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Measurements of vertical and horizontal distributions of ozone over Beijing from 2007 to 2010

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HIGHLIGHTS

- The vertical and horizontal distributions of O_3 over Beijing region was analyzed.
- A peak O_3 concentration occurring at ~1 km over Beijing is observed.
- O_3 concentration in the downwind of the city plumes was enhanced.
- A transition of O_3 formation from VOC-limited to NOx-limited at ~1 km is found.

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ABSTRACT

The vertical distributions of ozone (O_3) over a mega city (Beijing, China), and the horizontal O_3 distributions in the lower troposphere (2–3.6 km) over Beijing and its surrounding areas located in the North China Plain (NCP), were analyzed based on the aircraft measurements from 159 flights during 2007–2010. The results are highlighted as follows: (1) There was a peak of O_3 concentration occurring at ~1 km over Beijing, and the peak values ranged between 60 and 120 ppbv. (2) There was an O_3 minimum at the surface. The minimum was largely caused by the chemical reaction of NO + O_3. This process produced about 30 ppbv of the O_3 reduction below 0.5 km in the morning (9:00–10:00). (3) There was a transition altitude (~1 km), below which the ozone formation was in a VOC-limited condition, and above which the ozone formation was in a NOx-limited condition. (4) The analysis of the horizontal distribution shows that O_3 concentrations were enhanced in the downwind of the city plumes. This result suggests that there was an important regional O_3 chemical production in the NCP region.

1. Introduction

Ozone is a critical trace gas in the troposphere because it plays important roles in atmospheric chemistry, air quality, and climate change (Bojkov, 1988; Akimoto, 2003). The O_3 formation is significantly affected by several important chemical precursors, including volatile organic compounds (VOC) and oxides of nitrogen (NOx) (Sillman, 1995), meteorological factors (Benjamin and Winer, 1998; Tie et al., 2009a), and atmospheric aerosols (Tie et al., 2005).

China's rapid economic growth in recent years has resulted in large increases in pollutant emissions (Ohara et al., 2007; Zhang et al., 2007; Tang et al., 2009; Yang et al., 2011). The NCP region is located in central of China, and the region includes several large cities, such as the mega city of Beijing. The region is one of the most populated and industrialized regions in China and is suffered from the impacts of air pollution in many aspects (Hao and Wang, 2005; Shao et al., 2006; Quan et al., 2011). The O_3 problem is among the most stubborn environmental issues. For example, the concentration of O_3 has frequently exceeded the national air quality standard (Lu et al., 2010; Wang et al., 2009), with a peak hourly averaged concentration of 286 ppbv (Wang et al., 2006). Previous studies focused mainly on the spatial and temporal variations of the surface ozone and its precursors in the NCP region. Several observation data indicated that the surface O_3 concentration in suburb was higher than that in urban over NCP (e.g. Wang et al., 2006; Xu et al., 2011). The analysis of relationship between the rate of

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photochemical ozone production (P[O_3]) and it's precursors (NOx and VOCs) indicated that O_3 production in concentrated urban plumes is often in a VOC-limited regime (Shao et al., 2009; An, 2006; Duan et al., 2008; Xu et al., 2011; Lu et al., 2010; Tang et al., 2010). As a result, the high NOx concentration in NCP lead to a strong inhibitory effect on O_3 formation, and the reduction of NOx emissions might produce in accelerating ozone production (Wang et al., 2009; Yang et al., 2011). In addition, the aerosol loadings are extremely heavy in the NCP region (Tie and Cao, 2009), and the heavy loading of aerosols over the NCP region also affects the O_3 formation by reducing solar radiation (Bian et al., 2007a; Han et al., 2009). However, there were only the surface measurements of ozone and its precursors, which were available and used in the previous studies. It is lack of O_3 vertical measurements in the troposphere in NCP. Although satellite and ozonesonde can give some insights of the vertical structure of ozone (Bian et al., 2007b; Wang et al., 2012), the low spatial and temporal resolutions of the above data cannot provide the details of O_3 formation and transport in the lower troposphere.

In this paper, we show the in-situ aircraft measurements of O_3 and its important precursor (NOx) in the NCP region. The time resolution of the measurements is 1 s and the vertical resolution is several decameters. The analysis focuses on the following issues: (a) the vertical distribution of O_3 in the lower troposphere over Beijing; (b) the effect of NOx concentrations on O_3 formation over Beijing; and (c) the horizontal distribution O_3 in the lower troposphere over the NCP region.

