CHAPTER 6

Regional and Global Transport and Distribution of Trace Species Released at the Earth's Surface

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INTRODUCTION

Most atmospheric trace constituents, including pesticides and other toxics, are transported as mere passengers and do not influence the motions of the winds. Their time-dependent behavior is influenced by surface or near-surface emissions, the atmospheric winds, physical removal processes, and chemical destruction and production. This is summarized in the following descriptive equation:

$$\frac{\partial \text{Tracer}}{\partial t} = \text{Source} + \text{Transport} - \text{Sink} - \text{Chemical Destruction} + \text{Chemical Production}$$

where SOURCE is some form of surface or near-surface emission, TRANSPORT is the result of three-dimensional winds acting on gradients in the TRACER mixing ratio (mole fraction), SINK is a physical removal process that is a function of TRACER solubility and surface reactivity, CHEMICAL DESTRUCTION can be either direct chemical reaction or photodissociation, and CHEMICAL PRODUCTION converts an emitted compound into a toxic. If the trace gas or particle is uniformly mixed throughout the globe (i.e., there are no gradients in its mixing ratio or mole fraction), TRANSPORT will be 0. However, this is not the case for modern pesticides. The long-lived ones have only been released for a few decades and have not had time to become well mixed, and the short-lived ones, although they have reached a balance between emission and destruction, have large spatial gradients in their distribution. In all cases, the atmospheric winds play a key role in the global distribution of pesticides and other toxics.

SOURCE

We will first consider a number of observations and model simulations that demonstrate the impact of source nonhomogeneity on tracer transport and distribution.

In the first simulation, a tracer with no sinks (i.e., an infinite lifetime) was uniformly released for 10 days from a 10° midlatitude belt stretching around the globe in the Northern Hemisphere boundary layer. The simulated time evolution of the resulting distribution, averaged around latitude circles, is presented in Figure 1 as a function of latitude on the horizontal axis, and pressure (mb) on the left or height (km) on the right vertical axis. After 10 days of integration, the tracer remains in the northern midlatitudes, though it has started to mix into the free troposphere. After 3 months, it is relatively well mixed throughout the Northern Hemisphere, but very little has been carried across the equator. In 1 year, the tracer has mixed into both hemispheres, though there is still an interhemispheric gradient, which should disappear in approximately 10 years.

When the emissions of very long-lived chemicals are continuous, though not uniform, around the globe (both nitrous oxide and chlorofluorocarbons are good examples), we still find gradients and are able to observe transport events. Chlorofluorocarbons, with their very long lifetimes ($\approx$ 100 yrs), have distributions that have not yet equilibrated in the nearly 40 years of their release. They still show an excess ($\approx$5%) in the Northern Hemisphere.[1] Nitrous oxide, which has been released naturally for millions of years, has a very small interhemispheric gradient ($\approx$0.3%) that appears to be the residual result of a Northern Hemisphere bias in the source distribution. Both trace gases show considerable variability in the neighborhood of strong local sources,[2] as the simulated time series in Figure 2 demonstrate. Even with the very well-mixed background of nitrous oxide, one can simulate transport events in the vicinity of strong local sources, such as Panama, where the surface mixing ratio varies between 290 ppbv (parts per billion by volume) and 315 ppbv. However, remote regions with no sources, such as Samoa, show almost no variability in the simulated mixing ratio.

TRANSPORT

In this chapter, long-range transport refers to transport from continental source regions to the oceans and to other continents, as well as transport between the hemispheres.

