965	Oct. 1965	Jan. 1966	April 1966	July 1966
					At 20 km					
Observed	580	520	500	520	400	340	340	300	300	280
CALJPL	560	380	330	300	280	200	190	200	185	150
					At 32 km					
Observed	325	340	300	400	340	320	340	330	300	280
CALJPL	450	520	500	460	420	390	360	325	305	290

Table 9. Excess Carbon 14 Mixing Ratio Units Measured as a Function of Latitude and Altitude for November 1970 [*Telegadas et al.*, 1972]

65°N		42°N		30°N		9°N		34°S	
Altitude, km	Units	Altitude, km	Units	Altitude, km	Units	Altitude, km	Units	Altitude, km	Units
31.0	120	36.0	125	36.3	96	31.5	106	32.3	120
30.0	130	31.5	130	35.5	110	30.6	110	27.3	120
27.0	140	30.8	134	34.0	120	27.2	111	27.2	130
26.9	141	27.4	138	33.0	130	23.7	120	27.0	134
24.0	139	27.2	140	32.8	128	23.0	110	24.2	135
22.3	140	24.0	143	32.5	116	22.0	100	23.9	130
20.3	130	23.3	140	31.2	130	21.4	90	21.7	120
		21.3	130	27.6	142	21.0	80	21.1	128
		20.9	125	27.2	131	20.8	77	20.9	119
		19.6	120	26.5	140			20.0	120
				24.4	139				
				24.3	140				
				23.9	142				
				22.2	130				
				21.2	128				
				21.0	135				
				20.3	120				
				19.6	110				

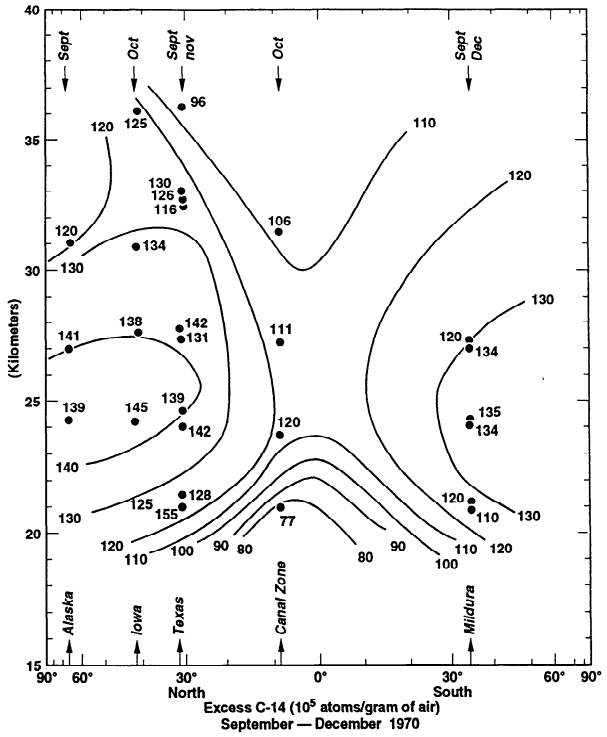

Figure 7. Altitude-latitude contour plot of mixing ratio units of carbon 14 in the fall of 1970, based on vertical profiles at five latitudes [*Telegadas et al.*, 1972].

the qualitative relation between measured and modeled vertical profiles is the same as that of Figure 6; that is, all models are in qualitative disagreement with the shape of the observed carbon 14 vertical profiles.

2.5.2. Measurements and model calculations using method 3 initial conditions. In section 2.1.3 we describe how we produced initial conditions for carbon 14 for October 1963 (Figure 1e) from four measured vertical profiles of strontium 90 for October 1964, where these method 3 initial conditions have much less carbon 14 above 28 km than the standard method 1 initial conditions. In the model simulations regarding strontium 90 as a gas, all models in effect carried out a carbon 14 simulation for a period of 27 months using method 3 as initial conditions. Italy gives the longest first-year carbon 14 lifetime of all the models, and AER gives the shortest first-year carbon 14 lifetime of the models that participated in the strontium 90 test (Table 2). For these two models, Figure 8 compares the model output for carbon 14 at 30°N, according to method 1 and method 3 initial conditions,

