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# Digital Simulation of the Global Transport of Carbon Monoxide

**Abstract:** A numerical model of the general atmospheric circulation is used to simulate the global transport of carbon monoxide. The sources are estimated from gasoline consumption data. Since the strongest sources lie in the northern hemisphere belt of strong prevailing westerly winds, "chaining" effects are quite pronounced. Emissions generated in one populous area are soon superimposed upon those of the next area downwind. Subscale cumulus convection, which is parameterized in the model, accounts for a significant fraction of the vertical CO transport.

## Introduction

The concentration and distribution of atmospheric carbon monoxide is a matter of current interest. Its role as one of the principal pollutants in regional air quality deterioration has long been known, and there is now some concern over a possible long-term buildup in the atmosphere at large. The current global rate of CO emission is estimated to be 200 million tons per year, and there appears to be no strong sink operating in the troposphere.

Robinson and Robbins [1] have documented rather high concentrations of carbon monoxide occurring far from populated areas. They reported measurements taken in north-central Greenland in which the mass fraction of CO varied during a four-day interval by a factor of 13 [50 to 650 parts per billion (ppb)]. A trajectory analysis revealed that the high concentrations were found in air masses that had become contaminated by previous travel over urban areas of North America. Robinson and Robbins also reviewed CO concentration data taken during an Antarctic cruise in January 1967. These data were somewhat scattered, but showed a definite and unexpected poleward increase of concentration. The 80-ppb mass fraction observed at latitude 76°S, longitude 174°E on January 20, 1967 is not readily explained by simple air-mass advection from populous areas.

On the more encouraging side, Pressman and Warneck [2] have pointed out that measurements taken over the past 20 years reveal considerable short-term fluctuations of the CO concentration in the atmosphere, but no apparent long-term buildup. The globally averaged CO mass fraction seems to persist at about 150 ppb. Since the estimated CO sources would double that value in three years, the apparent absence of tropospheric sinks cannot be the entire story. Some scavenging mechanism appears to be operating, either on the surface or in the stratosphere, with tropospheric CO migrating to the sinks sufficiently fast to offset the known sources.

Pressman and Warneck conjectured that a major sink lies in the lower stratosphere. The obvious mechanism of oxidation to $CO_2$ by stratospheric ozone is too slow, but they pointed out that there is another oxidation reaction, viz.,

$$OH + CO \rightarrow CO_2 + H, \tag{1}$$

which is highly effective. Hydroxyl radicals are abundantly present in the lower stratosphere because of the interaction of water vapor with photochemically generated excited oxygen atoms. Stratospheric photochemistry produces other species that could oxidize CO, but the reaction (1) appears to be dominant. Whether it is sufficient to account for the apparent absence of long-term CO buildup is not clear because of uncertainty in the migration rate of CO into the lower stratosphere. Citing current estimates of troposphere-stratosphere mass exchange, Pressman and Warneck [2] expressed the view that their sink mechanism "contributes significantly, but only partially to the overall removal of CO from the atmosphere."

During the last few years, the IBM authors of the present paper have been investigating digital simulation of the general atmospheric circulation [3]. In its present form the general circulation model cannot test the sink mechanism proposed by Pressman and Warneck [2] because it specifically neglects troposphere-stratosphere interaction. Nevertheless the model can address the problems raised by Robinson and Robbins [1] concerning the transport of CO to remote areas of the globe.

## Summary description of the general circulation model

During the last decade, long-term simulation of large-scale atmospheric motion has been undertaken by several research groups. With present-day computers it is marginally feasible to do a proper job of this. However, it is necessary to accept some compromises. For example, the present study uses a spatial resolution that is rather coarse, especially in the vertical direction. The price of grid refinement is not cheap: This short paper represents 18 hours of central-processing-unit time with the program running in a 500-kbyte partition of an IBM 360/91 computer.

Other general circulation models reflect the computer limitation in alternate ways, e.g., by simplifying the topography or the hydrologic cycle, or by restricting the simulation to a single hemisphere. By accepting these approximations one can afford a high-resolution vertical layering of the atmosphere that permits, for example, a detailed calculation of the radiative transfer. This latter calculation is carried out in the nine-layer model developed at the Geophysical Fluid Dynamics Laboratory (GFDL) of the Environmental Science Services Administration. A recent version of this model has been described by Manabe et al. [4]. The GFDL model can be used to investigate the detailed vertical variation of atmospheric temperature, which is completely beyond the scope of a two-layer model. On the other hand, the global coverage and detailed cumulus parameterization of the two-level model we use make it well suited for the present study.