Winds

Not only are atmospheric motions three-dimensional, but they occur on spatial scales ranging from meters for local turbulence, to kilometers for cloud

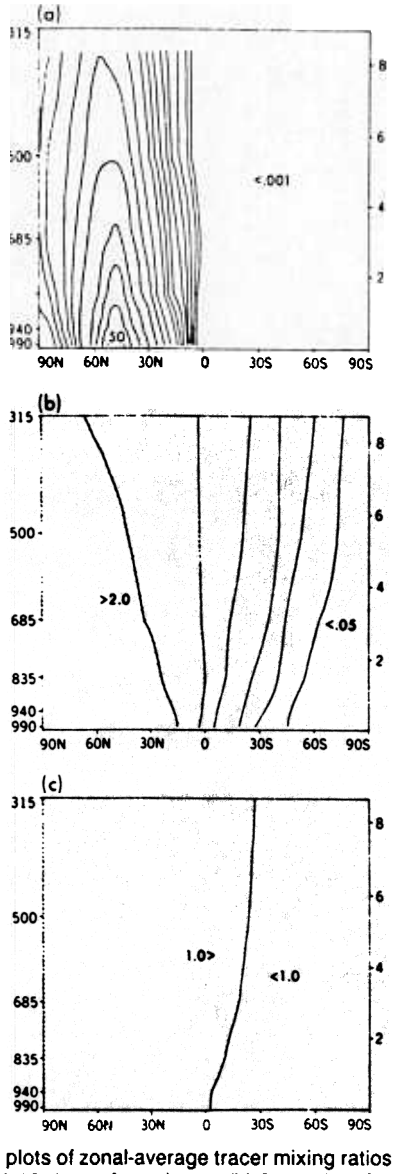

Figure 1. Latitude-altitude plots of zonal-average tracer mixing ratios (in dimensionless units) are shown for (a) 10 days after release, (b) 3 months after release, and (c) 1 year after release. The contours are in logarithmic intervals (1–2–5) with the dotted area > 1.0.

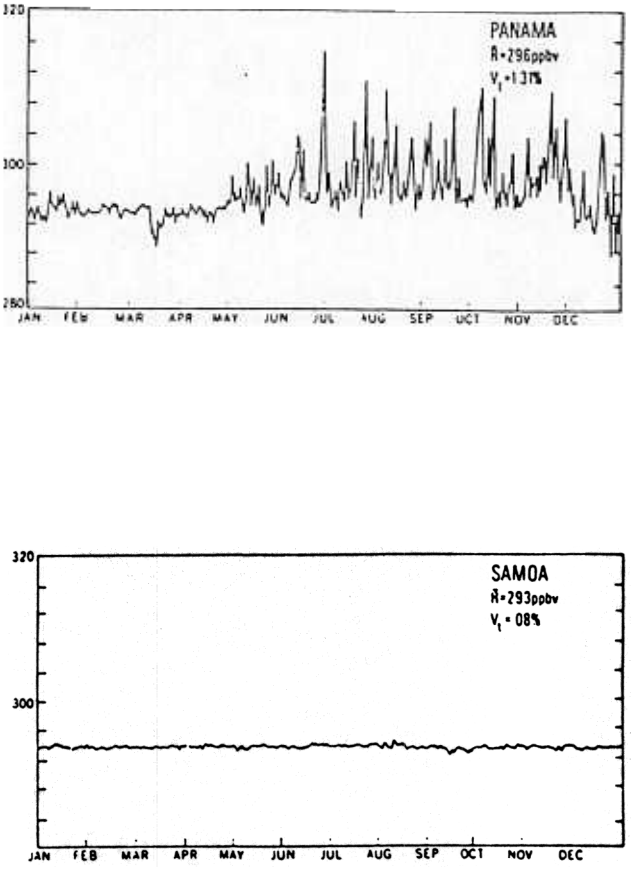

Figure 2. One-year simulated surface time series of nitrous oxide (ppbv) for two tropical locations, one that has nearby emission sources (Panama) and one that is near no sources (Samoa). *Source*: Levy, H., II, J. D. Mahlman, and W. J. Moxim. "Tropospheric N_2O Variability," *J. Geophys. Res.* 87:3061–80 (1982).

convection, and up to planetary-scale waves that circle the globe. To simplify these time-dependent three-dimensional motions and identify basic transport processes, we average the atmospheric winds over space and time. Having identified potential transport paths from this highly averaged picture, we will see that the average winds do not quantitatively reproduce the average transport and, in some cases, may be quite misleading.

