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A SUMMARY OF RESEARCH ON THE NASA-GLOBAL ATMOSPHERIC SAMPLING PROGRAM PERFORMED BY THE ATMOSPHERIC SCIENCES RESEARCH CENTER

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INTRODUCTION

This report summarizes activities at the Atmospheric Sciences Research Center (ASRC) in support of the Global Atmospheric Sampling Program (GASP), from January 1977 through July 1978. The role of ASRC has been generally three-fold;

to assist in calibration of, and suggest modifications to,
 GASP trace constituent instrumentation;

2. to provide consulting services to NASA on questions which might concern the quality of GASP trace constituent data, and in situations requiring meteorological expertise; and,

3. to develop methods of analyzing GASP trace constituent data which would provide useful information on the constituents themselves, and their relation to the meteorology of the upper troposphere and lower stratosphere.

Specific activities under roles 1 and 2 are described in Pratt and Falconer (1978). Progress on the third objective has been slower than anticipated because of technical problems involving GASP instrument deployment. Thus, much less data than originally expected was available for study, especially data for aerosol and particle constituents. Furthermore, the simultaneous measurement of trace species is of great interest for reasons discussed in the GASP review meetings of September 1977 (Ames Research Center) and January 1978 (Lewis Research Center). This type of approach was not possible using ozone data alone.

Nevertheless, the ozone data constitutes a unique and relatively large number of measurements near the tropopause. The remainder of this report describes activities under role 3. The GASP data were provided

-1-

to ASRC by NASA-Lewis Research Center on nine magnetic tapes (Holdeman, <u>et al</u>, 1978 and references therein). In June of 1978 ASRC received GASP data from the Pan American Flight 50, which include near-continuous measurements of carbon monoxide, particles, and ozone. A summary of the analyses, to date, is included here.

DATA ANALYSIS

Annual Cycle of Ozone in the Upper Troposphere

(i) Background and Analysis Technique

As part of ASRC's early involvement with the GASP program, an analysis of the annual ozone cycle beneath the tropopause was initiated. The preliminary findings (Falconer, 1978) suggested that an annual tropospheric ozone cycle (cf. Falconer, Pratt and Mohnen, 1978) is apparently a feature which can be determined from long term aircraft records.

These early analyses seem to further indicate that during the warm season, between March and July, two ozone maxima might be anticipated in the upper troposphere, the second perhaps as a result of the tropopause lifting phenomenon originally described by Staley (Staley, 1962) and subsequently included in a model of the tropospheric ozone budget (Junge and Czeplak, 1968). Since Falconer's original work we have extended the ozone data series through January 1977.

The location of the tropopause associated with each ozone data record was obtained from a space-time interpolation of tropopause pressure fields produced by the National Meteorological Center. The analysis schemes used by NMC to produce gridded, global tropopause pressure records twice daily have been based upon procedures developed by Flattery (1971) and Gustafson (1965). Importantly, for the GASP data set the objective analysis scheme used to derive the tropopause location was changed from

-2-

the Flattery technique to the Gustafson technique in mid December of 1975. The differences between these methods and the solution which they generate have been described by Holdeman <u>et al.</u> (1976). Despite the observation that the Gustafson method consistently seems to locate the analyzed tropopause below that which would have been anticipated from the Flattery technique, no attempts were made in this study to eliminate this discrepancy.

(ii) The Findings

The biweekly ozone records from middle latitudes at flight altitudes located from 150 to 250 hPa below the NMC tropopause are reproduced in Figures 1-3. The anticipated annual ozone periodicity (cf. Falconer, Pratt and Mohnen, 1978) is rather well-expressed for latitudes 20°N-30°N, 30°N-40°N, and 40°N-50°N. However, the secondary maximum which Falconer had earlier described for a "typical year" is an ambiguous feature of 1975 and 1976 in view of the standard deviations associated with the warm season, bi-weekly means. Although the tropopause lifting phenomenon probably contributes to the ozone burden of the upper troposphere, these data cannot be effectively used to quantify the importance of this feature.

