Influence of a middle-latitude cyclone on tropospheric ozone distributions during a period of TRACE A

Robert O. Loring Jr.,1 Henry E. Fuelberg,1 Jack Fishman,2 Mark V. Watson,1 and Edward V. Browell2

Abstract. A middle-latitude cyclone occurring during the Transport and Atmospheric Chemistry near the Equator-Atlantic (TRACE A) experiment is examined to determine its influence on distributions of tropospheric ozone over the South Atlantic Ocean. A maximum of tropospheric ozone is located in the vicinity of this cyclone on October 3, 1992. Flight level data and meteorological analyses indicate a downward protrusion of dry, ozone-rich stratospheric air near the cyclone, i.e., a tropopause fold. Forward trajectories show that air parcels arriving in the upper troposphere of the cyclone originate in the stratosphere. Forward trajectories are calculated from these locations having stratospheric histories. They indicate that some air is transported as far north as 22°S, subsiding into the middle troposphere along the southern fringes of a region of enhanced tropospheric ozone that is located west of Africa on October 6. Backward trajectories then are computed along the Greenwich meridian over much of the South Atlantic Ocean. This axis passes through the tropospheric ozone maximum west of Africa and the region of strong horizontal ozone gradients along its southern border. Results indicate that most air parcels arriving north of 20°S (in the ozone-rich region) originate over Africa. Conversely, most parcels arriving south of 20°S (where there is less ozone) originate from the west, passing over the southern half of South America. Thus the tropospheric ozone maximum west of Africa on October 6 appears to be attributable to outflow from Africa, with stratospheric transport being much less important. Formerly stratospheric air near the cyclone on October 3 also is transported forward into the middle troposphere near Madagascar where there is a second maximum of tropospheric ozone on October 6. Backward trajectories from this region indicate that middle-latitude systems exert a much greater influence here than over the South Atlantic. This area experiences relatively little outflow from Africa during our period of study.

1. Introduction

A major objective of the Transport and Atmospheric Chemistry near the Equator-Atlantic (TRACE A) experiment was to investigate a climatological maximum of tropospheric ozone that has been observed at low latitudes over the South Atlantic Ocean [Fishman et al., this issue (b), 1986, 1990; Fishman and Larsen, 1987; Levy, 1988]. The maximum is most pronounced during the months of September–October when it is centered near 10°S, 0°E [Fishman et al., 1990, 1991; Hudson et al., 1995]. It has been hypothesized to result from biomass burning in both South America and Africa [Cros et al., 1988; Crutzen et al., 1985; Fishman et al., 1990, 1991; Logan and Kirchhoff, 1986] and from biogenic emissions which are nearly coincident in time and space with the burning [Harris et al., this issue; Levine et al., this issue; Swap et al., this issue]. The burning is a common agricultural practice that is most widespread during the relatively dry months of August–October [Setzer and Pereira, 1991; Cahoon et al., 1992]. Precursors of ozone from the burning and biogenic emissions are photooxidized in the generally cloud-free tropical atmosphere and thought to be transported to the South Atlantic by the three-dimensional flow [e.g., Fishman and Larsen, 1987].

Additional sources of tropospheric ozone include tropopause folds that are associated with extratropical cyclones [Danielsen, 1968; Reed, 1955; Reed and Danielsen, 1959; Shapiro, 1976, 1978, 1980; Sechrist et al., 1986; Browell et al., 1987; Uccellini et al., 1985], transverse circulations occurring with jet streaks [Allam and Tuck, 1984; Sechrist et al., 1986; Shapiro et al., 1982], and subsidence associated with anticyclones [Krishnamurti et al., 1993].

General circulation models have been used to estimate the transport of ozone from the stratosphere into the troposphere [Mahlman et al., 1980; Levy et al., 1985]. Results indicated that large-scale processes are responsible for most of the cross-tropopause flux. Austin and Follows [1991] used data from ozonesondes to conclude that only 25% of the ozone mixing ratio at 300 mbar for Payerne, Switzerland, was due to stratospheric intrusions. The remainder was attributed to in situ photochemical production. Since tropospheric ozone is both transported from the stratosphere and produced photochemically [Levy, 1988], it is important to understand their relative roles in producing the ozone anomaly over the South Atlantic Ocean.

The current paper examines the influence of a middle-latitude cyclone on horizontal distributions of tropospheric ozone over the tropical South Atlantic and southwest Indian Oceans. We focus on a cyclone that occurred during TRACE
various meteorological parameters are at 12-hour intervals.

Mission 9, on October 3, 1992, was a trans-Atlantic flight from Rio de Janeiro, Brazil, to Johannesburg, South Africa, that traversed a middle-latitude wave cyclone located over the central South Atlantic Ocean.

Several types of data were used to investigate this case. The DC-8 "flying laboratory" sampled numerous meteorological parameters and chemical constituents at flight level. Dropsondes were released at regular intervals, providing temperature, dew point, and wind velocity as functions of pressure. The airborne differential absorption lidar (DIAL) remotely sensed the vertical distribution of ozone and aerosols above and below flight level.