Table 11. Carbon 14 Mixing Ratio Units at 30°N and 40 km

Date	Method 1 (Thesis Case B)	Method 2* (Thesis Case F)	Method 2† (Thesis Case E)
Oct. 1963	250	400	700
Oct. 1964	400	510	680
Oct. 1965	420	430	560
Nov. 1970	140	150	170

**Seitz et al.* [1968].
†*Peterson* [1970].

0, 6, 12, and 24 months after the initial time. Each model at each time, whether based on method 1 or method 3 initial conditions, shows nearly the same vertical profile above 15 km even though there are large differences in initial carbon 14 above 28 km. Over the period of 2 years, the vertical profile of each model unfolds upward much faster than the measured vertical profiles, which is the same qualitative difference as that shown by simulations based on model 1 and model 2.

3. Strontium 90

As with carbon 14, strontium 90 data were measured by balloon sondes and a large number of aircraft flights. *Telegadas* [1967] averaged the data over a 3-month period and produced altitude-latitude contour plots in units of disintegration per minute in 1000 cubic feet (28 m^3) of dry air, which is proportional to mixing ratio. Typically, four latitudes (64°N, 31°N, 9°N, and 34°S) were measured from October 1964 to January 1967 with an altitude range from the surface to 33 ± 3 km. *Johnston* [1989] used these plots to produce the October 1964 initial conditions for this study, shown by Figure 1d.

Since strontium 90 coalesces on aerosol particles, and the settling velocity of aerosol particles is proportional to the square of their radius, the removal rate of strontium 90 is dependent on the size distribution of aerosol particles in the atmosphere. In situ measurements of aerosol size in the early 1960s are sporadic. *Mossop* [1964] reported the size distribution of aerosol particles at 20 km from early 1963 to July 1964. Large-size particles with diameters up to 8 μm were collected in early 1963; the median diameters of aerosol particles gradually reduced to 0.2 μm by July 1964. Volcanic dust from the eruption of Mount Agung on March 17, 1963, had largely settled out of the lower stratosphere by the end of 1964. After 1963, no major volcanic eruptions were reported until the eruption of Fuego in October 1974. Since

Table 10. Excess Carbon 14 Mixing Ratio Measurements at 30°N Latitude and an Altitude of 26 km During November 1970

		Calculated									
	Observed	NCAR	MPI	GSFC	AER	LLNL	CALJPL	LLNLN	CAMED	DuPont	Italy
		Without Tropospheric Value (50 Units) Subtracted From Stratospheric Value									
Value	140	68	72	72	76	82	86	96	102	118	127
Ratio	1.00	0.49	0.51	0.51	0.54	0.59	0.61	0.69	0.73	0.84	0.91
		With Tropospheric Value (50) Subtracted From Stratospheric Value									
Value	90	18	22	22	26	32	36	46	52	68	77
Ratio	1.00	0.20	0.24	0.24	0.29	0.36	0.40	0.51	0.58	0.76	0.86

Date is almost 8 years after cessation of the 1961–1962 series of atmospheric nuclear bomb tests. At latitude of 30°N the observed carbon 14 mixing ratio was a maximum at the 26-km altitude.

