11.1 OBSERVATIONS OF TEMPORAL AND SPATIAL VARIABILITY OF OZONE DURING THE TEXAS AIR QUALITY STUDY 2000

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1. INTRODUCTION

During a six-week period beginning in mid-August 2000, the Texas Natural Resource Conservation Commission [now the Texas Commission on Environmental Quality (TCEQ)] coordinated a major field campaign to understand the production and transport of air pollution in Houston and the surrounding areas of Texas. The campaign, called the Texas Air Quality Study 2000 (or TexasAQS 2000), involved numerous research and regulatory organizations. Comprehensive surface-based and airborne measurements of pollutants, their precursors, and the associated meteorology were made during numerous intensive observing periods (IOPs) during the campaign.

The particular goal of our research is to explore the degree to which local meteorological circulations such as thermals or sea breezes may impose time or spatial scales on boundary layer variations of ozone. To do so, we are combining ozone observations from airborne and surface-based lidars, from a network of surface stations, and from ozonesondes with meteorological observations from wind profilers and radiosondes.

In this abstract, we describe the structure of ozone on 28 August 2000. This day was selected for initial analysis because it had moderate, steady winds in the boundary layer during the period of aircraft measurements. This is useful in attempting to relate aircraft-derived wavenumber spectra to surface measurements through Taylor's hypothesis. As it happened, this day was also a day of low overall ozone near the surface, except for the ozone plume that appears to originate near the ship channel in Houston. Significantly larger ozone values above the boundary layer also allowed us to draw conclusions regarding the importance of vertical mixing on surface ozone concentrations.

2. THE DATA

TCEQ maintains an operational network of surface monitoring sites across the state of Texas, including the Houston area. These stations measure a variety of variables, including ozone. The averaging interval for which ozone measurements are available from these sites is 5 min.

During the field campaign, the National Oceanic and Atmospheric Administration (NOAA) operated a DC-3 aircraft with a downward-facing lidar to measure ozone profiles over approximately the lowest three kilometers of the atmosphere. This differential absorption lidar provided 10-s samples of ozone with 90-m range resolution and a detection limit of 4–10 ppb. The sampling airspeed of the DC-3 was 60 m s^{-1}, giving a spatial resolution of 600 m. The characteristics of this instrument are more fully described in Alvarez II et al. (1995).

While ozone soundings were generally made once per day during TexasAQS 2000, no sounding was available on 28 August. Atmospheric temperature and moisture structure was measured, however, using AIR (Atmospheric Instrumentation Research) equipment provided by Pacific Northwest National Laboratory (PNNL). Finally, five wind profiling radars (see White et al., 2002) provided hourly measurements of the horizontal wind profiles across the Houston area during the field program.

3. ANALYSIS

We have used two analytical tools to investigate the structure of ozone variations in the boundary layer. The first, spectral analysis, is intended to determine whether there are any dominant temporal or horizontal scales of ozone variability. The second tool, the variogram, helps to identify layers over which ozone characteristics are relatively uniform and altitudes at which these layers become decoupled.

We calculated power density spectra from both the five-minute surface observation and the airborne lidar data. In the surface data, the primary temporal variability occurs on the diurnal period. However, our interest was directed more toward identifying variations in ozone that might occur as a result of organization by thermals or other boundary-layer-scale circulations. As a first attempt to spectrally analyze the surface data, we divided the daytime into four three-hour periods beginning at 6 a.m. LST. We then subtracted a simple linear trend from the ozone data to minimize the effect of the diurnal cycle in each segment and computed the power spectrum via the Fast Fourier Transform. For the airborne lidar data, we applied an analagous computation to selected flight segments that were oriented more or less along-wind and that did not contain the ozone plume. The along-wind criterion was to facilitate a comparison with surface data through Taylor's hypothesis. The second criterion was to avoid having an ozone plume originating from localized sources dominate the spectrum and obscure organization by boundary layer structure. To reduce effects of large-scale spatial variability for the airborne lidar data, we removed a linear trend at each altitude and over each flight leg prior to computing the spectra.