Our analyses of tropospheric ozone were based on a technique described by Vukovich et al. [1995] and applied by Fishman et al. [this issue (a)] to determine the utility and accuracy of using satellite measurements for studying the distribution of tropospheric ozone in the tropics. The scheme employs values of total column ozone from the total ozone mapping spectrometer (TOMS), and stratospheric ozone concentrations from the SBUV-II (solar backscatter ultraviolet) instrument. The tropospheric residual is obtained by subtracting the stratospheric component from the total column value. Fishman et al. [this issue (a)] demonstrate that the technique captures synoptic scale features of tropospheric ozone extremely well over ocean surfaces. Although they describe the potential limitations of the technique over land, the current study only considers ozone distributions over water. Low-level clouds can cause errors in TOMS data retrievals. However, Fishman et al. [this issue (a)] have shown them to have little effect on the accuracy of the technique during the TRACE A period. Thus we believe that the ozone maxima that we discuss during our 5-day study period collectively contribute to the climatological maximum described by Fishman et al. [1990].

Large-scale meteorological conditions were described using global gridded analyses prepared by the European Center for Medium-Range Weather Forecasting (ECMWF) and the National Center for Atmospheric Research (NCAR). These analyses were assimilated from surface observations, radiosonde data, aircraft reports, satellite-derived soundings, and cloud drift winds. The various meteorological parameters are at 12-hour intervals (0000 and 1200 UTC), and their spatial resolution is 2.5 × 2.5° latitude/longitude with 15 vertical levels between 1000 and 50 mbar. We used the lowest 12 levels (1000, 850, 700, 500, 400, 300, 250, 200, 150, 100, 70, and 50 mbar).

Potential vorticity (PV) was calculated using the isobaric form of the Ertel equation [Crum and Stevens, 1988]; that is,

\[
PV = \left( \frac{\partial \omega}{\partial \phi} \right) - \left( \frac{\partial}{\partial \phi} \left( \frac{\partial \theta}{\partial y} + \frac{\partial v}{\partial x} \right) \right),
\]

where \( (\xi + f) \) is absolute vorticity, \( (\partial \omega/\partial \phi) \) is static stability, and \( (\partial u/\partial \phi)(\partial \theta/\partial y) + (\partial v/\partial \phi)(\partial \theta/\partial x) \) is temperature advection due to vertical wind shear. This advection term does not appear when PV is calculated on isentropic surfaces. Calculations were made at 50-mbar intervals between 1000 and 50 mbar. The 12 levels of ECMWF data had been interpolated to these levels using a cubic spline procedure.

Forward and backward three-dimensional trajectories were calculated using a kinematic trajectory model in which parcels are advected by the three wind components without employing the isentropic assumption. The vertical motions needed in these computations were supplied with the ECMWF data set. Moosm [1990] has emphasized the importance of three-dimensional trajectories in studies of long-range chemical transport. Our kinematic scheme used cubic splines to interpolate the ECMWF data to 5 mbar vertical intervals, with linear interpolation within that interval. Bilinear interpolation was used for horizontal interpolation. The data also were linearly interpolated to 5 min time intervals. With this small time step, parcels were advected using

\[
x(t + dt) = x(t) + V[x(t)] dt
\]

where \( x(t + dt) \) is the parcel's position at time \( t + dt \), \( x(t) \) is its previous position, \( V \) is the three-dimensional wind vector, and \( dt \) is the time step. Trajectories that intersected the Earth's surface were terminated at that location. Fishberg et al. [this issue (a)] give additional details about this procedure and compare trajectories from it with those from an isentropic scheme.

3. Synoptic Discussion

3.1. Meteorological Analyses

Although Bachmeier and Fuelberg [this issue] provide a meteorological overview of the entire TRACE A experiment, it is useful to describe our study period in greater detail. The wave cyclone under study had its origins near the southeast coast of South America on September 30, 1992 (not shown). Its lowest pressure of 992 mbar occurred at 1200 UTC October 1 (Figure 1a) when located near 45°S, 25°W. The cyclone moved north-eastward and weakened; so by 1200 UTC October 4 (Figure 1d), all that remained was a trough near 42°S, 7°W. A second wave cyclone was centered southeast of the system of primary interest on October 1 (Figure 1a). This low became very intense (964 mbar) while advancing eastward (e.g., Figure 1b). Additional cyclones traveled along the southern edge of the analysis region throughout the period. Broad areas of high pressure developed over the South Atlantic and southwest Indian Oceans between October 4 and 6 (Figures 1d–1f). A frontal trough extended south of Africa toward a low center near 60°S, 40°E on October 6, while a developing cyclone was located just east of South America.

Three-dimensional atmospheric structure on the day of the trans-Atlantic flight (October 3) is depicted in Figures 2a–2c. The sea level pressure analysis (Figure 2a) shows the wave cyclone near 40°S, 15°W with a central pressure of 1009 mbar. Its associated surface cold front (not shown) extended north-westward from the center. The NASA aircraft traversed the northern portion of this system (Figure 2a). At 850 mbar (Figure 2b) the streamlines show the wave cyclone to be a major circulation feature. In addition, a broad region of anticyclonic flow stretched equatorward of the cyclone. In the upper troposphere (300 mbar, Figure 2c) a closed cyclonic circulation was centered near 40°S, 15°W. The polar jet stream (not
Three-dimensional structure on October 6, the last day of the study period, is shown in Figures 2d-2f. Major oceanic anticyclones flanked by wave cyclones dominated the surface flow (Figure 2d). These anticyclones also dominated the circulation at 850 mbar (Figure 2e). The anticyclones were displaced equatorward at 300 mbar (Figure 2f), with trough/ridge patterns influencing the middle latitudes. The prominent trough near Madagascar was associated with the surface cyclone that had dissipated several days earlier over the central South Atlantic but was followed by additional cyclones farther south (Figure 1). The polar jet streak had merged with the subtropical jet stream (not shown).

