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Impact of the vertical mixing induced by low-level jets on boundary layer ozone concentration



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HIGHLIGHTS

- ▶ The nocturnal O₃ maxima occurred concurrently at multiple sites along the corridor.
- ▶ The residual layer (RL) is leaky in the presence of a strong low-level jet (LLJ).
- ► The role of LLJ-induced vertical mixing on boundary layer O₃ is identified.
- ▶ Mixed-layer O₃ concentration on the next day is reduced as a result of a leaky RL.

ARTICLE INFO

Article history: Received 5 June 2012 Received in revised form 16 December 2012 Accepted 19 December 2012

Keywords:
Ozonesonde
Low-level jet
Atmospheric boundary layer
Ozone
WRF/Chem
Residual layer

ABSTRACT

After sunset, a stable boundary layer (SBL) develops close to the ground, while the upper region of the daytime mixed layer becomes the residual layer (RL). Mixing between the SBL and RL is often quite limited and the RL is thought to be a reservoir for daytime mixed-layer pollutants under such conditions. However, ozone (O₃) profiles observed in Maryland, U.S. suggest that the RL is not always a reservoir of O₃ in that region. Nocturnal low-level jets (LLJs) and/or other mechanisms are speculated to enhance vertical mixing between the SBL and RL, which influences the vertical O₃redistribution. Nocturnal surface O₃ maxima, an RL with reduced O₃ levels, and a concurrent strong LLJ were observed in Maryland on the night of August 9-10, 2010. Surface O₃ measurements in the region and three-dimensional air quality simulations suggest that horizontal advection cannot explain the nocturnal O₃ maxima and concurrent decrease of O₃ levels within the RL. A sensitivity study with a single column (1D) chemistry model was performed to investigate the role of LLIs in generating turbulent mixing within the nighttime boundary layer and to identify related impacts on O₃ concentrations at night and on the following day. The strong shear associated with the LLI enhanced turbulent mixing and weakened the decoupling of the RL and SBL substantially. Ozone was actively mixed down from the RL to the surface, causing secondary nocturnal surface O₃ maxima. Near the surface, O₃ was efficiently removed by chemical reactions and dry deposition, which resulted in lower O₃ peak values on the next day.

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1. Introduction

Following the traditional picture of the diurnal evolution of the atmospheric boundary layer, radiational cooling after sunset results in the development of a stable boundary layer (SBL) near the

Corresponding author. E-mail address: yuanfangcan@gmail.com (X.-M. Hu). surface that is typically quite shallow. Above the SBL is a residual layer (RL) with characteristics similar to those of the previous day's mixed layer (Stull, 1988). In the absence of strong disturbances, mixing and dispersion of pollutants between the RL and SBL become limited, and within the RL the concentration of pollutants remains at similar levels as in the mixed layer before its decay, which is why the RL is often viewed as a reservoir of pollutants (Stull, 1988). The pollutants trapped within the RL from the previous day can be entrained downward into the re-developing mixed

layer on the following day. In places such as the northeastern United States, such downward mixing of ozone (O₃) and its precursors is shown to contribute substantially to ground-level O₃ buildup in the morning in addition to chemical production (Zhang et al., 1998; Zhang and Rao, 1999). The downward transport of the RL O₃ in the morning also contributes to the maximum O₃ levels observed near the surface during daytime (Neu et al., 1994; Aneja et al., 2000; Yorks et al., 2009; Morris et al., 2010; Tong et al., 2011). Accurate information regarding the RL O₃ is thus critical for correctly simulating the daytime O₃ near the surface (Herwehe et al., 2011). Due to its relative inaccessibility, the actual detection of the properties of the RL at high temporal and spatial scales has been limited in the past. Recent field experiments (e.g., Balsley et al., 2008) showed that the classical view of a quiescent RL may have been oversimplified. Sporadic turbulence exists at night, weakening the decoupling between the RL and SBL, and the vertical mixing in the nighttime boundary layer may be significant, even compared to that in the daytime convective boundary layer (Poulos et al., 2002; Tjernström et al., 2009). Enhanced nighttime turbulence may be triggered by mesoscale motions such as low-level jets (LLJs), Kelvin-Helmholtz instabilities, gravity waves, wake vortices, and density currents (Sun et al., 2002, 2004; Salmond and McKendry, 2005; Fritts et al., 2009). Such intense turbulence can affect the vertical structure of the nighttime boundary layer and vertical distribution of pollutants. The view of the quiescent RL as a reservoir of pollutants may be challenged under such conditions. Recent observations (Hu et al., 2012) have suggested that the RL is leaky at times, i.e., active vertical exchange of air exists between the RL and the SBL. As a result, the O₃ levels in the RL may be highly variable and surface O₃ may not decrease as fast as anticipated based on the assumption of having a completely decoupled RL and SBL. In some cases, there are even secondary nighttime O₃ maxima reported, which were typically associated with periods of enhanced mixing (Corsmeier et al., 1997; Reitebuch et al., 2000; Salmond and McKendry, 2002; Talbot et al., 2005; Hu et al., 2012). Therefore, it is important to further investigate the dynamics and mixing of nocturnal boundary layers to better understand the temporal variability, absolute levels, and deposition rates of surface layer O₃ concentrations.