The variogram provides a quantitative measure of

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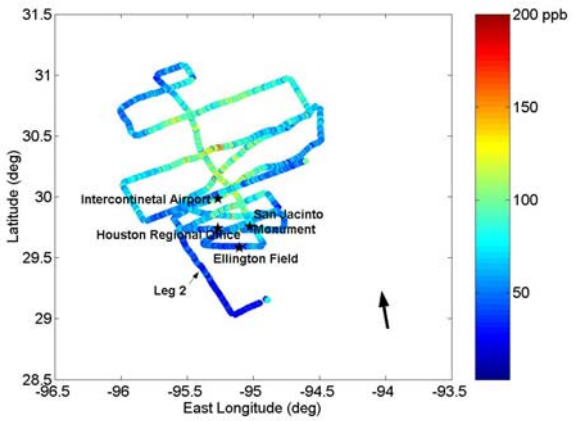

Figure 1. Flight path of NOAA DC-3 colored according to mean lidar-derived mean ozone concentrations between 200 m and 500 m MSL. The bold arrow indicates the general boundary layer wind direction during the flight, which began at approximately 1800 and ended about 2400 UTC.

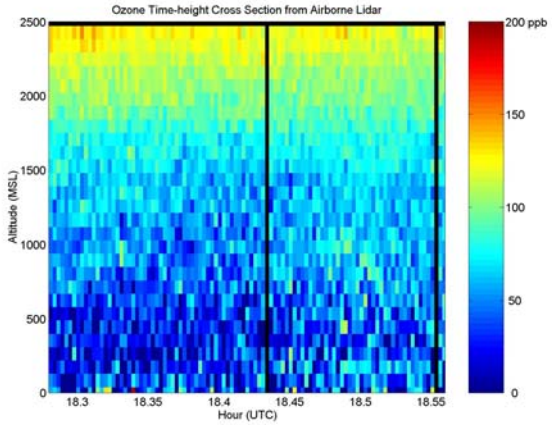

Figure 2. Time-height cross section of lidar-derive ozone concentration for the second leg of the flight shown in Fig. 1. Black areas are missing data. This figure is consistent the very low values of ozone observed by surface station on 28 August.

the degree to which (in our case) a layer of the atmosphere is related to those above and below it. Its definition incorporates the height-dependent structure function and is interpreted much like a correlation coefficient. Unlike the structure function, however, it includes the effect of variations in the mean value as well as fluctuations about the mean. We have used a slight modification of the variogram suggested by Eckman et al. (1992), which we define for a variable *u* as follows:

$$\gamma(z,\Delta z) = 1 - \frac{D(z,\Delta z)}{\overline{u^2(z)} + \overline{u^2(z+\Delta z)}} \qquad (1)$$

where

$$D(z,\Delta z) = \overline{[u(z) - u(z+\Delta z)]^2} \qquad (2)$$

$D(z,\Delta z)$ is the structure function, *u* for our purposes represents ozone, and the overbar is an average. The variogram as defined above has the property that

$$-1 \le \gamma(z,\Delta z) \le 1$$

To create a variogram for a flight leg, we have computed lidar-derived ozone differences over height for each 10-s profile and then averaged over the flight leg. To emphasize the variation with height of the mean field, we have subtracted an overall time-height mean for each leg prior to the computation.

4. RESULTS

4.1 Spatial Distribution of Ozone

Fig.1 shows both the flight path of the DC-3 and the distribution of ozone in the boundary layer as measured by the lidar. In this plot, we have followed a similar procedure to Senff, et al. (2002) in averaging ozone values between 200 m and 500 m MSL. The figure clearly shows a plume of ozone in the boundary layer, surrounded by ozone values that were quite low—often less than 50 ppb. During the period of the flight, the wind profiler array indicated winds from the south-southeast at about 5 m s^{-1}, which is consistent with the orientation of the observed plume. An inspection of the figure shows that for the most part, flight legs oriented along the mean wind were outside the ozone plume.