3.2. Ozone Analyses

The analysis of tropospheric ozone on October 3 (Figure 3a) indicates several regions of enhanced values. A maximum of 55 Dobson units (DU) off the west coast of central Africa (labeled M1) is consistent with a climatological feature hypothesized to be associated with widespread biomass burning [e.g., Fishman et al., 1990] and biogenic emissions [e.g., Harris et al., this issue]. A second, weaker maximum exceeding 45 Dobson units (DU) (labeled M2) is located near the transient cyclone at 35°S, 15°W. This ozone feature is described in detail by Fishman et al. [this issue (a)] where it is shown to result from large amounts of ozone in a layer between 430 and 270 mbar. Finally, there is enhanced tropospheric ozone near Madagascar (labeled M3). This feature also is consistent with a climatological area of enhanced tropospheric ozone [e.g., Fishman et al., 1990].

The tropospheric ozone maximum near the cyclone (M2) appears to weaken and move eastward through October 4 (Figure 3b), while the anomaly off the west coast of Africa (M1) expands and becomes more intense. This combination produces an elongated zone of relatively large tropospheric ozone along the Greenwich meridian. The anomaly just south of Madagascar (M3) has changed little.

Ozone maximum M2 that is associated with the wave cyclone is not readily apparent on October 5 (Figure 3c). Its remnants likely are located near 35°S, 0°E. M1 has changed little, while values south of Madagascar in M3 have increased to 55 DU. A new maximum (not labeled) is located near 50°S, 10°W, northwest of a wave cyclone that is south of the analysis region (Figure 1d).

On October 6 (Figure 3d) there is a broad area of enhanced tropospheric ozone (M1) with dual centers of approximately 58 DU located west of Africa. The maximum south of Madagascar (M3) continues, while the newer maximum south of 40°S between 10°W and 25°E has moved eastward. Unlike feature...
4. Results

Our objective is to investigate relationships between horizontal distributions of tropospheric ozone (Figure 3) and the wave cyclone located over the South Atlantic Ocean (Figures 1 and 2). We first document a tropopause fold near the cyclone using a combination of flight data and ECMWF meteorological analyses. We then estimate heights of the tropopause and calculate backward trajectories to locate parcels having stratospheric histories. Forward trajectories are used to examine the movements of this present or formerly stratospheric air. Finally, backward trajectories are calculated from regions of enhanced ozone.

4.1. Flight Data

Middle-latitude wave cyclones are accompanied by undulations in tropopause height. Stratospheric air above the tropopause is characterized by low humidity, strong static stability, large values of PV, and large ozone concentrations [Danielsen, 1968]. The thin line in Figure 4a depicts the concentration of flight level ozone as a function of time. The major increase in ozone after 0940 UTC (30°S, 30°W) occurs as the aircraft approaches the cyclone. The concentration reaches 300 parts per billion by volume (ppbv) by 1047 UTC (35°S, 22°W) and then fluctuates slightly for the following 55 min. Values decrease as the aircraft exits the cyclone. The vertical distribution of ozone along the flight track (Plate 1) was obtained from the airborne DIAL instrument [Browell et al., this issue; Browell, 1989]. The important feature is the local decreased altitude of the largest ozone values near the cyclone. Specifically, values of 80 ppbv extend as low as 6 km near 37°S, 15°W. Finally, the thick line in Figure 4a represents water vapor mixing ratio at flight level. The driest air (0.018 g kg⁻¹) is encountered during the period of maximum flight level ozone when the aircraft is near the center of the cyclone.

We calculated PV along the flight track using the ECMWF data at 2.5° x 2.5° resolution. Values of flight level PV (thick line in Figure 4b) range from 0.1 to 4.6 x 10⁻⁵ K s⁻¹ mbar⁻¹ (hereinafter called PV units). The abrupt increases at 0950 and 1315 UTC represent changes in flight altitude. One should note the increasing PV as the aircraft approaches the cyclone’s center. Values are greatest (4.6 PV units) near 1130 UTC.
(38°S, 14°W) and then decrease as the aircraft exits the cyclone. These variations in PV are similar to those observed with flight level ozone (thin line in Figure 4b).

The dry, ozone-rich air with large PV at flight level (Figure 4, Plate 1) indicates passage into a tropopause fold. These variations agree with earlier studies [e.g., Danielsen et al., 1987]. Although the spatial and temporal resolutions of the aircraft and ECMWF data differ greatly, their good agreement suggests that ECMWF data can be used to represent the large-scale meteorological environment of the South Atlantic region.

4.2. Meteorological Cross Sections

Vertical cross sections are valuable tools for diagnosing tropopause folds and are most revealing when orientated perpendicular to them. The tropopause fold associated with the cyclone on October 3 should slope from higher altitudes in the south toward lower altitudes in the north. Since the west to east flight track is not oriented perpendicular to the fold, we used ECMWF data to construct cross sections from southwest (47.5°S, 30°W) to northeast (20°S, 0°E) (see axis in Figure 2a).

The depression of isentropic surfaces above 350 mbar between 42°S, 24°W and 31°S, 12°W (Figure 5a) represents the center of the cyclone at these levels. The upper level frontal zone is indicated by downward sloping isentropic surfaces extending from the depression toward the northeast (the right). These features are similar to previous examples of upper level frontal zones [e.g., Shapiro et al., 1980, 1981].

Greatest values of PV above 700 mbar occur near the center of the cyclone (Figure 5b). Although the coarse resolution of the ECMWF data does not permit small-scale structures to be resolved, this downward extension of enhanced PV is consistent with a tropopause fold. The protrusion of dry air (mixing ratios <0.05 g kg⁻¹) extending to 450 mbar near 35°S, 17°W (Figure 5c) also suggests a stratospheric intrusion.