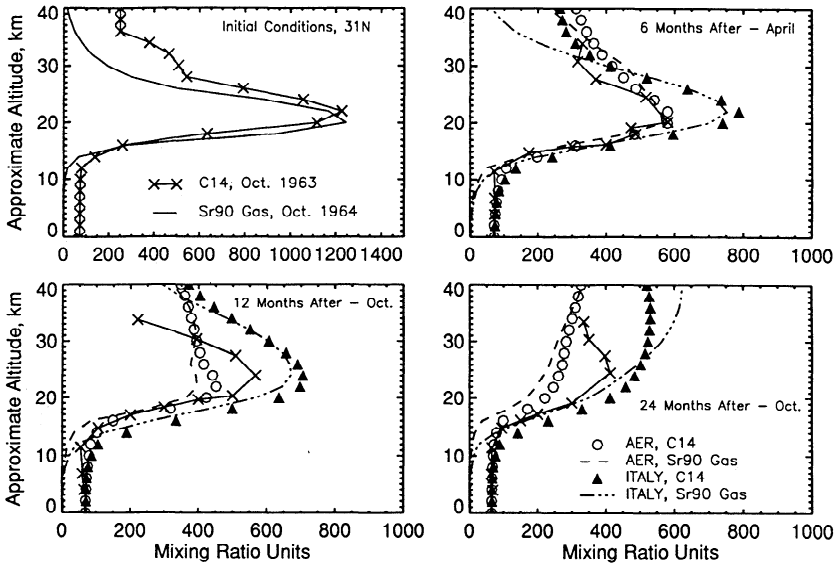

Figure 8. Comparison of measured and model-derived carbon 14 profiles based on method 3 initial conditions and measured profiles at 31°N for 0, 6, 12, and 24 months after the initial time.

Table 12a. Southern Hemisphere Settling Velocities of Aerosol Particles Used in the Strontium 90 Simulation

	Latitude Zones, deg											
Altitude, km	60–55	55–50	50–45	45–40	40–35	35–30	30–25	25–20	20–15	15–10	10–5	5–0
0–2	6.9	7.3	9.0	9.9	8.9	8.0	9.9	10.3	6.7	13.1	17.5	3.8
2–4	7.1	7.4	9.1	9.9	9.0	8.1	10.0	10.4	6.8	13.2	17.4	4.0
4–6	8.9	9.4	12.1	13.1	12.1	11.2	13.4	13.2	10.9	16.3	21.3	6.5
6–8	10.9	10.3	11.4	11.2	12.0	11.9	13.0	13.2	14.4	19.3	35.2	11.5
8–10	13.9	14.3	14.9	17.5	18.2	15.5	12.9	19.4	17.9	19.3	35.2	20.8
10–12	25.1	24.4	24.4	25.6	22.5	21.4	22.9	27.0	21.1	24.4	35.2	29.6
12–14	32.7	32.6	32.6	32.1	29.0	29.9	31.2	34.0	34.1	29.7	43.0	25.2
14–16	47.6	49.7	46.1	44.8	44.3	44.3	43.5	42.5	42.3	41.4	45.5	42.5
16–18	64.4	65.2	62.6	61.5	56.7	56.6	54.4	53.5	54.0	47.3	40.5	54.4
18–20	82.4	82.1	81.1	79.6	77.2	74.8	73.8	75.9	72.1	66.2	63.5	65.7
20–22	100.7	102.1	100.0	100.7	101.5	100.7	99.4	103.4	102.3	97.7	97.6	91.9
22–24	118.5	123.4	120.7	123.3	125.7	128.0	133.7	142.8	146.9	143.3	139.5	126.9
24–26	132.4	143.6	142.1	148.0	152.0	161.9	170.8	189.9	199.6	197.7	197.8	186.3
26–28	143.4	170.1	166.6	175.2	189.1	202.2	220.5	240.9	261.7	269.7	268.5	263.6
28–30	184.2	213.6	202.3	203.9	220.6	239.8	268.0	288.5	328.9	346.4	349.2	341.2
30–32	209.9	281.6	251.6	269.2	266.1	273.9	299.7	315.0	368.2	394.0	383.9	402.7

Velocities are $\times 10^{-4}$ cm s^{-1}.

