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Nocturnal ozone enhancement in the lower troposphere observed by lidar☆

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ABSTRACT

An ozone enhancement in the nocturnal residual layer was observed by the Huntsville ozone lidar from the late evening to midnight on 4 October 2008. The well-correlated ozone, aerosol, water vapor, and wind structures suggest a low-level jet is responsible for this ozone enhancement. HYSPLIT backward trajectories support this conclusion with southerly transport suggesting Birmingham, AL as the source. Correspondingly, the higher increasing rate of surface ozone observed in the morning of 5 October can be explained by the entrainment into the mixed layer of higher ozone aloft on this day as compared with 4 October. This case study demonstrates the importance of continuous high-resolution lidar profiling for capturing short-duration ozone variations in the lower troposphere.

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1. Introduction

Ozone variations within the planetary boundary layer (PBL) are complex and often rapid (Ancellet and Ravetta, 2005; Thompson et al., 2007) because of multiple factors such as surface emission and deposition, interaction with the free-troposphere (FT), photochemistry, and horizontal transport. Relative to the well-characterized diurnal variations of the surface ozone (Zhang and Rao, 1999), aloft ozone is more complicated and is sometimes observed with much higher concentrations than seen in surface measurements (Berkowitz et al., 2004). During daytime, ozone within the convective boundary layer (CBL) can be homogeneous due to solar-driven convective mixing; however, highly inhomogeneous structures are often observed (Ancellet and Ravetta, 2005) and require high-resolution measurement of ozone profiles to characterize. After the stable nocturnal boundary layer (NBL) starts to develop in the evening, ozone generally has a positive gradient from the surface toward the top of the NBL (Geyer and Stutz, 2004; Stutz et al., 2004) because of surface NO_x titration and insufficient downward mixing due to the nocturnal capping. Since the removal mechanism typically dominates near the surface during nighttime, a reasonable explanation for elevated ozone in the nocturnal PBL is the regional or small-scale transport associated with mechanisms such as the low-level wind jet (Banta et al., 1998; Corsmeier et al., 1997), topographic forcing (Zhu et al., 2006), or sea–land interaction (Sun et al., 1998). Among those mechanisms of particular interest are low-level jets (LLJ) which have been extensively discussed in conjunction with the Southern Oxidants Study (SOS) (Banta et al., 1998; Baumann et al., 2000; Meagher et al., 1998). For this case, the pollutant plume can form during the day, when the meteorological conditions are favorable to pollutant accumulation, and be transported downwind during the nighttime affecting the background levels on the second day.

Ozone is the most important primary pollutant for Huntsville. After the Environmental Protection Agency (EPA) lowered the ground-level ozone standard from 0.08 ppm to 0.075 ppm in March 2008, Huntsville failed to meet the new air quality standard having a three-year average of the fourth-highest 8-h average of 0.078 ppm (Natural Resources and Environmental Management Division, 2009). Previous model studies demonstrate that air stagnation regime in the Southeast appears to have more influence on the episode days than synoptic-scale transport (Hidy, 2000). However, Huntsville is surrounded by several larger cities: Birmingham, Memphis, Nashville, and Atlanta, which have

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numerous industrial emission sources that can potentially affect Huntsville's air quality. The impact of pollution transport on Huntsville has not been previously studied. The objective of this paper is to report on an elevated nocturnal ozone event measured over Huntsville and discuss its formation mechanism and impact on the surface ozone.

2. Instruments

2.1. Ozone DIAL

The tropospheric ozone Differential Absorption Lidar (DIAL), developed jointly by the University of Alabama in Huntsville and NASA/Goddard Space Flight Center, retrieves ozone profiles from 0.5 to ~8-km altitude at 10-min intervals with a vertical resolution of better than 750 m. This instrument is described elsewhere (Kuang et al., 2010), so only a short description will be included in this paper. The transmitter consists of two 20-Hz Nd: YAG-pumped dye lasers generating ~4 mJ pulse^{-1} at 285 and 291 nm. The receiving system has two telescopes, a 40-cm Newtonian telescope, which covers the altitudes from 3 to 8 km with a precision better than 20%, and a 10-cm Cassegrain telescope for altitudes between 0.5 and 5 km with a precision better than 10%. The ozone retrievals of the two altitude-channels join between 3.3 and 4.4 km, typically. Due to the lack of a third wavelength, an iterative aerosol correction is applied to reduce the retrieval error arising from differential aerosol backscatter in the lower troposphere. This procedure iteratively substitutes the ozone profile from the DIAL algorithm back to the off-line (291-nm) signal to calculate the aerosol-extinction profile by assuming an appropriate lidar ratio. Then, the correction terms due to differential aerosol backscatter and extinction can be derived using Browell's approximation (Browell et al., 1985) by assuming an Ångström exponent.

