Atmospheric Transport and Dispersion of Pollutants and Related Meteorological Studies, July 1971 - June 1972

ROBERT J. LIST, Editor
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ATMOSPHERIC TRANSPORT AND DISPERSION
OF POLLUTANTS AND RELATED METEOROLOGICAL STUDIES,
JULY 1971 - JUNE 1972

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Preface

This is the second in a series of NOAA Technical Memoranda describing the work done by the Air Resources Laboratories for the U.S. Atomic Energy Commission, Division of Biomedical and Environmental Research (Formerly Division of Biology and Medicine). The work for Fiscal Year 1972 under Contract AT(49-7)-4 is reported here. NOAA Technical Memorandum ERL ARL-36, March 1972, described the work for Fiscal Year 1971.

1.1 Introduction

This presentation is offered from the viewpoint of a meteorologist concerned with predicting the possible future climatic change caused by an increase of CO₂ in the atmosphere. There are two aspects of this climatic prediction. First, the rate of the added CO₂ in future years and, second, the possible climatic change caused by CO₂ increases. We shall primarily discuss the first of these topics and touch only lightly on the second. Future CO₂ in the air depends upon two factors: first, the added fossil-fuel CO₂ injected into the air and, second, the apportioning of this CO₂ between the atmosphere and the reservoirs with which it can exchange.
PART 1

PREDICTION OF CO₂ IN THE ATMOSPHERE

L. Machta and K. Telegadas

Predictions of future and past carbon dioxide (CO₂) concentrations have been derived from a simple multireservoir exchange model: stratosphere, troposphere; mixed layer of the ocean, deep ocean; short-term land biosphere, long-term land biosphere, and marine biosphere. All exchanges are prescribed from literature or personal estimates except the troposphere-mixed ocean layer, which is obtained from a trial-and-error procedure using bomb C¹⁴O₂ as a tracer. The CO₂ content predicted for the year 2000 is about 385 ppm compared with the present 320 ppm, if we assume the estimates of the growth in the use of fossil fuel are reasonable. The uncertainties in the model are probably less significant when using the above procedure than the assumption that the oceans and biosphere will continue to behave in the future as they did in the 1960's. The so-called "greenhouse effect," using a simplified one-dimensional (vertical) model of the atmosphere, calls for about a 0.5°C warming from the increase of CO₂ to 385 ppm. But this estimate is on even shakier ground than the forecast of future CO₂ concentrations.

1.1 Introduction

This presentation is offered from the viewpoint of a meteorologist concerned with predicting the possible future climatic change caused by an increase of CO₂ in the atmosphere. There are two aspects of this climatic prediction. First, the fate of the added CO₂ in future years and, second, the possible climatic change caused by CO₂ increases. We shall primarily discuss the first of these topics and touch only lightly on the second. Future CO₂ in the air depends upon two factors: first, the added fossil-fuel CO₂ injected into the air and, second, the apportioning of this CO₂ between the atmosphere and the reservoirs with which it can exchange.

We will discuss the observed data for the growth of CO₂ in the atmosphere at Mauna Loa. Other data such as those collected by Bischof (1970) in Sweden and by NOAA aboard Ocean Weather Ship Charlie and in the western United States (Machta, 1972) support the trends in figure 1.1, the mean monthly CO₂ concentrations at Mauna Loa. There are two very obvious features in this graph. The first is a long-term increase, the second a strong seasonal variation. It is the long-term increase projected into the future that is discussed. Between 1958 and 1971, the growth of CO₂ in the air is about one-half that put into the air by the fossil-fuel CO₂ if it is uniformly distributed throughout the atmosphere. The other half has gone into the biosphere or into the oceans. A prediction of this partitioning into the other reservoirs is needed to forecast future CO₂ concentrations.

1.2 The Model

Figure 1.2 shows the model we have used. It contains a troposphere, a stratosphere, a mixed and deep layer of the ocean, and a long-term and short-term biosphere, as well as a marine biosphere. The exchange between atmosphere and ocean is based on first-order kinetics while
the uptake of carbon by the biosphere is the net primary production. The exchange between stratosphere and troposphere is based on radioactive material in the stratosphere having a residence time of about 2 years, and the exchange between the deep and mixed layer of the ocean is based on the average age of the carbon in the deep ocean being about 1600 years. The values for the net primary production are taken either from the estimates that were given to us by Woodwell (personal communication) and Olson (personal communication) or from Lieth (1970). The last, and most important, exchange is that between the atmosphere and the mixed layer of the ocean; this calculation is based on the C\textsuperscript{14}O\textsubscript{2} from nuclear tests.

Figure 1.3 shows the history of carbon-14 atoms produced by nuclear tests in the troposphere and in the entire atmosphere to about 30 km. This figure was derived from data collected by the U.S. Atomic Energy Commission (AEC) mainly over North and South America. The sharp increases of these curves reflect periods of active testing. After about 1963, only a few injections into the atmosphere occurred and small corrections have been made for these.

Figure 1.2. The model of the three reservoirs of CO\textsubscript{2}. The quantity \( \lambda \) denotes the fraction of the CO\textsubscript{2} in one reservoir being transferred to an adjacent reservoir according to the arrow.
After about 1963, the C\textsuperscript{14}O\textsubscript{2} content of the entire atmosphere decreased. This implies a transfer to other reservoirs, either into the ocean or the biosphere. We have specified the transfer into the biosphere; hence, the remaining atmospheric decrease must be the result of CO\textsubscript{2} entering the oceans. This reasoning and figure 1.2 form our basis for the exchange coefficient between the atmosphere and the ocean using a trial-and-error procedure. The mean residence time in the atmosphere for a CO\textsubscript{2} molecule becomes about 5 years. The trial-and-error procedure also provides a mean residence time of carbon and the carbon content of the mixed layer of the ocean. This carbon content corresponds to the depth of the mixed layer of 110 m. It represents the global average depth into which relatively rapid mixing of CO\textsubscript{2} occurs. The 5-year atmospheric residence and 110-m mixed-layer depth represent significant changes from the original model (Machta, 1971) and are caused by programming errors.

The amount of fossil-fuel CO\textsubscript{2} expressed in grams of carbon that have been added to the atmosphere since the last UN Report (1968) has been followed by an annual growth rate of 4 percent per year between 1969

![Figure 1.3. The history of C\textsuperscript{14} atoms produced by nuclear tests in the troposphere and in the whole atmosphere (up to 30 km). (Source of data, Telegadas, 1971.)](image)
and 1979 and $3\frac{1}{2}$ percent between 1980 and 1999. When this fossil-fuel $\text{CO}_2$ is added to the atmosphere via the model, one obtains a prediction of the changes in $\text{CO}_2$ content from 1860 to 2000. This prediction appears in figure 1.4 using the Mauna Loa concentration of about 322 ppm in 1970 as the base. The concentration of $\text{CO}_2$ will be about 385 ppm in 2000 and was about 290 ppm at the beginning of the industrial era. This later value is approximately correct when compared with the poorly measured data in the latter part of the 19th century.

The partitioning of fossil-fuel $\text{CO}_2$ molecules between the three reservoirs shows that as of 1970, about 65 percent of the fossil-fuel $\text{CO}_2$ remained airborne; half of the remainder appeared in the oceans, mostly in the mixed layer, and the other half in the biosphere; almost entirely in the long-term land biosphere. This model assigns a surprisingly large fraction of the fossil-fuel $\text{CO}_2$ to the biosphere relative to the oceans. During 1958-70 the average airborne fraction of the fossil-fuel $\text{CO}_2$ was between 60 and 65 percent, which is about 10 percent more than is suggested by the Mauna Loa and other stations.