Tropopause folds extend equatorward from jet streams [e.g., Shapiro, 1980], and both polar and subtropical jet streams are
indicated in the cross section of total wind speed (Figure 5d). The polar jet is located on the southwest (left) side of the diagram between 300 and 400 mbar, while the subtropical jet is located farther north around 200 mbar. The polar jet is directed northward (into the page) at 31 m s⁻¹, while the subtropical jet stream flows southeastward (out of the page) at 38 m s⁻¹. The position and orientation of the polar jet stream is consistent with a tropopause fold near 35°S, 17°W.

Sinking motion occurs beneath the upper level frontal zone (Figure 5e), while rising motion exists farther northeast ahead of the front. The greatest subsidence (2 μbar s⁻¹) is located just southwest of the fold near 37°S, 18°W. This descent transports dry stratospheric air into the upper and middle troposphere (Figure 5c).

To summarize, each meteorological parameter along the cross section indicates a tropopause fold. This evidence is consistent with that from the flight level data (Figure 4) and vertical cross section of ozone (Plate 1).

**4.3. Tropopause Evaluation**

Tropopause heights were needed over the entire TRACE A region. It is informative to examine the several procedures for obtaining these heights since they can lead to different values.

The World Meteorological Organization (WMO) defines the tropopause as the base of the lowest layer in which the temperature lapse rate becomes less than 2°C km⁻¹ and does not exceed that value for at least 2 km [e.g., Reiter et al., 1969]. Thermal definitions are difficult to apply near baroclinic disturbances and jet streams because of the numerous stable layers associated with upper level fronts [Danielsen 1959, 1968; Reiter, 1975]. In the current case, a dropsonde sounding in the northern sector of the cyclone at 38°S, 14.5°W (Figure 6) exhibits a nearly isothermal layer between 490 and 520 mbar. Although this feature likely represents the lowered tropopause, the lapse rate remains less than 2°C km⁻¹ for only 1.6 km. Thus the tropopause would be reported at a much higher altitude (~300 mbar) based on the WMO definition.

The tropopause also can be located using a dynamical approach [e.g., Reed, 1955; Reed and Danielsen, 1959; Shapiro, 1978]. Specifically, it is denoted as a zero-order discontinuity in potential vorticity (PV) separating relatively small values in the troposphere from larger values in the stratosphere. The PV-defined tropopause exhibits greater spatial and temporal continuity than one defined thermally [Danielsen, 1968, Danielsen and Hipskind, 1980].
Figure 5. Vertical cross section from 47.5°S, 30°W to 20°S, 0°E (see axis in Figure 2a) for 1200 UTC October 3. (a) Isentropes (K). (b) Potential vorticity where $1 = 1.0 \times 10^{-5}$ K s^{-1} mbar^{-1}; values ≥1 (solid) are contoured at intervals of 1; values <1 (dashed) are contoured at intervals of 0.1. (c) Mixing ratio (g kg^{-1}); values ≥1 (solid) are contoured at intervals of 1; values <1 (dashed) are contoured at intervals of 0.1; additionally, the 0.05 g kg^{-1} isopleth is indicated. (d) Total wind speed, values >10 m s^{-1} are contoured at intervals of 5 m s^{-1}. (e) Vertical motion, isolines are at intervals of 0.5 µbar s^{-1}; arrows denote ascent or descent.

There is no general agreement regarding the PV threshold that represents the tropopause. Suggested thresholds have ranged from 1.0 to 3.5 PV units [Reed, 1955; Staley, 1962; Danielsen, 1964, 1968; Shapiro, 1980; WMO, 1986; Browell et al., 1987; Danielsen et al., 1987; Hoerling et al., 1991]. We calculated surfaces of constant PV for values between 1.0 and 3.5 units on October 3 and analyzed pressures on each. We then used the thermal definition to locate the tropopause at 11
The two dynamical criteria (PVs of 1.0 and 1.5) have developed a more sophisticated
sounding sites in the middle latitudes. The various thermally derived tropopause heights did not correspond to any single
PV threshold; however, most occurred between 1.0 and 1.5 PV
units.

The tropopause also has been defined on the basis of chemical
criteria. The downward protrusion of enhanced ozone into the middle troposphere seen in Plate 1 corresponds to the
tropopause fold. More specifically, an ozone concentration of
100 ppbv often is used to denote the tropopause. In addition,
Browell et al. [this issue] have developed a more sophisticated
technique using DIAL-derived ozone concentrations.

Figure 6 depicts estimates of tropopause heights along the flight track based on the various techniques discussed above.
Data for a given technique were not available at all times. The
lowest ozone-based tropopause heights are 450 mbar (100
ppbv threshold) and 400 mbar (DIAL procedure). When the
thermal definitions are applied at the dropsonde site near the
cyclone (Figure 6), the tropopause is indicated at either 300
mbar (WMO definition) or 520 mbar (thermal discontinuity).
As noted earlier, these heights, especially the 300-mbar value,
may be unreliable since the location is close to the polar jet
streak and upper level frontal zone [Danielsen, 1959, 1968;
Reiter, 1975]. The two dynamical criteria (PVs of 1.0 and 1.5
units, Figure 7) yield tropopause heights that differ less than 25
mbar, except near the center of the cyclone (1130 UTC) where
they differ approximately 65 mbar. These PV-derived heights
show good agreement with those inferred from the two ozone
procedures.

