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A study of elevated pollution layer over the North China Plain using aircraft measurements

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ABSTRACT

An elevated pollution layer (EPL) at altitude ~1700 m was observed over the North China Plain (NCP) in November 2016. The vertical profiles of aerosol loadings, chemical compositions and meteorological parameters were in-situ measured at both ground and aircraft platforms. The EPLs were observed simultaneously over Beijing and Baoding city (~150 km distance between) with similar aerosol concentration and size distribution, indicating the impact of the EPL at regional scale. The synoptic and remote sensing analysis suggest the pollutants in the EPL may result from regional transport from the polluted southwest, and then elevated by the influence of anticyclone circulation and surrounding terrain. The descent air mass next day may lead to EPL entrainment and contribute to increased aerosol concentration at lower level. The non-refractory compositions measured by aerosol mass spectrometer showed more significant fraction of nitrate and secondary organics in the EPL compared to the other layers. The pollutants in the EPL was then mixed into the developed planetary boundary layer (PBL), leading to uniform distribution of aerosol composition. Such atmospheric stratification at high level and its subsequent impact on the lower level needs to be considered for the future radiative forcing study over this region.

1. Introduction

Elevated pollution layers have been reported in many cities around the world (Chen et al., 2009; Padmakumari et al., 2013; Shi et al., 2011; Viskanta and Daniel, 1980; Wakimoto and McElroy, 1986). The layers can be formed in a number of ways, such as transport process, evolution of planetary boundary layer (PBL) and local circulation. EPL can be produced by pollutants from tall smoke stacks as high as 380 m (Lusis and Wiebe, 1976). Roger et al. (1986) reported an EPL over Los Angeles with lidar observation, indicating that upper-level winds within the inversion, orographic effects, and thermally induced changes in the depth of the mixed layer controlled the evolution of these layers. Aircraft observations near the foothills of Himalayas suggested that the EPL sources can be from local anthropogenic activities as well as long-range transport (Padmakumari et al., 2013). An EPL at altitudes above 10 km was also observed over Europe due to long-range transport, which originated from the Pagami Creek forest fire in Minnesota, USA (Dahlkötter et al., 2014). Shi et al. (2011) observed a residual nocturnal layer in the urban atmosphere caused by PBL evolution. EPLs also occurred in China, especially in the North China Plain. Chen et al. (2009) reported an EPL at the altitude of 2500–3500 m observed by aircraft measurement in Beijing, and suggested that the mountain-valley breeze induced Chimney Effect on EPL formation. Previous studies suggest that EPLs have significant influences on temperature structure and dispersion in the PBL due to their prevention of solar radiation reaching the surface as well as the heating effects in lower atmosphere.

Severe haze events have been frequently observed in the North China Plain (NCP), especially during autumn and winter (Guo et al.,

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2014; Li et al., 2011; Quan et al., 2013; Zhang et al., 2015; Zheng et al., 2015). Although substantial efforts to reduce anthropogenic emissions during the last decade, Beijing is still experiencing frequent severe haze events (Sun et al., 2016b; Tang et al., 2015; Tao et al., 2016). Previous studies indicated the air pollution in Beijing was not only a local issue but also a regional problem (Hua et al., 2016; Wang et al., 2016; Wu et al., 2017).

The increase of $PM_{2.5}$ during haze events could be rapid in Beijing. Sometimes the mass concentration of $PM_{2.5}$ could be elevated by an order of magnitude in less than one day (Sun et al., 2016a; Zheng et al., 2015; Zhong et al., 2017). However, the reasons for the rapid formation of such severe haze episodes are still not well understood. Ground observations were found not to be able to fully explain the pollution process, while vertical profiling on the meteorological parameters and aerosol properties could improve the understanding of regional transport influence on heavy pollution events (Hua et al., 2016). By aircraft in-situ measurements, Liu et al. (2009) analyzed 152 vertical profiles of aerosol number concentration and size distribution observed by aircraft-based optical spectrometer probe in Beijing, China, indicating the vertical profiles of aerosol number concentration were influenced by the structures of PBL significantly (Liu et al., 2009). In addition, aerosol vertical profiles could be influenced by weather condition, reginal transport, local circulation, and secondary aerosol formation (Chen et al., 2009; Morgan et al., 2009, 2010; Zhang et al., 2009).

The objective of this study is to investigate the formation mechanism of the EPL and its impacts on ground aerosol mass concentration and composition, using both ground and aircraft observations, and provide an in-depth insight to the high-level regional transport process. Findings of this study would guide the air quality management strategies in the North China Plain.