Table 12b. Northern Hemisphere Settling Velocities of Aerosol Particles Used in the Strontium 90 Simulation

	Latitude Zones, deg											
Altitude, km	0–5	5–10	10–15	15–20	20–25	25–30	30–35	35–40	40–45	45–50	50–55	55–90
0–2	11.5	9.3	11.1	10.4	9.7	10.5	13.4	8.6	6.5	9.7	8.4	10.2
2–4	11.6	9.4	11.1	10.5	9.8	10.6	13.5	8.7	6.7	9.8	8.5	10.2
4–6	15.4	13.8	14.6	13.1	13.6	13.7	16.6	12.1	9.7	13.1	10.6	13.6
6–8	16.9	13.6	15.2	13.6	14.7	13.8	13.1	14.3	13.2	13.2	13.1	13.1
8–10	20.8	19.2	22.6	21.4	20.9	20.6	16.8	19.0	16.4	17.0	13.7	14.8
10–12	25.8	27.6	30.3	24.2	25.1	27.7	23.6	26.7	25.7	27.1	21.9	24.3
12–14	30.3	34.5	35.4	34.8	31.7	32.4	33.6	31.9	32.4	34.9	33.3	32.1
14–16	39.1	44.3	45.0	45.5	47.1	43.4	44.6	44.3	42.3	50.5	44.7	43.3
16–18	54.2	45.9	49.1	50.4	54.3	55.9	58.2	58.0	59.4	59.5	58.8	57.2
18–20	69.6	70.5	61.9	66.6	68.6	72.6	76.2	77.2	79.7	77.6	78.6	77.3
20–22	92.4	94.3	93.7	98.3	94.0	98.2	99.1	99.8	100.9	98.7	98.8	99.1
22–24	123.1	131.8	136.5	140.4	128.3	131.2	126.6	127.2	127.1	126.7	126.5	126.8
24–26	179.8	192.7	198.7	192.6	174.4	174.0	155.5	154.2	155.0	157.0	160.2	155.6
26–28	265.1	270.2	264.7	251.8	225.2	220.2	178.9	173.6	186.9	179.3	176.2	183.2
28–30	352.9	353.7	339.7	318.2	297.8	267.9	223.8	211.5	202.1	234.5	239.7	234.1
30–32	394.9	385.7	379.3	348.3	322.8	291.9	302.2	272.0	265.1	284.0	273.9	282.5

Velocities are $\times 10^{-4}$ cm s^{-1}.