This is perhaps a convenient time to explain some of the special details of the model. First, using the crude estimates provided by Woodwell (personal communication), we have assumed that the increase in

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**Figure 1.4.** The history, as predicted by the model, of global atmospheric $\text{CO}_2$ from 1860 to 2000.
photosynthesis amounts to 5 percent for each 10 percent increase in the CO₂ content of the troposphere. However, this applies only to one-half of the land biosphere, since the other half is assumed nutrient or water limited. This factor (0.5 x 0.5 = 0.25) allows for an increase in the CO₂ content of the biosphere; without it a steady state would exist in the biospheric carbon irrespective of the atmospheric growth. Second is a buffering effect of the oceans. Suppose that the exchangeable carbon mass in the mixed layer (isolated from the deep ocean) equals that of the air. Then for every 11 units of CO₂ added to the atmosphere, 10 remain airborne, and only one goes into the ocean instead of the 50-50 split expected by their carbon masses. However, the C¹⁴O₂ is added in exceedingly low quantities so that its buffering effect is unimportant and has been neglected.

1.3 Sensitivity Tests

Sensitivity tests have been performed on various doubtful components of the model. In these tests, however, one must remember that whatever model is chosen it must account for the full decrease in the C¹⁴O₂ evident in the nontesting period of figure 1.3. Should the biosphere be eliminated, for example, the oceans must then compensate for its omission. Later when the model is applied to the fossil fuel CO₂, the oceans, in our example, will partially fulfill the role of the neglected biosphere.

In the sensitivity tests, the biosphere has been omitted, the net primary production doubled, the long-term biosphere altered from a 40- to 20-year return time, and the growth limitation reduced to 0.05 rather than 0.25. In the oceans, the buffering factor has been reduced to 6 and up to 14 rather than the 10 in the model and the deep oceans assumed only 400 rather than 1600 years old. The changes in the forecast of the CO₂ concentration in the year 2000 generally lie within ± 10 ppm, which is insignificant in the light of the uncertainties to be discussed below converting the CO₂ predicted concentration into a climatic forecast.

But these sensitivity tests of changes in the model reflect but the tip of the iceberg of uncertainty in predicting future levels of CO₂. They naively assume that the history of the atmosphere-biosphere-ocean transfers found in the 1950's and 1960's have and will be applicable from 1860 to 2000. This is hardly likely to be the case. In fact, year-to-year differences exist between the simple model predictions and the Mauna Loa observations. Put another way, the model presumes, in effect, that about 60 to 65 percent of the fossil fuel CO₂ will remain airborne each year. But in the mid-1960's, this observed fraction dipped below 40 percent and in very recent years rose to well over 60 percent if we assume that Mauna Loa annual increments represent those for the globe. If these changes in the airborne fraction are indeed valid, significant year-to-year changes do occur in the uptake by the biosphere and oceans.
This point may come across more clearly by comparing the CO₂ photosynthetic uptake with fossil-fuel emissions. From figure 1.2 there are 5.6 or 7.6 x 10¹⁶ g of carbon taken up (and presumably released by respiration and decay) each year; this figure depends on whether one wants to include the very uncertain marine net primary production. This is about 15 to 20 times greater than the 1969 fossil-fuel CO₂ releases. Thus, a 1 percent decrease in the photosynthetic uptake due to droughts or other factors would appear as a 15 to 20 percent increase in the fossil fuel CO₂ airborne fraction. Do we know that the year-to-year values of global net primary production will not change by 1 percent.

Assuming that the atmosphere comes into equilibrium with the sea surface partial pressure of CO₂, one can argue that each 1°C warming of the ocean surface will increase the CO₂ content by as much as 10 ppm. A global warming of the ocean temperature of only 0.1°C could possibly equal the current annual increment of CO₂ in the air.

These comparisons suggest strongly that the biosphere and oceans play so vital a role in the changes in atmospheric CO₂ content that even small changes in their behavior can easily modify the predictions of the simple model calibrated from conditions in the 1950's and 1960's.

1.4 Climate Prediction

An even larger measure of uncertainty must be attached to the prediction of climatic changes from an increase in atmospheric CO₂. It is well known that CO₂ absorbs short-wave solar wavelengths poorly, but does absorb the long-wave terrestrial wavelengths effectively. Thus, solar radiation can reach the earth unattenuated by an increase in CO₂ concentrations but the long-wave terrestrial radiation will be absorbed and partly emitted back to the lower atmosphere. The result is the so-called "greenhouse effect." But the analogy between an atmosphere with enhanced absorbers of long-wave radiation and a greenhouse is not accurate because the glass inhibits turbulent transfer of heat. Further, CO₂ in the air always cools the atmosphere, but more CO₂ cools the lower atmosphere less while cooling the high atmosphere more.

The response of the atmosphere to changes in the radiation distribution is very complex, and a fully evolved model to simulate the climatic change does not now exist. What can be reported are only approximations to the true atmospheric adjustment to increasing CO₂ concentrations. Radiative transfer theory is reasonably well established. But the dynamic response, air movements, and energy transfers are much more difficult to recreate. Perhaps most important, the changes in cloudiness, which represent a dominant influence on the global heat budget, may be modified in a manner as yet unpredictable.
Manabe and Wetherald (1967) are most often quoted as the best available study on the change in temperatures from the addition of CO₂ from a one-dimensional (vertical) model. Figure 1.5 shows the relationship between atmospheric CO₂ content in parts per million to change in global surface temperature away from the current 320 ppm. The prediction of 385 ppm in 2000 calls for a warming of about 0.5°C. A slightly different radiative treatment reduces the warming by about 30 percent. The model upon which this forecast is based allows for new radiative temperature changes for increasing CO₂ but a constant relative humidity to account for the added moisture that warmer air can hold. However, the average cloudiness employed in the calculations does not change when the air warms or cools. The model also produces much greater cooling in the high atmosphere than warming near the ground.

Manabe (1971) has reported on some calculations using a three-dimensional model comparing climatic conditions with a doubling of the current CO₂ atmospheric composition. No ocean circulation is admitted and a perpetual winter is prescribed. Further no change in the cloudiness occurs, even when the atmospheric humidity changes in response to the heating by the added CO₂. The result suggests a slightly greater global warming in the lower atmosphere than Manabe and Wetherald (1967) found in their one-dimensional case. But the most dramatic change is the much larger warming of the Arctic than might have been expected. The high latitudes warm twice as much as the globe as a whole. This is because of the marked thermal stability of the Arctic lower atmosphere in winter and the recession of the ice with the added warmth of the air. This calculation should not be treated literally but only as suggestive of the fragility of the Arctic region to climate changes.

Figure 1.5. Surface air temperature change due to increased atmospheric CO₂ with average cloudiness and assuming a convective-radiation equilibrium and a fixed humidity (adapted from Manabe and Wetherald, 1967).
Indeed the cooling that has been in progress since the mid-1940's in the Northern Hemisphere has been concentrated almost entirely in the Arctic (north of 50°) in the winter. This cooling has proceeded despite the rapid increase in CO₂ concentration evident in the atmosphere. One must recognize that other factors besides CO₂ are responsible for the global climate; in all likelihood natural fluctuations, as yet not understood, play a dominant role since climatic fluctuations have occurred long before man had his present technological powers.

There are at least two feedback mechanisms that have so far been omitted in quantitative treatments of climatic changes from increased CO₂. First, the added moisture may increase the cloudiness. Clouds play a dominant role in controlling the earth's albedo, or reflecting power. It has been estimated that the net effect of an increase of only 0.6 percent in low cloudiness over the globe will cool the lower atmosphere by 0.5°C the same amount that the 385 ppm is predicted to warm the lower atmosphere (Wilson et al., 1971). Thus, if the added warmth and moisture by the so-called greenhouse effect also increase low cloudiness, much of the warming can be negated.