We will use the dynamical approach to locate the tropo-
pause because it appears suitable for use with ECMWF data,
and there is precedent for its selection [Hoerling et al., 1991,
1993]. Instead of using a single PV threshold, we will utilize the range 1.0–1.5 units, thereby acknowledging the uncertainties
described above.

4.4. Conditions on October 3

We now investigate the histories of air parcels arriving near
the cyclone and associated tropospheric ozone maximum M2
on October 3 (Figures 2 and 3), seeking to identify those
parcels which have stratospheric histories. Our procedure is
patterned after that of Danielsen [1980] who utilized the 8en-
tropic coordinate system. We calculated 72-hour backward tra-
jectories beginning at 1200 UTC October 3 and ending at 1200
UTC September 30. Trajectories that encountered PV > 1.5
units during the 72-hour period were considered likely to have
had stratospheric histories. Similarly, those trajectories which
encountered PV between 1.0 and 1.5 units were possibly of
stratospheric origin.

The limitations of this methodology should be noted. Spe-
sifically, intense small-scale mixing occurs near jet streams
and tropopause folds [Danielsen, 1968, 1980; Shapiro, 1978, 1980;
Browell et al., 1987; Danielsen et al., 1987]. Therefore as strato-
spheric air ascends, it acquires a progressively greater tropo-
spheric component, causing values of stratospheric markers
such as ozone and PV to decrease. Although the integrity of air
parcels is questionable in these situations [Vaughan, 1988],
trajectories nevertheless have provided valuable information
about the histories of such parcels [Danielsen, 1980].

Our backward trajectories originated at intervals of 2.5ø
latitude/longitude on and within a box bounded by 30øS to 50øS
and 0øE to 30øW (Figure 8a). This box encompasses the cy-
clone (Figure 2a) and tropospheric ozone maximum M2 (Fig-
ures 3a, 8a). Trajectories were calculated to arrive at 50-mbar
intervals between 450 and 200 mbar, with 117 trajectories at
each level. Trajectories arriving at 350 mbar (Figure 8b) are
similar to those arriving at the other levels. Specifically, most of
them originate from the west, with many exhibiting major
horizontal undulations due to the strongly curved and rapidly
changing upper level flow (Figure 2).

Figure 9 indicates those grid points of the trajectory box
where arrivals appear to have been within the stratosphere
during the previous 72 hours. Arrivals at the five levels are
shown separately. The various symbols denote the value of PV
that was encountered (either exceeding 1.5 PV units or be-
tween 1.0 and 1.5 PV units) along with the time that the parcel
last encountered that value (at the current time (1200 UTC
October 3) or during the previous 72 hours). Grid points that
are blank never encountered PV of 1.0 units during the pre-
ceeding 72 hours. The analysis of tropospheric ozone for Octo-
ber 3 (isopleths $\geq 45$ DU from Figure 3a) is superimposed on

Figure 6. Vertical profile of temperature and dew point from
the dropsonde released at 1133 UTC October 3 (38øS,
14.5øW). Sounding lines represent dew point (left) and tem-
perature (right).

Figure 7. Tropopause heights along the flight track between
0930 UTC (29.1øS, 31.2øW) and 1330 UTC (35.5øS, 7.3øE)
based on the simple thermal (bottom dash) and WMO defini-
tions (top dash) at the dropsonde location, 1.0–1.5 PV units
(stippled), 100 ppbv ozone concentration (O3), and DIAL-
derived ozone technique (D).
Figure 8. (a) Locations for trajectory arrivals (square enclosing M2) superimposed on tropospheric ozone valid for October 3 (as in Figure 3a). (b) Horizontal projections of 3-day backward trajectories arriving at 350 mbar. Trajectories end at 2.5° latitude/longitude intervals on and within the box in Figure 8a. They end at 1200 UTC October 3 and begin at 1200 UTC September 30. (c) As in Figure 8b but only for those trajectories displaying stratospheric characteristics. (d) As in Figure 8b but for 5-day backward trajectories arriving at 700 mbar.

Each panel to the extent legible, and the complete analysis is shown in Figure 9f. Ozone data were not available south of 50°S. Table 1 contains statistics for each level.

All parcels (trajectories) arriving near ozone maximum M2 (the box) at 200 mbar (Table 1, figure not shown) and 97% of those arriving at 250 mbar (Table 1, Figure 9a) either are within the stratosphere at 1200 UTC October 3 or encountered stratospheric PV during the previous 72 hours. These percentages decrease steadily for arrivals at the lower altitudes (greater pressures). One should note that the locations of parcels having apparent stratospheric histories correspond closely to the location of M2. For example, at 400 mbar (Figure 9d) the center of M2 corresponds to parcels which encountered PV > 1.5 units during the previous 72 hours, whereas the periphery of M2 corresponds to parcels encountering PV between 1.0 and 1.5 units during that period. Only 23% of the parcels arriving at 450 mbar (Table 1, Figure 9e) had stratospheric histories. However, each is located near the center of enhanced tropospheric ozone. This agreement between trajectory and ozone data lends credibility to our methodology, demonstrating that ECMWF analyses and satellite-derived ozone data can be used together to examine this study period.

The trajectories with apparent stratospheric histories originate south and west of the surface cyclone. For example, the 72 trajectories arriving at 350 mbar and encountering PV > 1.0 unit during the preceding 72 hours are shown in Figure 8c. The surface cyclone was located near 40°S, 15°W on the arrival date (October 3, Figure 2a). Mean pressure altitudes for these trajectories are shown as a function of time in Figure 10. The parcels descend an average of 55 mbar in 36 hours as they approach the cold region behind the cyclone and frontal zone (Figure 2).

