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The variability of free tropospheric ozone over Beltsville, Maryland (39N, 77W) in the summers 2004–2007

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ABSTRACT

Ozone profiles are often used to investigate day-to-day and year-to-year variability in origins of free tropospheric ozone. With this in mind, more than 50 ozonesonde launches were conducted in Beltsville, MD, during the summers of 2004 through 2007. Budgets of free tropospheric ozone were calculated for each ozone profile in the four summers using a laminar identification (LID) method and unusual episodes were analyzed with respect to meteorological variables. The laminar method showed that stratosphere-to-troposphere transport (ST) accounted for greater than 50% of the free tropospheric ozone column on 17% of days sampled, a more pronounced influence than regional convective and lightning (RCL) sources. The ST origins were confirmed with trajectories, and tracers (water vapor and potential vorticity). The amount of free tropospheric ozone from ST and RCL sources varied from year-to-year (up to 13%) and can be explained by differences in mean meteorological patterns. On average, almost 30% of the free tropospheric column was attributed to ST influence, about twice as much as RCL, although the LID method may not capture weeks-old lightning influences as in a chemical model. The prevalence of ST ozone in summertime Beltsville soundings was similar to six sounding sites in the IONS-04 campaign [Thompson, A.M., et al., 2007b. Intercontinental Transport Experiment Ozonesonde Network Study (IONS, 2004): 1. Summertime upper tropospheric/lower stratosphere ozone over northeastern North America. J. Geophys. Res. 112, D12S12; Thompson, A.M., et al., 2007c. Intercontinental Transport Experiment Ozonesonde Network Study (IONS, 2004): 2. Tropospheric ozone budgets and variability over northeastern North America. J. Geophys. Res. 112, D12S13.] and to statistics from a 30 year climatology of European soundings [Collette, A., Ancellet, G., 2005. Impact of vertical transport processes on the tropospheric ozone layering above Europe. Part II: Climatological analysis of the past 30 years. Atmos. Environ. 39, 5423–5435]. The Beltsville record also demonstrated the value of soundings for air quality forecasting in an urban area. The 22 nighttime soundings collected over Beltsville in 2004–2007 can be divided into distinct polluted and unpolluted subsets, the former 20 ppbv higher in residual layer ozone (1 km) than the latter. These distinctions propagated to daytime differences of 10 ppbv at the surface in the Washington, DC, area, with the high-ozone residual layers leading to non-attainment of the National Ambient Air Quality Standard for ozone. More frequent ozone observations aloft appear essential for better understanding ozone variability and for enabling air quality modelers to achieve more accurate ozone forecasts.

1. Introduction

The U.S. Environmental Protection Agency (USEPA) classifies ozone as one of six criterion pollutants. It is important to track ozone and to understand the causes of its variability because in order to protect against its harmful effects on human and agricultural health, ozone sources must be correctly characterized so that accurate air quality forecasts can be issued and proper regulation can be enacted. Monitoring of ozone at the surface is routine over wide regions within the U.S. For example, in the mid-Atlantic region, 19 stations report concentrations of ozone in the Washington, DC, area, eight in the Baltimore, Maryland region, and 19 in metropolitan Philadelphia. The observations from the stations that report to EPA are archived in the AIRNOW system: http://www.airnowtech.gov/. Surface measurements and air quality models are used to forecast chemical weather and to evaluate potential control scenarios. However, surface observations alone are not sufficient to evaluate air quality conditions because the chemical composition of the surface layer depends largely on mixing from

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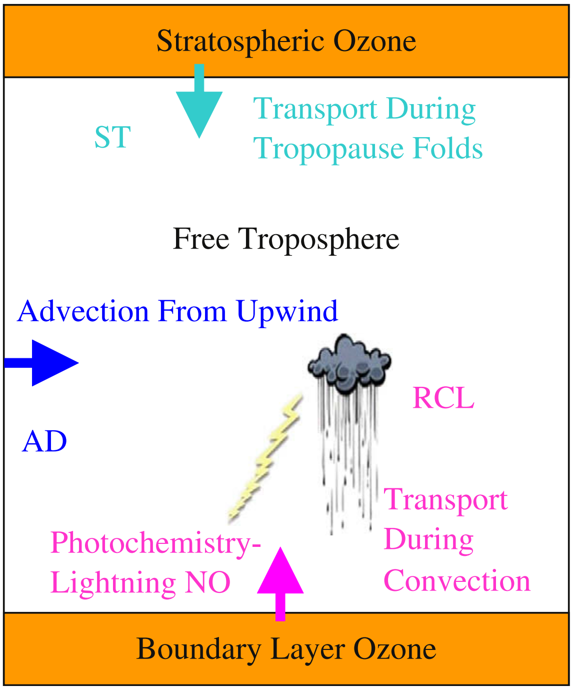

Fig. 1. Schematic of free tropospheric ozone sources resolved in LID method (Thompson et al., 2007b, 2008).

above (Zhang and Rao, 1999). Air quality forecast models can incorrectly estimate surface ozone due to poor ozone estimations aloft and inaccurate vertical mixing schemes (Ryan et al., 2005). New integrated observing strategies call for a combination of satellite data and profiles from aircraft, ground-based remote sensing devices and balloon-borne sondes (Thompson, 2006). Tropospheric ozone profiles obtained from ozonesonde instruments lead to a better understanding of ozone variability above the mixed layer and may enable air quality forecasters to achieve more accurate ozone forecasts.