The second is a positive rather than a negative feedback. If the warming of the lower atmosphere should also warm the surface layers of the waters, additional oceanic CO₂ would be released. This, in turn would intensify the atmospheric warming and so forth.

At this time, we feel in a less confident position to forecast the future climate due to the combustion of fossil fuels than we are to estimate the increase in atmospheric CO₂ concentrations.

1.5 Acknowledgment

We are indebted to Jerry Olson (Oak Ridge National Laboratory) and George Woodwell (Brookhaven National Laboratory) for most of the details of the biosphere but we, rather than they, must take the blame for any of the inaccuracies.
1.6 References


PART 2

SOURCES AND SINKS FROM PROFILE DATA

L. Machta

2.1 Introduction

A knowledge of the sources and sinks of air pollutants is necessary for the fullest understanding of the cycle of man-made pollutants in nature. This information should, most properly, derive from first principles such as: emission rates, physical and chemical conversions within the atmosphere, removal from the atmosphere by impaction or precipitation scavenging, photochemical reactions, and other processes. Unfortunately much of the above information is often unavailable or confusing. In many cases, such as carbon monoxide (CO) and methane (CH₄), both man-made and natural sources contribute to atmospheric concentrations.

What follows is an empirical approach. We presume to know the observed distribution of the concentration of the pollutant. This distribution results both from the presence of sources and sinks and from atmospheric readjustment due to turbulent motions, organized circulations, settling of particles, and other processes. Since the three tracers used in this paper are gaseous, particle settling is absent. This method assumes the atmospheric motions and steady-state concentrations to be known and leaves only the sources and sinks as the unknowns. Some of the reasons for the uncertainties in the method are discussed in section 2.5.

The trial-and-error best fit procedure employed below is, in principle, the same as another method that has been used by many other investigators. If, say, there is a decrease of global concentration with height (an upward pointing gradient), then, in steady state, the upward flux is the net loss above the altitude in question. If we calculate the flux through two altitudes, their difference represents the sink (or source) in the layer bounded by the altitudes. In the trial-and-error modeling, we will try to reproduce the profile and, in effect, end up with the same information on fluxes. When two- or three-dimensional data become available later, the trial-and-error method (or an equivalent procedure) must be used since transport can occur in two or three directions. For the moment, the trivial advantage in this analysis, based on a trial and error fit to the profile, over a more simplified approach lies in being able to graph the results and in using the model's finite difference scheme to smooth irregular data results.
We do not now have full three-dimensional distribution of the two substances whose sources and sinks are being sought (CO and CH₄). Instead very limited vertical profiles in the north temperate latitude are now available. Thus, a one-dimensional analysis will replace the three-dimensional case. The extrapolation of the source-sink values from limited information in the north temperate latitude to the globe adds to the uncertainty.

2.2 Trial and Error Method Applied to Ozone (O₃)

To calibrate the diffusion coefficients in the vertical direction and to test the method for a substance whose sources and sinks are known, the procedure has been applied to ozone (O₃). The transport in the model employs the flux-gradient relationship: the flux of a gas lies in the direction of and is proportional to the gradient of mass concentration or mixing ratio of the gas. The proportionality constant is the coefficient of diffusion and reflects the state of atmospheric turbulence. The meteorological literature, and the author's experience in fitting various tracer substances by similar models, have led to the diffusion coefficients that appear in the figures. They reflect about an order of magnitude higher coefficients of diffusion in the troposphere than in the stratosphere. The diffusion coefficients vary with season, weather, time (at low levels) and other factors, but these variations are neglected here. In the troposphere, the true mean value may lie between 3 and 20 x 10⁴ cm²/sec; our mean value is 6 x 10⁴ cm²/sec. In the stratosphere, the range of correct numbers may lie between 2 and 10 x 10³ cm²/sec with the mean value of 5 x 10³ cm²/sec or perhaps a little lower.

The method is tested by using O₃ profiles. Although there are very many O₃ profiles derived from ozonesonde flights and Umkehr (Dodson spectrophotometer measurements) data, the results of the vertical profiles in Seiler and Warneck's (1972) flights are used, since these are the same flights that will later be used for carbon monoxide (CO) profiles. The source of O₃ lies above the topmost altitude (12 km) of the flight profile, or at 13 km. Machine runs with equivalent sources at 20 instead of 13 km show identical results, as might be expected. The sink occurs within the bottom box of the model or at the ground. Some loss of O₃ may occur within clouds and hence above the ground, but in my opinion this omission will not seriously disturb the answers. The computer program fits the observed profile by a trial-and-error procedure by trying different values for the ground-level sink.

Figure 2.1 illustrates the best fit passing through the rough average of the two observed profiles, the solid lines. The theoretical curves arbitrarily match the observed profile at an altitude of 8 km. The variability between profiles of O₃ is in accord with other profiles of O₃. The fit for two other cases with different sink values is also shown; the match is obviously worse. The source and sink must be equal for the steady state assumed in this computation.
The destruction of O\textsubscript{3} at the earth's surface is known approximately and can be compared with the estimates from the above profile method. The daily loss of 15 percent of the O\textsubscript{3} in the first box, 500 m deep, with an air density of about 1x10\textsuperscript{-3} g/cm\textsuperscript{3} results in the destruction of about 4.5x10\textsuperscript{14} g of O\textsubscript{3} per year over the entire earth, or about 3x10\textsuperscript{10} molecules/cm\textsuperscript{2}/sec. Estimates of O\textsubscript{3} loss in New Mexico at the ground are about 3x10\textsuperscript{11} molecules/cm\textsuperscript{2}/sec over land and 3x10\textsuperscript{10} molecules/cm\textsuperscript{2}/sec over a lake (Aldaz, 1960). These latter numbers have large uncertainties themselves. The source-sink value from the profile method, thus, lies within the correct range.

### 2.3 Carbon Monoxide (CO)

Carbon monoxide (CO) is formed and destroyed at the earth's surface and in the stratosphere. The observed profile (Seiler and Warneck, 1972) decreases through the tropopause, indicating a net formation near the ground and a destruction aloft. But the unique feature of the two CO
profiles is the constant concentration some short distance above the tropopause as is evident in figure 2.2. This means no net vertical flux of CO occurred through the 11 km level. Carbon monoxide destruction above the top of the observed profile would result in an upward gradient of concentration instead of the uniform vertical distribution in the 11 to 12 km layer.

Figure 2.2 indicates that the sink must be at the base of the stratosphere or near 10 km in this case. This loss of CO in the lower stratosphere amounts to about $2 \times 10^{11}$ g/yr or $2.6 \times 10^{10}$ molecules/cm$^2$/sec. This is also the net ground level source. Seiler and Warneck (1972) calculated an upward flux at the tropopause of $4 \times 10^{10}$ molecules/cm$^2$/sec. This compares with $2.6 \times 10^{10}$ molecules/cm$^2$/sec. The difference arises from my stratospheric diffusion coefficient of $5 \times 10^3$ cm$^2$/sec compared with Seiler and Warneck's $3 \times 10^3$ cm$^2$/sec (which I would accept as equally good). Their smaller diffusion coefficient should produce an even smaller flux or destruction rate than mine. But the main reason for our difference lies in the solution of the diffusion equation; I fit the observed data while Seiler and Warneck, following Junge et al. (1971), assume an exponential decrease of concentration above the tropopause. I feel that the two values are equally good.