It is fortunate that computer limitations can be accepted in various ways without grossly distorting the main features of the general circulation. Models can be developed with specific numerical experiments in mind. The examples cited are but two of many possible illustrations. It is worth noting in this regard that the studies of Leith [5] contributed significantly to general circulation research even though he was limited to the computers available in the early 1960's. An interesting approach to the problem has been reported by Oliger et al. [6]: The general circulation model developed at the National Center for Atmospheric Research was deliberately coded with a "modular" structure, permitting its users a choice of ways to accept computer limitations.

Our general circulation model is a version of that developed at UCLA by A. Arakawa and Y. Mintz, with the collaboration of A. Katayama. The essential features of the model are described in their paper [7]; a detailed description is set out in the series of reports by Langlois and Kwok [3].

In this model the troposphere is divided into two horizontal layers of equal mass. Specifically, "$\sigma$ coordinates" are used, i.e., the vertical coordinate is

$$\sigma = (p - p_T)/(p_S - p_T), \tag{2}$$

where $p_S$ is the surface pressure and $p_T$ is the tropopause pressure, which is taken to be a constant 200 mbar. Thus the upper tropospheric layer corresponds to $0 \leq \sigma < \frac{1}{2}$ and the lower layer to $\frac{1}{2} \leq \sigma < 1$. The earth's surface, which follows the elevation of the large-scale mountain systems, corresponds to $\sigma = 1$.

Horizontal differencing is carried out in the longitude-latitude plane. In this plane the image of the earth's surface is a rectangle of height $\pi$ and width $2\pi$. The finite-difference grid is constructed by subdividing this rectangle into a network of congruent rectangular cells. For the study reported here, each cell's dimensions are 5° of longitude by 4° of latitude (the "fine-grid" version documented in the reports by Langlois and Kwok [3]).

The surface underlying each grid cell is specified as being ice-free ocean, sea ice, ice-free land, glacier, or snow-covered land. The temperature of open ocean surface is a specified function of season and position. Land surface is regarded as a thermal insulator with no capacity to store heat; its temperature is determined by balancing incoming and outgoing thermal fluxes. If the land is ice- or snow-covered, this temperature is constrained not to exceed the melting point of ice. Sea ice is treated like ice-covered land, except that there is some heat conduction through the ice.

The dependent variables are the temperatures and the horizontal wind velocities of the two layers, the surface pressure, and the mixing ratio of the lower layer (the moisture content of the relatively cold upper layer is neglected).

Radiative heating and cooling depend on the mixing ratio and on the nature and extent of the cloud cover. Three types of clouds are distinguished: stratus deck, which results from large-scale condensation occurring when the mixing ratio exceeds its saturation value; cumulus towers, which result either from middle-level convection or from penetrating convection; and low-level cumulus clouds, which result from boundary-layer convection and which produce no rain. The physical theories of radiation and cloud parameterization used in the model are described in the appendices of the paper by Arakawa, Mintz and Katayama [7]. Insolation at the top of the atmosphere depends on latitude, season and local time of day.

The moisture source in the model is evaporation from the open sea, from ice- or snow-covered surfaces, and from ice-free land that has previously been moistened by rain. The evaporation rate depends on the surface wind speed and on the vapor pressure difference between the air and the surface. For ocean, ice and snow, the surface vapor pressure is the saturation value for the surface temperature; for ice-free land it depends on the wetness of the ground, which in turn depends on the history of precipitation, evaporation and runoff.

### Large-scale transport of carbon monoxide

In any atmospheric-contaminant transport study, one should allow for the possibility of feedback because the presence of the contaminant may disturb the weather processes to a significant extent. In the case of a gaseous pollutant, weather disturbances usually result from 1) specific heat or effective molecular weight differences between polluted air and clean air, and 2) variations in radiative transfer [8].

Mechanism 1 can be rejected at once. The effects of a trace constituent on the specific heat and effective molecular weight of air are directly measured by the mass fraction of the contaminant—a few ppm at most. In the case of CO one has, *a fortiori*, a gas whose specific heat and molecular weight coincide with those of atmospheric nitrogen to four significant figures.

