I Investigating the impact of regional transport on PM_{2.5}

2 formation using vertical observation during APEC 2014

3 Summit in Beijing

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17 ABSTRACT

18 During APEC (Asia-Pacific Economic Cooperation) Economic Leaders' 2014 Summit in Beijing, 19 strict regional air emission control was implemented, providing a unique opportunity to investigate the transport and formation mechanism of fine particulate matter ($PM_{2.5}$). This study explores the use 20 21 of vertical observation methods to investigate the influence of regional transport on PM_{2.5} pollution in 22 Beijing before and during the APEC Summit. Vertical profiles of extinction coefficient, wind, 23 temperature and relative humidity were monitored at a rural site on the border of Beijing and Hebei Province. Three PM_{2.5} pollution episodes were analysed. In episode 1 (October 27th to November 1st), 24 25 regional transport accompanied with the accumulation of pollutants under unfavourable meteorological conditions led to the pollution. In episode 2 (November 2nd to 5th), pollutants left from 26 27 episode 1 were retained in the boundary layer for 2 days in the region and then settled down to the surface, leading to an explosive increase of PM_{2.5}. The regional transport of aged aerosols played a 28 crucial role in the heavy PM_{2.5} pollution. In episode 3 (November 6th to 11th), emission from large 29 30 point sources had been controlled for several days while primary emissions from diesel vehicle might 31 lead to the pollution. It is found that ground-level observation of meteorology condition and air quality could not fully explain the pollution process while vertical parameters (aerosol optical properties, winds, relatively humidity and temperature) improved the understanding of regional transport influence on heavy pollution process. Future studies may consider including vertical observations to aid investigation of pollutant transport especially during episodic events of rapidly increasing concentrations.

1 1. Introduction

2 With a rapid economic development and increases in energy consumption, large quantity of emissions 3 has caused serious particulate matter pollution in China. Monitoring data show that Beijing-Tianjin-4 Hebei (BTH) region is one of the most polluted region in China (Zhao et al., 2013; Wang et al., 2014). 5 The region was home to eight out of the top 10 most polluted Chinese cities in 2014 (MEP-Ministry 6 of Environment Protection, 2015). In 2014, the annual average PM_{2.5} (particulate matter with 7 aerodynamic diameter less than 2.5 μ m) concentration reached 95 μ g/m³ in the BTH region. With 8 21.5 million residents and 5.3 million vehicles, Beijing has been burdened with severe pollution 9 episodes frequently in recent years (Beijing Municipal Bureau of Statistics, 2014). The capital is 10 surrounded by mountains in three directions (north, west and east). The top three most polluted cities 11 in China (Baoding, Xingtai and Shijiazhuang) are located in the south to Beijing. Polluted air mass 12 from the south contributes to PM_{2.5} pollution in Beijing (Wang et al., 2015). Source apportionment by 13 Beijing Environmental Protection Bureau indicates regional transport contributed 28%-36% to PM_{2.5} 14 in Beijing in 2012-2013. During some severe pollution periods, regional contribution was more than 50% (http://www.bjepb.gov.cn/bjepb/413526/331443/331937/333896/396191/index.html). Quite a 15 few researches have studied the causes of heavy polluted episodes in BTH region and show regional 16 17 transport plays an important role in pollution formation. The sharp PM_{2.5} build-up events in Beijing 18 were unique while accumulation pollution process occurred at other cities in the region. This 19 indicated that PM_{2.5} was probably transported to Beijing from other cities (Zheng et al., 2015; Ji et al., 20 2014; Tao et al, 2014; Zhao et al., 2013). In the meanwhile, most severe pollutions are under stable 21 synoptic meteorological conditions in Beijing (Sun et al., 2015; Zheng et al., 2015; Zhao et al., 2013). 22 The low wind speed and stable synoptic meteorological condition at ground level cannot explain the 23 reason that regional transport makes significant contribution to severe pollution. A previous study has 24 shown the secondary aerosol in Beijing probably mainly formed over regional transport according to a 25 vertical observation from the ground to 260m height. (Sun et al., 2015). Therefore, vertical profiles of 26 meteorology and air quality might help us to understand the impacts of regional transport to heavy 27 pollution during stagnant conditions.