Figure 2.2. Carbon monoxide profiles.
2.4 Methane (CH₄)

Figure 2.3 displays, again, the observed and "best fit" profiles; this is to 30 km since Ehhalt (1972) was able to obtain high altitude balloon samples. In addition, Ehhalt et al. (1972) have measured an atmospheric methane (CH₄) sample collected at 44 to 62 km, which is plotted on the figure at 50 km following their suggestion. The observed concentration at 30 km exceeds the value at 26 km; however, in view of the lower concentration at 50 km, and on advice from Dr. Ehhalt, a smooth decrease is presumed to represent the vertical profile in the stratosphere.

The results of the trial-and-error fit indicate, as with CO, a greater destruction rate in the lower stratosphere than above. Thus, as seen in the figure, the loss of CH₄ just above the tropopause (16.5 km)
is greater than near the top of the model (30 km). From the gradient above about 30 km, one can readily calculate the upward flux of CH₄ through that level. In the assumed steady state this same amount of CH₄ must be destroyed above 30 km. The loss of CH₄ from 16.5 to 30 km greatly exceeds that lost above 30 km. The total CH₄ destruction in the stratosphere is similar to that calculated by Ehhalt (1972) from the same data and using the same concept; the differences lie in the selection of diffusion coefficients and gradients of CH₄ concentration.

2.5 Uncertainties

This method of finding the sources and sinks of atmospheric constituents should, in principle, provide reliable estimates. But limitations in observed constituents and meteorological data greatly influence the confidence in the present results.

The first — and possibly suspect — assumption in the above analysis is how representative the very few vertical profiles are in time and space. There is no way of knowing how different the answer would be if the full distribution were available. The only test, the prediction of O₃ destruction with similar single profiles, suggests the error is no more than and probably less than a factor of, say, five. Seiler and Warneck (1972) point out the weather conditions during the two European sampling programs for CO were not typical. It is conceivable that the sharp decrease and constancy at the top of the CO profiles are a consequence of the special conditions. Second, the transport processes and magnitudes are known imperfectly. But these errors, even the omission of transport other than "down the gradient," probably contribute a much smaller source of uncertainty than the representatives of the available data.

2.6 Conclusions

In light of the above and other uncertainties in applying the profile method to CO and CH₄, what can be claimed from the present state of knowledge?

1. The method gives the correct order of O₃ destruction at the ground by using only the two European profiles. This suggests that the diffusion coefficients are not grossly wrong.

2. Destruction of CO occurs just above the tropopause in temperate latitudes. When extrapolated to the entire earth, about $2 \times 10^{11}$ g/yr are destroyed in this region.

3. The constant CO profile from 11 to 12 km indicates no net formation or destruction above 11 km; this does not preclude formation and destruction rates above 11 km so long as the two approximately balance one another.
4. Most of the CH₄ is also destroyed just above the tropopause. The total destruction of methane in the stratosphere is estimated to be about 5x10¹³ g/yr.
2.7 References


Modeling of local fallout deposition is continuing on a low priority basis, as described in our previous annual report. This work includes (1) analyzing fallout from cratering detonations to provide modeling data on the amount and distribution of radioactivity in the initial nuclear cloud for this type of event, and (2) improving the computer program to increase accuracy and efficiency and to adapt the fallout prediction code for easier use in possible future nuclear tests or Plowshare events.

The fallout pattern resulting from the Schooner detonation (31 kiloton (kt) device detonated 108 m below the surface at the AEC Nevada Test Site in December 1968) has been analyzed to determine the fraction of the total local fallout that was deposited each hour (normalized) after detonation. Results are shown in table 3.1, which is a revision of table 2.1 in our previous annual report. Also added are results for Cabriolet (2.5 kt detonated at 52 m) and the hourly deposition rates produced by the present computer code for cratering detonations. This cratering model was based on preliminary analyses of near-surface detonations (Johnnie-Boy and Smallboy) and subjectively modified for cratering detonations. Although the cratering model had been adjusted in the proper sense (compare with deposition rates in the "Tower" model) further adjustments are needed to bring the model into closer agreement with cratering data; for example, fallout should be increased slightly for the first hour and decreased for the second and third hours.

The Schooner results indicate a rather high deposition fraction during the second hour and low deposition fractions beyond the fourth hour (low by about a factor of 2). These differences appear to be real, but they may be due, in part, to uncertainties in the fallout data or analysis techniques. In any case, the differences among all cratering detonations are small enough so that the cratering model can be adjusted to provide a reasonably good fit to all the data.

Work has been completed on a revised radioactive decay module to permit either $t_n$ or $e^{-\lambda t}$ decay calculations and summation for up to 12 nuclides, as discussed in the previous annual report. Other program revisions must be completed before we have a completely satisfactory operational system for routine fallout prediction. This work will be done as time and priorities permit. A report documenting the fallout prediction program and describing its modes of operation is about 70 percent complete.
### Table 3.1. Percent of Local Fallout Deposited Each Hour

<table>
<thead>
<tr>
<th>DETONATION</th>
<th>Tower Model</th>
<th>Cratering Model</th>
<th>Small-boy</th>
<th>Johnnie-Boy</th>
<th>Teapot ESS</th>
<th>Sedan</th>
<th>Danny Boy</th>
<th>Cabriolet</th>
<th>Buggy</th>
<th>Schooner</th>
</tr>
</thead>
<tbody>
<tr>
<td></td>
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<td></td>
<td></td>
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</tr>
<tr>
<td></td>
<td></td>
<td>-8</td>
<td>2</td>
<td></td>
<td>63</td>
<td>164</td>
<td>141</td>
<td>131</td>
<td>131</td>
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</tr>
</tbody>
</table>

<table>
<thead>
<tr>
<th>HOURS</th>
<th>PERCENT DEPOSITED</th>
</tr>
</thead>
<tbody>
<tr>
<td>0-1</td>
<td>64.0 81.4 80.0 81.0 83.0 84.0 86.0 88.0 88.0 86.0</td>
</tr>
<tr>
<td>1-2</td>
<td>20.0 12.1 9.7 8.8 6.0 6.6 6.8 4.8 4.8 8.7</td>
</tr>
<tr>
<td>2-3</td>
<td>8.7 4.0 4.2 4.7 4.2 3.6 3.7 2.2 2.5 2.4</td>
</tr>
<tr>
<td>3-4</td>
<td>4.4 1.5 2.2 2.4 2.3 2.1 2.0 1.6 1.3 1.3</td>
</tr>
<tr>
<td>4-5</td>
<td>2.5 0.6 1.6 1.2 1.5 1.0 1.4 1.5 0.80 0.47</td>
</tr>
<tr>
<td>5-6</td>
<td>1.0 0.92 1.1 0.73 0.70 0.72 0.62 0.37</td>
</tr>
<tr>
<td>6-7</td>
<td>0.65 0.63 0.73 0.70 0.72 0.62 0.37</td>
</tr>
<tr>
<td>7-8</td>
<td>0.43 0.44 0.65 0.46 0.57 0.32</td>
</tr>
<tr>
<td>8-9</td>
<td>0.30 0.37 0.63 0.44 0.24</td>
</tr>
<tr>
<td>9-10</td>
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</tr>
<tr>
<td>10-11</td>
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<tr>
<td>11-12</td>
<td>0.12</td>
</tr>
<tr>
<td>12-13</td>
<td>0.09</td>
</tr>
</tbody>
</table>

1 Normalized to 12,000 ft cloud top.  
2 Scaled depth = D/y^{0.294} where D is depth of burial in feet and Y is yield in kilotons.
Trajectory studies by the Air Resources Laboratories were initiated in Las Vegas to support various programs conducted at the Nevada Test Site. Of special interest were forecasting trajectories for 700 and 600 mb. A post facto trajectory program was then started, so that post facto and forecast trajectories could be compared and confidence intervals assigned to the forecast trajectories for operational and planning purposes.

