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Diurnal variation of surface ozone in mountainous areas: Case study of Mt. Huang, East China

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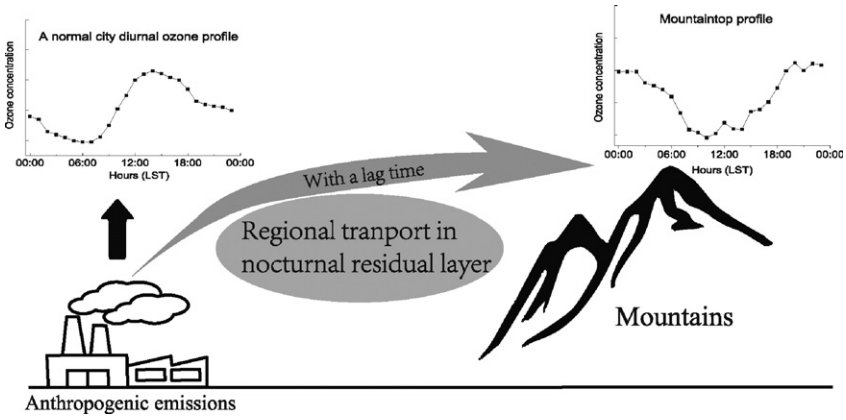
HIGHLIGHTS

- A distinct diurnal cycle of O_3 at the summit of Mt. Huang in East China.
- The diurnal O_3 pattern at the summit was driven by the regional O_3 transport.
- The diurnal O_3 change at high-altitude sites was associated with residual layer.

GRAPHICAL ABSTRACT



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ABSTRACT

To explore the variations in atmospheric environment over mountainous areas, measurements were made from an intensive field observation at the summit of Mt. Huang (30.13°N, 118.15°E, 1841 m above sea level), a rural site located in East China, from June to August 2011. The measurements revealed a diurnal change of surface O_3 with low concentrations during the daytime and high concentrations during the nighttime. The causes of diurnal O_3 variations over the mountain peak in East China were investigated by using a fairly comprehensive WRF-Chem and HYSPLIT4 modeling approach with observational analysis. By varying model inputs and comparing the results to a baseline modeling and actual air quality observations, it is found that nearby ozone urban/anthropogenic emission sources were contributing to a nighttime increase in mountaintop ozone levels due to a regional transport lag and residual layer effects. Positive correlation of measured O_3 and CO concentrations suggested that O_3 was associated with anthropogenic emissions. Sensitivity modeling experiments indicated that local anthropogenic emissions had little impact on the diurnal pattern of O_3. The diurnal pattern of O_3 was mainly influenced by regional O_3 transport from the surrounding urban areas located 100–150 km away from the summit, with a lag time of 10 h for transport.

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1. Introduction

Tropospheric O_3 has an important effect on air quality, tropospheric chemistry, climate change, as well as human health and vegetation growth (Gao et al., 2005; Lippmann, 2000; Sitch et al., 2007; Solomon et al., 2000; Yang et al., 2012). O_3 and other atmospheric constituents over remote mountains could represent the atmospheric background of environmental change (Aneja et al., 1994; Tan et al., 2015; Gheusi et al., 2011; Puxbaum et al., 1991; Ribas and Penuelas, 2006; Skelly et al., 1997; Van Ooy and Carroll, 1995). The O_3 variation over mountainous areas is more complex, reflecting the interactions of physical and chemical processes, especially the terrain effect on dynamic transport on different scales and the exchange of atmospheric constituents between the boundary layer and the free troposphere.

It is well-known that diurnal changes of surface O_3 between low daytime concentrations and high nighttime concentrations were observed at many mountaintop sites over the world (Gallardo et al., 2000; Oltmans, 1981; Oltmans and Komhyr, 1986; Ryan, 1997), which is the opposite of that observed for surface O_3 over plains and flat terrain. The distinct pattern of diurnal O_3 changes measured at high-altitude observatories of mountaintops was thought to be a consequence of terrain thermally induced circulations, for example, mountain-valley breezes (Cristofanelli et al., 2013; Oltmans and Komhyr, 1986; Oltmans and Levy, 1994; Zaveri et al., 1995) in association with the vertical distribution of O_3 in the lower troposphere (Bonasoni et al., 2000; Forrer et al., 2000; Gheusi et al., 2011; Zellweger et al., 2000, 2003). In remote mountainous areas, the O_3 vertical distribution shows a positive correlation with altitude within the troposphere because of less impact of anthropogenic sources (Chevalier et al., 2007). In these areas, up-slope winds during the day drive the transport of O_3-poor air masses from the foothill regions to the mountaintop, and downdraft winds during the night bring O_3-rich air aloft towards the mountaintop, driving the diurnal O_3 changes (Yang et al., 2012). The terrain height of mountains was found to be a key factor in driving the exchange of O_3 between boundary layer and free tropospheric, with a critical altitude of 1.0–1.2 km a.s.l (Chevalier et al., 2007; Monteiro et al., 2012).