The simplest picture of the winds, generated by averaging over many years and over longitude (i.e., around latitude circles), does identify some important features of atmospheric transport. The yearly averaged E-W and N-S wind velocities are shown in Figures 3a and 3b as a function of latitude and pressure. Pressure is given in db, 1/10 of the surface atmospheric pressure; thus, 10 db is the surface, and 1 db is a standard height of ≈ 16 km, the

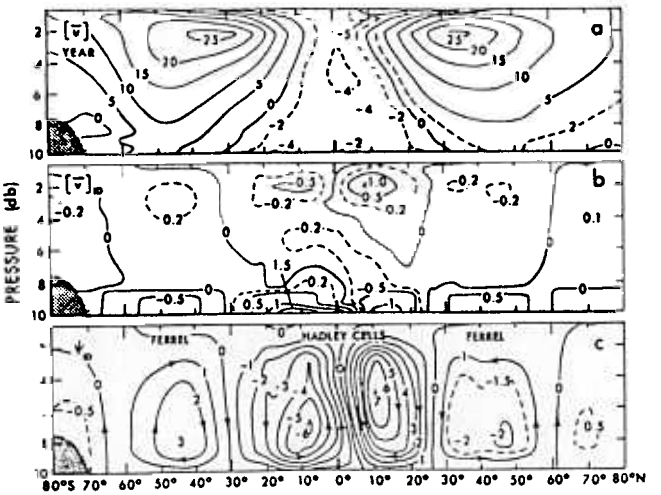

Figure 3. Zonal mean cross sections of (a) the east-west yearly-average wind in m/sec where negative values are from the east, (b) the north-south yearly-average wind in m/sec where negative values are towards the north, and (c) the yearly-average atmospheric mass flow in 10^{10} kg/sec. *Source*: Peixoto, J. P., and A. H. Oort. "Physics of Climate," *Rev. Mod. Phys.* 56:365–429 (1984).

approximate top of the troposphere. Note that the E-W velocities are from the east (easterly) in the tropics, from the west (westerly) at higher latitudes, and increase with height, reaching 25 m/sec in the upper troposphere of the midlatitudes (the jet stream). In contrast, the N-S velocities, though providing a more complex picture, are much weaker and seldom exceed 1 m/sec.

Because the average E-W winds are so strong, we can, as a first approximation, assume that an atmospheric tracer is well mixed in the E-W direction and focus on the slower transport that is driven by the time-averaged vertical and N-S winds. A standard two-dimensional meteorological picture of the time-averaged mass flow is shown in Figure 3c. A strong upward flux in the tropics and subsiding flux in the subtropics is produced by the thermally direct Hadley cells. The subtropics is that region lying between the tropics, a 20–30° latitude belt centered at the equator, and midlatitude. As a result of a statistical artifact, there is rising at midlatitudes and sinking in the subtropics. There is also very weak sinking at the Poles. This highly averaged picture, although statistically correct for the ensemble of atmospheric motions over the years, does not exist for the motion of a particular air parcel. Moreover, the time-averaged transport is even less complex. On the average, trace species are lifted in the tropics; they are carried poleward and descend in the subtropics, midlatitudes, and polar regions; and they are carried back to the tropics in the lower troposphere.

However, from these highly averaged wind fields, we can tell that

1. transport in the E-W direction will increase with height
2. air parcels will subside in the subtropics
3. air passing between hemispheres will rise in the tropics and be exposed to precipitation produced by the lifting of moist tropical air

Although soluble tracers, in the absence of chemical sinks, should be relatively long-lived in the dry and stable subtropics, their transport between the hemispheres is quite unlikely.