Mechanism 2 warrants more discussion. Low concentration does not guarantee negligible influence on the radiative transfer, ozone being an obvious counterexample. However, the meteorologically important part of the CO molecular absorption spectrum consists of only two narrow bands in the near infrared region, viz., the fundamental vibration-rotation band at 4.47 to 4.89 micrometers and the relatively weak first overtone at 2.31 to 2.39 micrometers. Only one-half percent of the solar radiation, and virtually none of the terrestial radiation, falls within these bands. Higher overtones are far too weak to have any meteorological significance. The only electronic absorption bands that might be important, the fourth positive group, are superposed by the Lyman–Birge–Hopfield bands of molecular nitrogen. Thus it is clear why Kondratyev [9] dismisses the effect of carbon monoxide on atmospheric radiation as "of no practical importance."

In view of these observations we proceeded on the assumption that carbon monoxide is meteorologically passive. There is no significant way it can perturb the principal mechanisms of its large-scale transport.

Advection by the simulated wind is the only mechanism in our model for the horizontal transport of CO. Horizontal CO diffusion by sub-grid-scale eddies is neglected.

Vertical transport is also effected by large-scale advection. In addition, we account for another mechanism: vertical redistribution by cumulus convection. This is a sub-grid-scale effect, but it can be simulated in a consistent way. As we indicated in the preceding section, the moisture—and hence the latent heat—is carried entirely by the lower layer. However, when this heat is released during precipitation it is divided between the upper and lower layers in a ratio determined by the cumulus parameters. This ratio provides a measure of the overturning of air by cumulus activity, and hence of the vertical CO redistribution.

The importance of cumulus overturning, relative to large-scale vertical advection, was tested by repeating part of the simulation with cumulus CO redistribution suppressed. The results of this test are described in the final section.

### Carbon monoxide sources

According to Jaffe [10], emissions from gasoline-powered road vehicles account for 91 percent of all CO from major technological sources (based on 1966 data for the United States). Moreover, the geographic distribution of the other nine percent probably correlates well with that of the automotive sources. Hence it was essential to the study that we determine the global distribution of carbon monoxide sources from automobiles. The primary problem came in matching the significant variables (number of automobiles and miles driven per year as reported only by areal administrative units) to the cellular grid superimposed over the terrestrial model. This was achieved by first plotting the cell boundaries on large-scale maps showing population, economic activities, surface relief, generalized land use and transportation networks. By this process all parts of a country, e.g., densely populated, highly developed sections or sparsely populated areas of low development, could be delimited and an appropriate number of the country's total automobiles assigned to each cell. By multiplying the number of vehicles by the average distance driven per year in the country or state involved [11], and by multiplying this product by the CO production rate per vehicle [12], we obtained the annual CO production for the cell. When portions of two or more countries fell within a cell, the procedure was modified in an obvious manner.

Quite recently Swinnerton, Linnenbom and Lamontagne [13] pointed out that the oceans may act as a background source for carbon monoxide. They cited references which report that marine algae, the Portuguese man-of-war and some siphonophores produce carbon monoxide. Their own data, taken at two stations in the tropical western Atlantic Ocean, revealed concentrations of dissolved CO sufficiently high for a net transfer from ocean to atmosphere. However, if their results are typical of the world's oceans, the total oceanic source amounts to only five percent of the known anthropogenic source. Our *a priori* guess is that the source strength is lower still, since the data were taken in a region of atypically high biological activity. In view of its presently uncertain magnitude and apparently small relative importance, the oceanic source was omitted from the present study.

The carbon monoxide sources operate in the lower layer only. The total mass of lower-layer air overlying

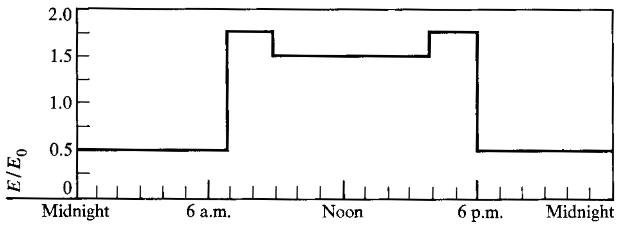



**Figure 1** Model of the diurnal variation of CO source emissions.

a grid cell of area $A$ is $A(p_S - p_T)/2g$, where $g$ is the acceleration due to gravity. Thus if $E$ denotes the mass emission rate of the sources operating in the grid cell, the contribution of these sources to the CO mass-fraction tendency is given by $2gE/[A(p_S - p_T)]$.

Seasonal variation of $E$ was neglected. However, we attempted to model the diurnal variation that results from heavier vehicular traffic during the daytime, especially at peak commuting hours. If $E_0$ denotes the average emission rate, we use $E = \frac{1}{2}E_0$ during the night, $E = \frac{7}{4}E_0$ during commuting hours (taken as 7 to 9 a.m. and 4 to 6 p.m. local time), and $E = \frac{3}{2}E_0$ during the day (see Fig. 1).