In the case of ozone, balloon-borne ozonesonde with electrochemical concentration cell (ECC) devices are a popular method for measuring profiles. The instruments are sufficiently accurate and precise (∼5–10% on both counts; Smit et al., 2007; Thompson et al., 2007a; Deshler et al., 2008) and easy to deploy. Also, near-real time data can support field activities, forecasting, and assimilation for decision-making. Summertime ozonesonde launches at Beltsville, Maryland (39N, 77W), Houston, Texas (30N, 95W), and Narragansett, Rhode Island (41.5N, 71W) were part of the IONS-04 (intercontinental transport experiment (INTEX) Ozonesonde Network Study; Singh et al., 2006) and IONS-06 activities that operated, respectively, in July–August 2004 (Thompson et al., 2007b,c) and August 2006 (Thompson et al., 2008), in support of multi-platform pollution-oriented aircraft and ground campaigns. IONS sondes allow us to obtain profiles to the stratosphere (5–10 hPa normal balloon burst), so stratospheric influences can be determined. Thompson et al. (2007b) reported that, on average, 25% of free tropospheric ozone over northeastern North American sites (Virginia through Nova Scotia) during IONS-04 was of stratospheric origins. IONS observations also pinpointed areas where air quality models and satellite instruments may need improvement (Chai et al., 2007; Pierce et al., 2007; Tarasick et al., 2007; Yu et al., 2007). When stratospheric ozone influences the composition of the upper and middle free troposphere (Vukovich, 1994), tropospheric ozone may be under-predicted in air quality models because the model's upper boundary or stratosphere-to-troposphere flux is not accurate (Tarasick et al., 2007; Tong and Mauzerall, 2006). Surprisingly, only two ozone sounding sites in the U.S. have multi-decadal records, Boulder, Colorado (40N, 105W) and Wallops Island, Virginia (38N, 76W) (WMO, 1999). In addition to these stations, ozonesonde operations began in 2004 over Beltsville in the high-ozone months, May– September. The suburban location of Beltsville makes it a suitable site for monitoring ozone variability aloft, for measuring surface ozone, and for examining free tropospheric ozone sources and their influence on the surface.

A high degree of day-to-day variability in free troposphere ozone has emerged from IONS analyses. In July 2004, for example, Houston, Texas, was affected by Alaskan and Canadian wildfires (Morris et al., 2006), stratosphere-to-troposphere injection (ST, Thompson et al., 2007c) and lightning (Cooper et al., 2006). For 2006 analyses to date, similar mixtures of sources were identified over Houston (Morris et al., submitted for publication). Using FLEX-PART trajectory mapping (Stohl et al., 2005) with lightning flash data, Cooper et al. (2007) argue that persistent high -ozone in the upper troposphere over Houston is lightning-related. In Thompson et al. (2008), the laminar identification (LID) method and meteorological analyses indicate that lightning accounts for much of upper tropospheric ozone in the first half of August–September 2006 over Houston; in the second half, stratospheric influences became more dominant. The free troposphere over Beltsville and Narragansett in summer 2004 also displayed a complex mixture of stratospheric, lightning and pollution influences (Thompson et al., 2007b,c). When selected ozone profiles for Narragansett, the Gulf of Maine and maritime Canada region were examined, the interpretation of ozone structure from LID was consistent with meteorological observations. In this study, 4 years of Beltsville summer soundings are used to quantify contributions to free tropospheric ozone using the LID method. Stable laminae, classified in relation to potential temperature gradients as described in Teitelbaum et al. (1994) and Pierce and Grant (1998), are refined with tracers and meteorological information (winds, potential vorticity, cloud cover, lightning flashes; Thompson et al., 2007b,c, 2008) to assign free tropospheric ozone to three sources: (1) the stratosphere, designated in our analyses as ST; (2) regional convective redistribution of ozone and/or ozone precursors, with lightning, referred to as RCL; and (3) advection of pollution together with background ozone, collectively denoted as AD. Fig. 1 shows the free troposphere categories along with boundary layer ozone. In order to assess the influence of this free tropospheric ozone on the boundary layer, lower tropospheric ozone statistics are also analyzed. Free tropospheric ozone may have a significant impact on boundary layer ozone through downward mixing under many meteorological situations. This study will demonstrate the importance of frequent ozonesondes by characterizing the variability of ozone sources in the free troposphere and providing insight on the influence this ozone aloft has on surface ozone so that more accurate air quality forecasts can be issued.

2. Experimental methods of analysis

Ozone soundings were taken in summers 2004 through 2007 at Beltsville, Maryland, as part of several campaigns and under

Table 1
Beltsville ozonesonde data 2004–2007.

Year	Project	Time frame	Day launch	Night launch
2004	IONS-04	July 8–Aug 11	9	0
2005	Howard University/ MDE Pollution Study	July 18–Aug 25	9	11
2006	IONS-06, WAVES 2006	June 30–Aug 28	11	13
2007	WAVES 2007	June 27–Aug 8	15	10