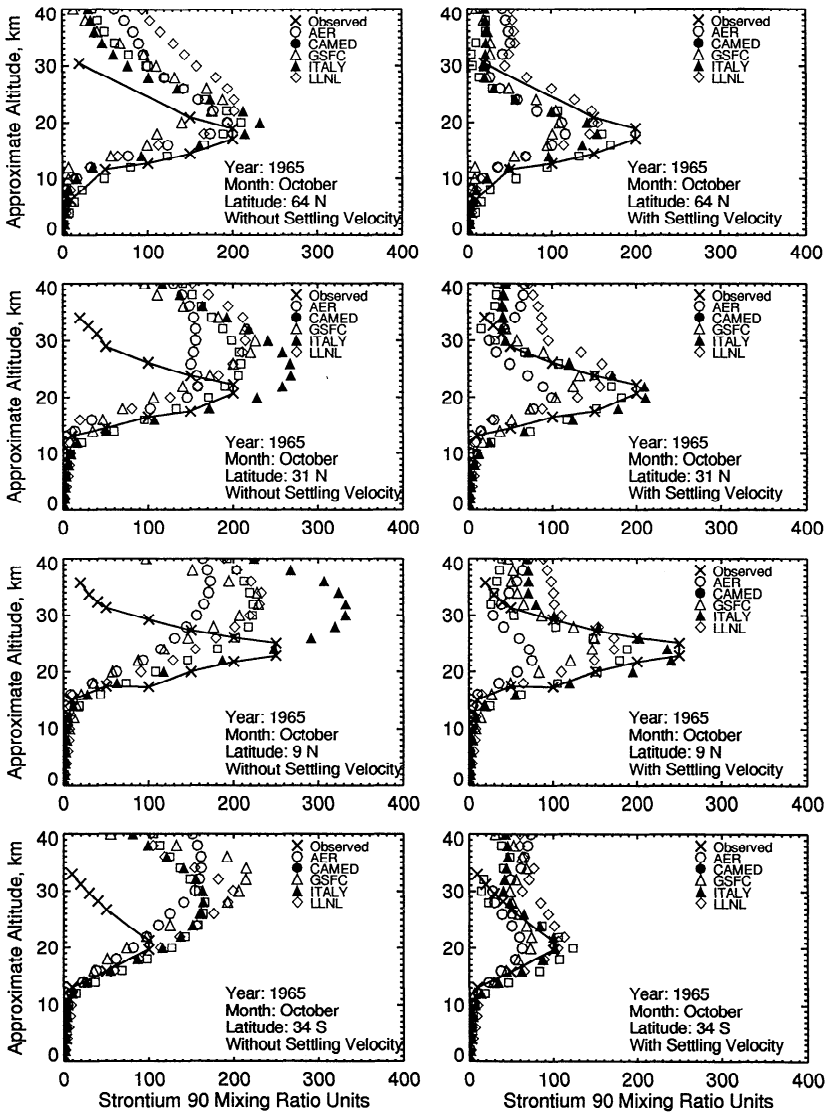

Figure 9. Model-derived strontium 90 based on initial conditions of observed strontium 90 in October 1964 (Figure 1d), which are compared with observed data at 64°N, 31°N, 9°N, and 34°S. For each time period, results are shown for strontium 90 treated as a gas and as an aerosol particle, that is, with and without settling velocities. Profiles are in units of disintegrations per minute per 1000 cubic feet (28 m^3) of standard air. These units are proportional to mixing ratio. Multiply by 0.013 to obtain picocuries per kilogram of air.

this radionuclide study is to simulate the concentration of strontium 90 from October 1964 to January 1967, we assume that in this period there are only "background" aerosol particles in the stratosphere.

The satellite-borne Stratospheric Aerosol and Gas Experiment (SAGE) II provides global coverage of the properties of aerosols and some tracer gases. The multiwavelength aerosol extinction measured by SAGE II can be used to deduce aerosol size [*Yue et al.*, 1986]. Analysis of SAGE II data shows the loading of aerosol particles in the stratosphere has gradually decreased since the eruption of Nevado del Ruiz in November 1985. In this study, we assume the aerosol particles have returned to "background" levels by 1989. We used the January and February 1989 SAGE II data to deduce the global distribution of aerosol size. The settling velocities were then calculated as a function of altitude and latitude from 0 to 31 km (Tables 12a and 12b), and we assumed that the aerosol above 31 km has a negligible settling velocity, as if a gas above 31 km. (Note: Our decision to treat the aerosol as if a gas above 31 km was a mistake; many model vertical profiles show an unreal change of slope at 31 km and some show an unreal secondary peak above 31 km [*Prather and Remsberg*, vol. 1, pp. 62–90] (see Figure 9). This mistake does not limit the validity of the data for comparisons below 25 km).

In Figure 9 a small sample of the strontium 90 profiles derived by the models (October 1965; 64°N, 31°N, 9°N, and 34°S) is compared to the observed profiles cataloged by *Johnston* [1989]. For each time period, results are shown for strontium 90 treated as a gas (left-hand panels) and as an aerosol particle below 31 km (right-hand panels) for the 7 models that participated in this study. Figure 9 suggests two significant comparisons. First, comparison of the left-hand panels versus the right-hand panels shows that the strontium 90 profiles derived by the models disagree in a major way at all altitudes above 20 km with the observed data when

Table 13. Strontium 90 Global Stratospheric Residence Times Based on Linear Least Squares Regression Analysis

Model	Stratospheric Residence Time, years	
	With Settling Velocities	Without Settling Velocities
AER	0.9	1.8
CALJPL	1.4	NA
CAMED	1.6	2.9
GSFC	1.1	1.8
Italy	2.1	3.9
LLNL	1.5	2.3

The model-derived burden was integrated globally from 16 to 60 km. NA, not available.

settling velocities are not incorporated into the scenario (compare *Johnston* [1976] and *Kinnison* [1989]). When the settling velocities are used, the agreement between model-derived and observed distributions of strontium 90 below 30 km is comparable to that between the models and carbon 14 data. The conclusion is that particulate settling is important for aerosols below 31 km altitude. Second, in the right-hand column of panels in Figure 9, the disagreement with models and measurements above 31 km is a result of our false assumption that strontium 90 attached to aerosols does not undergo particulate settling below 31 km. These large discrepancies give an additional conclusion: Aerosol settling is important even above 31 km.

