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Peroxyacetyl nitrate observed in Beijing in August from 2005 to 2009

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ABSTRACT

Measurements of peroxyacetyl nitrate (PAN) were made at a Beijing urban site each August from 2005 to 2009. Over this 5-year period, the average PAN concentration for August in each year increased from 3 (2005) to 11.7 μg/m^3 (2007); however, it decreased rapidly in 2008 (4.1 μg/m^3). Generally, the variation over the 5 years showed a rise in the first part of the study period, followed by a decline. We considered two categories of local and regional air masses in this study, which revealed that the PAN concentration in Beijing was affected mainly by southeastern air masses. The August PAN variation was influenced predominantly by local air masses in 2005, but by 2009 regional air masses had become more important. This study showed the level and variation of PAN in the month of August in 5 consecutive years for the first time, and proved that control measures are useful in decreasing photochemical pollution; hence, these measures are probably feasible for other megacities too. Furthermore, this method of analyzing regional and local impacts might be useful for other studies as well.

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Introduction

Peroxyacetyl nitrate (PAN, $CH_3C(O)OONO_2$) is an important photochemical oxidant in the atmosphere (Stephens, 1969) that can irritate human eyes, damage plants (Temple and Taylor, 1983), and even lead to genetic mutations (Kleindienst et al., 1990). Previous studies have shown that PAN is ubiquitous in the troposphere (Singh and Salas, 1983; Singh et al., 1986; Rappenglück et al., 1993; Grosjean, 2003; Dassau et al., 2004; Marley et al., 2007; Lee et al., 2008; Wang et al., 2010c) and originates solely from photochemical reactions. Its precursors are $RC(O)O_2$ radicals and NO_2. PAN is a better indicator of photochemical processes than ozone due to its low natural background concentration and its thermal instability (Rappenglück et al., 1993). Thermal decomposition is the major loss mechanism for PAN in the lower troposphere (Grosjean et al., 2001). During thermal decomposition, PAN releases NO_2 and $CH_3C(O)OO$ radical, and is therefore an important NOx reservoir (Singh and Salas, 1983; Singh et al., 1986). Although PAN decomposes quickly at high temperatures, at low temperatures it can be transported for thousands of miles before releasing NOx, which influences the photochemical reaction capacity in other locations (Roberts et al., 2004).

As the capital of China, Beijing has achieved an average annual GDP growth of 10% (Feng et al., 2012). However, with the rapidly developing economy, the city's environmental problems have intensified. Air pollution, especially photochemical pollution (Shao et al., 2006), which includes PAN, is the most significant environmental problem that the government must manage. The earliest studies of PAN in Beijing were made in the 1990s (Zhang and Tang, 1994). The CareBeijing Campaign has been undertaken since 2006, resulting in the publishing of a series of articles regarding Beijing's air pollution, including PAN (Wang and Zhang, 2007; Yang et al., 2009; Liu et al., 2010; Zhang et al., 2011). The previous studies of PAN have been made over a single year or month, with very little information regarding the year-to-year variation of PAN in August available. A study on the year-to year variation of PAN in August would therefore provide a better understanding regarding the variation in photochemical pollution in Beijing during recent years. Furthermore, it would also provide

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support for the government's pollution control policies (Grosjean, 2003; Marley et al., 2007; Zhang et al., 2011).

Owing to the high temperatures and sunlight, summer is the most appropriate season to study photochemical reactions. Hence, in this study, we measured PAN in August in each year from 2005 to 2009 (18 days in 2005 and 2006, 30 days in 2007–2009) at an urban site in Beijing. Moreover, the effect of transport on the PAN concentration in Beijing and typical pollution periods were analyzed using the HYSPLIT (HYbrid Single-Particle Lagrangian Integrated Trajectory) model.

1. Methods

1.1. Sampling site

Beijing is located in the northwest of the North China Plain and has a continental monsoon climate. It is surrounded by mountains in three directions, and owing to this terrain, air pollutants tend to accumulate in Beijing. The sampling site was located in the northwest of urban Beijing (Fig. 1), on the roof of a 25-m-high building in Peking University (39.99°N, 116.31°E). The inlet of the instrument, which was made of Teflon and contained a replaceable filter membrane that was used to prevent particular matter from blocking the mass flow controller in the instrument, was deployed 4 m above the roof level, and the filter membrane was replaced about every 5 days. All the other instruments mentioned later in this study also sampled from the same location on the roof.