A more complex picture of the atmospheric motions is provided by Figures 4a and 4b, which show the seasonal average surface pressure and associated surface winds for January and July. Air flows from the subtropics into the tropics (i.e., converges in the tropics); areas of high pressure form in the subtropics and move north and south 10–15° with the sun. In the Northern Hemisphere winter, areas of high pressure also develop over the North American and Asian continents, while areas of low pressure form in the North

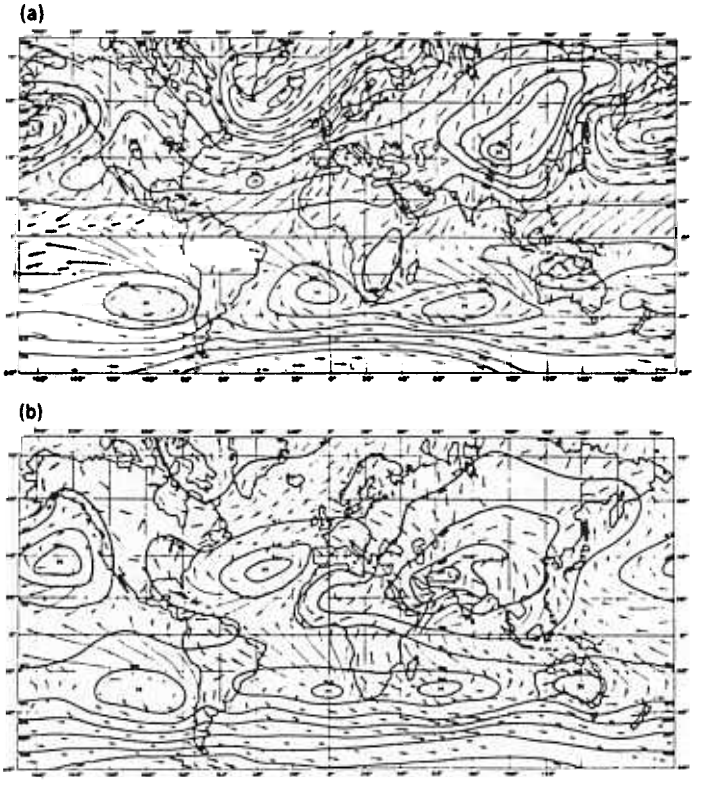

Figure 4. Surface pressure (mb) and surface wind direction and speed. The wind flows with the arrows; the darker the arrows, the faster the wind. (a) January averages; (b) July averages. *Source*: Hasse, L., and F. Dobson. Introductory Physics of the Atmosphere and Ocean (Dordrecht, Netherlands: Reidel, 1986).

Atlantic and North Pacific. These low pressure areas are a time-averaged result of the dominant winter storm tracks off the eastern coasts of North America and Asia. The winter storms promote lifting out of the atmospheric boundary layer, the lowest layer (1–2 km thick) of the troposphere, and northeastward transport of emissions from North America and Asia out over the ocean. There is a strong surface flow from India and Southeast Asia into the Southern Hemisphere and a strong westerly flow in the Southern Hemisphere midlatitudes.

In the northern summer, the flow in midlatitudes is weaker, and high surface pressure in both the subtropical Atlantic and Pacific becomes important. Rather than large-scale lifting of continental emissions by winter storms, the emissions are lifted into the free troposphere, that portion of the troposphere lying above the atmospheric boundary layer, by small-scale convection, both dry and wet. The surface flow in the Indian Ocean is now toward India and helps maintain that region's summer monsoons, while the converging flow in the rest of the tropics has moved north of the equator. A major feature of both seasons is this converging surface flow in the tropics that is coupled to a vigorous rising motion in the region and results in strong precipitation. This region is called the intertropical convergence zone (ITCZ) and forms a strong barrier to transport of soluble tracer between the hemispheres.

Time-Averaged Transport vs Time-Averaged Winds

Even the relatively complex picture presented by the time-averaged winds in Figure 4 fails to capture the time-averaged transport resulting from the time-dependent three-dimensional motions of the winds. As an example, winter averages of the simulated distributions of a water soluble tracer released in North America, in this case emissions of nitrogen oxides from fossil fuel combustion, are shown in Figures 5a and 5b.[3] In both cases there is a constant release of emissions in the two lowest model levels. In Figure 5a, the emissions are transported by constant winds that are the winter average of the time-dependent winds generated by the parent climate model, and the climate model's average precipitation is used in SINK. In Figure 5b, the transport is provided by the climate model's time-dependent winds, and the model's self-consistent and time-dependent precipitation is used in SINK.