As a further aid to our Las Vegas Laboratory, a forecast isentropic trajectory program is now being developed. With the recent general interest in air pollution problems, a post facto trajectory program for the boundary layer is also being developed.

This section summarizes all the trajectory programs to date and emphasizes those not discussed in earlier reports.

Three studies related to the movement of particles in the free atmosphere—for several hours to several days—have been undertaken; they resulted in the development of computerized trajectory programs for the CDC 6600 computer located at the National Meteorological Center (NMC), Suitland, Maryland. These studies are (1) post facto trajectory programs, (2) forecast trajectory programs, and (3) a trajectory comparison program.

For this report, discussions include only those portions of the computer programs necessary to familiarize the reader with program concepts. Thus far, program output is based on only enough data to check out the programs and give a potential user an idea of the type and form of information available.

4.1 Post Facto Trajectory Programs

The post facto trajectory programs consider trajectories (1) at constant pressure levels (isobaric), (2) in boundary layers, and (3) at constant heights. Each type of trajectory can be computed forward or backward in time for any desired duration.

4.1.1 Data input. Input to the isobaric trajectory program consists of wind reports from 60 stations in the western U.S. (see fig. 4.1) extracted
Figure 4.1. Upper air wind report stations (black dots) used in the isobaric trajectory program.
for 0000, 0600, 1200, and 1800 GMT from 700-mb charts for October 1971 through February 1972. These data were punched on cards, transferred to tapes, and are readily available at ARL.

Input data to the boundary layer program and constant height program are obtained in a special format of upper air reports packed on a magnetic tape available from the USAF Environmental Technical Air Command (ETAC). Each report contains a heading, the station identification, location, and elevation as well as the time of observation and type of report for all reporting stations in the world. Pibal and raob wind observations are combined in a separate, synthetically constructed "pibal" report. Temperature and humidity observations are included in a "raob" report.

The body of the "pibal" report consists of groups of four numbers — speed, direction, height, and pressure — plus indicators showing whether the data are observed directly, corrected, or synthetic. Data are quoted to the nearest $10^{-1}$ m/sec, degree, meter, and $10^{-1}$ mb. The ETAC tape is available organized by time, or organized by station number.

Before use, the data are unpacked and reduced by creating an intermediate tape consisting of whichever portion of the data is considered necessary for a particular investigation; e.g., blocks 72 through 74 for levels lower than 5000 m.

Once the intermediate tape is created, the data may be used in calculating trajectories directly by computer, or they may be displayed on maps drawn by an in-house program on a CALC0MP plotter. Background maps can be created in arbitrary scales, sizes, and orientations using either the polar stereographic, Mercator, or Lambert conformal projections. A scale of nautical miles versus latitude is provided, and coastlines are drawn using a tape provided by NOAA's National Oceanographic Data Center (N0DC). Station models may be placed on these background maps. At present, this includes a circle for the station, an arrow in the downwind direction with the speed in knots at the tip, and the height in decameters of a particular pressure level. Date, time, and pressure level notations are also provided.

A sample ETAC tape for 15 to 26 September 1971 was made available to ARL. The status of tapes for other periods and ARL access to them are as yet to be determined.

4.1.2 Trajectory techniques. To draw the best possible post facto trajectories, we would have to investigate various techniques and use varying parameters in each technique; these results would then be compared with observed trajectories of tracer effluents, constant-level balloons, etc. Since a very limited number of these observed trajectories are available for comparison, the following method was used as an alternative.
Three techniques — described below — for estimating winds at a point from surrounding wind reports were considered; we assumed that the best one of these could be applied to determine the most reasonable post facto trajectories. Two techniques use wind persistence with current wind observations and the third uses only the current observations. Wind persistence was included to see if comparisons would be improved for periods of sparse observations.

Consider wind report $V_i$ at distances $d_j$ from a given location and $N$ reports within a radius $R$ of the same location (fig. 4.2). The wind at that location, $V'$, can be estimated from a wind at a previous time, $V_p$, as a first approximation, and then by applying a correction to that wind, we get

**Technique 1:**

$$V' = V_p + WE$$

where

$$W = \frac{R^N - \prod d_i}{R^N + \prod d_i}$$

$$E = V_c - V_p, V_c = \frac{\sum_{i=1}^{N} \frac{V_i^2}{d_i}}{\sum_{i=1}^{N} \frac{1}{d_i^2}}$$

**Technique 2:**

$$V' = V_p + \frac{\sum \left( W_i E_i \right)}{N}$$

Figure 4.2. Determining a wind $V'$ at a point (with a previous wind $V_p$) using reported winds $V_1$ and $V_2$ at stations whose distances $d_1$ and $d_2$ are within radius $R$ of the point.
where \( W_i = \frac{R^2 - d_i^2}{R^2 + d_i^2} \), \( E_i = V_i - V_p \),

or using only the current observations

**Technique 3**: \( V' = V_c \).

One of these techniques is used to estimate the winds at each of the reporting stations, and then the estimated winds are compared with the observed winds, \( V_0 \). The observed wind at a station is, of course, not included in the estimation process.

Sample computer printouts of comparisons using Technique 3 are given in figures 4.3 to 4.5.

Table 4.1 compares the three techniques as applied at 700 mb over the western U. S. The small range of values in the last column (standard error) seems to indicate that no significant difference in results can be expected from any of the three techniques. Therefore, most of the trajectory computations employed Technique 3 because of its comparative simplicity.

The effects of varying certain parameters within Technique 3 are shown in table 4.2. Apparently the choice of the minimum number of reporting stations required, and the radius within which these stations must be located, have little effect on the results! Furthermore, varying the power to which \( d_i \) is raised (\( d_i^1 \), \( d_i^2 \), and \( d_i^3 \) were assumed) made no appreciable difference in the results.

The comparisons were limited to 700-mb winds over the western U. S.; therefore, results obtained may not apply to other atmospheric levels or other geographic areas. A similar study should be applied to the boundary layer over areas where air pollution trajectory programs are being considered.