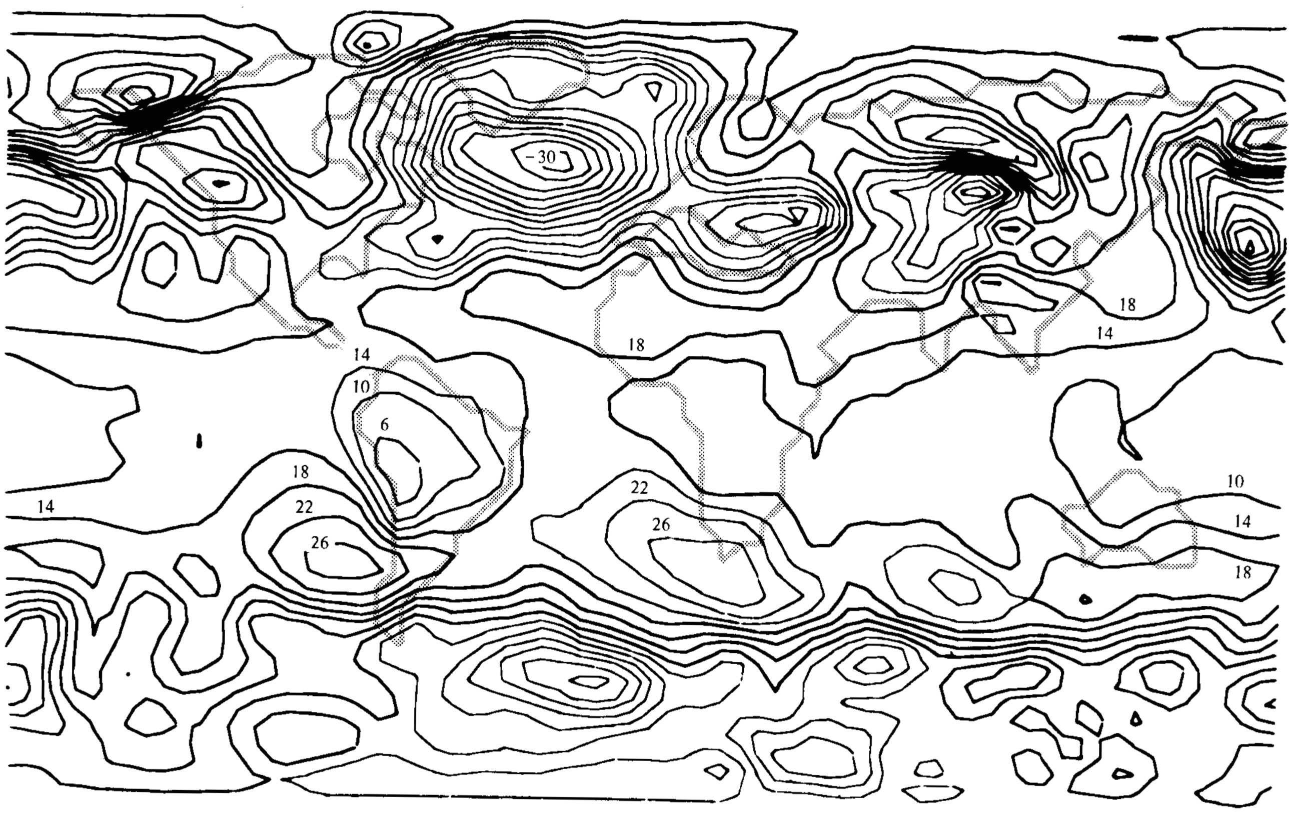
There is a data-dependent variation of the sources that was omitted from this study but which could be incorporated into a more sophisticated treatment. The emissions in a grid cell may be temporarily trapped under a subsidence inversion. The grid cell then suffers a "smog attack." Since the emissions do not escape to the free atmosphere, the sources in the cell are inoperative as far as the general circulation is concerned. When the inversion breaks up, several days' emissions are released in a relatively brief period of time.

Since our numerical simulation begins with a "clean" atmosphere, and since atmospheric sinks of CO are weak, rather diffuse and poorly understood, we made no attempt to model the carbon monoxide sinks. In a very-long-term simulation—three simulated years or so—some sink mechanism would be needed to prevent an unrealistically high buildup of CO. For the four-week simulation reported here, this consideration was unimportant.