1.2. Equipment

PAN was measured in the month of August of the years 2005–2007 using a gas chromatography–electron capture detector (GC–ECD) system designed by the National Oceanic and Atmospheric Administration (NOAA) (Williams et al., 2000) and measured in the month of August of the years 2008–2009 using a similar system designed by Peking University (intercomparison of results showed that it performed the same as that of NOAA), having the same instrumental parameters as the NOAA system. The instrument had a detection limit of 0.03 μg/m^3 and a time resolution of 5 min, up to 10% accuracy (A) and 5% precision (P), and was calibrated every week (Williams et al., 2000; Wang et al., 2010c; Zhang et al., 2011). A standard PAN gas was made by the photolysis of CH_3COCH_3 and NO and then diluted to desired concentrations using PAN-free air (Volz-Thomas et al., 2002). In this system, PAN-free air was produced by the thermal decomposition of ambient air (Williams et al., 2000). NO and CH_3COCH_3 were purchased from Scott-Marrin, Inc., Riverside, California, USA. All of the other pollutants such as O_3, NO, NO_2, CO, $PM_{2.5}$, propene and meteorological data were measured simultaneously with TECO (Thermo Electron Corporation, Waltham, Massachusetts, USA) models 49C (1% A; 2% P), 42CTL (1% A; 1% P), 48C (1% A; 1% P), and TEOM (Tapered Element Oscillating Microbalance) 1400a (0.75% A; 1% P), a gas chromatograph equipped with a quadrupole mass spectrometer and a flame ionization detector (5% A; %5 P) (Wang et al., 2010c) and Peking University's automatic meteorological station, respectively.

1.3. HYSPLIT model

The HYSPLIT model was developed by the National Oceanic and Atmospheric Administration (NOAA) and Australia's Bureau of Meteorology (Draxler and Rolph, 2003; Rolph, 2003) and is widely used for simulating the motion of air masses at the regional scale (Rappenglück et al., 2003; Traub et al., 2003; Wolfe et al., 2007; Zhang et al., 2009; Fischer et al., 2011). In the model, meteorological data are derived from the Global Data Assimilation System (GDAS) archive, which has a 1° × 1° spatial resolution. To study the specific influence of transport on the PAN concentration in Beijing, we analyzed the relationship

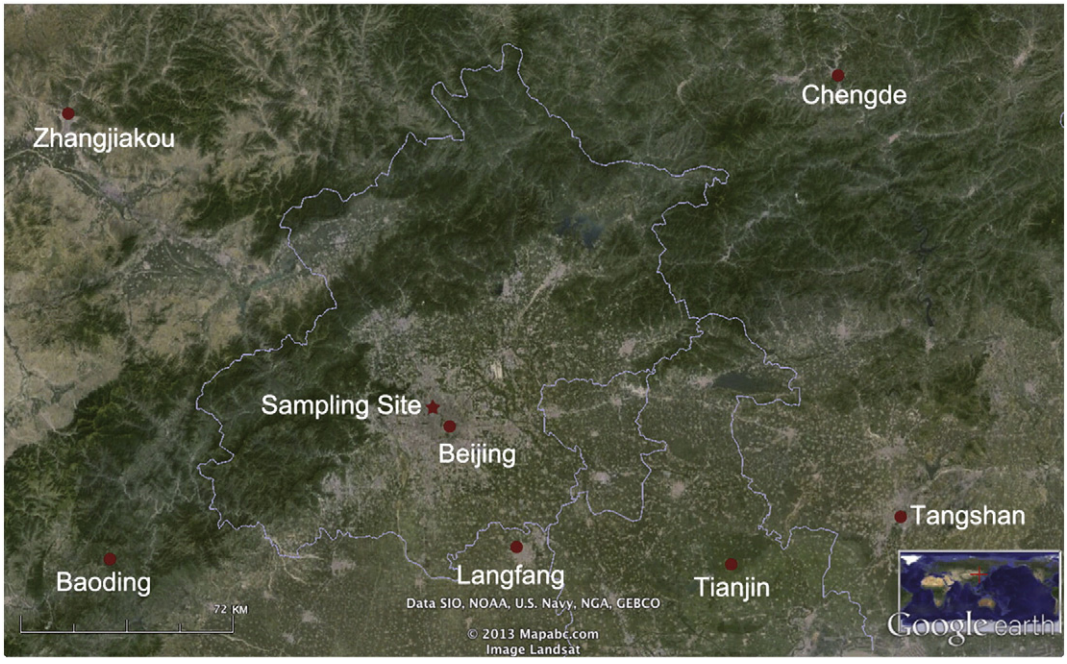

Fig. 1 – Location of the sampling site in Beijing.

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