2. Description of the measurement

2.1. Instruments on the aircraft

Several commercial instruments were mounted on the aircraft (Y-12) to measure the concentrations of O_3, NOx (NO + NO_2), and other atmospheric pollutants. Ozone was measured using a commercial UV photometric analyzer (Model 49iTL, TEI Inc.). The time resolution was set to 1 s during the aircraft measurement in order to get more spatial information, which might affect the detectable limit of the measurements. For example, the detectable limit decreases from 0.05 ppbv to 0.5 ppbv when the time resolution increases from 120 s to 1 s. The measured O_3 concentrations range from 0.5 to 200 ppbv, with temperature and pressure correction. NO–NO_2–NOx was measured with a chemiluminescent trace level analyzer (Model 42iTL, TEI Inc.). The analyzer has a detection limit of 0.025 ppbv. These analyzers were calibrated before the field campaigns by injecting a span gas mixture in scrubbed ambient air generated by a TEI model 111. The zero check was also conducted. The inlet system was placed in front of the aircraft to avoid the influence of the aircraft exhausts for measurements. Aerosol particles were measured by a passive cavity aerosol spectrometer probe (PCASP-200, DMT Inc.), with the particle size ranging from 0.10 to 3.0 μm in diameter. The meteorological measurements included location, temperature, relative humidity, barometric pressure and wind using an aircraft integrated meteorological measurement system (AIMMS-20, Advantech Research Inc.). The PCASP-200 and AIMMS-20 instruments were amounted under the wing of aircraft.

2.2. Flight information and data process

Fig. 1 displays the horizontal flight routes during 2007–2010. Three airports were used during the experiments, including Shahe (SH), Zhangjiakou (ZJK), and Dingxing (DX) airports. The geographical locations (latitude and longitude) of the three airports are indicated in Fig. 1. The three airports are not for commercial use, and there are only several flights in each day. The effect of the emissions of airplanes is very small, and the measured vertical distributions of gaseous pollutants were insignificantly perturbed by the aircraft emissions. The detailed information for measuring O_3 vertical profiles is described as follows:

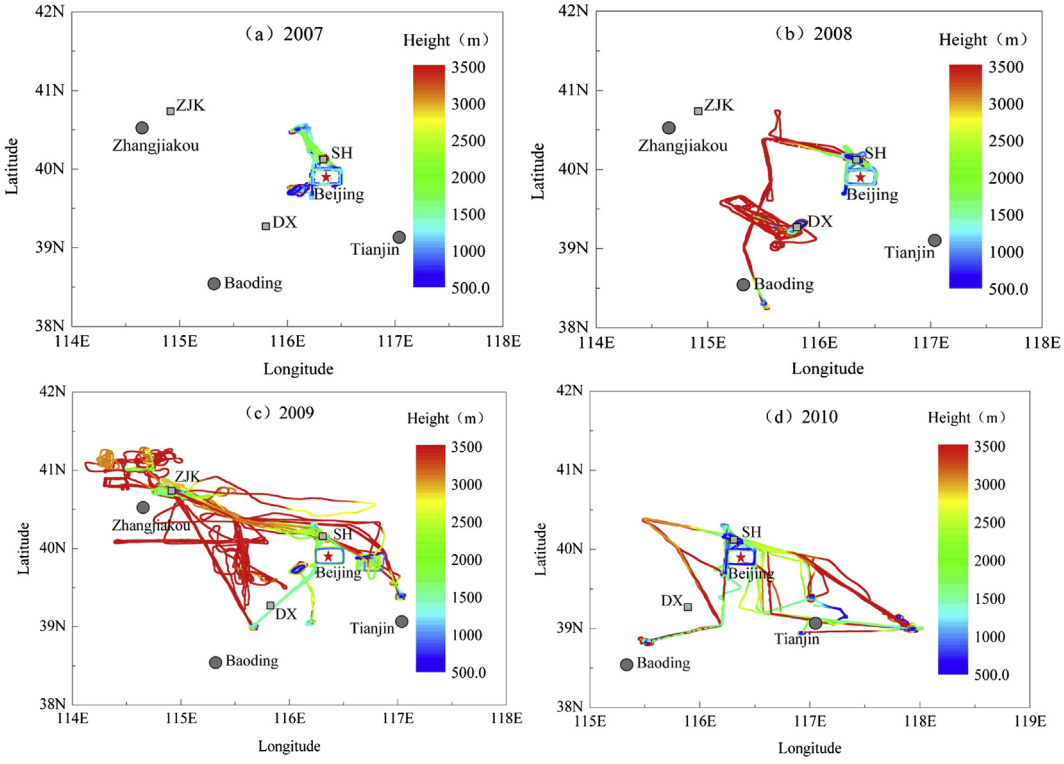

Fig. 1. The horizontal flight routes over NCP during 2007–2010.