2. Description of the measurements

2.1. Ground-level observation site

Comprehensive measurements were conducted in a field campaign at the Baolian meteorological station, China Meteorological Administration (CMA) (39°56′N, 116°17′E). The Baolian station locates between the western 3rd and 4th ring highways in Beijing. The station is about 400 m away from the main road without significant point sources. A high-resolution time-of-flight aerosol mass spectrometer (HR-ToF-AMS, Aerodyne, USA) was used to measure non-refractory submicron aerosol compositions (NR-PM_1) with a time resolution of 5 min, including organics, nitrate, sulfate, ammonium, and chloride. Gaseous pollutants including NO_x, SO_2, O_3 and CO were measured simultaneously. All instruments were calibrated in the beginning and middle of the project following the standard protocols. A micro-pulse lidar (MPL-4B, Sigmaspace Co., USA) was employed to investigate the evolution of the PBL. The pulse repetition frequency of the MPL is 2500 Hz, at the wavelength of 532 nm. The peak value of the optical energy of laser beam is 8 mJ. The pulse duration was set to 100 ns, and the pulse interval was 200 ns, corresponding to a spatial resolution of 30 m. The measurements of Wind-Profile-Radar and automatic meteorological observation (Vaisala Milos520, Finland) were conducted to obtain wind profiles and meteorological parameters (pressure, temperature, humidity, wind).

2.2. Details of flight observation

2.2.1. Flight routes

The aircraft (Yun-12 and KingAir-350) were used to carry out vertical profiles in November 2016. The typical aircraft speed is about 180 and 250 km h^{-1} for Yun-12 and KingAir-350 respectively, and the ascent and descent rates during profile was ~2–5 m s^{-1}. The surrounding terrain and three-dimensional flight routes during this campaign are shown in Fig. 1. The Yun-12 aircraft took off and climbed to about 3000 m over the Shahe airport, which located in the north of Beijing city with distance of 35 km. Then the aircraft flew towards southwest for about 150 km reaching Baoding area and performed vertical profiles in step levels of 300 m (from 2700 to 600 m). Meanwhile, the KingAir-350 performed the vertical profiles over Beijing. Before taking off, all instruments were operated for 1–2 h to measure aerosols on ground. As the Shahe airport is not for commercial use, there are only a few flight operations per day. The effect of aircraft emissions was therefore minor in contributing to the measured vertical distributions of aerosols. The detailed information of vertical profiles in this study is summarized in Table 1.

2.2.2. Instruments on the aircraft

In this study, the aerosol number concentration, size distribution and chemical composition were all measured. A passive cavity aerosol spectrometer probe (PCASP, DMT, USA) was mounted on the Yun-12 aircraft wingtip to measure aerosol size distribution ranging 0.1–3.0 μm in 1 Hz. The PCASP was maintained every year. and was calibrated using polystyrene latex (Duke Scientific Corporation) on monthly basis during field observations. A Compact time-of-flight aerosol mass spectrometer (C-ToF-AMS, abbreviated as AMS) was mounted in the KingAir-35er cabin to measure submicron non-refractory aerosol (NR-PM_1) chemical compositions with the time resolution of 1 min, including nitrate (NO3), sulfate (SO4), ammonium (NH4), chloride (Chl) and organics (Org). The sample air was introduced into the aircraft cabin through the isokinetic aerosol sampling inlet (Model 1200, BMI CO.) and was split to the AMS sampling line using dedicated stainless-steel flow splitters (Hermann et al., 2001). The operation of AMS is detailed in previous publications (DeCarlo et al., 2006; Jimenez et al., 2003). An integrated meteorological measurement system (Aimms-20, Advantech Research Inc.) was used to measure aircraft location, temperature, relative humidity, barometric pressure and wind.

In addition, the ambient pressure may have impacts on the AMS performance, such as the sample flow rate, particle transmission in aerodynamic lens and flight velocity in sizing chamber. To avoid these effects, a pressure controller was mounted upstream of the AMS inlet and maintained at fixed pressure during flight (Bahreini et al., 2008). To keep the flow rate constant, the fixed pressure should be lower than the pressure at the maximum flight height. In this work, the pressure controller was set to 650 hPa and all calibrations (flowrate, particle velocity, ionization efficiency) were performed under this pressure before and after each flight.

2.2.3. AMS data process

Standard ToF-AMS data analysis software packages (SQUIRREL version 1.50) were used to deconvolve mass spectrum and obtain mass concentrations of chemical components. Mass concentrations derived from the AMS are reported as micrograms per standard cubic metre (T = 273.15 K, p = 1013.25 hPa), with the time resolution of 1 min. The AMS collection efficiency (CE), which accounts for the incomplete detection of aerosol species due to particle bounce at the vaporiser and/or the partial transmission of particles by the lens (Canagaratna et al., 2007), is significantly modulated by particle phase (Matthew et al., 2008). In this study, we used a CE correction following the principle developed by Middlebrook et al. (2012). Ionization efficiency (IE) calibrations were performed regularly by using size-selected (300 nm) pure ammonium nitrate particles before and after each flight during the observation period. The comparison of submicron aerosol mass concentrations derived by AMS and PCASP showed high consistency (Fig. S1).

A Positive Matrix Factorisation (PMF) analysis was performed on the organic mass spectral dataset following the procedures by Ulbrich et al. (2009). The application of PMF to AMS OA spectra has been described in detail previously (Ulbrich et al., 2009; Zhang et al., 2011). In this study, organic aerosol (OA) were differentiated into hydrocarbon-like organic aerosol (HOA) and oxygenated organic aerosol (OOA),