2.2. Mobile Integrated Profiling System

A Mobile Integrated Profiling System (MIPS), which consists of a 915-MHz wind profiler (Radian LAP-3000), a 12-channel microwave profiling radiometer (MPR; Radiometrics TP/WVP-3000), a lidar ceilometer (Vaisala CT-25k), and various other instrumentation (Karan and Knupp, 2006; Knupp et al., 2006) collected the local meteorological data used in this paper. The coaxial ceilometer measures the atmospheric backscatter from the ground to 7.6 km with a 30-m vertical range resolution and 15-s temporal resolution and covers the bottom 500 m of the PBL, a region that is not currently covered by the ozone DIAL. The 915-MHz profiler provides horizontal winds up to a 4-km altitude with a 1-m s^{-1} accuracy, 105-m vertical range resolution, and 60-s temporal resolution. The MPR measures atmospheric microwave emissions at twelve discrete frequency bands to derive the water vapor density with an error less than ~1.1 g m^{-3} and temperature with an error of less than ~2.0 K below 7 km (Güldner and Spänkuch, 2001). The percentage uncertainty of the derived relative humidity (RH) is mostly determined by the actual water vapor and is also related to the uncertainties of water vapor and temperature in a complicated manner. An estimated uncertainty for a derived 50% RH when ambient temperature equal to 10 °C (saturated water vapor density is 9.4 g m^{-3}) is about 25%. The vertical resolution of the MPR measurement for either water vapor or temperature retrieval decreases from ~50 m at surface to ~1 km between 700 m and 4 km.

2.3. Ozonesondes

UAHuntsville has been routinely launching ozonesondes (EN-SCI model 2Z with unbuffered 2% cathode solution) weekly since 1999 (Newchurch et al., 2003). The ozonesondes measure ozone profiles with a ~100-m vertical resolution from the surface to 35-km altitude at an accuracy better than 10% (Komhyr et al., 1995; Oltmans et al., 1996) and a co-located radiosonde (Vaisala RS-80-15) measures temperature, pressure, and RH.