In Table 13, the global stratospheric residence times (between 16 and 60 km) for strontium 90 are shown for the time period of October 1964 through October 1966. Observed global stratospheric residence times are not available, but *Johnston* [1989] evaluated residence times of the vertical columns at 64°N, 31°N, 9°N, and 34°S, where the values ranged from 1.0 to 1.6 years. The models yield global stratospheric residence times between 0.9 and 2.1 years when settling velocities are included. Without settling velocities, the range of model-derived global stratospheric residence times is from 1.8 to 3.9 years.

With only model results that include particle settling, the ratios of calculated strontium 90 to observed strontium 90 are presented in Table 14 for each of six models, at four latitudes, and for the eight times when data above 20 km were reported. At each latitude the altitude is that of the observed maximum strontium 90 mixing ratio: 17 km at 64°N, 20 km at 31°N and 34°S, and 24 km at 9°N. A systematic decrease of this ratio with time indicates model removal of tracer gas faster than that observed. The individual models are discussed here one by one.

The AER model shows the ratio ^{90}Sr (calculated)/^{90}Sr (observed) to decrease strongly with time at all four latitudes (Table 14). The interpretation is that this model sweeps strontium 90 out of the lower stratosphere at a rate much faster than that observed. To a less extreme extent, the GSFC model shows the same pattern. The CALJPL model shows the calculated/observed ratio to decrease at 31°N and 9°N and to remain about constant at 64°N and in the southern hemisphere. On balance, the evidence is that these three models remove strontium 90 from the stratosphere at a rate faster than that observed.

The LLNL model shows a similar pattern: a systematic decrease of the calculated/observed ratio at 31°N, and weak or no trends at 64°N, 9°N, and 34°S. At the three latitudes with no distinct trends, the average value of the calculated/observed ratio is close to 1.

The CAMED model shows no trend of the calculated/observed ratio with time, and the average value of this ratio is close to one at all three latitudes. The interpretation is that this model removes strontium 90 from the stratosphere at the same rate as that observed.

The Italy model shows an increasing trend of the calcu-

Table 14. Ratio ^{90}Sr (Calculated)/^{90}Sr (Observed) at the Altitude of Maximum Observed ^{90}Sr for Each of Four Latitudes, Including Settling Velocities of Stratospheric Junge Particles

Model	Date	64°N/17 km	31°N/20 km	9°N/24 km	34°S/20 km
AER	Oct. 1964	1.00	1.00	1.00	1.00
	Jan. 1965		0.86		
	April 1965	0.97	0.83	0.43	1.00
	July 1965	0.78	0.53		0.78
	Oct. 1965	0.56	0.51	0.29	0.62
	Jan. 1966		0.46	0.25	
	April 1966	0.57	0.39	0.22	0.72
	July 1966	0.38	0.31		0.30
	Oct. 1966		0.30	0.22	0.44
CAMED	Oct. 1964	1.00	1.00	1.00	1.00
	Jan. 1965		1.06		
	April 1965	1.03	1.17	0.77	1.08
	July 1965	0.95	0.87		1.08
	Oct. 1965	0.85	0.93	0.76	0.97
	Jan. 1966		1.00	0.80	
	April 1966	1.14	1.00	0.87	1.26
	July 1966	0.95	1.00		0.74
	Oct. 1966		0.94	1.00	1.40
CALJPL	Oct. 1964	1.00	1.00	1.00	1.00
	Jan. 1965		0.80		
	April 1965	1.36	0.93	0.41	1.08
	July 1965	0.95	0.54		1.08
	Oct. 1965	0.70	0.63	0.32	0.97
	Jan. 1966		0.75	0.34	
	April 1966	1.45	0.83	0.39	
	July 1966	0.87	0.63		0.74
	Oct. 1966		0.60	0.43	1.20
LLNL	Oct. 1964	1.00	1.00	1.00	1.00
	Jan. 1965		1.02		
	April 1965	0.98	1.12	0.71	1.00
	July 1965	0.86	0.95		1.12
	Oct. 1965	0.69	0.66	0.70	1.05
	Jan. 1966		0.80	0.69	
	April 1966	1.08	0.86	0.71	1.24
	July 1966	0.82	0.77		0.72
	Oct. 1966		0.64	0.84	1.20
GSFC	Oct. 1964	1.00	1.00	1.00	1.00
	Jan. 1965		0.77		
	April 1965	0.68	0.75	0.69	0.95
	July 1965	0.65	0.52		0.90
	Oct. 1965	0.50	0.53	0.59	0.72
	Jan. 1966		0.54	0.55	
	April 1966	0.54	0.51	0.53	0.84
	July 1966	0.46	0.48		0.42
	Oct. 1966		0.42	0.53	0.60
Italy	Oct. 1964	1.00	1.00	1.00	1.00
	Jan. 1965		0.92		
	April 1965	0.86	1.17	0.82	1.19
	July 1965	0.91	0.67		1.10
	Oct. 1965	0.75	1.08	0.96	1.00
	Jan. 1966		1.13	0.96	
	April 1966	0.98	1.20	1.12	1.80
	July 1966	0.94	1.28		0.87
	Oct. 1966		1.29	1.42	1.46

