# Airborne DIAL Ozone and Aerosol Trends Observed at High Latitudes Over North America from February to May 2000

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## ABSTRACT

Ozone ($O_3$) and aerosol scattering ratio profiles were obtained from airborne lidar measurements on thirty-eight aircraft flights over seven aircraft deployments covering the latitudes of 40°-85°N between 4 February and 23 May 2000 as part of the TOPSE (Tropospheric Ozone Production about the Spring Equinox) field experiment. The remote and in situ $O_3$ measurements were used together to produce a vertically-continuous $O_3$ profile from near the surface to above the tropopause. Ozone, aerosol, and potential vorticity (PV) distributions were used together to identify the presence of pollution plumes and stratospheric intrusions. The number of observed pollution plumes was found to increase into the spring along with a significant increase in aerosol loading. Ozone was found to increase in the middle free troposphere (4-6 km) at high latitudes (60°-85°N) by an average of 4.3 ppbv/mo from about 55 ppbv in early February to over 72 ppbv in mid-May. The average aerosol scattering ratios in the same region increased at an average rate of 0.37/mo from about 0.35 to over 1.7. Ozone and aerosol scattering were highly correlated over entire field experiment. Based on the above results and the observed aircraft in-situ measurements, it was estimated that stratospherically-derived $O_3$ accounted for less than 20% of the observed increase in mid tropospheric $O_3$ at high latitudes. The primary cause of the observed $O_3$ increase was found to be the photochemical production of $O_3$ in pollution plumes.

## 1. Introduction

The TOPSE field experiment was conducted at high-latitudes over North America from February to May 2000 to investigate the cause of the springtime maximum in tropospheric $O_3$[1]. The NASA Langley Research Center's (LaRC) UV airborne differential absorption lidar (UV DIAL) system was flown on the NCAR (National Center for Atmospheric Research) C-130 aircraft for remote measurements of $O_3$ and aerosols during TOPSE. The specific objectives of the UV DIAL investigation were to: 1) provide large scale vertical profiles of $O_3$ and aerosol distributions across the troposphere and into the lower stratosphere; 2) investigate the distribution and trends in tropospheric $O_3$ and aerosols during the winter and spring transition; 3) use real-time data from the UV DIAL system to remotely identify atmospheric layers of interest in order to select aircraft altitudes and geographic locations for optimal in situ sampling; 4) determine the relative contribution to the $O_3$ budget of stratospheric–tropospheric exchange and photochemical $O_3$ production/destruction; and 5) combine the UV DIAL data with in situ measurements of gases and aerosols to aid in the interpretation of the large-scale chemical and dynamical evolution of the tropospheric composition during TOPSE. This paper discusses the trends in DIAL measurements of $O_3$ and aerosols observed in the free troposphere at high latitudes during TOPSE and relates these trends to changes found in tropospheric potential vorticity (PV), stratospheric-tropospheric exchange, and photochemical $O_3$ production.

## 2. Instrumentation and Methodology

TheTOPSE airborne UV DIAL system used two frequency-doubled Nd:YAG lasers pumping two dye lasers that were frequency-doubled into the UV to produce the on-line (288.2 nm) and off-line (299.6 nm) wavelengths at 8.6 Hz for the DIAL $O_3$ measurements. The residual 1064-nm and ~590-nm beams from the frequency-doubling process of the Nd:YAG and dye lasers, respectively, were also transmitted with the UV DIAL wavelengths. As a result, four laser beams are transmitted simultaneously into the atmosphere below (288.2, 299.6, 576.4, and 1064 nm) and above (288.2, 299.6, 599.2, and 1064 nm) the aircraft for lidar measurements of $O_3$, aerosols, and clouds from near the surface to about

3 km above the tropopause. The accuracy of the UV DIAL measurements of $O_3$ with a vertical resolution of 300 m and an averaging time of 5 minutes (about 38-km horizontal resolution at C-130 ground speeds) has been previously demonstrated to be better than 10% or 2 ppbv (parts per billion by volume), whichever is greater, with a measurement precision of 5% or 1 ppbv[2]. Intercomparisons between in situ and UV DIAL $O_3$ measurements were made throughout the course of TOPSE to ensure that the accuracy of the measurements was being maintained. The aerosol scattering ratio distributions are determined from the relative lidar backscattering distribution using an analyzed cross section of the atmospheric density along the flight track and a normalization of the lidar backscatter distribution to a clean (nearly aerosol free) region, usually found near the tropopause. The vertical and horizontal resolutions for the aerosol scattering ratio measurements are 60 m and 3.8 km (30-s averaging time), respectively.