As in other megacities with local sources and regional transport, air quality in Beijing are affected by many factors, including emissions inside the city, formation of secondary pollutants, atmospheric mixing, and regional transport. It has been well known that the strength of each factor varies according to emissions and/or weather conditions. Therefore, it is challenging to pin point the major contributors in any given time periods, either clean or polluted episodes. This is especially difficult in BTH region considering the complicated emission sources and transport processes.

Emission control measures implemented during some events provide a unique opportunity to 1 2 investigate the impact of various factors influencing air quality. One of them was APEC (Asia-Pacific Economic Cooperation) Economic Leaders' 2014 Summit held in Beijing from November 5th to 11th, 3 4 2014. A strict emission pollution control plan was carried out in the BTH Region to improve air quality in Beijing from November 3rd to 11th for APEC. According to a conservative estimate by 5 MEP, production of 9,289 plants were paused and 3900 plants were running at reduced capacity in six 6 7 provinces (Beijing, Tianjin, Hebei, Shanxi, Shandong and Inner Mongolia); and more than 40 8 thousand construction sites were shut down temporally 9 (http://www.zhb.gov.cn/gkml/hbb/qt/201411/t20141115_291482.htm). Other measures include traffic 10 control (50% of private passenger vehicles and 70% of buses were off-road) and frequent road 11 sweeping and cleaning in Beijing. More detail emission control measures are supplied in the supporting information. Studies have found that regional emission control effectively reduced air 12 13 pollutant concentrations during the Summit (Wen et al., 2015; Tang et al., 2015; Han et al., 2015; 14 Chen et al., 2015; Sun et al., 2016a). The significantly reduced local emissions led to reduced 15 complexity of particulate matter pollution process, thus providing a unique opportunity to investigate 16 the influence of transport events on PM_{2.5} levels in Beijing.

17 The objective of the study is to investigate the impact of regional transport on $PM_{2.5}$ in Beijing using both ground-level and vertical observations. Field observation was conducted at a rural site (Liulihe) 18 19 in southwest Beijing before and during the control period of the APEC 2014 Summit. Vertical profiles 20 of temperature, RH (relative humidity), wind speed and direction, and extinction coefficient were 21 observed as well as pollutants concentration and meteorological parameters on the ground. The 22 characteristics of three PM_{2.5} pollution episodes were analysed. Findings of this study will help 23 explore vertical observation methods for in-depth analysis of the meteorological and transport 24 influence. Furthermore, it can aid the development of future air quality management strategies in BTH 25 and other regions around the globe, including emission control and air surveillance.

26 **2. Field observation and analysis methods**

27 **2.1 Field observation site and sampling methods**

Beijing is surrounded by mountains in the west, north and east directions, which blocks the pollutants from spreading. The open air corridor in the south exposes the capital to air mass passing Hebei Province (Fig. S1) a heavily polluted area in China. To investigate the impact of regional transport on Beijing, a rural site (Liulihe site, 116°2′E, 39°36′N) was chosen in the southwest of Beijing. It was located on the border of Beijing and Hebei Province (Fig. S1).

The field campaign was conducted from October 27th to November 12th, 2014, including both ground-1 2 level and vertical observations. Detailed information of instruments at Liulihe site is provided in 3 Table S1. Ground-level observations included meteorological parameters, mass concentration of PM_{2.5}/PM₁₀, SO₂, NO_x and O₃ as well as physical and chemistry properties of PM. PM_{2.5}/PM₁₀ mass 4 5 concentration was determined by the TEOM method. Particle size distribution from 3nm to 10µm 6 were measured by a spectrometer assembled in-house including one Nano scanning mobility particle 7 sizers (NSMPS), one scanning mobility particle sizers (SMPS), and one aerodynamic particle sizer 8 (APS) (Liu et al., 2014).