These same data records, obtained well below the tropopause show a surprising inter-latitudinal similarity of their respective mean ozone amounts ($45 \pm 3 \text{ ppbV}$) and average annual amplitudes ($20 \pm 5 \text{ ppbV}$). This is in contrast to the data of Chatfield and Harrison (1978) for selected North American stations during 1966-69, and the aircraft-derived tropospheric ozone measurements made during 1970-73 in the European-African sector (Pruchniewicz <u>et al.</u>, 1974), both of which suggest distinctive differences in the annual ozone cycle between the subtropics and northern

-3-

middle latitudes. We judge such discrepancies to be real and undoubtedly related to the year-to-year differences in the efficiency of stratospheric tropospheric exchange rates and north-south ozone transport within the troposphere. The GASP data would be consistent with rapid meridional advection but much more aerological documentation would be necessary to close this issue. Recent data from the Global Atmospheric Measurement Experiment on Tropospheric Aerosols and Gases (GAMETAG) indicate that episodic ozone transport across the middle latitudes occurs in the midtroposphere as well. Based upon the GASP data, the recent GAMETAG results, and other evidence for long range, meridional transport (cf. Danielsen, 1967 or Mani <u>et al</u>., 1977), we may hypothesize that ozone is easily exchanged in some years between the subtropics and middle latitudes through the action of meridional motions associated with mid-latitude wave disturbances.

The GASP data records indicate the mean amount and variation of ozone content increases across the tropopause. In Figs 3-5 we indicate how rapidly this change occurs at latitudes 40° N-50°N. The 1975-76 average ozone mixing ratios suggest that the free tropospheric ozone content well-below the tropopause (150-250 hPa) is perhaps one-third the amount found surrounding the tropopause itself (±50 hPa). We estimate the mean abundance and variability to be 50 ± 20 ppbV, 65 ± 27 ppbV, and 145 ± 90 ppbV at 150-250 hPa and 50-150 hPa below the tropopause, and within 50 hPa of the tropopause, respectively. Apparent interannual differences of the mean ozone levels; particularly at the tropopause level, are primarily the result of differences between tropopause analysis schemes described previously. However, real year to year variations in the tropopause height can be expected. In general, Figs. 3-5 are consistent

-4-

with the concept of an ozone reservoir above the tropopause which frequently supplies air to the upper troposphere.

Preliminary Ozone Climatology Along Global Flight Routes

(i) Background and Analysis Technique

As part of a recent effort to assess the basic ozone climatology at aircraft cruise altitudes, we have examined nearly 41,000 ozone observations from the participating GASP B-747 airliners and, from these records, constructed a preliminary map series indicating the probability that various, pre-assigned ozone mixing ratio levels may be exceeded in certain locations and at different times of the year. The data records from March 1975 through January 1977 were collected into bi-monthly periods for the region 250 hPa below the tropopause to 150 hPa above the tropopause. No explicit correction was applied to the data to account for known differences in the tropopause analysis schemes. We judge that these statistics do not suffer any serious deficiencies because of such an omission.

For each observing period and altitude region the cumulative frequency distributions were computed for 10° latitude by 10° longitude (Marsden square) areas provided that at least ten independent data points were available. Continuous (limit) mode data have been excluded. The probabilities of equalling or exceeding a given ozone level within each Marsden square were determined, and correspond to the fraction of observations in the Marsden square which exceed the specified level. Contours of equal probability were drawn.

The ozone levels selected for these analyses are not entirely arbitrary. Extensive experimental testing of the relative abundance of cabin (inside) ozone to ambient (outside) levels in various airliners indicated that ozone

-5-

destruction due to thermal and line losses is apparently not a unique number (Table 1). For example, recent NASA statistics (Perkins, <u>et al.</u> 1978) indicate that under average circumstances the B-747 aircraft equipped with a charcoal filtration system (D = 95%) is 16 times more efficient in removing ozone from the cabin airstream that an unmodified

AIRCRAFT	MODIFICATIONS TO SYSTEM	AVERAGE DESTRUCTION OF AMBIENT OZONE (D)	MAXIMUM AMBIENT OZONE LEVEL FOR CABIN ATTAINMENT of 80 ppbV
B-747	none	61%	205 ррЪV
B-747S P	none	20%	100 ppbV
B-747 SP	reduced cabin air recircu- lation rate	42%	138 ppbV
B-747 SP	fifteenth stage compressor	81%	421 ppbV
B-747 SP	charcoal filter- ing system	95%	1600 ppbV

Table 1. Recent estimates of the average percent destruction of atmospheric ozone within the cabin environment of various B-747 aircraft (cf. Perkins et al., 1978).