The distinct pattern of diurnal O_3 changes over mountainous areas was also found related to air mass transport from pollutant emission sources. For example, polluted air masses from L'Aquila were transported to Mt. Portella (Italy) through breeze circulation at night (Cristofanelli et al., 2013). The diurnal O_3 pattern at Mt. Fuji (Japan) was built by regional transport and vertical mixing of thermal convection (Tsutsumi and Matsueda, 2000). Regional pollutant transport is believed to form the diurnal O_3 variations at Mt. Abu (India) as well (Naja et al., 2003). At Mt. Tai (northern China), the air pollutant transport within the boundary layer to the summit resulted in increases in afternoon O_3 concentrations (Li et al., 2008), however, the cause for the nighttime peak of O_3 at Mt. Tai is only speculated to be the effect of local mesoscale dynamic transport (Gao et al., 2005). The regional transport of O_3 from the surrounding emission sources to the receptor regions at mountaintops and its influences on the diurnal O_3 changes at mountaintops are complicated by the pathway and period of regional transport on different scales, physical and chemical processes as well as the exchange between the boundary layer and the free troposphere over the mountaintops, which is required to further understand.

The Mt. Huang is located in East China (Fig. 1), a major pollutant emission source region connecting with economic boom and dense population over past decades in China. The atmospheric environment over Mt. Huang is found to be affected by air pollutant emissions over the surrounding areas. An O_3 contribution of 20–50% from the Yangtze Delta and East China led to a high O_3 event at the summit of Mt. Huang (Wang et al., 2006b). Regional transport was a key factor in driving the seasonal changes of O_3 concentrations at Mt. Huang (Li et al., 2007). However, little is known about the mechanism of regional transport forming the diurnal variation in surface O_3 over the Mt. Huang. As such, this study aimed understanding causes on diurnal O_3 variation over mountainous areas in connection with regional environment change.

2. Experiment

2.1. Measurement site and O_3 concentrations

An intensive field observation including hourly meteorology and concentrations of O_3 and CO were simultaneously measured at the summit of Mt. Huang in summer 2011. The measurement site was located at the summit of Mt. Huang, at an elevation of 1841 m a.s.l., in the southern rural area of East China. The summit is approximately 280 km southwest of the Yangtze River Delta and overlooks the city of Huangshan, 45 km to the south. Around Mt. Huang, a number of cities are located within 100–150 km, such as Anqing and Chizhou to the northwest, Jingdezhen and Nanchang to the southwest, Quzhou and Jinhua to the southeast (Fig. 1). As a popular tourism destination in China, Mt. Huang receives a large number of tourists during summer (June, July, and August). Local pollutant emissions, such as carbon monoxide, $PM_{2.5}$ and volatile organic compounds from combustion sources are produced from small restaurants and temples.

Climatologically, the area around Mt. Huang is a typical monsoon region with significant changes in winds, temperature, humidity, and

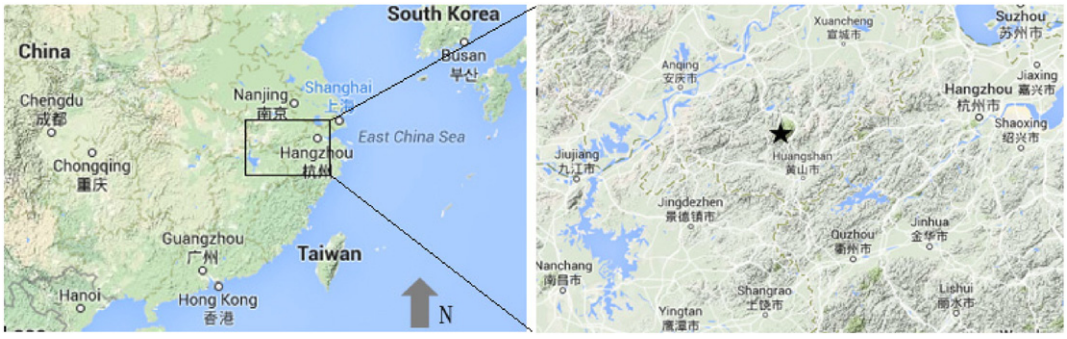

Fig. 1. Location of Mt. Huang (black star) and its neighboring cities in East China. (https://www.google.co.uk/maps/@32.2128013,112.3352055,5z).