Note that the transport by the time-averaged winds in Figure 5a corresponds to what might be expected, based on the previous section. In the boundary layer, the tracer is confined to the average North Atlantic storm track. It is confined to a midlatitude belt in the free troposphere, where, on average, the westerly winds are quite strong. In neither case is there transport to the Arctic. However, the actual average distribution—a result of time-dependent winds acting throughout the winter—is quite different (see Figure 5b). Although at the surface there is still a maximum along the North Atlantic storm track, the actual amount transported to Europe is much smaller. Furthermore, tracer is carried into the subtropics of both the Atlantic and Pacific, and there is

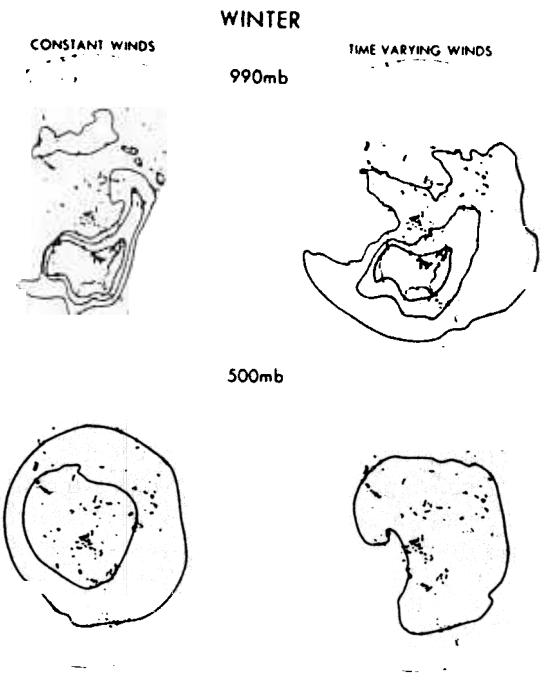

Figure 5. Average winter mixing ratios (ppbv) for a soluble tracer released at the surface in North America and transported by both (a) constant winter-average winds and (b) fully time-dependent winds. The clear contour contains mixing ratios in the range 0.01–0.1 ppbv, the dotted area is for the range 0.1–1.0 ppbv, and the dark area is 1.0–10.0 ppbv. *Source*: Levy and Moxim.[3].

significant transport into the Arctic. In the middle of the atmosphere, the tracer spreads throughout the Arctic, though the actual time-dependent winds do not sweep the tracer completely around the globe as was the case when they were time-averaged.

The inability of time-averaged winds to produce time-averaged transport is easily demonstrated by a simple thought experiment. Consider emissions in the eastern United States and a very simple wind pattern that blows from the west for half the year and from the east for the other half. Over the year, the winds over the eastern United States cancel and produce no transport by the average wind. The resulting distribution is a blob over the eastern United States. In fact, the actual transport would be towards the east for half the year and towards the west for the other half. The resulting average distribution would be a midlatitude belt of emissions around the globe, not a blob over the eastern United States.

Table 1. Percentage Contribution to NOy in the Arctic by Source Region.

Source	80 m	0.5 km	1.5 km	3.1 km
Eurasia N of 60° N	49	25	10	6
Europe S of 60° N	30	44	50	46
North America S of 60° N	10	15	26	37
Asia S of 60° N	11	16	14	11
Global	100	100	100	100

Source: Levy and Moxim.[3]

Regional and Global Transport

It should be clear from the preceding discussion that the average winds are only part of the story. For that reason, we have developed comprehensive global transport models that use time-dependent three-dimensional winds and require very powerful computers. Such a model, which has been discussed in considerable detail,[4] was used to simulate the examples of tracer transport presented in this chapter.

The wintertime transport of combustion products to the Arctic is simulated by a global transport model using time-dependent three-dimensional winds and a constant emission source in the lowest two model levels. The percentage contributions of different source regions to the mixing ratios at different heights in the Arctic atmosphere are summarized in Table 1. These results show the influence of two important transport mechanisms: the strong high pressure system over western Asia and the North Atlantic storm track. As a result of the northward transport of pollution by this Asian high pressure system, the surface concentration of combustion products in the Arctic is dominated by emissions from the European Arctic and northern Europe as a whole. The simulated distribution of the nitrogen component of arctic haze in Figure 6 shows a strong asymmetry between surface concentrations over the North American and European Arctic. North American emissions do become quite important in the free troposphere as they are lifted and carried northeastward by the prevailing storms over the North Atlantic (i.e., the North Atlantic storm track).