B-747 SP (D = 20%). We may use these destruction loss values in conjunction with a prescribed cabin ozone attainment level of, say, 80 ppbV to arrive at a maximum ambient ozone level beyond which the attainment ozone mixing ratio is likely exceeded (cf. last column, Table 1). We have assumed three representative destruction constants, namely 60%, 73%, and 80%, such that the corresponding ambient, threshold ozone levels are 200-, 300-, and 400 ppbV; the probabilities of encountering greater ozone amounts have been computed (without assuming a probability distribution function) from the empirical cumulative ozone frequency ogives.

The frequency distributions of ozone levels for individual Marsden squares exhibited by GASP data cannot in general be approximated by a normal or other analytic distribution. A combination of sampling time and location and meteorological conditions lead to various unimodal or bimodal distributions in different locations and times of year. We therefore feel that is is appropriate to base calculations of exceedance statistics on <u>empirically</u> determined histograms of GASP ozone values. An alternative approach has been adopted by Belmont <u>et al</u>. (1978).

(ii) The Findings

The probability charts of March-April are perhaps of greater interest than those of other months because they generally coincide with the ozone episode season. Figure 6 is typical of these charts and represents the probabilities associated with encountering ozone at levels of at least 200 ppbV in the upper troposphere and lower stratosphere. For those aircraft whose characteristic ozone destruction losses are on the order of 60%, flight routes across the midwestern United States, the northern Pacific, between northern Laborador and Iceland, and over much of the European Continent hold the most potential for cabin ozone encounters.

Although this result is entirely consistent with meteorological considerations which we have described elsewhere in this report, we should point out that the greatest contribution to geographic variability seen in the probability isopleths comes from characteristics of the ozone samples themselves. It is important to recognize that the spatial structure of the isopleths in Fig. 6 is determined by a combination of three factors which are implicit in the GASP data. These are the spatial structure of the mean tropopause height, the location of the

-7-

particular flight routes on which data was taken, and the non-uniform distribution of GASP data throughout individual time periods. For instance, the extent to which stratospheric ozone values enter into a sample must be carefully accounted for. This, in turn, is a function of both the typical cruise altitude along the flight path and the average tropopause pressure along the path.

If we compare the percentage of GASP ozone data obtained in the stratosphere (Fig. 7) with the probability isopleths shown in Fig. 6 we are struck by the great similarity between the two. Other meteorological parameters such as the geopotential heights of upperlevel pressure surfaces do not reveal an analogous pattern, although certainly upper level waves in the tropopause heights and in the ozone distribution itself influence the isopleth patterns to some extent. The similarity of Figs. 6 and 7 therefore largely results from the fact that the GASP aircraft experienced stratospheric flight in preferred locations (such as the central U.S.), and the probability distributions were established accordingly. Although this situation could be a problem for establishing climatologically representative fields, it is in fact appropriate for investigating the probabilities of scheduled airline encounters with high ozone. The complete set of bi-monthly, probability isopleths for ozone mixing ratios in excess of 200-, 300-, and 400 ppbV, and the corresponding total number of GASP observations are given in Appendices A and B.

-8-

Limit Mode Analysis

A survey of all occurrences of continuous ozone recordings found on the first seven GASP tapes was undertaken in order to assess the frequency, duration, and location of such data for possible further analysis. In particular, it was of interest to determine whether the segments were long enough to permit the computation of small scale space spectra of ozone.

Continuous or limit mode recording can be initiated by turbulence or a sample bottle exposure, and results in effectively three independent ozone measurements per minute. Approximately 55 continuous record segments were found which are at least 40 km in length, or approximately 5 minutes duration. Very few were longer, and the longest was 122 km (9 min.). A scan of some of the data indicated relatively little variability in most of the segments. In view of this characteristic and the relatively short length of the records, it was decided not to pursue a spectral analysis. Fig. 8 shows the geographical distribution of segments at least 40 km in length.