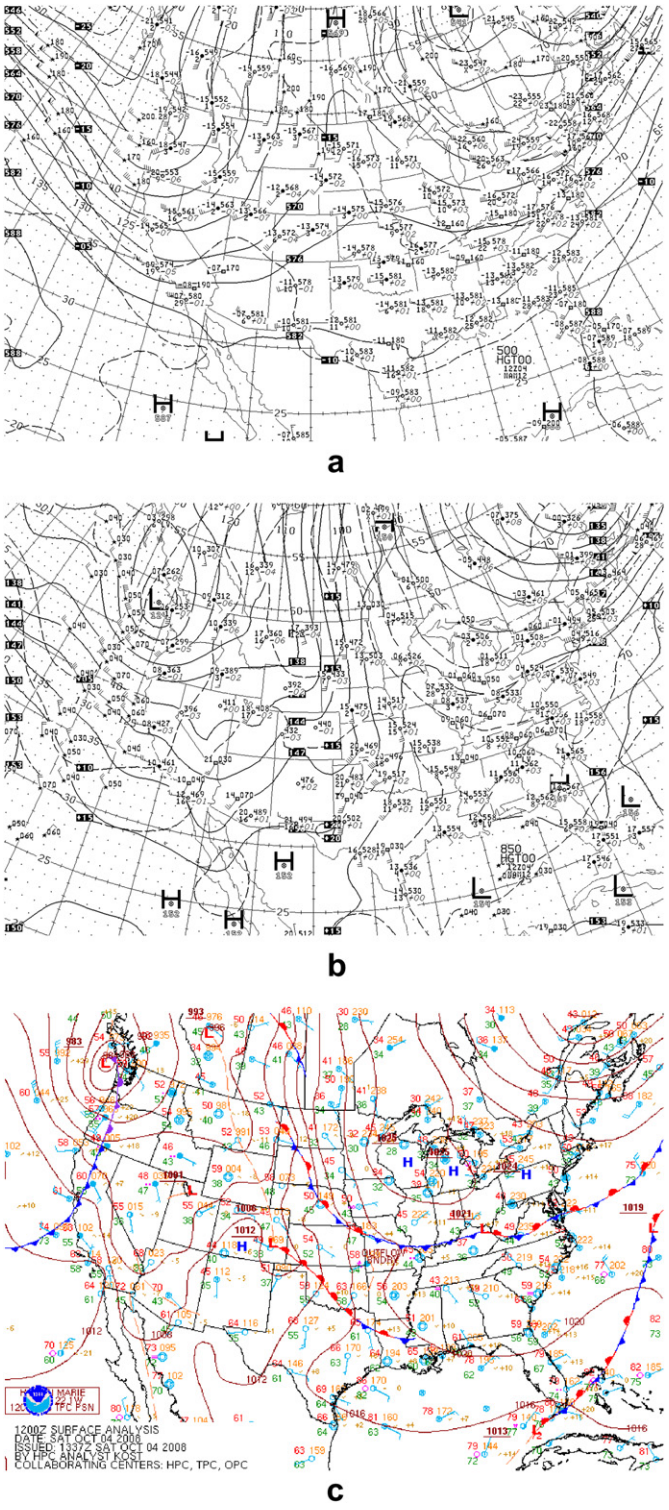

Fig. 1. Weather charts for 7:00 local time (12:00 UTC) 4 October, 2008 (source: National Climatic Data Center of NOAA, http://nomads.ncdc.noaa.gov/ncep/NCEP). (a) 500-hpa height contours. (b) 850-hpa height contours. (c) Surface weather map.