A summertime simulation, using the same global transport model and global emission source, finds that surface emissions from the southwestern United States are carried far out into the subtropical Pacific (see Figure 7). The observation of a similar plume of pesticides and other organics is reported in Chapter 11. This long-range transport in the free troposphere results from the subtropical high that moves north of Hawaii in the summer. The transport path is fed by emissions in the southwestern United States that are intermittently lifted from the surface by convection over the arid land. As a result, episodes of continental pollution are then observed in the eastern Pacific. A more complex transport process results in the transport of Asian emissions out over much of the North Pacific, particularly in the spring and the fall.[5]

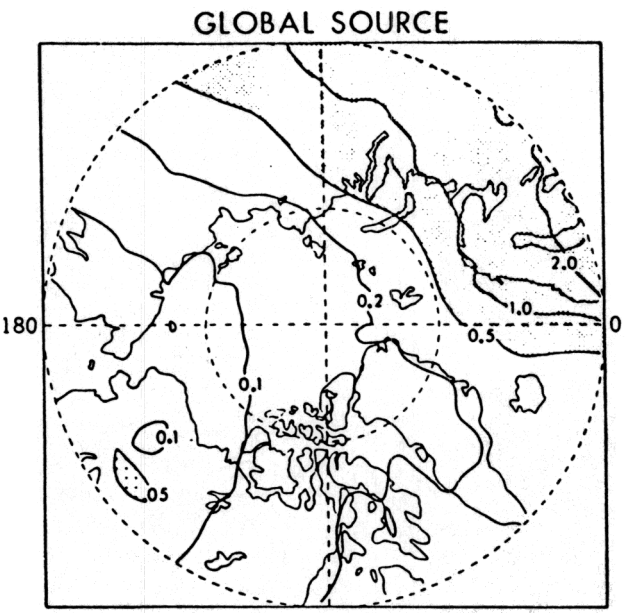

Figure 6. Simulated average winter mixing ratios (ppbv) for the nitrogen component of arctic haze in the atmospheric boundary layer. The light dots are for mixing ratios less than 0.05 ppbv and the dark shading is for mixing ratios that exceed 0.50 ppbv. Log contours 1–2–5 are used. *Source*: Levy and Moxim.[3].

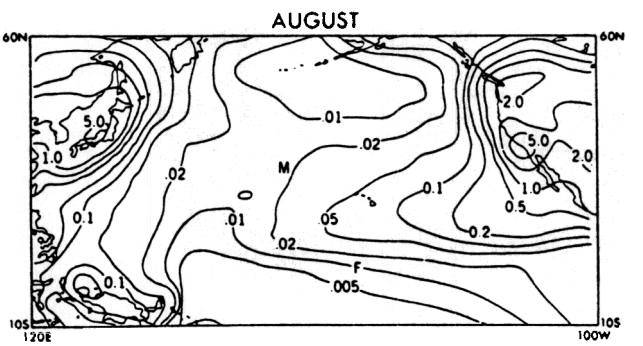

Figure 7. The simulated August average plume of the nitrogen component of North American pollution (ppbv) that has been transported out over the eastern North Pacific. M represents Midway Island; F, Fanning Island. The Hawaiian Islands are drawn on the map. *Source*: Levy and Moxim.[4].

REMOVAL

Sink

Pesticides and other toxics that react with and stick to surfaces are readily removed in the atmospheric boundary layer and must be lifted into the free troposphere for effective long-range transport. If these substances are also soluble, their rapid removal by intermittent precipitation inhibits transport out of the boundary layer. As a general rule, the long-range transport of "sticky" or soluble chemicals is greatly reduced by their short atmospheric lifetimes. The stable dry conditions found in the winter Arctic and discussed in the previous section are special cases that do promote long-range transport.