Fig. 2 shows a time-height cross section of ozone from the second flight leg. This flight leg occurred shortly after local noon on 28 August, and shows several interesting characteristics of the day. As we noted above, the ozone values very near the surface are infrequently above 50 ppb. However, there is a general increase in ozone with height. At altitudes

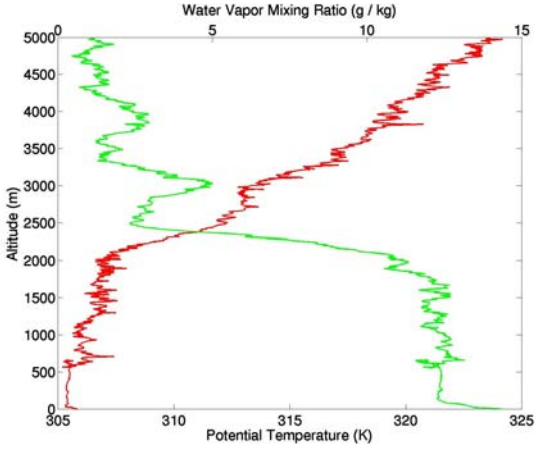

Figure 3. Sounding showing temperature and humidity structure over downtown Houston at approximately 1400 LST.

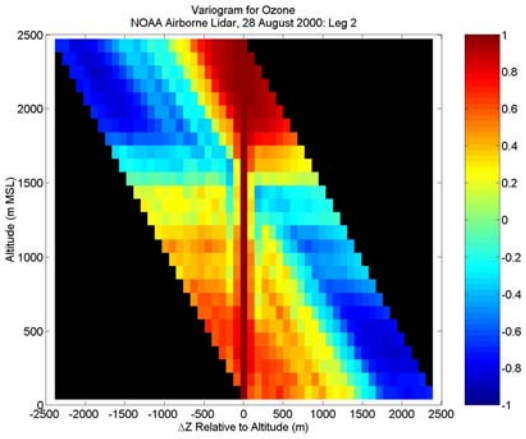

Figure 4. Variogram for the ozone data of Fig. 2. The interpretation of this figure is given in the text.

approaching 2.5 km, the ozone values are regularly 125 ppb.

It is interesting to compare the data in Fig. 2 with the thermodynamic structure as revealed in the radiosonde launch from downtown Houston shortly before 3 p.m. LST (Fig. 3). The potential temperature and water vapor both show sharp vertical changes between 2000 m and 2500 m. This is conventionally the altitude that would be identified as the top of the convective boundary layer and is approximately where the largest values of ozone occurred.

Fig. 4 shows the variogram computed for the data of Fig. 2. The ordinate in this figure is altitude, while the abscissa indicates height differences relative to that altitude. For example, at the lowest altitude value, comparisons can only be made with the same or higher altitudes; therefore, values exist only for non-negative Δz. Similarly, at the highest altitude, 2500 m, variogram values exist only for non-positive Δz. At all altitudes, the variogram value is unity for $\Delta z = 0$, since a time series is perfectly correlated with itself. The predominance of red and orange colors in the lower left portion of Fig. 4 indicates that the lowest 1000 m of the boundary layer are very similar with respect to the ozone values. The blue colors in the lower right region of the variogram reflect the substantially different ozone values at higher altitudes. The figure is consistent for the higher altitudes, where the red colors in the upper right part of the variogram indicate great similarity in ozone values from 2500 m down to 1800 m or so. A revealing feature in the variogram occurs at 1500 m, where the generally green colors (near-zero variogram values) for both positive and negative $\Delta z's$ indicate that the ozone at that level is not very much related to the levels either above or below it. This would suggest that this is the best estimate for the altitude at which the decoupling occurs between the ozone values aloft and those lower in the boundary layer. This is notably lower than the altitude of 2200 m or so where one would place the top of the boundary layer from thermodynamic considerations.