Ozone and $S_A$ data from each flight were binned into 0.25-degree latitudinal bins and averaged to arrive at an average latitudinal distribution for $O_3$ and $S_A$ for each flight. The latitudinal $O_3$ and $S_A$ distributions below the tropopause from each flight were combined with the other flights in a deployment to produce an overall latitudinal distribution for $O_3$ and $S_A$ for the deployment. In this process, the data were averaged together when the flights had overlapping data in the same latitudinal bin. The overall $O_3$ and $S_A$ trends observed during TOPSE were derived from these deployment-average latitudinal distributions.

## 3. Results and Discussion

### *3.1 Ozone Trends*

To better quantify the trend in $O_3$ in the free troposphere, the data were binned into two latitudinal regions (40°-60°N and 60°-85°N) and two altitude regions (2-4 km and 4-6 km). The average $O_3$ value and the standard error of the average are shown in Figure 1 for each deployment in each region. A linear correlation for the first four deployments and the last four deployments as well as for the entire TOPSE field experiment are shown The observed trends in $O_3$ were larger in both altitude regions at 40°-60°N compared to the respective 60°-85°N regions. In the first three deployments at 40°-60°N, $O_3$ was rather constant at ~49 ppbv with very little vertical gradient. Starting in the fourth deployment, $O_3$ increased abruptly in the 4-6 km region to ~66 ppbv, and it stayed near that level for the rest of the field experiment. This transition was also seen in the 2-4 km region, but the change was not as large.

In the higher latitudinal region (60°-85°N), there was no similar abrupt increase in $O_3$ seen in the fourth deployment; in fact, it appears like there was no increase at all during the first four deployments. Ozone steadily increased from the fourth to seventh deployments. The intermediate correlation fits show the break in the trends between the two periods. The average increase in $O_3$ over the TOPSE field experiment in the free troposphere was found to be 2.8 ppbv/mo at low altitudes (2-4 km) increasing to 4.3 ppbv/mo in the mid troposphere (4-6 km). Examining the trend over the final four deployments, the rate would be as large as 4.7 and 6.7 ppbv/mo, respectively, with very high correlation coefficients ($R > 0.96$).

### *3.2 Aerosol Trends*

The average latitudinal distributions of aerosol scattering ratios ($S_A$) were constructed for each deployment using the procedures discussed above. They were further limited to the same regions where the $O_3$ data existed for each deployment. To better quantify the changes in $S_A$ over TOPSE, the aerosol data were binned in the same manner as the $O_3$, and this is shown in Figure 2.

The scatter in the $S_A$ values for the first three deployments was very large in all regions shown in Figure 2, with the possible exception of the 60°-85°N, 4-6 km region where the scatter was low and the $S_A$ trend for the first four deployments was consistent with the overall trend for the field experiment. In the 40°-60°N region, the $S_A$ values increased very rapidly from the fourth to seventh deployments at all altitudes in the free troposphere. In the 60°-85°N region, there was no significant change in $S_A$ values in the fourth to seventh deployments in the lower free troposphere, but in the 4-6 km region there continued to be an increase which could have even started in early February. The rate of increase in $S_A$ in the 60°-85°N, 4-6 km region was found to be 0.37/mo over the entire field experiment or 0.38/mo over the last four deployments. This compares to the 40°-60°N, 4-6 km region with 0.50/mo overall and 0.81/mo over the last four deployments.

### *3.3 $O_3$/Aerosol Correlation*

Both $O_3$ and aerosol scattering were found to increase during TOPSE in the 60°-85°N, 4-6 km region (Figures 1 and 2), but to investigate whether they were increasing together, it is necessary to examine their correlation to each other.

Figure 3 presents the correlation between $O_3$ and $S_A$ for the 60°-85°N, 4-6 km region. The deployment averages and standard error of those averages are shown along with the best fit linear correlation line and the correlation coefficient. The $O_3$ and $S_A$ data from all the deployments show very good correlation (R=0.951) with a positive regression slope of $O_3/S_A$=10.6 ppbv and intercept of 54.4 ppbv (Figure 3). The high positive correlation between $O_3$ and aerosols is usually found in association with continental pollution plumes, which is opposite to the negative correlation between $O_3$ and aerosols that is found in association with stratospherically influenced air observed in the troposphere[3].