9 ACSM (Aerosol Chemical Speciation Monitor), a low-maintenance aerosol mass spectrometer, was 10 used to measure non-refractory (NR) particulate matter with aerodynamic diameters smaller than 1µm 11 (PM₁) (Ng et al., 2011). The ACSM data was calibrated with a collection efficiency (CE) value to 12 compensate for the particle loss. The CE value of 0.45 recommended by Middlebrook et al. (2012) 13 based on the monitoring site condition (see supporting information) was used in this study. The NR-14 PM1 concentration measured by ACSM tracks well with PM2.5 measured by the TEOM (R2=0.91) and 15 the regression slope is 0.43 (Fig. S2). Positive matrix factorization (PMF) with the PMF2.exe 16 algorithm was used to distinguish different components of OA measured by ACSM (Paatero and 17 Tapper, 1994). The PMF was performed and evaluated following the PMF analysis guide (http://cires1.colorado.edu/jimenez-group/wiki/index.php/PMF-AMS_Analysis_Guide). Three factors 18 19 were distinguished (Fig. S3), i.e., HOA (hydrocarbon-like organic aerosol), SVOOA (semi volatile 20 oxygenated organic aerosol) and LVOOA (low volatile oxygenated organic aerosol).

21 Beyond ground-level concentrations of routinely monitored air pollutants and meteorological 22 parameters, the assessment was aided by vertical observations including vertical extinction coefficient 23 profile, as well as vertical wind, RH and temperature profiles. The vertical extinction coefficient 24 profiles depict the distribution of PM, which could be used to infer mixing process of particles 25 transported in from high evaluations and those near the ground. Vertical wind profile indicate the 26 transport direction. Vertical RH profiles reflect the strength of heterogeneous reaction at different 27 layers. Vertical temperature profiles provide information on the stability of and mixing in the 28 boundary layer. Lidar was used to observe the vertical optical properties of atmospheric aerosols at 29 Liulihe site. The lidar consists of three parts, including emitting system, receiving system and signal 30 analogue system (Chen et al., 2015). The laser source emitted pulse at 355/532nm. The pulse energy 31 is 30MJ at 355nm and 20MJ at 532nm. The pulse repetition is 20Hz. The telescope for receiving 32 system is based on a Cassegrain design. Diameter of the telescope is 200mm with a vertical resolution 33 of 7.5m. The particle backscatter coefficient and extinction coefficient was retrieved by Fernald method (Frederick et al., 1984). CFL-03 phased array wind profile radar was used to monitor the vertical wind speed and direction with resolutions of 50 m (0-1 km) and 100 m (1-5.5 km). Parameters of these instruments can be found in another paper (Wang et al., 2013). There are 300m blind area for CFL-03. Vertical profiles of atmospheric temperature and humidity were derived by profiling radiometers. The channel centre frequencies were 22-32 GHz (K-Band) and 51-59 GHz (V-Band). The vertical resolutions were 60 m (0-4 km) and 120 m (4-10 km).

7 2.2 Back trajectory analysis

8 Trajstat, a GIS-based software into which the HYSPLIT (Hybrid Single Particle Lagrangian 9 Integrated Trajectory) model was loaded (Wang et al., 2009), was used to calculate the back 10 trajectory. The model was run every 6 hours in a 24-hour mode back-trajectory mode at 1000 m above 11 sea level from Liulihe site to identify the origins and path way of air mass. The meteorology data used 12 in the mode was obtained from the Global Data Assimilation System (GDAS) model 13 (http://www.ready.noaa.gov/READYamet.php).

14 **2.3 Quantification of regional transport contribution**

A novel technique was used to quantify the contribution of regional transport (Jia et al., 2008). The diurnal trend of $PM_{2.5}$ in Beijing often exhibit "Saw-tooth cycles" with a smoothly increasing or decreasing baseline upon which daily cycles are superimposed. Ancillary measurements around Beijing show that the baselines represent regional aerosols, while the daily cycles represent local aerosols. Following Jia et al. (2008), the total contribution is defined as the area under the concentration line (A_t), while its regional component is defined as the area under the baseline curve (A_r). Both areas are approximated using trapezoid numerical integration as Eq. (1):