Some of the longer data segments exhibited a regular wavelike variation in ozone. For the example in Fig. 9, one wavelenth was spanned in about 1.3 minutes, corresponding to a wavelength of about 20 km. This is probably a manifestation of a gravity wave such as that seen by Danielsen and Mohnen (1977) in very high resolution data. The data in Fig. 9 is from the NASA CV 990, which recorded several long segments in continuous mode during the Interhemispheric Survey flights (NASA 1977). It demonstrates clearly the ability of the GASP system to record accurately relatively small scale and low amplitude fluctuations. Two of the GASP aircraft have recently been modified to record long continuous data segments on some flights.

-9-

Flight Route Analysis

Characterization of various properties along individual flight routes has been proposed as a means of dealing with the peculiar distribution of GASP data in both space and time.

An initial phase of this research was carried out in Spring of 1978. All flights across the North Atlantic, and from the U.S. West Coast to Hawaii, were isolated for analysis. These particular routes were chosen for their meteorological interest and relatively high frequency of data. One goal was to determine to what extent geographic variations of several parameters were apparent, as well as variations in the vertical as measured by ambient pressure, or pressure distance from the NMC tropopause. It was also of interest to study the northward horizontal ozone flux near the tropopause, especially the degree to which longitudinal variations in this quantity are important. The parameters examined were temperature, wind components, and ozone mixing ratio, as well as the northward fluxes of sensible heat, zonal momentum, and ozone, through correlations involving wind. Heat and momentum fluxes computed from GASF data were compared with existing computations of these quantities based on much more extensive meteorological data sets. This was done to assess the degree to which the calculation of these quantities from GASP data was representative in a climatological sense. This would serve at least as a necessary condition for accepting the ozone fluxes as representative, due to the extreme variability of ozone at GASP levels.

The study thus far has shown that on the North Atlantic and Hawaii flight routes, and probably for much of the rest of the GASP data, the data is too sparsely and irregularly distributed to permit the calculation of stable correlation quantities such as horizontal fluxes. Ozone itself

-10-

is sufficiently variable that this holds true for simple means of that quantity as well as its horizontal flux at GASP levels. This implies that horizontal fluxes which have been computed ignoring the longitudinal distribution of GASP data may be misleading, since sampling variability is large compared to the quantity being measured. Nastrom (1978) has also expressed this concern after having analyzed GASP data.

In view of this problem of distribution, GASP data does not lend itself to fixed-location estimates of fluxes or other correlations over a particular time period. However, each individual flight record contains information on the horizontal distribution of quantities at a fixed time. Future work with the flight route approach will attempt to exploit this fact. Also, although meteorological variability obscured most of the geographic variations on the two flight routes already examined, other routes (such as those crossing the Rocky Mountains) are worth examining for topographically-related features. We feel that there remains considerable room for attempting many other analysis techniques with the two-year GASP ozone data set.

Compositing Studies

The relation of ozone distribution near the tropopause to meteorological structures on the synoptic scale is well established (Nastrom, 1977, 1978; Reed, 1950), as are some aspects of its sub-synoptic scale distribution (Danielsen <u>et al.</u>, 1975). Most of the GASP data consists of horizontal profiles of ozone from a range of meteorological situations. With careful analysis of the large ozone data set provided by GASP, it is possible that our overall knowledge of the nature and frequency of cross-tropopause transfer of ozone (stratospheric air) can be improved. Compositing data obtained from similar meteorological features may prove useful in this

-11-

regard although preliminary attempts at compositing ozone data from several flights through a persistent upper-level trough served primarily to define the problems of compositing rather than resulting in a meaningful and scale-consistent, average distribution for ozone in a typical trough.