lated/observed ratio at all four latitudes. The interpretation is that this model removes strontium 90 from the lower stratosphere more slowly than the measured values.

The strontium 90 data test two things at once, the model for settling velocity and the model of stratospheric air motions, all models using the same settling velocities. If the model for settling velocities is correct, the test gives fairly well focused information about removal of material from the lower stratosphere. Two models, CAMED and LLNL, that agree well with the strontium 90 data do not agree nearly so well with the carbon 14 data, especially the long-term and middle stratospheric tests. These differences may arise from features that control exchange between the lower and middle stratosphere.

4. Discussion

The carbon 14 data provide a unique inert gaseous tracer for following stratospheric motions, and it is desirable to get as much information out of the data as is cost- and time-effective. However, the sampling in the stratosphere is sparse, each profile is the average over a 3-month period, and as for any observation there is random error and possibly systematic error. The model results are compared to the stratospheric measurements, not as if the observed data are absolute standards, but in an effort to obtain new insight about the models and about the atmosphere.

No single model even approximately tracks all the carbon 14 data from October 1963 to November 1970, but among the 11 models almost all the carbon 14 measurements are included within the spread of model results, except for a few points: at 31°N, April 1964 to January 1965; at 42°S, 20 km, January 1964 to April 1965; and at 42°N to 34°S, 20 to 30 km, November 1970 (*Prather and Remsberg* [1993, vol. 3, chap. 1]) (compare Figures 4–6).

During the first year after the initial conditions, much of the carbon 14 number density (not mixing ratio) is in the high-latitude, low-altitude lobes of the stratosphere where there are lines of constant potential temperature connecting the stratosphere and troposphere, and the various models show widely different carbon 14 stratospheric residence times, from 1.3 to 3.2 years (Table 2). Two to seven years after initial conditions, most of the carbon 14 number density is at higher stratospheric altitudes where there are no lines of constant potential temperature connecting the stratosphere and troposphere, and the various models show a narrow spread of carbon 14 stratospheric residence times, from 4.3 to 5.4 years (Table 2). This comparison indicates that the models differ substantially among themselves with respect to stratosphere-troposphere exchange, but the differences among the models are small with respect to transport between the middle stratosphere and the lower stratosphere.