Backward trajectories arriving at levels below 450 mbar also were examined. Since wind speeds at these altitudes generally are slower than those aloft, these trajectories were run for 5 days. Arrivals at 700 mbar (Figure 8d) are similar to those at the other lower levels; that is, all originate from the west or southwest, crossing the southern portion of South America where biomass burning was minimal during TRACE A [Fishman et al., this issue (b)]. None has a stratospheric history (not shown).

In summary, there is a close relation between the locations of parcels having stratospheric histories and the position of tropospheric ozone maximum M2 located near the middle-latitude cyclone. Air near M2 does not originate over Africa. These results and those of the previous sections indicate that this tropospheric ozone maximum is due to downward transport of stratospheric air.

4.5. Forward Trajectories to October 6

We next investigate the future locations of those parcels near M2 on October 3 which had stratospheric histories. Specifically, forward trajectories were calculated for all parcels that encountered PV > 1.0 unit during the preceding 72-hour period (those in Figure 9). These trajectories began at 1200 UTC October 3 and ended 72 hours later at 1200 UTC October 6. Figure 11 depicts them for each of the six upper tropospheric levels used earlier.
Figure 9. Trajectories with stratospheric histories (based on PV) superimposed on analysis of tropospheric ozone (contours at 5-DU intervals for values ≥45 DU) for 1200 UTC October 3 at (a) 250, (b) 300, (c) 350, (d) 400, and (e) 450 mbar. (f) Complete ozone analysis (as in Figure 3a) where M2 denotes the cyclone-related area of enhanced ozone. Boxed asterisk represents >1.5 PV units at 1200 UTC October 3, solid circle represents >1.5 PV units during the past 72 hours, asterisk represents PV between 1.0 and 1.5 units at 1200 UTC October 3, and open circle represents PV between 1.0 and 1.5 units during the past 72 hours.

Table 1. Statistics Describing Backward Trajectories Originating Within the Tropospheric Ozone Feature on October 3 (see Figure 9)

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Figure 10. Mean pressure altitude as a function of time for 3-day backward trajectories having stratospheric histories. The mean is based on all trajectories in Figure 8c.
Strong upper level westerly flow (Figure 2c, 2f) causes most parcels leaving 200 mbar (Figure 11a) to travel eastward and pass south of Africa. Most parcels departing 250 mbar (Figure 11b) follow similar paths. However, some that originate behind the cyclone and cold frontal zone head northeastward and arrive off the west coast of southern Africa. This path is more evident for parcels departing the 300- and 350-mbar levels (Figure 11c and 11d). Trajectories departing northeast of the cyclone's center travel southeastward toward 45°S, 10°E. Some then turn northeast, arriving east of South Africa near Madagascar. The remaining parcels at 45°S, 10°E continue to fan out toward the northeast. There are fewer formerly stratospheric parcels at 400 and 450 mbar; however, their paths are similar to those leaving 300 and 350 mbar.

It is informative to examine these forward trajectories from near M2 in greater detail, relating their motions to locations of tropospheric ozone maxima M1 and M3 that were described earlier (Figure 3). Figure 12 depicts 72-hour forward trajectories (through 1200 UTC October 6) that depart 350 mbar on October 3 (i.e., those in Figure 11d). Each of these parcels has a stratospheric history (PV > 1.0 unit) during the preceding 72 hours (Figure 9c). The analysis of tropospheric ozone for October 6 (from Figure 3d) is superimposed. Figure 13 contains average pressure altitudes as a function of time for two groups of trajectories: those arriving off the west coast of Africa near the southern fringes of M1 (solid line) and those arriving south of Madagascar near M3 (pluses).

The formerly stratospheric parcels arriving at the southern edge of tropospheric ozone maximum M1 on October 6 (Figures 12, 13) have traveled generally northeastward. They decelerate considerably during the 72-hour period while slowly subsiding. Although they arrive at an average pressure of 710 mbar, some individual parcels descend 450–500 mbar during the 3-day period, sinking deep into the lower troposphere to 800–850 mbar. Some of these parcels travel well into the tropics, reaching near 20°S. Extending the trajectory period
beyond 3 days (not shown) produces little additional movement into the tropics.

The formerly stratospheric parcels arriving south of Madagascar near tropospheric ozone maximum M3 undergo considerable change in both direction and altitude along the way (Figures 12, 13). These parcels travel southeastward during the initial 24 hours while ascending to an average pressure of 325 mbar. Some individual parcels reenter the stratosphere, while others remain within the troposphere. The relative minimum of tropospheric ozone near 45\øS, 10\øE on October 4 (Figure 3b) corresponds to the mean location of these parcels after 24 hours. The trajectories then veer northeastward, traveling around the southern tip of Africa while descending. This subsidence into levels of slower winds retards their forward motion during the final 24 hours of calculation. They arrive near M3 at an average pressure of 570 mbar, although some individual parcels descend to as low as 650–700 mbar.

Our forward trajectories (Figures 11-13) show excellent agreement with those of Danielsen [1964, 1980] during a tropopause folding event. Some of his trajectories descended into the lower levels and turned anticyclonically toward the tropics. In the current case, these trajectories arrive west of southern Africa (Figure 12) at latitudes near 20\øS. Others of his trajectories remained within the fold in the upper troposphere, turning cyclonically while ascending back into the stratosphere. Some then could reenter the troposphere during the next folding event. In the current case, these trajectories turn cyclonically toward the southeast and then back to the northeast, arriving near Madagascar (Figure 12). These trajectories move through the cyclone faster than the phase speed of that cyclone.