### 4.1.3 Isobaric

Using Technique 3, we computed isobaric trajectories and grouped them by day, month, or season, starting four times daily (0000, 0600, 1200, and 1800 GMT) from any origin. A trajectory is composed of 3-hr segments, determined as illustrated in figure 4.6, vectorially added. Each 3-hr segment is computed assuming all winds reported closest to mid-segment time persist for 3 hr. The computed trajectories are presented in two forms: (1) printed latitude and longitude 6-hourly endpoint positions, and (2) computer plotted on a grid with map overlay available. (The latitudes and longitudes are "retained" by the computer program for comparison with forecast trajectories, etc.) A sample plot of 700-mb trajectories starting at Hanford, Washington, for October 1971.
<table>
<thead>
<tr>
<th>OCT 71</th>
<th>00Z</th>
<th>06Z</th>
<th>12Z</th>
<th>18Z</th>
</tr>
</thead>
<tbody>
<tr>
<td>DAY 1</td>
<td>0</td>
<td>0</td>
<td>0</td>
<td>0</td>
</tr>
<tr>
<td>DAY 2</td>
<td>15</td>
<td>10</td>
<td>29</td>
<td>23</td>
</tr>
<tr>
<td>DAY 3</td>
<td>21</td>
<td>17</td>
<td>27</td>
<td>16</td>
</tr>
<tr>
<td>DAY 4</td>
<td>24</td>
<td>20</td>
<td>30</td>
<td>24</td>
</tr>
<tr>
<td>DAY 5</td>
<td>22</td>
<td>18</td>
<td>31</td>
<td>25</td>
</tr>
<tr>
<td>DAY 6</td>
<td>23</td>
<td>19</td>
<td>29</td>
<td>23</td>
</tr>
<tr>
<td>DAY 7</td>
<td>26</td>
<td>22</td>
<td>28</td>
<td>22</td>
</tr>
<tr>
<td>DAY 8</td>
<td>17</td>
<td>5</td>
<td>30</td>
<td>23</td>
</tr>
<tr>
<td>DAY 9</td>
<td>19</td>
<td>15</td>
<td>31</td>
<td>26</td>
</tr>
<tr>
<td>DAY 10</td>
<td>19</td>
<td>5</td>
<td>29</td>
<td>20</td>
</tr>
<tr>
<td>DAY 11</td>
<td>23</td>
<td>15</td>
<td>30</td>
<td>24</td>
</tr>
<tr>
<td>DAY 12</td>
<td>21</td>
<td>13</td>
<td>29</td>
<td>24</td>
</tr>
<tr>
<td>DAY 13</td>
<td>22</td>
<td>17</td>
<td>30</td>
<td>23</td>
</tr>
<tr>
<td>DAY 14</td>
<td>15</td>
<td>10</td>
<td>29</td>
<td>25</td>
</tr>
<tr>
<td>DAY 15</td>
<td>20</td>
<td>11</td>
<td>29</td>
<td>25</td>
</tr>
<tr>
<td>DAY 16</td>
<td>11</td>
<td>3</td>
<td>29</td>
<td>25</td>
</tr>
<tr>
<td>DAY 17</td>
<td>8</td>
<td>0</td>
<td>29</td>
<td>25</td>
</tr>
<tr>
<td>DAY 18</td>
<td>7</td>
<td>0</td>
<td>29</td>
<td>21</td>
</tr>
<tr>
<td>DAY 19</td>
<td>15</td>
<td>10</td>
<td>30</td>
<td>25</td>
</tr>
<tr>
<td>DAY 20</td>
<td>14</td>
<td>5</td>
<td>29</td>
<td>23</td>
</tr>
<tr>
<td>DAY 21</td>
<td>17</td>
<td>6</td>
<td>24</td>
<td>22</td>
</tr>
<tr>
<td>DAY 22</td>
<td>21</td>
<td>9</td>
<td>30</td>
<td>25</td>
</tr>
<tr>
<td>DAY 23</td>
<td>16</td>
<td>6</td>
<td>31</td>
<td>27</td>
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<td>DAY 24</td>
<td>15</td>
<td>5</td>
<td>29</td>
<td>24</td>
</tr>
<tr>
<td>DAY 25</td>
<td>16</td>
<td>9</td>
<td>28</td>
<td>20</td>
</tr>
<tr>
<td>DAY 26</td>
<td>14</td>
<td>7</td>
<td>31</td>
<td>27</td>
</tr>
<tr>
<td>DAY 27</td>
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<td>22</td>
</tr>
<tr>
<td>DAY 28</td>
<td>13</td>
<td>5</td>
<td>31</td>
<td>27</td>
</tr>
<tr>
<td>DAY 29</td>
<td>11</td>
<td>2</td>
<td>27</td>
<td>19</td>
</tr>
<tr>
<td>DAY 30</td>
<td>9</td>
<td>4</td>
<td>29</td>
<td>21</td>
</tr>
<tr>
<td>DAY 31</td>
<td>13</td>
<td>8</td>
<td>29</td>
<td>24</td>
</tr>
<tr>
<td>TOTAL</td>
<td>322</td>
<td>36</td>
<td>305</td>
<td>725</td>
</tr>
<tr>
<td>TOTAL FOR ALL TIME PERIODS</td>
<td>865</td>
<td>688</td>
<td>522</td>
<td>384</td>
</tr>
</tbody>
</table>

Figure 4.3. Computer printout comparing the frequency of estimated and observed wind for October 1971 using Technique 3. The numbers give the number of comparisons made at the indicated times and days. The conditions are reported wind at the observing (0) station, condition C1, and at least three reported winds less than 300 n miles from the observing station, condition C2.
Figure 4.4. Example computer printouts (same period and conditions given in fig. 3) comparing estimated post facto winds (DP or P) with projections of the vector error between observed and post facto winds along (DA) the estimated winds. The numbers shown are comparison frequencies; LLL indicates a frequency greater than 99. A key is shown in the insert and the computed statistics are at the lower left.
Figure 4.5. Same as figure 4.4 except with projection of the vector error normal to (DN) the estimated winds.
Table 4.1. Comparisons of Estimated ($V'$) and Observed ($V_o$) 700-mb Winds at 60 Stations in the Western U.S. Using Three Techniques. Minimum number of reporting stations per comparison ($NMIN = 3$) within a radius ($R = 300$ n miles).

<table>
<thead>
<tr>
<th>Technique</th>
<th>Number of Comparisons</th>
<th>SE (knots)</th>
</tr>
</thead>
<tbody>
<tr>
<td>October 1971</td>
<td></td>
<td></td>
</tr>
<tr>
<td>1</td>
<td>2001</td>
<td>8.2</td>
</tr>
<tr>
<td>2</td>
<td>2001</td>
<td>8.8</td>
</tr>
<tr>
<td>3</td>
<td>2428</td>
<td>8.4</td>
</tr>
<tr>
<td>Total Possible</td>
<td></td>
<td>3112</td>
</tr>
<tr>
<td>November 1971</td>
<td></td>
<td></td>
</tr>
<tr>
<td>1</td>
<td>1779</td>
<td>8.3</td>
</tr>
<tr>
<td>2</td>
<td>1779</td>
<td>9.0</td>
</tr>
<tr>
<td>3</td>
<td>2225</td>
<td>8.5</td>
</tr>
<tr>
<td>Total Possible</td>
<td></td>
<td>2894</td>
</tr>
</tbody>
</table>

$$SE = \sqrt{\frac{SE_{DA}^2 + SE_{DN}^2}{2}}$$

$SE_{DA} = \text{standard error of DA (comparison along $V'$)}$

$SE_{DN} = \text{standard error of DN (comparison normal to $V'$)}$

is shown in figure 4.7. The grid scale and starting position on the grid can be designated by the user. In these examples, as well as all succeeding examples, a trajectory segment was computed only if the criteria $NMIN = 2$ for $R = 300$ n miles was satisfied. Otherwise, the trajectory was terminated (indicated by a latitude, longitude of 99.8, 999.8).

4.1.4 Boundary layer. An investigation of trajectories in the boundary layer has just begun. The first step has been to consider effluent movement by studying averaged winds in designated layers above the ground. The procedure for determining the average wind in a layer at a reporting station is outlined below.
Table 4.2. Comparisons When Varying R and NMIN Using Technique 3.

<table>
<thead>
<tr>
<th>Radius (n miles)</th>
<th>Number of Comparisons</th>
<th>SE (knots)</th>
</tr>
</thead>
<tbody>
<tr>
<td></td>
<td>NMIN = 2</td>
<td></td>
</tr>
<tr>
<td>200</td>
<td>972</td>
<td>9.2</td>
</tr>
<tr>
<td>250</td>
<td>2088</td>
<td>9.5</td>
</tr>
<tr>
<td>300</td>
<td>2618</td>
<td>9.7</td>
</tr>
<tr>
<td>350</td>
<td>2709</td>
<td>9.8</td>
</tr>
<tr>
<td>400</td>
<td>2755</td>
<td>9.9</td>
</tr>
<tr>
<td>450</td>
<td>2780</td>
<td>9.9</td>
</tr>
<tr>
<td></td>
<td>NMIN = 3</td>
<td></td>
</tr>
<tr>
<td>200</td>
<td>343</td>
<td>8.4</td>
</tr>
<tr>
<td>250</td>
<td>1076</td>
<td>9.0</td>
</tr>
<tr>
<td>300</td>
<td>2110</td>
<td>9.3</td>
</tr>
<tr>
<td>350</td>
<td>2535</td>
<td>9.7</td>
</tr>
<tr>
<td>400</td>
<td>2683</td>
<td>9.8</td>
</tr>
<tr>
<td>450</td>
<td>2736</td>
<td>9.9</td>
</tr>
</tbody>
</table>

Total number of possible comparisons = 2806

Terrain heights averaged over 1° x 1° areas were obtained from Reap (1968). Figure 4.8 illustrates the relationship, at a wind reporting station, between station height, average terrain height, and three layers whose thicknesses can be designated. The average wind in any layer is computed from the reported winds weighted according to their vertical spacings, as shown in figure 4.9. Trajectories in a layer can then be computed using these average layer winds at reporting stations. For a better understanding or as a check on a computer trajectory, information on reporting levels, layer bases and tops, average computed winds, and those winds used in the averaging procedure can be printed out.