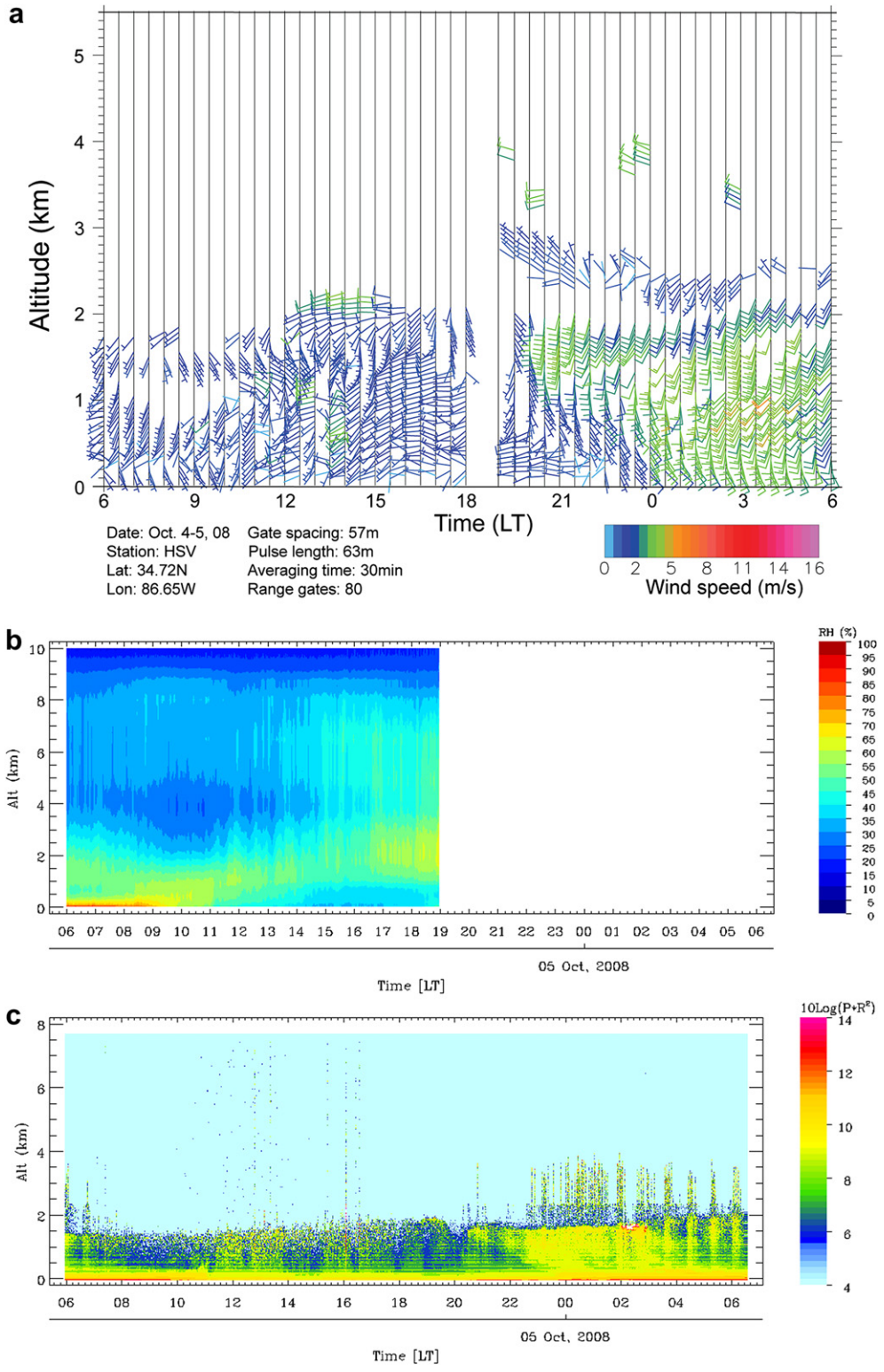

Fig. 2. The UAHuntsville MIPS measurements on 4–5 October 2008. (a) wind structure measured by the 915-MHz wind profiler; (b) RH retrieval by MPR (RH data after 18:59 is not available due to instrument issue); (c) the ceilometer backscatter.

3. Meteorological conditions

It is well known that meteorological conditions play an important role in the formation, transport, and deposition of pollution plumes (Solomon et al., 2000). The synoptic condition responsible for the event described here was a high-pressure system moving slowly eastward that stalled over the United States during this period. The 500 and 850-hpa height-contour charts in Fig. 1 illustrates a ridge tilted westward with height and protruding over the central United States on 4 October 2008.

The upper atmosphere over Alabama displays a weak westerly to southwesterly wind flow. Associated with the upper-air system was a surface high centered over Lake Michigan and elongated to the southeastern United States. The weak pressure gradients in the southeastern U.S. resulted in light surface winds (<1.5 m s^{-1}) over a broad area. In Fig. 2(a), the wind profiler measurement reveals the details of the wind variations up to 4 km in Huntsville on 4 October. The winds in the boundary layer during the daytime were weak, generally less than 2 m s^{-1} with variable direction from westerly to southerly. However, a southerly nocturnal low-level jet (LLJ) developed at 20:00 local time (LT) with the jet nose starting from ~1.5 km and extending downward to ~0.5 km. The jet turned clockwise with height and reached a maximum speed of ~10 m s^{-1} between 0.5 and 1 km until it diminished when the mixing layer of the second day formed. Such northerly or southerly jets, especially during the nighttime, have been observed and studied extensively in the central United States (Banta et al., 2002). Two major mechanisms contributing to the formations of LLJ are synoptic-scale forcing, associated with the mesoscale circulation features, and boundary layer forcing when the airflow near the top of the nocturnal boundary decouples from the surface friction especially under a stable atmosphere condition (Walters et al., 2008). From 20:00 to 06:00 in Fig. 2(a), the distinct clockwise rotation in the direction of the jets (turning from southerly to southwesterly with time) and the contrary turning direction of the winds in the boundary layer and FT (winds between 2 and 3 km turning from northwesterly to southwesterly with time) is evidence that the boundary layer forcing is primarily responsible for this LLJ (Whiteman et al., 1997).