4.6. Backward Trajectories From October 6

Backward trajectories were calculated from tropospheric ozone maxima M1 and M3 on October 6 to better understand these features. We first describe 6-day backward trajectories arriving along the Greenwich meridian at 2.5\ø latitude intervals between 50\øS and 10\øN (Figure 14a). This axis intersects the full north-south extent of tropospheric ozone maximum M1 and much of the second maximum located south of 40\øS. Arrival pressures were at 50-mbar intervals between 200 and 950 mbar. Trajectories from all 16 arrival levels are superimposed in Figure 14b.

The latitude band north of ~20\øS corresponds to tropospheric ozone maximum M1 (Figure 14a). Most trajectories...
arriving in this area originate over Africa (Figure 14b). The outflow from Africa in this region during September–October 1992 has been described in recent studies by FueUberg et al. [this issue (b)], Swap et al. [this issue], and Garstang et al. [this issue]. Since almost none of these trajectories has a stratospheric origin (not shown), these results suggest that M1 is attributable to photochemical processes as previously hypothesized [Cros et al., 1988; Crutzen et al., 1985; Fishman et al., 1990, 1991; Logan and Kirchhoff, 1986; Harris et al., this issue; Levine et al., this issue; Swap et al., this issue]. There was widespread biomass burning over south central Africa during TRACE A [Fishman et al., this issue (b)].

Trajectories arriving south of ~20°S come from the west or southwest (Figure 14b). Some pass over southern Brazil, while many others take an even more southerly track. Most of these parcels have passed over regions of little biomass burning [Fishman et al., this issue (b)].

The southern extent of M1 is located between 20° and 25°S on October 6 (Figure 14a). The strong gradient of tropospheric ozone in this region apparently is due to the different origins of the air parcels arriving there (Figure 14b). Some parcels arriving between 20° and 25°S have stratospheric histories. For example, previously stratospheric air processed by the middle-latitude cyclone of October 1–3 reaches 20°S (Figure 12). In addition, other trajectories arriving between 20° and 25°S (not shown) have descended from the stratosphere in the anticyclonic flow behind the wave cyclone. Nonetheless, relatively few trajectories have either of these stratospheric origins. Thus their contribution to tropospheric ozone between 20° and 25°S appears to be small.

The tropospheric ozone maximum south of 40°S on October 6 (Figure 14b) corresponds to the location of an upper level trough (Figure 2f). Tropopause folding may be occurring here as observed with the cyclone on October 3. Trajectories arriving in this region have traveled far south over the South Atlantic Ocean and the extreme southern part of South America. Many more of them have stratospheric histories than observed between 20° and 25°S (not shown). Therefore this maximum may be attributable to meteorological processes instead of being an artifact of the satellite technique for deriving tropospheric ozone at latitudes poleward of 45°S [Fishman et al., 1990].

We next examine tropospheric ozone maximum M3 in greater detail. One should recall that forward trajectories with stratospheric histories arrive near M3 (Figure 12). To investigate further, we calculated 10-day backward trajectories from the region bounded by 20°–30°S, 10°E–20°W. This region includes M3 and the forward trajectories arriving there on October 6 (see dotted square in Figure 12). Arrivals at 800, 600, and 400 mbar are shown in Figure 15a, where Figures 15a–15c contain all trajectories, while Figures 15d–15f contain only those that encountered PV > 1 unit sometime during the 10-day period, thereby suggesting a stratospheric history.

The backward trajectories from M3 are very revealing (Figure 15). One should note that very few pass over Africa at these three levels (or the other levels not shown). Those trajectories that do, tend to skirt the coastline instead of traveling well inland where the biomass burning is most prevalent [Fishman et al., this issue (b)]. Furthermore, most of the few trajectories that reach South America within 10 days pass over the southern part of the continent where there is little biomass burning.

The arrivals at 800 mbar (Figure 15a) follow two basic paths across the South Atlantic Ocean. The northernmost path passes near the cyclone traversed by the DC-8 on October 3. The surface positions of this cyclone ranged between 40°–50°S, 30°–5°W during October 1–4 (Figure 1). None of these arrivals at 800 mbar has a stratospheric history (Figure 15d). The second major group of arrivals at 800 mbar follows a more southerly track that extends beyond 60°S. Many of these parcels previously have been within the stratosphere (Figure 15d), mostly near their southermost locations. The tropopause is relatively low at these higher latitudes (not shown), and it is a region of numerous surface cyclones and upper level cyclonic flow during the period of interest (Figures 1, 2).

Most trajectories arriving near M3 at 600 mbar (Figure 15b) have followed the same two basic paths as those arriving at 800 mbar. Now, however, many trajectories along both general paths have stratospheric histories (Figure 15e), encountering either the lowered tropopauses associated with the cyclone of October 1–3 near 40°S or the cyclones located even farther south. The 3-day backward trajectories from the cyclone and M2 (Figure 12) are similar to the portion of these backward trajectories east of 20°W, thereby emphasizing the role of the middle-latitude cyclone on tropospheric ozone feature M3. Finally, some trajectories exhibit anticyclonic curvature near 10°W, 25°S (Figure 15b). These parcels have been influenced by the subtropical oceanic anticyclonic flow that is a climatological feature of the area. A case study by Krishnamurti et al. [1993] demonstrated that a similar anticyclone during October 1989 helped to shape ozone distributions over the South Atlantic Ocean.