The most difficult problem to overcome, if this approach is to prove useful, is a means of establishing references in the horizontal and vertical to apply to each meteorological situation. An intermediate step toward this goal is to arrange the data in such a form that it can be readily compared with the meteorological setting. This now being done by cataloging the flights by geographical region and time period, and by producing computer line printer plots of ozone, pressure altitude, and the meteorological variables for ready reference to, for instance, the contemporaneous NMC 200 hPa analysis of geopotential height. Presentation of the data in this manner (Fig. 10) expedites the investigation of the ozone data in a wide range of situations, whether or not they are potential candidates for compositing. The more recent data, which includes species other than ozone will also be plotted in this manner, for inspection prior to correlation computations.

Flight 50 Analysis

Data from Pan American's around-the-world flight has recently been analyzed at ASRC. This flight represents the first occasion when ozone, carbon monoxide, condensation nuclei, and light scattering particle data were available on a single flight. Moreover, the flight itself was only the second such circumglobal research expedition in recent times (Machta <u>et al.</u> 1967). ASRC analysis of Flight 50 data has resulted in two research papers, the results of which are described below.

-12-

(i) High resolution GASP system data taken in a variety of meteorological situations has been examined. Despite various instrument limitations, the system as a whole appears capable of providing a consistent and sometimes quite detailed description of several trace constituents in their meteorological setting. (cf Figs. 11-14)

(ii) Stratospheric ozone and condensation nuclei concentrations across the north and south polar caps are generally anticorrelated as would be expected for constituents with opposite vertical gradients through the upper troposphere and lower stratosphere.

(iii) A clear anti-correlation between ozone and carbon monoxide is only obvious in those situations where stratospheric and tropospheric air masses were sampled in succession. Within the stratosphere or the troposphere, such a correlation is not obvious in this data.

(iv) The first measurements of carbon monoxide in the Antarctic lower stratosphere obtained on Flight 50 ranged from 35 to 51 ppbV, averaging 44 ppbV.

(v) The horizontal distribution of tropical, upper tropospheric ozone was generally uniform. However, evidence has been found of relatively abrupt changes in concentration which coincide with transitions between air masses of different meteorological origin.

(vi) Carbon monoxide levels were substantially higher in the tropics
over Africa (89 ppbV) than over the open waters of the Pacific Ocean
(66 ppbV), even after excluding a region of anomalously high CO (115 ppbV).
(vii) Despite the apparent longitudinal differences in tropical CO, a
pronounced latitudinal uniformity was observed on the tropical segments
of the flight.

(viii) A local region of elevated CO was encountered in the tropics over Africa. Available synoptic data indicates that a reasonable explanation is the generation of CO by hydrocarbon reactions over vegetated land, and its subsequent transport to flight level by deep convection. (ix) Particles sufficiently large to scatter light were rather uniformly distributed $(1.0 \pm 0.5 \text{ cm}^{-3})$ throughout the upper troposphere and lower stratosphere at all latitudes (Fig. 15). Concentrations greater than 1.0 cm^{-3} were found only over the Pacific, particularly near the storm zone $(50^{\circ}\text{S} - 60^{\circ}\text{S})$ and Intertropical Convergence Zone. These regions may be the major source regions and transport routes of particles to the upper troposphere.

(x) Smaller particles were rather symmetrically distributed about the globe at altitudes between 160 and 250 hPa. A maximum occurred over the ITCZ and number concentrations were nearly equal on the Greenwich and dateline sides of the world. Enhanced concentrations occurred over the boundaries between tropical and temperate air masses. Comparison of total and light scattering particle data at several latitudes enforces Junge's conclusion that coagulation exceeds sedimentation as a sink mechanism for particles in the higher layers. Where fresh injections are rare, and coagulation times are long, as over the Arctic and Antarctic, small particles are few.

(xi) Generally, aerosol concentrations decrease with increasing latitude, but higher concentrations are sometimes found just above strong inversions, in the vicinity of high winds, indicative of a possible transport route for particles from troposphere to stratosphere. There are small interhemispheric differences in total aerosol concentration; but these may be seasonal or even due to chance in a single observation.

-14-

Concluding Remarks

The GASP data base now includes a substantial, and still increasing, base of multiple-species observations. Many details involving data organization, evaluation, and interpretation have been worked out at the Atmospheric Sciences Research Center. Major work now in progress includes a study of the time dependence and spatial character of all tropical tropospheric ozone data, and analysis of trace constituent distributions and dynamical quantities for a number of subtropical jet stream crossings. We expect that data availability and quality in the third year of GASP analysis will be suitable for fruitful study of the mutual variation of constituents, as well as the particle data alone, over a wide variety of situations.