### *3.4 Potential Vorticity (PV),Trace Gas, and Long Range Transport Trends*

Key trace gases and aerosols were measured in situ on the C-130 during TOPSE, and their observed trends over this field experiment were used to further investigate the relative contribution of photochemistry and stratosphere-troposphere exchange in determining the $O_3$ trends across the spring equinox at high latitudes in the mid troposphere. The results from the average in situ $O_3$ measurements are shown in Figure 4, and it indicates an overall increase of 6.3 ppbv/mo, which is very close to the 6.6 ppbv/mo seen in the last four deployments from the combined UV DIAL averages. The in situ measurements of aerosol number densities in the 0.1-3.0 μm range are also shown in the Figure 4, and the aerosol number density trend reflects the same strong positive trend as was observed in the aerosol scattering data. As an indicator of stratospheric air, $^7$Be should show a significant positive trend if the stratosphere-troposphere exchange was increasing over this period of time, but the $^7$Be data showed only a small (9.2 fCi $m^{-3}$/mo) positive trend over TOPSE [4].

The variation in PV during TOPSE was examined across the same regions as for $O_3$ and aerosol scattering, and the results were evaluated for an indication of any general trend in stratosphere-troposphere exchange that occurred during TOPSE. No statistically significant positive trend in PV was found for any of the model results in the entire 40°-85°N, 2-6 km region, and in general the average trend in PV over the field experiment was slightly negative. These results indicated that instead of there being an increase in stratosphere-troposphere exchange during TOPSE, there may have actually been a slight decrease in exchange.

To investigate the source of the air reaching the TOPSE experiment region in the mid troposphere, we used 10-day backward isentropic trajectories for air masses reaching the TOPSE flight tracks at altitudes from 4 to 6 km. The later deployments (4-7) showed significant ascending air masses from continental sources into the upper troposphere when the remote measurements showed elevated ozone and aerosol in the upper troposphere. Very few backward trajectories ending in the mid troposphere at high latitudes exhibited significant descent, which would have indicated a possible stratospheric source within the last 10 days.

## 4. Summary

The combination of the UV DIAL and in situ $O_3$ measurements made from the C-130 over the seven deployments of TOPSE conducted between 4 February and 23 May 2000 showed an average increase in $O_3$ at all altitudes across the free troposphere in the latitude range of 40°-85°N. The largest average $O_3$ increases were found in the 40°-60°N region with 3.4 ppbv/mo in the 2-4 km region and 5.8 ppbv/mo in the 4-6 km region. At higher latitudes (60°-85°N), the average $O_3$ increase ranged from 2.8 ppbv/mo to 4.3 ppbv/mo in the 2-4 km and 4-6 km regions, respectively. A higher rate of $O_3$ increase was observed over the last four deployments of TOPSE in the higher latitudes with 4.7 ppbv/mo and 6.7 ppbv/mo in the 2-4 km and 4-6 km regions, respectively.

Aerosol scattering ratios showed a dramatic increase across the free troposphere in all latitudinal regions investigated during TOPSE. In the 60°-85°N, 4-6 km region, $S_A$ increased over 400% during TOPSE at an average rate of 0.37/mo. The average $O_3$ and aerosol scattering ratio levels were highly positively correlated over the TOPSE experiment with a correlation coefficient of 0.95 and an $O_3/S_A$ slope of 10.6 ppbv. As has been observed in previous field experiments, if the source of additional $O_3$ was predominantly from the stratosphere, the correlation between $O_3$ and aerosol scattering would be negative since at the time of TOPSE the stratospheric air had low aerosol loading compared to normal tropospheric conditions. This was the first indication that the predominant reason for the springtime $O_3$ increase at high latitudes was due to photochemistry and not stratosphere-troposphere exchange.