22
$$A_N = \sum_{n=1}^{N-1} A_i = \sum_{n=1}^{N-1} \frac{(C_i + C_{i+1})}{2} \times (t_{i+1} - t_i)$$

23 (1)

Where N is the total number of hourly $PM_{2.5}$ concentrations in a specific time period, C_i is total concentration (for A_t) or baseline concentration (for A_r) value at time t_i (i=1, N-1). The baseline concentration curve is the line connecting daily afternoon minimal values. The percentage regional contribution (*R*) is expressed as following Eq. (2):

$$1 \qquad R = \frac{A_r}{A_t} \times 100\%$$

$$2 \qquad (2)$$

3 The uncertainty evaluation mainly includes systematic errors, random errors and sensitivities. The 4 major systematic errors depend on the calibration of instruments for PM2.5 concentration measurement. 5 Minor systematic errors might be from the judging the location and height of the daily minima and the 6 sensitivities analysis suggests these errors are less than 10%. Random errors include data 7 measurement and quantification step, such as identifying the daily minima properly, dealing with days 8 without less-obvious afternoon minima and using linear interpolation between the daily minima. All 9 these errors are evaluate by Jia et al. (2008). As a whole, this technique has an uncertainty of 40%-50% 10 for results of daily regional transport.

11 **3. Results and discussion**

12 **3.1** General characteristics of atmospheric pollution before and during APEC summit

To investigate the changes in air quality during APEC summit, average pollutant concentrations and the rates of changes were calculated. Concentrations of $PM_{2.5}$, SO_2 and NO_2 decreased significantly during the emission control (November 3^{rd} to November 12^{th}) compared to the period before control (October 27^{th} to November 2^{nd}) as shown in Fig. S4 (a). The large rates of reduction were observed for NO_2 (37%) and SO_2 (36%), while the reduction in $PM_{2.5}$ was smaller (21%) but still significant (Fig. S4 (b)).

19 Three pollution episodes were selected to discuss the pollution characteristics during the observation 20 (Fig. S5). Episode 1 (October 27th to November 1st) represents the period before the emission control. Episode 2 (November 2^{nd} to 5^{th}) was the first pollution episode during the emission control. Episode 3 21 (November 6th to 11th) was the second pollution episode during the emission control. PM2.5 22 23 concentration at Miyun site (locate in northern Beijing, shown in Fig. S1, data source: Beijing EPB) is 24 shown in Fig. S5 alongside Luilihe to demonstrate the synchronism of PM_{2.5} levels at different sides in Beijing. At Luilihe, $PM_{2.5}$ concentration was the highest in episode 1 (140±70µg/m³) before 25 26 implementation of emission control, whereas the mean values were close during the last two episodes 27 $(91\pm75\mu g/m^3 \text{ and } 89\pm61\mu g/m^3).$

The average concentration of online non-refractory PM_1 chemical components was shown in Fig. 1. Average concentrations of OM (organic matter), NH_4^+ , SO_4^{2-} and NO_3^- were the highest in episode 1 before emission control. During episode 2, those compounds decreased by 32-60%. In episode 3, the average concentrations remained similar except NH4⁺ which decreased by 12%. HOA (related to
 primary emission), LVOOA and SVOOA were distinguished. Compared with episode1, the HOA,
 LVOOA and SVOOA decreased by 22%, 58% and 28% in episode 2. After that, LVOOA kept
 decreasing by 10% in episode 3 while HOA and SVOOA increased by 39% and 5%.

5 Overall, most meteorological parameters changed little during the three episodes except RH (Fig. 6 S6)). The average ground-level RH (69%) in episode 1 was higher compared with those in episode 2 7 (50%) and in episode 3 (58%). Wind speed remained low during the entire observation. The average 8 wind speed was 0.5m/s, 0.8m/s and 0.7m/s in episode 1, episode 2 and episode 3, respectively. The 9 dominant wind direction was southwest during the 17 days observation. The frequency of southwest 10 wind was above 60% during each of the three episodes, with the highest occurrence of 81% observed 11 during episode 2.