4. Upper-air ozone

Fig. 3 shows the ozone DIAL retrievals on 4 October 2008, which agree very well with the simultaneous ozonesonde measurement at 13:01 LT shown in Fig. 4. The most interesting FT ozone feature is a 2-km thick, low-ozone layer (>30 ppbv) at ~3.5 km during the daytime. This layer, which was captured by both the DIAL and ozonesonde, gradually increased to ~55 ppbv in the afternoon as shown by the DIAL curtain plot. This increase is likely due to physical mixing processes. These observations demonstrate that the tropospheric ozone is highly variable in both space and time and a high-resolution lidar is needed for capturing these rapid ozone variations. Two thermal inversions at 1.5 and 4.0 km appear on the temperature profile in Fig. 4 reflecting stable air layers. The first strong temperature inversion at 1.5 km represents the daytime CBL top which blocked convection between the CBL and FT. The second weak temperature inversion was caused by large-scale subsidence associated with a subtropical high-pressure air mass. This was confirmed by the radio-sonding profiles in Nashville and Birmingham at 7:00 LT (not shown). Daytime ozone mixing ratios within the boundary layer are typically about 64 ppbv, somewhat higher than the yearly autumn average in Huntsville of about 55 ppbv (Newchurch et al., 2003), and are related to the meteorological conditions. The weak horizontal-pressure gradients and consequent weak winds during the daytime suppressed the local dissipation of the pollutants. This air stagnation condition began on 3 October when a high-pressure system was moving over the continental U.S. The large ozone gradients just above the boundary layer top before 18:00 suggest a stable FT, and limited interaction

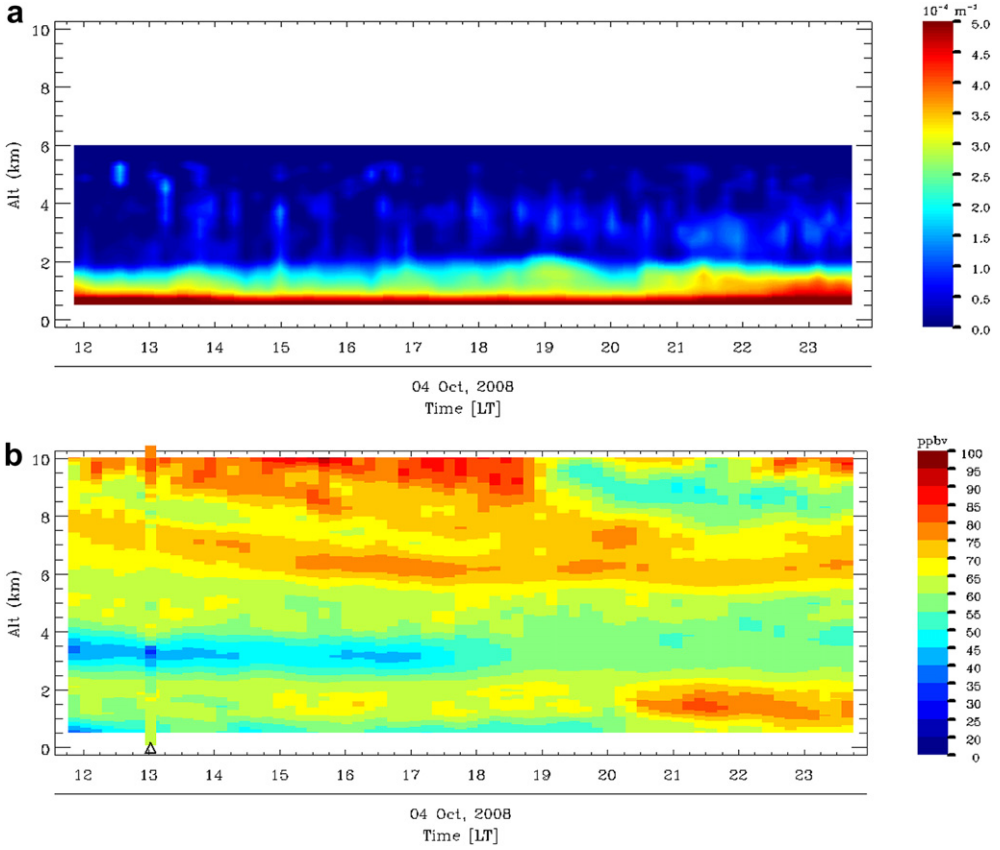

Fig. 3. Ozone DIAL measurements on 4 October 2008. (a) Calculated aerosol-extinction coefficient at 291 nm from the ozone DIAL. (b) Ozone DIAL retrieval with 10-min temporal integration and 750-m vertical resolution. The coincident ozonesonde measurement is marked with a triangle at the *x*-axis.