In Beltsville, Maryland (MD), nighttime vertical O₃ profiles have been measured during the summertime since 2004 (Yorks et al., 2009; Hu et al., 2012). Beltsville is located between Washington, D.C. and Baltimore, MD, in the middle of the Mid-Atlantic urban corridor of the United States. Heavy emissions of O₃ precursors and favorable meteorological conditions frequently lead to extreme O₃ events in this area (Ryan et al., 1998). Within the RL, O₃ levels at the Beltsville site at times resemble those found in the free troposphere with concentrations that are significantly lower than those in the previous day's atmospheric mixed layer. Ozone in the RL inherited from the daytime mixed layer appears to be readily mixed down to the surface, contributing to elevated O₃ at night (Hu et al., 2012). Previous studies have shown that LLIs occur frequently in the Mid-Atlantic region of the United States during the period between 1900-0600 h local time (LT), with peak winds ranging from 8 to 23 m s⁻¹ (Ryan, 2004; Zhang et al., 2006). Such strong LLJs may cause the RL to become leaky (Banta et al., 2007). Thus, the LLJs are hypothesized to contribute to the formation of leaky RL and nighttime surface O₃ maxima in the Mid-Atlantic region.

In the current study, the impacts of an LLJ observed in the Beltsville area on surface O₃ and vertical O₃ profiles are investigated in detail using different numerical modeling approaches. Initially, a three-dimensional model is employed to examine the spatial extent of the LLJ and to diagnose its role in modulating boundary layer O₃. A single column model is then applied to isolate the impacts of the LLJ on boundary layer mixing, nocturnal O₃ dispersion,

and O_3 built-up on the subsequent day. This study, for the first time, provides direct modeling evidence that LLJs induce substantial turbulence and reduce the RL O_3 significantly; as a result, the O_3 level in the daytime boundary layer on the following day is lowered.

2. Methods

During summer 2010, a research field campaign (Hu et al., 2012) was conducted at Howard University's Atmospheric Research Site in Beltsville, MD (39.06°N, 76.88°W). The meteorological variables and mixing ratios of chemical species (O₃, NO, NO₂, CO, SO₂) were measured at 5 m above ground level (AGL). The mixing ratios of chemical species were recorded every second. During several intensive observation periods, balloon-borne meteorological and O₃ sondes were used to obtain vertical profiles of temperature, humidity, wind speed and direction, and O₃. On the night of August 9-10, 2010, a secondary O₃ maximum and a concurrent LLI were observed at this research site. This event will be the focus of the current study. In addition to the night of August 9–10, O₃ sondes were also launched in the afternoon of August 9, which provided the unique opportunity during the field campaign to investigate the O₃ variation from the daytime convective boundary layer to the nighttime boundary layer. LLJs commonly occur in the Mid-Atlantic region (Zhang et al., 2006). This case study demonstrates potential impacts of the frequently occurring phenomenon of nocturnal LLJs on boundary layer O₃.

Three-dimensional (3D) air quality simulations, using the Weather Research and Forecasting model with Chemistry (WRF/Chem, Grell et al., 2005), for the 2010 summer campaign were applied in Hu et al. (2012) to investigate regional transport of O_3 and illustrate certain caveats in 3D air quality simulations. As part of the current study, output from these WRF/Chem simulations along with hourly O_3 data recorded at the AIRNow sites in the region were first used to examine the spatial extent and the potential causes of elevated nocturnal surface O_3 concentrations during the night of August 9–10, 2010. Details about the model set-up and domains of the WRF/Chem simulations can be found in Hu et al. (2012).

To further investigate the impacts of the strong LLJ observed on the night of August 9–10, 2010 near the Beltsville site on the vertical distribution of O₃, a single column photochemical model CACHE (Forkel et al., 2006) is employed in this study. In the vertical direction, 40 model layers extend from the surface to the 2.64-km height, with a vertical grid spacing of 1 m for the lowest layer and 500 m for the uppermost layer; such a setup appears to adequately capture the boundary layer structure during both nighttime and daytime. The multi-layered photochemical model solves the following system of equations:

$$\frac{\partial \chi_{i,j}}{\partial t} = E_{i,j} + D_{i,j} + C_{i,j} + \frac{\partial}{\partial z} \left(K \frac{\partial \chi_{i,j}}{\partial z_j} \right)$$
 (1)

where subscripts i and j denote the ith chemical species and the jth model layer, respectively, with χ being the concentration of a chemical species. Terms E, D and C are the rates of change due to emissions, dry deposition, and chemical reactions, respectively. The estimation method of emissions of volatile organic compounds, E, was updated to use the formula of Guenther et al. (2006). The dry deposition, D, is treated using the methods of Wesely (1989) and Gao et al. (1993). The chemical reaction rate, C, is computed using the Regional Atmospheric Chemistry Mechanism (RACM) gasphase mechanism (Stockwell et al., 1997). The atmospheric turbulent transport term, i.e., the last term of (1), is parameterized using a first-order closure scheme. The eddy diffusivity K is described

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