Figure 4.10 shows the computer plotted trajectories starting at Idaho Falls and printed endpoint positions in layer 1 with a top 300 m above the average terrain.
Figure 4.6. Determining a 3-hour trajectory segment $\bar{P}$. Observed winds (closest to the mid-segment time) $V_1$ at position 1 and $V_2$ at position 2 are assumed to persist for the 3-hours. Trajectories $\bar{P}_1$ and $\bar{P}_2$ are 3-hour segments determined, respectively, from $V_1$ and $V_2$. $d_1$ and $d_2$ are distances to the mid-position on $\bar{P}_1$ and $\bar{P}_2$.

As mentioned earlier, the use of Technique 3 with criteria NMIN = 2 for $R = 300$ n miles, which was suggested from 700-mb observations, was used for these computations. Trajectory technique development using average winds has not as yet been fully explored. We think that this is a necessary step toward understanding effluent movement in the boundary layer, because terrain influences at different geographical locations might be an important factor in choosing which technique to use for a study of the area.

A further and most necessary extension of determining trajectories in the boundary layer will be to investigate the mixing of an effluent between layers. No work has, as yet, been done on this aspect of the problem.

4.1.5. Constant height. The program for computing boundary layer trajectories was modified to compute trajectories at a constant height. This was done by choosing a thin layer enveloping the desired height and then proceeding as with the boundary layer program. The output is identical with that described above.

The trajectory programs described above can be run backward in time as well as forward in time. Thus it is possible to compute a trajectory that ends at a given location. Figure 4.11 shows some computer-plotted trajectories ending at Idaho Falls, Idaho, and printed starting positions in layer 2 of thickness 900 m.
Figure 4.7. Computer plot of 700-mb trajectories that start four times daily at Hanford, Washington, for 6 October 1971. The trajectory starting positions within the grid are indicated by the grid point surrounded by zeros.
An additional form of output from the trajectory programs is available which gives monthly or seasonaly frequencies that trajectories of varying durations will pass over geographic areas of interest. The present program computes frequencies within squares on the grid; the grid scale and starting position within the grid can be designated by the user. An example is shown in figure 4.12 for 700-mb trajectories of 24 hrs duration starting four times daily at Yucca Flat, Nevada, during October 1971. The numbers are centered in the grid square and indicate the percent of total trajectories for the month (or season, if desired) that pass over the grid areas. The distance in nautical miles from the trajectory starting position is given at the lower and left grid boundaries. Hand analyses of the percentage have also been included. A modified version of the boundary layer trajectory program combined with this form of output would be especially effective for computing and showing average concentration values from a continuous effluent source.

Figure 4.8. Relationship between station height, average terrain height, and three layers of designated thickness.
Still another form of output shows trajectories that cross any chosen border. The trajectories are identified by their starting time and computer plotted on a grid of any selected state. Included is a plot of the borders that were chosen. An example is shown in figure 4.13 for 700-mb trajectories starting four times daily from Yucca Flat, Nevada, for October 1971.

4.2 Forecast Trajectory Programs

4.2.1. Isobaric. In support of various programs at the Nevada Test Site, isobaric trajectory forecasts are computed twice daily at NOAA's National Meteorological Center, Suitland, Maryland, and sent to ARL in Las Vegas over close-circuit teletype. These forecasts are based on wind information at standard pressure surfaces determined by the Primitive (PE) model. Forecasts at intermediate pressure levels are obtained by linear interpolation.
Figure 4.10. Trajectories starting at Idaho Falls, Idaho, for 15 September 1971, in layer 1 with a top 300 m above the average terrain.
Figure 4.11. Trajectories in layer 2 of thickness 900 m ending at Idaho Falls, Idaho, for 16 September 1971.
Figure 4.12. Frequencies with which 700-mb trajectories of 24 hours duration starting four times daily at Yucca Flat, Nevada, pass over 50 x 50 n mile areas for October 1971. Printed numbers are grid centered. Those along the axis indicate the distance in nautical miles from the trajectory starting position. Percentages have been hand analyzed.
Figure 4.13. Sample printout of those 700-mb trajectories starting four times daily at Yucca Flat, Nevada, for October 1971 that cross the border delineated by the printed letters B.
The program can calculate 30 trajectories during a single run. Any combination of starting locations, starting times (lag after initial map time), and pressure levels up to 30 can be specified. The current forecast message gives the latitude and longitude of endpoints at 6-hr intervals for trajectories starting at 12 locations in the western U.S. including six lag times and four pressure levels.

4.2.2. Isentropic. The isentropic forecast program presently under development is similar to the above program, but it also includes temperatures and isobaric heights as determined by the PE model. Trajectories are calculated on constant isentropic (θ) surfaces, rather than at constant pressure levels. The user still specifies a pressure level at which he wants the trajectory to start. The θ values at that level becomes the isentropic surface on which the trajectory is calculated.

This program, developed jointly with William Davis of Battelle-Pacific Northwest Laboratories (Hanford, Wash.) considers three stages: (1) the basic kinematic isentropic trajectory, (2) an energy balance correction, and (3) diabatic influences. The energy balance correction to the kinematic trajectories is currently being developed.

4.3 Trajectory Comparison Program

The technique for comparing forecast and post facto trajectories has been discussed in detail (List, 1972). Since then, additional 700-mb wind data have been processed, post facto trajectories computed, and the comparison program run with these trajectories and the forecast trajectories based on the PE model. A brief resumé of the parameters used in the comparison technique is given in figure 4.14. Examples of the comparison program computer printout are shown in figures 4.15 to 4.17.

The 700-mb post facto and forecast trajectories starting two times daily (0000 and 1200 GMT) from Yucca Flat, Nevada, for December 1971 were compared and some results are shown in figures 4.18 and 4.19. Figure 4.18 indicates that an expected post facto trajectory endpoint is generally behind and to the left of those for a given forecast trajectory for durations of 36 hr. Greater separations between endpoint occur as the lag increase. The 50 percent confidence radii, as shown in figure 4.19, increase with longer trajectory duration and with greater lag.

These results for 00 and 24 hr lags are shown in figures 4.20 and 4.21, respectively. Included are the envelopes about the expected trajectory endpoints within which 50 percent of the forecast trajectory endpoints would be contained (50 percent of confidence envelopes). The size of the confidence envelopes in figure 4.21 indicates that forecast trajectories of 24 hr lag must be used with extreme caution.
FORECAST
(End 2 to End 1
Normal to End 1)

EXPECTED
POSITION = \overline{Y_A} or \overline{Y_N}

50\% \text{CONFIDENCE RADIUS} = 1.18 \sqrt{\frac{SE_A^2 + SE_N^2}{2}}

SE_A = \text{STANDARD ERROR OF } Y_A
SE_N = \text{STANDARD ERROR OF } Y_N

Figure 4.14. An explanation of terms used in the computer trajectory comparison program. The computer printout labels a forecast trajectory "1" and a post facto trajectory "2."
Figure 4.15. Sample printout of the trajectory comparison program. The numbers are comparison frequencies along END1 of trajectory of 0 lag, 12 hr duration from Yucca Flat, Nevada, for December 1971.