The measured carbon 14 vertical profiles at 31°N show a maximum mixing ratio in the altitude range 20–25 km from October 1963 through July 1966, but all modeled profiles show mixing-ratio maxima that increase in altitude from 20 km in October 1963 to greater than 40 km by April 1966 (Tables 6 and 7; Figure 4). For three methods of generating initial conditions, each of which agrees reasonably well with carbon 14 measurements in October 1963 and each of which has strongly different values outside the range of carbon 14 measurements, the calculated 30°N vertical profiles above the mixing-ratio maximum open upwardly much faster in all

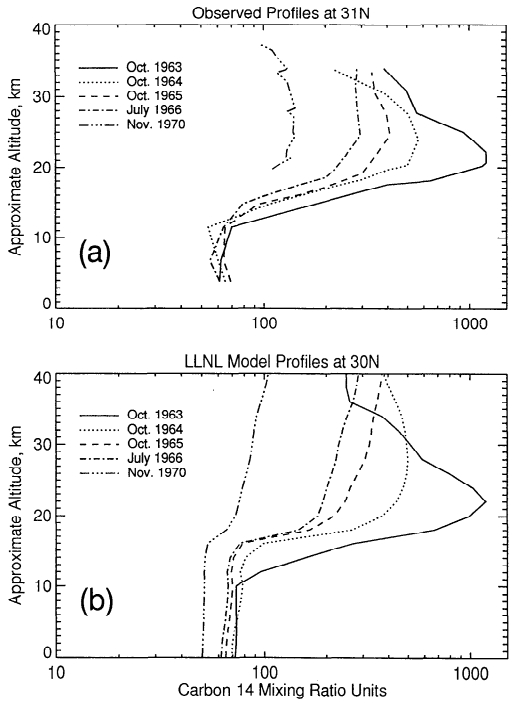

Figure 10. (a) Measured carbon 14 vertical profiles at 31°N between October 1963 and November 1970. (b) LLNL model-calculated vertical profiles for the same place and times as in Figure 10a. The other models show a similar pattern of qualitatively different shapes between the measured and modeled vertical profiles.

of the models than the carbon 14 observations (Tables 6 and 7 and Figures 4 and 8 [see *Kinnison*, 1989, pp. 178–183]). This difference in the shape of the vertical profiles at 31°N during almost 3 years is a qualitative difference between measurements and all the models.

The carbon 14 data for the fall of 1970 are the most extensive of the 1952–1971 series; they were taken almost 8 years after the end of high-altitude nuclear bomb tests, which distances them from assumed initial conditions. The analytical methods used to observe the carbon 14 represent the culmination of 20 years of practice and development within AEC. These data provide a unique, though limited, test of stratospheric motions using an inert tracer. In terms of magnitude, most of the measured data fall within the spread of the 10 models (Figure 6). The measured data show a decrease of carbon 14 mixing ratio with altitude above about 27 km at four latitudes of observation between 65°N and 34°S, and this decrease continues up to the topmost altitude, 36 km at 31°N (Figures 6 and 7). All the models at all latitudes are in qualitative disagreement with the shape of the measured carbon 14 vertical profiles for the fall of 1970.

Figure 10a gives the observed vertical profiles of carbon 14 at 31°N from October 1963 to November 1970, and Figure 10b gives the LLNL model profiles for the same cases. From this comparison it appears that the nature of the discrepancy between models and measurements at all four latitudes in the fall of 1970 is the same as the discrepancy in the upper part of the profiles at 31°N at almost all times.

The minimum with respect to latitude above the tropics (Figure 7) may be rationalized in terms of the Brewer model of stratospheric circulation, in that tropospheric air of rela-

tively low carbon 14 mixing ratio rises from the tropical troposphere into the stratosphere. The minimum with respect to altitude above midlatitudes seems to indicate a stream of the Brewer circulation carrying recent tropospheric air above 28 km or so overriding a midlatitude pocket of stratospheric air centered at about 25 km, which is subject to turbulent and other losses to the troposphere at its lower boundaries. The carbon 14 measurements seem to indicate that all models sweep carbon 14 out of the stratosphere too slowly at altitudes above about 28 km (Figures 6 and 7), and about three quarters of the models mix low stratospheric air into the troposphere too rapidly (Tables 4 and 5). It is possible that AEC had the same systematic error in both of its methods of sampling carbon 14 from balloons, but these data should not be rejected on the arbitrary assumption of this remote possibility. Atmospheric dynamicists should seriously consider what the carbon 14 measurements imply if their major features are correct.

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