12 The significant reduction in pollutant concentrations during APEC shown above implied that the 13 emission control was effective. However, the general characteristics derived from ground-level 14 observation are insufficient to identify the leading cause of particulate matter pollution, local 15 emissions, regional transport, or both. Furthermore, the significant differences of particle chemical components changes from episode 2 to episode 3 under similar ground-level meteorological 16 17 conditions and local emission intensity suggest different transport or formation mechanisms during 18 those two episodes. Therefore, vertical observations will be used to aid further investigation in each of 19 the three episodes in the following section.

3.2 Characteristics of heavy PM_{2.5} pollution episodes and contribution of regional transport

21 **3.2.1 Pollution process in episode 1**

Episode 1 (October 27^{th} to November 1^{st}) was before emission control. The average concentration of PM_{2.5} reached to $140\mu g/m^3$. The high level of PM_{2.5} is typical in Beijing during the autumn. There were two unique features in this episode. One is the continued increases of PM_{2.5} mass and PM₁ component concentrations during the first four days, with OM showing a more distinct diurnal cycle (Fig. 2, Fig. 3 and Fig. S5). Another is the rapid increase of OM on Oct 29^{th} (Fig. 3). Both suggest except secondary formation, other mechanisms might impact the OM growth and needs further investigation.

Various parameters collected during episode 1 are shown in Fig. 4. Combining the ground-level observation and vertical observation, it is evidenced that the pollution was caused by the regional

transport and pollutants accumulation later. Vertical extinction coefficient data observed at 1 2 Yongledian site (116°47′E, 39°43′N) near Liulihe site were used (Fig. 4(a)), because the optical lidar 3 at Liulihe didn't work in October. High level of PM appeared at approximately 2 km above ground 4 (Fig. 4 (a)) and retained there for 1 day. The air mass came from the southwest where emissions were 5 high (see horizontal wind direction profile, Fig. 4 (c)). Back trajectories also show air mass from southwest arrived in Liulihe, as well as Yongledian (Fig. S7). Then pollutants settled down (see 6 7 downward vertical wind direction in Fig. 4 (b)) and mixed with aerosols on the ground (Fig. 4 (a)). 8 The online particle size distribution also implied the transport process. During the same period (from 9 13:00 to 20:00 on October 28th), a new group of particles appeared and mixed with existing particles, 10 indicating the arrival of aged aerosols (Fig. 4 (e)). As mentioned above, except secondary formation, 11 other mechanisms might impact OM increase. The increase of OM might come from freshly-emitted 12 organic particles and transported to the site instead of aged particles. One evidence is that both HOA and OOA increased significantly. Another is that the OM peak appeared after the transport 13 14 occurrence, much earlier than SNA. It is noticed, even wind direction on the ground changed to north in the early morning on October 29th, it still retained in the southwest above 500m, indicating 15 16 significant influence of regional transport.

17 In the next two days (October 30th to 31st), vertical wind direction was downward which was unfavorable for the pollutants diffusion (Fig. 5(a)). Weather Research & Forecasting Model (WRF) 18 19 modeling results also show the whole region was under control of weak downward wind from late 20 night on October 30th. (Fig. S8, modeling parameters are provided in supplemental 21 information).What's more, both the atmosphere press and wind speed decreased at the same time 22 (Fig. S6). This indicates the site was probably in the rear of cold anticyclone. The steady weather 23 condition promote the pollutants accumulation. Meanwhile, high RH on the surface (Fig. S6) 24 enhanced the formation of SA (secondary aerosol) as pointed out by Pathak et al. (2009). Under this condition, NH_4^+ , SO_4^{2-} and NO_3^- concentrations increased at rates of $0.26\mu g/m^3/h$, $0.21\mu g/m^3/h$, and 25 26 0.58µg/m³/h, respectively. The peak of NH₄⁺, SO₄²⁻ and NO₃⁻ concentrations was two days later than 27 OM. This also proved the organic particles were transported to Beijing and reached to the peak on October 29th and secondary formation became severe later, both of which promoted the pollution 28 29 occurrence.