## Simulation

The general circulation model was run for 24 simulated days with no CO source operating. The purpose of this preparatory simulation was to build up a realistic circulation pattern. If this were not done, the CO transport

**Figure 2** Sea-level atmospheric pressure contours at the end of the preparatory 24-day simulation period. The contour interval is 4 mbar; isobar labels indicate pressures above 1000 mbar.



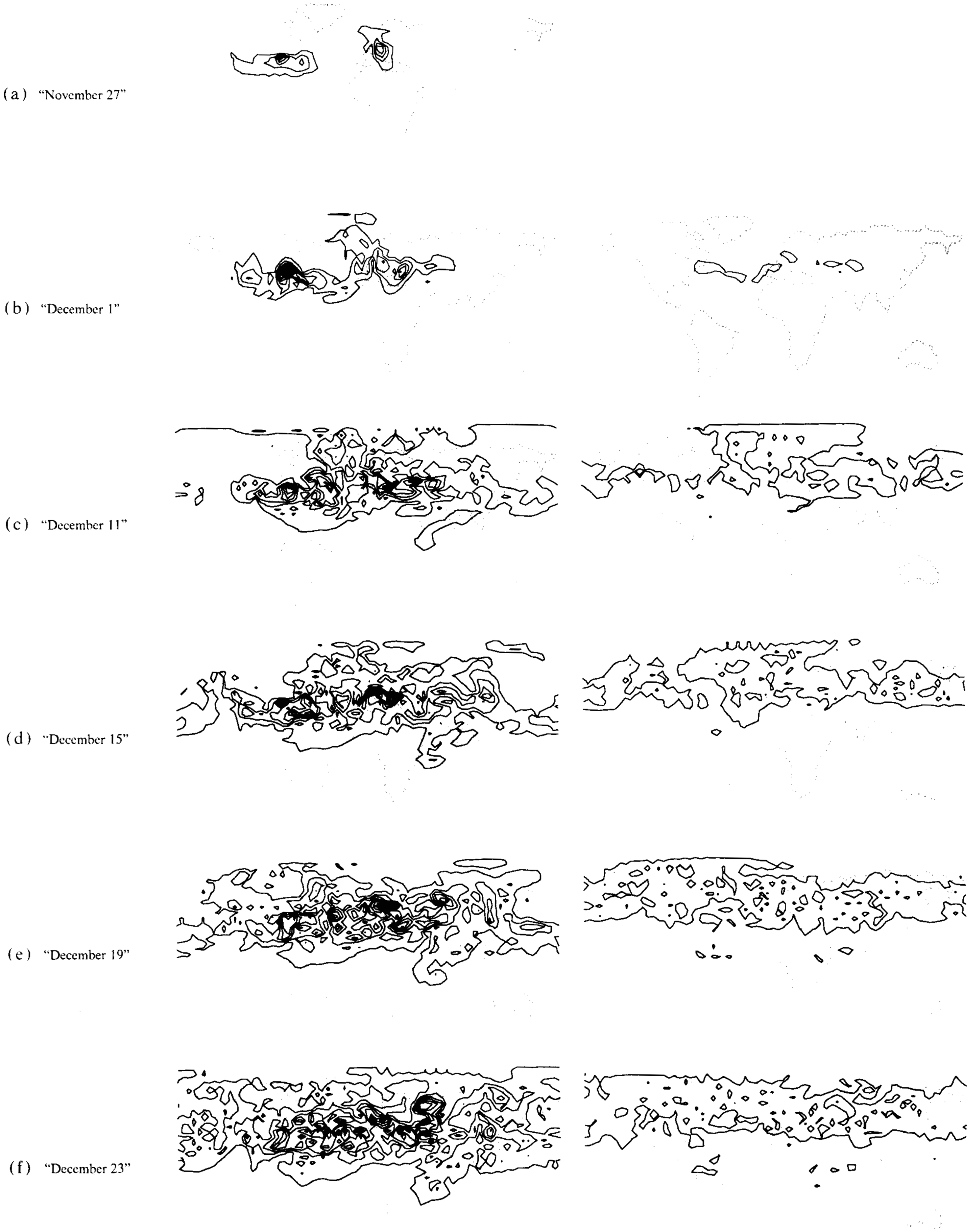



**Figure 3** Simulated CO concentration contours for the days indicated at 00:00 GMT; diagrams on the left are the lower-layer contours, those on the right are the upper-layer contours. The contour interval is 10 (CO) ppb.