Most trajectories arriving near M3 at 400 mbar (Figure 15c, 15f) have come from the northwest, at least during the preceding several days. Very few have followed the more southern track seen at the lower levels. Some trajectories have been influenced by the middle-latitude cyclone, while others exhibit more anticyclonic curvature. The almost tropical origins of some parcels (i.e., north of 20°S) should be noted. Many trajectories from the various regions have stratospheric histories.

To summarize, both forward (Figure 12) and backward (Figure 15) trajectories to/from tropospheric ozone maximum M3 on October 6 indicate that previously stratospheric air arrives in its vicinity. Some of this stratospheric air can be attributed to middle-latitude cyclones, while another portion is associated with anticyclonic flow. Although photochemical ozone production certainly cannot be discounted during this period of study, it is important to note that relatively few parcels pass over south central Africa or central South America.

5. Summary and Conclusions

A middle-latitude cyclone occurring during the Transport and Atmospheric Chemistry near the Equator-Atlantic (TRACE A) experiment has been examined to determine its influence on distributions of tropospheric ozone over the South Atlantic Ocean during October 3–6, 1992. NASA's DC-8 "flying laboratory" passed through the northern portion of the cyclone on October 3. A maximum of tropospheric ozone (denoted M2) was located in its vicinity. Flight level data as well as ECMWF analyses indicated a downward protrusion of stratospheric air near the cyclone, i.e., a tropopause fold. The fold area was characterized by dry air and enhanced values of tropospheric ozone and potential vorticity. Backward trajectories from this region indicated that the air had been transported downward from the stratosphere by circulation patterns.
Figure 15. Horizontal projections of 10-day backward trajectories arriving near ozone feature M3 at 800, 600, and 400 mbar. (a–c) All trajectories, (d–f) only those trajectories with stratospheric histories; that is, PV > 1.0 PV units. The dotted portions of the trajectories indicate locations within the stratosphere. The outline of the trajectory arrival box is shown relative to the tropospheric ozone analysis and those forward trajectories with stratospheric histories in Figure 12.

associated with the transient cyclone. None of these parcels originated over Africa, although some had passed over southern portions of South America.

Three-day forward trajectories were calculated from this cyclone/region of enhanced tropospheric ozone (M2) on October 3. Some trajectories turned anticyclonically toward the tropics. They subsided into the middle troposphere and reached as far north as 22°S, along the southern boundary of a large region of enhanced tropospheric ozone denoted M1. The location of this ozone maximum on October 6 corresponds to that of a climatological maximum that has been described previously [Fishman et al., this issue (b), 1986, 1990; Fishman and Larsen, 1987; Levy, 1988]. Other forward trajectories reentered the stratosphere and traveled southeast. They then turned northeastward and arrived in the middle troposphere near Madagascar, coinciding with another region of enhanced tropospheric ozone (denoted M3). The location of M3 on October 6 also is consistent with a climatological zone of enhanced tropospheric ozone [e.g., Fishman et al., 1990]. Both of these groups of forward trajectories agreed closely with Danielsen's [1964] schematic of trajectories during a tropopause fold event.

Backward trajectories were calculated along the Greenwich meridian on October 6. This axis intersected tropospheric ozone maximum M1 which was located west of Africa and north of 20°S. Specifically, a strong north-south gradient of tropospheric ozone at the southern edge of M1 (from 20° to 25°S) separated ozone-rich air to the north from smaller concentrations to the south. Most trajectories arriving north of ~20°S originated over Africa. Conversely, trajectories arriving south of ~20°S originated from the west in the middle latitudes. These results indicate that tropospheric ozone maximum M1 was not influenced appreciably by middle-latitude cyclones during our period of study. Instead, biomass burning and biogenic emissions from Africa appear to be the dominant mechanisms leading to its formation [Cros et al., 1988; Crutzen et al., 1985; Fishman et al., 1990, 1991; Logan and Kirchhoff,
Middle-latitude cyclones appeared to have a much greater influence on tropospheric ozone maximum M3 that was located near Madagascar on October 6. More of the forward trajectories from M2 on October 3 reached this area than those from M1 over the central South Atlantic. In addition, backward trajectory calculations from M3 on October 6 showed many parcels with stratospheric histories arriving from near M2. Other previously stratospheric parcels had traveled farther south, experiencing cyclonic flow associated with systems at these higher latitudes. Finally, some previously stratospheric parcels originated in the subtropics, being influenced by the semipermanent high-pressure centers in that area.

Relatively few trajectories from southern Africa arrived near tropospheric ozone maximum M3 south of Madagascar on October 6. These results suggest that M3 was influenced more by stratospheric transport than by photochemical processes. It must be emphasized that we have examined only a short time period (October 1–6, 1992). Thus our findings may not represent general conditions. We currently are examining the area near Madagascar in much greater detail, exploring the large temporal and spatial fluctuations in tropospheric ozone that occurred there during the 6-week TRACE A period.

This study has shown that transient middle-latitude cyclones and their transports can be adequately analyzed over the data-poor South Atlantic Ocean using the ECMWF global data sets. It also has demonstrated the combined use of several data sources, e.g., various in situ aircraft data, airborne DIAL-derived vertical profiles of ozone, satellite remotely sensed tropospheric ozone, and ECMWF global meteorological analyses. Nonetheless, these data and our analysis procedures cannot quantify the relative contributions of photochemical and stratospheric processes. That determination likely will require a global numerical meteorological model that includes photochemical processes. Such a model would yield valuable insights into many questions about atmospheric chemistry and transport.

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