To quantify the impacts of regional transport, the transport component is calculated with the method introduced in section 2.2. The baseline needs to be defined first especially for pollution end timing. The vertical observation and ground observation were combined to discuss when the pollution ended (see supporting information). The regional component is calculated based on the determination of

baseline. For episode 1, the regional component accounted for 75% of PM_{2.5} mass concentration 1 2 observed at Liulihe site, indicating the important influence of regional transport on the pollution. It 3 can be seen that episode 1 was a pollution episode influenced by transport process in Beijing. RH was 4 high, wind speed was continuously low and wind direction was dominated by southwest in the 5 surface. Vertical observation showed pollutants transported from southwest settled down. OM concentration increased significantly when the transport PM was observed. After that the low wind 6 7 speed, high RH can easily promoted the pollutants accumulation and downward vertical wind was 8 unfavorable for pollutants diffusion.

9 **3.2.2 Pollution process in episode 2**

10 Episode 2 (November 2nd to 5th) saw a lower mean PM_{2.5} concentration (91±75µg/m³) due to the 11 implementation of emission control since November 2nd. Unlike the gradual accumulation of PM observed in episode 1, PM2.5, OM and SNA had a sharp increase from November 4th to 5th. The 12 concentrations of NH4⁺, SO4²⁻ and NO3⁻ increased at rates from the lowest to the highest of 13 14 $0.88 \mu g/m^3/h$, $0.43 \mu g/m^3/h$, and $1.64 \mu g/m^3/h$, respectively, much faster than that in episode 1. OOA 15 also increased much more significantly during this episode. The explosively increases of PM 16 components mainly SA in such a short period of time is contrary to lower RH values in this episode 17 leading to less heterogeneous reaction. Thus, such rapid increases in PM levels could be transport of 18 aged aerosol from other regions, as hypothesized by previous studies where the transport process 19 wasn't observed directly (Yue, et al., 2009; Massling., et al, 2009; Sun et al., 2014; Sun et al., 2016b).

20 With the aid of vertical observation, an in-depth investigation revealed atmospheric processes leading to the peak concentrations during November 4th to 5th. Firstly, after the end of episode 1 at November 21 1st, relatively high PM levels still resided at 1000m (from November 2nd to 3rd) as shown in the 22 vertical extinction coefficient (Fig. 7). Furthermore, a band of high PM centered around 750 m were 23 24 observed ((Fig. S9) on November 3rd at another site (Baoding site, 115°31′E, 38°52′N, shown in Fig. 25 S1) in the BTH region, suggesting a wide-spread PM aloft in the region. During the next two days, the pollutants were transported in the region and the slow winds (average speed of 4.8m/s at 1000 m) 26 27 allowed aerosols ample time to age in their journey. Back trajectories showed transport of air mass from the southwest at the night of November 3rd (Fig. S10), consistent with the vertical wind profile 28 29 observed at Liulihe (Fig.5 (b) and Fig.9). On November 3rd and November 4th, the downward motion 30 of air mass around 1000 m above ground intensified, bringing the aged aerosols down and mixing 31 them with the aerosols on the ground. The well mixed boundary layer with regard to aerosol is 32 evidence in Fig. 8 with a fairly uniform distribution from the ground to 900 m. Consequently,

secondary chemical component concentrations of PM₁ (Fig. 2 and Fig. 3) started ascending with
 remarkably fast rates.

3 Dry and clean air mass from the north arrived in the early morning on November 5th. RH started to 4 increase significantly at 10:00 and wind speed became higher from 12:00. At the same time, $PM_{2.5}$ 5 concentration started to decrease. Based on the analysis, the pollution ended up at 12:00. The 6 calculation shows regional transport contributed 62%, relatively lower than that during episode 1 (Fig. 7 6).

8 Rather than chemical reaction, aged aerosols settled down and had important contribution to the high 9 PM_{2.5} concentration in episode 2. Vertical observations found that the aged aerosol settled down and 10 caused the explosive increase of SNA in such a short time, which can't be explained by the ground-11 level observations. It was also noticed that the high PM_{2.5} level appeared when the emission control 12 just started, which means this episode was partly caused by regional transport before control. Even 13 when local emission control was conducted effectively, the uncontrolled regional emission still led to 14 severe particulate matter pollution in Beijing.