Based on the above analysis, which examined the $O_3$-$S_A$ correlation, the estimate of stratospherically-derived $O_3$ trend from the PV analysis of three models and from the $^7$Be data, and the in situ trace gas and aerosol trends, we estimate that stratosphere-troposphere exchange contributed less than 20% to the observed $O_3$ trend at high latitudes in the mid troposphere across the spring equinox. The dominant (>80%) source of the free tropospheric $O_3$ trend was found to be due to photochemical $O_3$ production in the troposphere along with long range transport. A detailed manuscript on the topic is currently in press[5].

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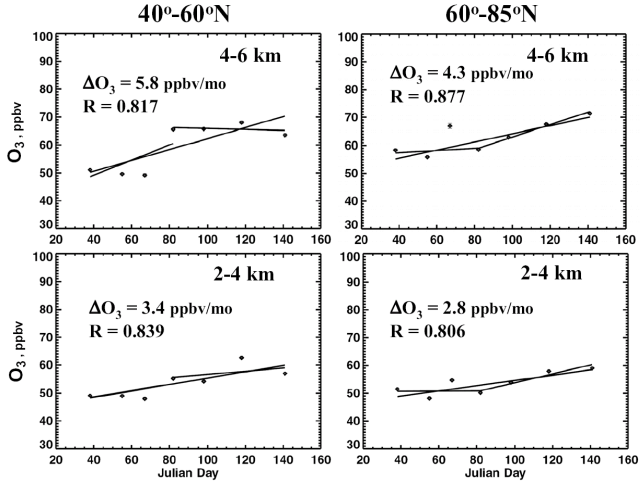


**Figure 1.** Average ozone trends during TOPSE

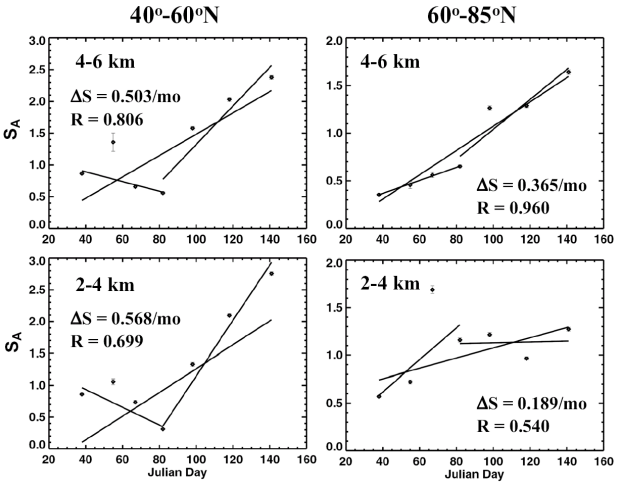


**Figure 2.** Average aerosol trends during TOPSE

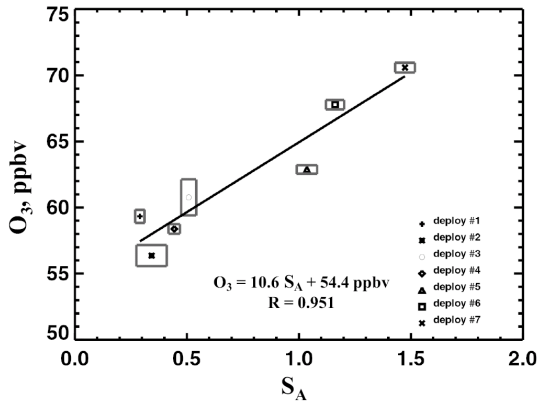


**Figure 3.** Correlation of ozone and aerosol scattering ratios (60-80N,4-6km)

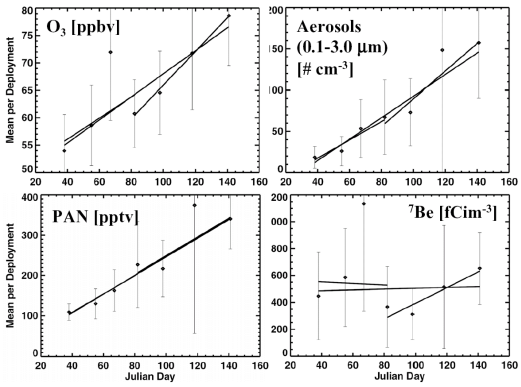


**Figure 4.** In situ measuremnets of $O_3$ [Ridley, NCAR], Aerosols [NCAR]; PAN [Flocke, NCAR], $^7$Be [Dibb, UNH]