4.2 Scales of Variability

Temporal and spatial scales of variability (apart from the diurnal cycle) are not as apparent in the data, at least in this preliminary analysis. Fig. 5 shows a power density spectrum from surface ozone data collected at the TCEQ Houston Regional Office from 12–3 p.m. on 28 August. There is a significant peak in the first harmonic (suggesting that perhaps an alternate filter to linear detrending should be investigated), but there are also a peaks at frequencies of 0.3 and 0.5 s^{-1} (corresponding to length scales of about 15 and 10 km). This is plausible although a bit large for boundary-layer-scale circulations in a 2-km deep boundary layer. However, if we look at spectra as a function of height from the airborne lidar (Fig. 6), a similar scale is not apparent. Here the spectra from the 90-m range gates have been averaged over three vertical range gates. It is apparent that the variance of the ozone fluctuations diminishes with height, but there is no apparent characteristic peak in the spectra that would indicate a preferred scale for ozone fluctuations. This casts some doubt on the significance of the peaks in the surface spectra as analyzed.

5. SUMMARY AND CONCLUSIONS

We have presented a preliminary analysis of the structure of ozone in the atmospheric boundary layer as measured from a variety of platforms on 28 August during TexasAQS 2000. This particular day was characterized by generally low ozone levels at the surface during an afternoon when winds were moderate and relatively steady from the south-southeast. Nevertheless, data from an airborne lidar operated by NOAA clearly showed a plume of ozone being transported by the mean wind, and larger values of ozone (not shown) were observed by surface stations in the path of this plume.

Outside the plume of ozone in the boundary layer, ozone levels remained quite low, despite ozone concentrations that were routinely as large as 125 ppb

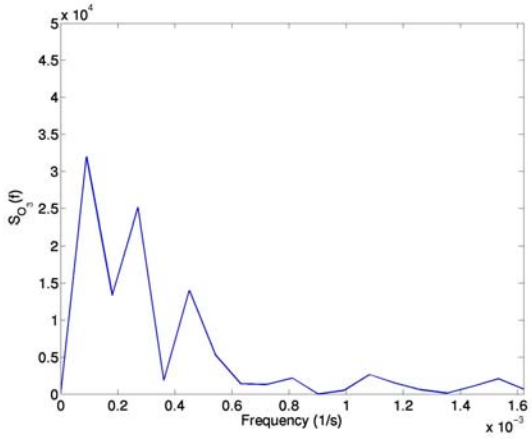

Figure 5. Power density spectrum of ozone for the period 1200–1500 LST on 28 August. The data were measured at the Houston Regional Office, indicated in Fig. 1.

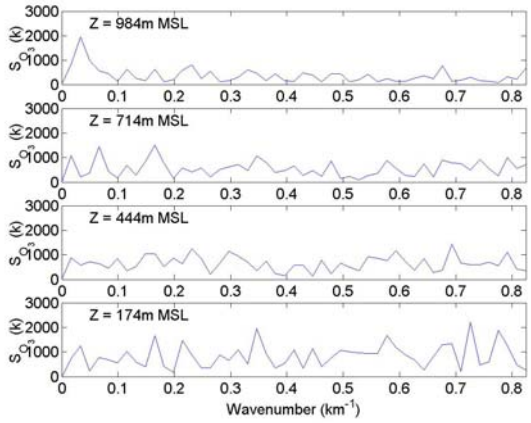

Figure 6. Power density spectra of ozone constructed from the data ozone data shown in Fig. 2. Under the conditions of these measurements, there was no dominant wavenumber that would indicate a characteristic scale of ozone fluctuation in the boundary layer.

above 2000 m. The variogram analysis indicated that the lower, relatively clean layer and the upper layer with higher ozone values were decoupled at an altitude of about 1500 m. This decoupling between the air aloft and the surface suggests that the larger surface ozone readings that occurred in Houston on this day were due to a boundary layer plume rather than to mixing down of elevated values from higher in the atmosphere.

Finally, variations in ozone with space and time, at least outside the ozone plume, did not suggest that ozone fluctuations depended significantly on the organizing effects of local boundary layer circulations. This may be a limitation of the relatively low space and time resolution of the surface stations and lidar compared with conventional boundary layer instrumentation. This may also be a reflection of the decoupling of the lower boundary layer and the layer with higher ozone. These conclusions, of course, are based on a very limited subset of the TexasAQS 2000 dataset, and the generality of our results will be determined as we expand our analysis to other days and meteorological conditions of the field campaign.

6. ACKNOWLEDGEMENTS

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