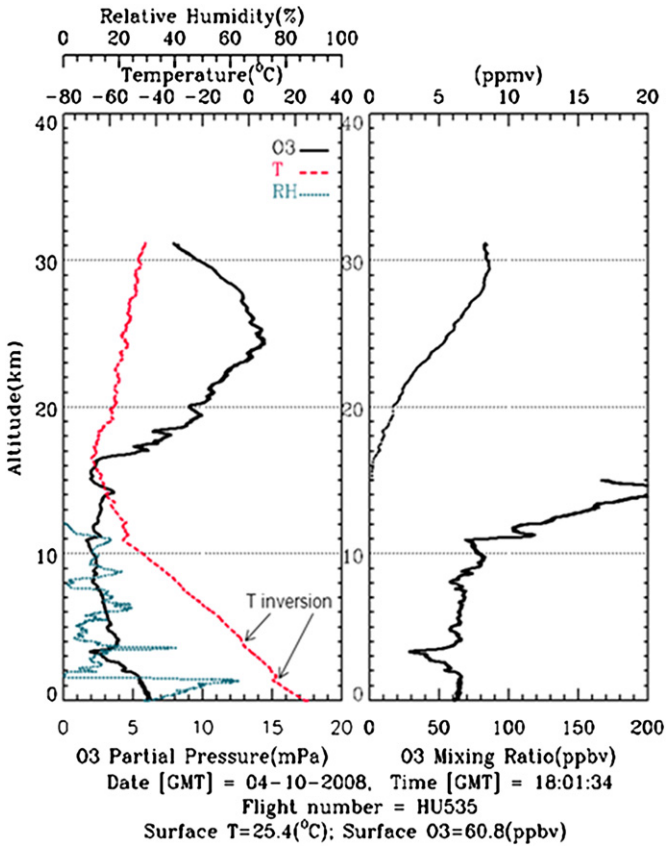

Fig. 4. Ozone, temperature (T), and RH profiles measured by ozonesonde at 13:01 LT 4 October at the same location as the DIAL and MIPS.

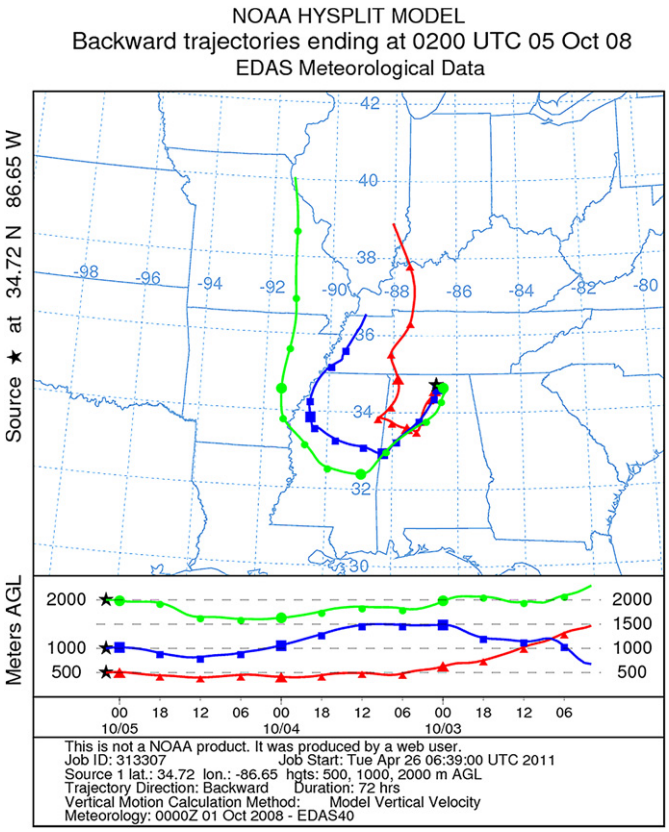

Fig. 5. 72-h HYSPLIT-model backward trajectories in Huntsville with a 6-h time interval ending at 21:00 LT Oct. 4, 2008 for the layers at 500, 1000, and 2000 m a.s.l.

between the boundary layer and FT although some vertical mixing within the boundary layer during the afternoon exists. A similar structure and mostly positive correlations between ozone and MPR RH retrievals (Fig. 2(b)) suggest tropospheric sources for those ozone laminae.