-15-

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Figure 1: Biweekly ozone mixing ratio averages obtained from participating GASP airliners at pressure distances of 150 to 250 hPa beneath the tropopause between latitudes 20°-30°N.



Figure 2: Biweekly ozone mixing ratio averages obtained from participating GASP airliners at pressure distances of 150 to 250 hPa beneath the tropopause between latitudes 30°-40°N.



Figure 3. Biweekly ozone mixing ratio averages obtained from participating GASP airliners at pressure distances of 150 to 250 hPa beneath the tropopause between latitudes 40°-50°N.



Figure 4: Biweekly ozone mixing ratio averages obtained from participating GASP airliners at pressure distances of 50 to 150 hPa beneath the tropopause between latitudes 40°-50°N.



Figure 5. Biweekly ozone mixing ratio averages obtained from participating GASP airliners at pressure distances of 50 hPa above to 50hPa beneath the tropopause between latitudes 40°-50°N.



Figure 6: Percent probability of exceeding an ozone mixing ratio of 200 ppbV in the upper troposphere and lower stratosphere during March and April. Based upon GASP records from 1975-1977. Cross hatching indicates regions where participating GASP airliners did not fly.



Figure 7: Percentage of GASP ozone data which was obtained in the lower stratosphere during March and April. Based upon GASP records from 1975-1977.



Figure 8: Number of continuous record cycles exceeding approximately 40 km (5 minutes) in the upper troposphere and lower stratosphere. Based on GASP data from 1975-1977.



Figure 9: Example of small ozone variation which suggests the presence of gravity waves. Data were obtained on 14 November 1976 at the 216 hPa level south of New Zealand.

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Figure 10: Sample computer line plot of ozone (0), aircraft cruise pressure (P), and tropopause pressure (-) for a flight between Bangkok (BKK) and Tehran (THR) on 9 August 1976. Time markers, latitudes, longitudes, wind speeds and directions are also indicated on the vertical axis.



Figure 11: Flight record obtained within the stratosphere between San Francisco (SFO) and London (LHR). Ozone (0_3) and carbon monoxide (CO) mixing ratios, condensation nuclei (CN) and light scattering particle $(1-2 \text{ cm}^{-3} \text{ indicated})$ by solid shading, $\geq 2 \text{ cm}^{-3}$ shown as hatched shading) concentrations obtained from the GASP sensors. Pressure altitude, ambient air temperature, windspeed and direction are derived from aircraft flight systems. All data have been averaged over 1-deg latitude intervals, except for CO which represents a several minute average.



Figure 12: Flight record obtained within the troposphere between London (LHR) and the South Atlantic Ocean, due south of Capetown, South Africa (CPT). Plotting conventions are described in Fig. 11.



Figure 13: Flight record obtained within the Antarctic stratosphere enroute to Auckland, New Zealand (AKL). Plotting conventions are described in Fig. 11.



Figure 14: Flight record obtained within the Pacific troposphere between Auckland (AKL) and San Francisco (SFO). Plotting conventions are described in Fig. 11.



Figure 15: A polar plot showing the aerosol concentrations detected by Pan Am Flight 50 while making a Polar circumnavigation of the Earth 29-31 October 1977. The flight level in millibars, and the presence of tropospheric or stratospheric air at the flight level are shown on the periphery of the figure. The mean values of total aerosol concentration, measured by a condensation nucleus counter, averaged over 10 degree latitude bands are shown along with mean values of larger particles found by light scattering techniques. APPENDIX A

Percent probability of exceeding an ozone mixing ratios of 200-, 300-, and 400 ppbV in the upper troposphere and lower stratosphere during the indicated, bi-monthly periods. Computations have been based upon combined GASP data records from 1975-1977. Cross hatching represents regions where no commercial flights were made.





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APPENDIX B

Number of ozone observations made from participating GASP airliners during the indicated bi-monthly periods in the upper troposphere and lower stratosphere. Compiled from combined data records from 1975-1977.



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