<table>
<thead>
<tr>
<th>END2 to END1</th>
<th>0</th>
<th>2</th>
<th>5</th>
<th>3</th>
<th>2</th>
<th>1</th>
</tr>
</thead>
<tbody>
<tr>
<td>END1 to END1</td>
<td>0</td>
<td>1</td>
<td>3</td>
<td>1</td>
<td>2</td>
<td>1</td>
</tr>
<tr>
<td>400</td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
</tr>
<tr>
<td>350</td>
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<td>300</td>
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<td>4</td>
<td>1</td>
<td></td>
<td>1</td>
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<td></td>
</tr>
</tbody>
</table>

| 500         |   |   |   |   |   |   |
| 450         |   |   |   |   |   |   |
| 400         |   |   |   |   |   |   |
| 350         |   |   |   |   |   |   |
| 300         |   |   |   |   |   |   |
| 250         |   |   |   |   |   |   |
| 200         |   |   |   |   |   |   |
| 150         |   |   |   |   |   |   |
| 100         |   |   |   |   |   |   |
| 50          |   |   |   |   |   |   |
| 0           | 5 | 1 | 1 |   |   |   |

0 50 100 150 200 250 300 350 400 450 500 550 600 650 700 750 800 850 900 950 1000
Figure 4.16. Same as figure 4.15 except normal to END1.
Figure 4.17. The statistics from figures 4.15 and 4.16.

<table>
<thead>
<tr>
<th>00 Hours Lag</th>
<th>12 Hours Duration</th>
</tr>
</thead>
<tbody>
<tr>
<td>ENU2 to FNID (A)</td>
<td></td>
</tr>
<tr>
<td>N = 42</td>
<td></td>
</tr>
<tr>
<td>XHAR = 24 &amp; 9</td>
<td></td>
</tr>
<tr>
<td>YHAR = -34 &amp; 9</td>
<td></td>
</tr>
<tr>
<td>SX = 10 &amp; 5</td>
<td></td>
</tr>
<tr>
<td>SY = 5 &amp; 7</td>
<td></td>
</tr>
<tr>
<td>CORR = -.170</td>
<td></td>
</tr>
<tr>
<td>SLOPE = -199</td>
<td></td>
</tr>
<tr>
<td>YINTER = -14 &amp; 7</td>
<td></td>
</tr>
<tr>
<td>XINTER = -21 &amp; 3</td>
<td></td>
</tr>
<tr>
<td>SE = 57 &amp; 9</td>
<td></td>
</tr>
</tbody>
</table>

ENU2 to FNID (A) |
N = 42 |
XHAR = 24 & 9 |
YHAR = -34 & 9 |
SX = 10 & 5 |
SY = 5 & 7 |
CORR = -.170 |
SLOPE = -199 |
YINTER = -14 & 7 |
XINTER = -21 & 3 |
SE = 57 & 9 |

50% CONFIDENCE RADIUS = 67.4 |
50% CONFIDENCE RADIUS/XHAR = .271

Figure 4.18. A comparison of endpoints of post facto to forecast trajectories at 700 mb (with lags of 00 and 24 hr) starting twice daily at Yucca Flat, Nevada, for December 1971. (A definition of terms is given in fig. 4.14.)
Figure 4.19. Endpoint scatter (radius of the circle within which 50 percent of the expected trajectory would end) for those trajectories given in figure 4.18.
Figure 4.20. Pictoral representations of comparisons in figure 4.18 and 4.19 for 00 hr lag.

Figure 4.21. Same as figure 4.20 except for 24 hr lag.
4.4 References


This project is to investigate the feasibility of conducting long-range atmospheric tracer experiments to study transport and diffusion of gaseous plumes over continental distances. The goal is to improve our understanding of plume behavior and thus enhance the ability to estimate location and strength of a source from air samples obtained at great distances.

One approach could be a series of experiments in which a tracer gas is released for 3 to 12 hours, then cross-country air sampling at ground level and aloft would determine the distribution of the tracer as a function of time and distance from the source. The feasibility of such large-scale experiments rests on the availability of a nontoxic tracer gas that can be released in large quantities and detected at very low concentrations by using relatively simple and inexpensive techniques. Ideally, the amount of tracer already present in the atmosphere should be below the level of detectability. Compounds investigated as potential tracers include sulfur hexafluoride (SF₆) and two halocarbons (C₂F₄Br₂ and CF₂Br₂). All are detectable by electron-capture gas chromatography at extremely low concentrations (parts in 10¹² or better). Pre-concentration of a sample will allow detection down to 1 part in 10¹⁴.

At first, SF₆ appeared to be the prime candidate for a tracer since sampling and analysis techniques have been under development for some time. However, it now appears that existing concentrations of SF₆ in air caused by leakage from electrical equipment and other industrial sources, could lead to ambiguous results in a long-range experiment. A background survey conducted last summer involved collecting over 100 air samples along the Eastern Seaboard (see previous semi-annual report). Many samples showed concentrations greater than 1 part in 10¹². In light of these results and the rapidly increasing commercial use of SF₆, attention was shifted to the other candidate tracers that are believed to be present in much lower concentrations.

¹ Much of this research was supported by the Advanced Research Projects Agency of the Department of Defense.
A contract was arranged with Dr. James Lovelock, a leading authority in sampling and analyzing halocarbons by gas chromatography, to develop prototype equipment for sampling and detecting two selected halocarbons, to determine proven limits of detection, and to report on the feasibility of using these compounds for a long-range tracer experiment. Dr. Lovelock's report indicates that both CF$_2$Br$_2$ and C$_2$F$_4$Br$_2$ are feasible tracers for long-range experiments. He has delivered a prototype of a portable dual-chromatograph capable of analyzing SF$_6$ and the two halocarbons. He has also suggested, and tested in the laboratory, a cryogenic sampling technique that permits analysis of concentrations as low as a few parts in $10^{14}$ by volume. Detailed plans are being developed for a field experiment in September 1972 to test a variety of equipment and techniques for release, sampling, and analyzing the chosen tracers. This test, at the Idaho Falls National Reactor Testing Station (NRTS), will also provide detailed data on plume behavior out to 90 km. Design of the experiment remains essentially as described in the previous semi-annual report. For comparison, several additional tracers will be released at no cost to this project. Also arrangements are being made with Brookhaven National Laboratory to mount their chromatograph in an aircraft for this experiment. Their instrument has the unique ability to obtain continuous SF$_6$ profiles across a plume.

Although the use of the halocarbons, CF$_2$Br$_2$ or C$_2$F$_4$Br$_2$, as tracers in cross-country plume tracking appears to be technically feasible, very large amounts of the tracer are required; therefore, the experiments will be costly and, at the farthest distances, the present detection capability would be pushed to its limit. Therefore, an alternative experimental approach is also being considered. In lieu of extensive cross-country sampling of plumes from a few releases of these tracers, a model of long-range plume behavior might be developed from routine (daily or twice daily) sampling of an existing radioactive plume originating at NRTS. It is proposed to set up sampling equipment at about 15 National Weather Service stations along an arc 1500 km downwind of NRTS. More extensive sampling would be impractical because of the relatively high cost of sampling and analysis. Maximum advantage might be gained by including occasional releases of SF$_6$ or halocarbons into the NRTS plume and combining detailed sampling data within the first hundred km with data from the 1500 km arc.