The aerosol retrieval at 291 nm (Fig. 3(a)) and ceilometer data (Fig. 2(c)) both indicate turbid air conditions below 2 km during the daytime. A distinct ozone enhancement occurred in the residual layer after sunset accompanied by a similar enhancement in the aerosol. The maximum nighttime ozone mixing ratio is 95 ppbv for altitudes below 2 km with strong aerosol activity in the same time–height region. This positive correlation between ozone and aerosol in the troposphere, which has also been observed in other measurements (Browell et al., 2003), suggests the same source is responsible for the enhanced ozone and aerosols. The temporal variation of the ozone and aerosol enhancement agrees well with the wind profiler measurement in Fig. 2, which clearly shows the pollutants were carried by the southerly LLJ.

Fig. 5 shows the 72-h NOAA's HYSPLIT-model backward trajectories (Draxler and Rolph, 2003) in Huntsville ending at 21:00 LT 5 October 2008 for three layers at 500, 1000, and 2000 m a.s.l. We have performed a time sensitivity test and a horizontal sensitivity test to increase the reliability of this analysis. These trajectories are not time sensitive within 4 h before this event and most of the 1-degree separated locations around Huntsville display similar trajectory characteristics. Therefore, the trajectory result is believed to be robust to some degree although we understand that the locations of the trajectory calculation still can be rather uncertain especially in the PBL due to complex turbulent mixing processes (Stohl, 1998). The recirculation backward trajectory pattern at 1000 m suggests two possible sources which may have substantial anthropogenic emission: the Birmingham metropolitan area 14 h previously and Memphis 60 h prior. Considering a daily-surface maximum ozone of 38 ppbv measured in Memphis (station ID: 47-157-0021, surface data come from EPA's Air Quality System (AQS)) on Oct. 2 and a 76-ppbv daily maximum measured at the closest Birmingham station (ID: 01-073-5003) to Huntsville on Oct. 4, which is also on the path of the backward trajectory, it appears that Birmingham is more likely responsible for this ozone enhancement within the residual layer. The emissions of photochemical precursors from upwind locations can impact the downwind region during a period of light winds (Baumann et al., 2000; Trainer et al., 1995). The nighttime acceleration flow above the stable NBL can carry the pollution plume away from the daytime buildup region and redistribute it over the downwind region (Banta et al., 1998; McNider et al., 1998). However, the LLJ is not indicated on the Birmingham 0Z 5 October radiosonde data (not shown), so it might play an important role only in the Huntsville area. Also, the calculated wind speed of $\sim$2.8 m s^{-1} from the estimated transport distance ($\sim$20 km) at the 1000-m altitude between 00 and 02 UTC in Fig. 5 is slower than that of the LLJ described foregoing and possibly suggests a discrepancy between the HYSPLIT database and local measurements. Despite the fact that the ozone DIAL cannot uniquely identify the pollution source even with the help of local meteorological observations, it does identify features that can be explored with other tools such as HYSPLIT or photochemical models.

5. Discussion on the surface ozone

Because stagnant-air conditions are favorable to pollutant accumulation, moderate ozone-polluted air quality was observed