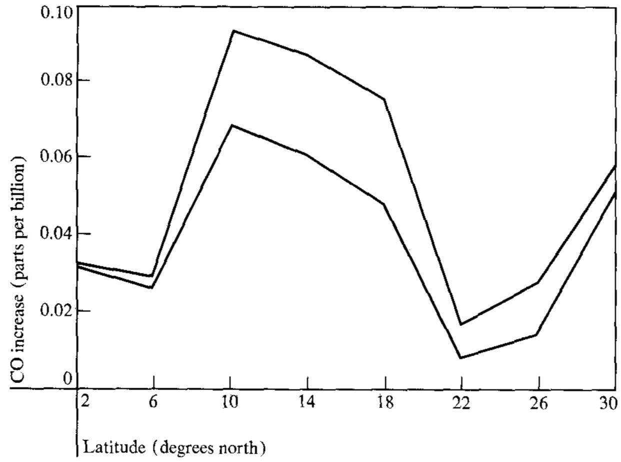



**Figure 4** Increase in the zonal-mean mass fraction of CO in the upper layer during the four-day test. The lower curve was obtained with cumulus redistribution suppressed.

would have been dominated by the Hadley circulation that persists for several days after a cold start. The seasonal parameters (latitudes of the north and south snow boundaries and declination of the sun) at the beginning of the preparatory simulation corresponded to their values for November 1. The surface temperatures for oceanic grid cells were assigned values for northern hemisphere winter and were left unchanged throughout both phases of the simulation. At the end of the 24-day simulation, the atmospheric pressure reduced to sea level had the pattern shown in Fig. 2. The contour plots were generated on an IBM 2250 Model III display unit using the plotting routine developed by Schreiber [14],

At this point the CO sources were turned on. During the next few simulated days, most of the CO remained near the major sources in North America and western Europe. However, the sources were in a strong belt of prevailing westerly winds. Hence the CO mass-fraction contours associated with the American sources extended well out over the northern Atlantic Ocean after only two days [Fig. 3(a)]. After six days the contours became chained to those of the European sources [Fig. 3(b), left]. At about the same time, discernable contours appeared in the upper layer [Fig. 3(b), right].

The next ten days of the simulation proceeded uneventfully, leading to the CO distribution shown in Fig. 3(c). The most noticeable feature in the lower layer at this time is the penetration of contours into the southern hemisphere in the region of the Asiatic winter monsoon. The upper-layer concentrations are of course weaker, but the stronger winds have spread the contours throughout most of the northern hemisphere westerly-wind belt. Four days later [Fig. 3(d)] the lower-layer contours also encircled the northern hemisphere.

In the final stages of the simulation [Figs. 3(e) and 3(f)] we observe in both layers closed isopleths in source-free regions such as the tropics. This condition appears to result, at least in part, from vertical exchange: In regions of lifting or subsidence, CO from one layer is superimposed on the CO already present in the other layer, producing a high local concentration. This effect suggests a similar explanation for the anomalous southern hemisphere data reported by Robinson and Robbins [1]: high-level transport to the Antarctic region, followed by subsidence in the polar anticyclone and return flow at lower levels.

## Significance of cumulus convection as a vertical exchange mechanism

To assess the relative importance of large-scale vertical advection and cumulus overturning as vertical CO exchange mechanisms, we deleted the portion of the simulation program that models the cumulus CO redistribution and repeated four days of the calculation—"December 11" through "December 14." Cumulus activity per se was not suppressed, for this would have changed the entire meteorological pattern; only the CO distribution was affected by the actual change.

During the four-day interval the global-mean mass fraction of CO in the upper layer increased by 21.6 percent with cumulus CO redistribution in the model and by 19.6 percent without it. Thus it appears that during the northern hemisphere winter about ten percent of the global-mean vertical CO transport results from cumulus overturning.

As one might expect, however, cumulus CO redistribution is more important in the tropics than the global-mean percentages indicate. At 18°N latitude cumulus overturning represented 35 percent of the zonal-mean increase in the upper-layer CO mass fraction (Fig. 4). This trend obtained to a lesser degree throughout the northern hemisphere tropics.

The special significance of cumulus overturning in the tropics has import with respect to inter-hemisphere CO exchange. In general, the trans-equatorial winds are stronger in the high troposphere and the four-day test indicates that the upper-layer air just north of the equator is appreciably more contaminated than it would be without cumulus CO redistribution.

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