In Figure 8, we show the simulated yearly average surface distribution of an anthropogenic trace chemical—in this case, nitrogen oxides and nitrates—that is soluble and surface reactive. As expected for such a tracer, there is a strong correlation between the distribution maxima and the continental source regions. Transport from Asia out over the North Pacific storm track and from North America out over the North Atlantic storm track shows up clearly. With less than .005 ppbv over much of the southern ocean, it is clear that little is transported from the source regions of the Northern Hemisphere through the tropics and the ITCZ into the Southern Hemisphere.[4] We see downwind transport at southern midlatitudes due to the prevailing westerlies, and easterly transport in the tropics and subtropics.

Chemical Destruction

If the pesticides and other toxics are neither soluble nor surface reactive ("sticky"), their atmospheric lifetime is determined by their rate of chemical destruction. Methane, a chemical tracer with a moderately long (5–10 yr) lifetime, has been released for a long time in the atmosphere and has an

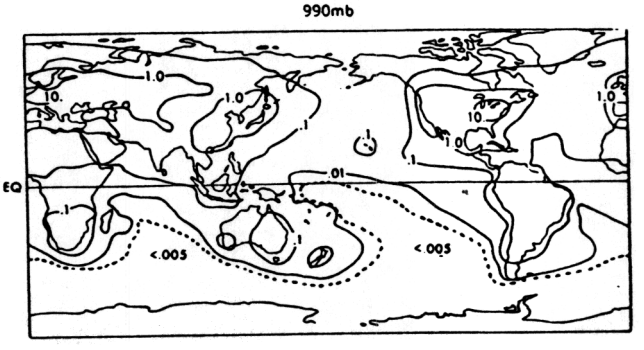

Figure 8. Latitude-longitude plot of the yearly-average surface mixing ratio (ppbv) of a soluble tracer released by fossil fuel combustion. The contour intervals are 0.01, 0.1, 1.0, and 10.0. *Source*: Levy and Moxim.[3]

equilibrated distribution. With most of its source in either the tropics or the Northern Hemisphere, it has a significant interhemispheric gradient ($\approx 10\%$) and a small seasonal variation.[6] As a result of this interhemispheric difference, one can identify air masses that have been transported from one hemisphere to the other.

Carbon monoxide, an atmospheric trace gas with an even shorter lifetime (≈ 3 months), has much higher variability. Near the surface, its mole fraction or mixing ratio ranges from a high of 50 ppmv in severely polluted urban areas, to 0.15–0.25 ppmv in the background Northern Hemisphere, and to a low of 0.050 ppmv in the very clean Southern Hemisphere.[7] The range of variation is much less, 0.05–0.15 ppmv, in the middle of the troposphere. Carbon monoxide has frequently been used as a tracer of anthropogenic pollution[8] as well as a tracer of interhemispheric exchange. There are a number of very short-lived (i.e., highly reactive) hydrocarbons whose presence signifies a nearby source. These compounds are only found at very low levels in remote regions distant from their sources.

SUMMARY

Although time-averaged wind fields may help identify preferred paths of transport, they are not adequate to explain either simulated or observed average transport. Time-dependent meteorology, ranging from small-scale turbulence and convection to large storms, plays an important role in lifting surface emissions into the free troposphere, where long-range transport can readily occur. Once in the free troposphere, atmospheric transport is still strongly influenced by large-scale fluctuations in the time-dependent wind fields.

Even very long-lived trace chemicals will have a variable distribution and display identifiable transport events if their surface sources are nonuniform over the globe. Those tracers that are insoluble and nonreactive with surfaces will have their atmospheric lifetime determined by their chemical reactivity. The more chemically reactive the tracer, the more its mixing ratio distribution is influenced by the distribution of its sources, and the less likely it is to be transported long distances. Soluble tracers have short atmospheric lifetimes and are only transported long distances under stable dry conditions, such as those found in the winter Arctic and over the subtropical oceans. Due to removal by precipitation in the ITCZ, their transport between the hemispheres is quite unlikely.

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