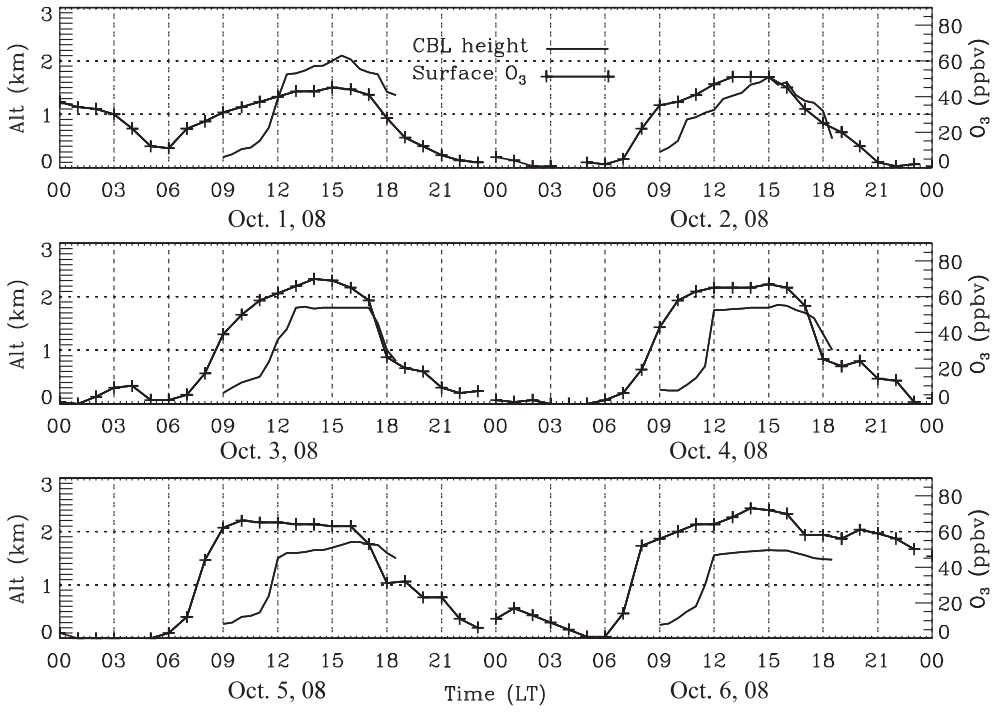

Fig. 6. Huntsville hourly surface-ozone variations and the CBL height derived from the UAHuntsville 915 MHz profiler measurement.

by EPA surface stations over a wide area in the southeast beginning 3 October and extending into the central eastern part of the U.S. by 6 October. Fig. 6 shows the hourly surface-ozone mixing ratio measured at Huntsville airport road station and the CBL height derived from the wind profiler (White, 1993) between 1 and 6 October, 2008. The surface-ozone maxima during the afternoon gradually increased from 45 ppbv on 1 October–70 ppbv on 3 October due to the moving high-pressure system. The surface ozone on the afternoon of 4 October is consistent with the lidar and ozonesonde observation in the CBL under clear sky conditions. The evening surface ozone on 4 October is only slightly higher than the previous days. However, the increasing rate of surface ozone between 6:00 and 9:00 LT October 5, 20.3 ppbv h^{-1}, is 49% higher than that of 4 October, 13.7 ppbv h^{-1}, with a similar CBL developing process. This suggests the aloft ozone in the morning on 5 October is higher than that of 4 October and has already affected the surface through the downward mixing process. However, on October 5, the surface ozone maximizes at 10:00 instead of in the afternoon as usual and slightly decreases from 10:00 to 04:00 possibly because of the partly cloudy conditions that cause insufficient mixing and weak photochemical production. Compared with 5 October the 10-ppbv higher daily-maximum surface ozone observed on 6 October might be due to more complete mixing and the active photochemistry associated with clear sky conditions.

6. Conclusion

The PBL ozone was measured at ~64 ppbv by the Huntsville ozone lidar during daylight hours on 4 October 2008, which was about 20% higher than the seasonal average due to a high-pressure system above the southeast U.S. The PBL ozone was further enhanced to ~95 ppbv beginning from late evening to midnight; this increase was associated with a southerly LLJ observed by the co-located wind profiler. The highly-correlated variations and vertical structure of the ozone, water vapor, and aerosols imply the same tropospheric source for the pollution plume, which was most likely the Birmingham area, as suggested by the HYSPLIT backward trajectory calculation. The high-pressure controlling meteorological conditions explains the observed stable atmosphere and resulted in distinct ozone laminae from the PBL to the FT, brought in by the air flows from different directions.

Air stagnation led to high daily-maximum surface ozone widely observed throughout the southeast between 3 and 6 October 2008. The higher rate of increase of surface ozone in Huntsville in the early morning of Oct. 5 could be explained by the higher aloft ozone caused by the transport in the night of Oct. 4. However, the observed daily-maximum surface ozone on Oct. 5 was not higher than the previous day possibly due to the insufficient mixing and weak photochemical production under cloudy sky conditions relative to clear sky conditions. Although ozone in the nocturnal residual layer largely explains the ozone variability in the second day afternoon CBL (Morris et al., 2010; Neu et al., 1994), the maximum values were not always observed at the surface and could occur aloft within the boundary layer depending on both photochemical and physical processes. These observations reinforce the need for ground based high-resolution ozone profiling of the lower troposphere.

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