15 **3.2.3 Pollution process in episode 3**

During episode 3 (November 6th to 11th), Luilihe site recorded a relatively high average $PM_{2.5}$ concentration of $89\pm61\mu$ g/m³. Furthermore, this episode is characterized by much more and faster increases in OM concentrations than SNA (Fig. 2 and Fig. 3). Specifically, concentrations of aerosol related with fuel combustion (HOA) increased significantly. While SNA increased slowly (NH₄⁺ and NO₃⁻) or changed little (SO₄²⁻). All of these indicate primary emission rather than the formation of SA was the dominant cause.

Vertical extinction coefficient shows pollutants appeared at 2000-2500m on November 7th. The air mass came from the northwest and the vertical convection bringing them down on November 7th and 8th (Fig. 7, Fig. 5(b) and Fig. 9). Air mass trajectories at 1000 m also show air mass arrived in Beijing from the south on November 7th but changing to the northwest on November 8th (Fig. S11). Because the northwest was less polluted and the effective emission control in BJH region during the APEC, the regional transport of PM was weakened. This is supported by an estimated regional contribution of 53% to PM_{2.5} in Beijing, much lower than in episode 1 (75%) and episode 2 (63%).

Figure 10 depicts black carbon (BC) concentrations measured by Aethalometer and OM
 concentrations measured by ACSM. They tracked each other well during this episode. Concentrations

1 of BC, a marker of vehicular emission in urban settings, had two peaks every day. One was in the 2 early morning and another was after morning rush hour of 10:00-11:00 am. The first peak might result 3 from diesel vehicle emissions (Westerdahl, et al., 2009). This is because transportation of goods to Beijing via heavy-duty diesel vehicles has been permitted at night only, and the number of trucks was 4 5 large. The second peak might be resulted from vehicles from outside coming into Beijing. Vehicles 6 not registered in Beijing are banned to come into Beijing in the rush hour (7:00 am to 9:00 am), which 7 reduces the morning peaks and smoothes the traffic flow. The vehicles coming into Beijing reach a 8 peak after morning rush hour 9 (http://wenku.baidu.com/link?url=SjtPVT1tgo4ON0KDQ5py8ehw1ZAzUr3k0mSd74D3F-

<u>8IOQZPPvedZiro6E5-MOeFFuww7VZjy3XwRqfU-mHXkg0_8kSy5p9FGyokfrFZX0e</u>). As a result,
 a second peak appeared in the late morning at Liulihe site where is close the entrance from Hebei
 Province into Beijing. When the regional emission control was conducted effectively and air mass
 was from relatively clean areas, traffic emissions in and around the city became the dominant source.

14 **4.** Conclusion

This study indicates that the meteorology condition on the ground sometime couldn't explain the air pollution process, especially the air pollutions episodes significantly impacted by regional transport of air pollutants. Vertical observation can provide the vertical meteorological and optical profile, which can help identify the regional transport episodes. Combining the ground-level observation with information from radars, we can determine the regional transport influence on air quality.

20 Three episodes of different types under similar ground meteorological condition were discussed in 21 this study. In episode 1, particle concentration accumulated under the unfavorable meteorological 22 condition after transport occurred. The transport pollutants brought organic aerosol and SNA 23 increased under high RH later. In episode 2, pollutants left from episode 1 was retained in the 24 boundary layer in the region. When vertical wind direction changed to downward, the pollutants were 25 settled down. As a result, OM and SNA increased much explosively. In episode 3, when the control 26 had been conducted for several days, SNA and OA concentration increased much less while HOA and 27 increased significantly. The pollution might be caused by the primary emission from diesel vehicles.

Our research suggests regional transport of air pollutants has significant contribution (up to 70%) to severe secondary particle pollution, even when local emission was controlled effectively (53%, such as in APEC summit). Although lots of efforts were paid to air quality management in Beijing, the

- 1 equal efforts need to be paid to regional emission to ensure the clean air. What's more, diesel vehicle
- 2 emission at night in Beijing might be an important pollution source and needs further investigation.

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1 Figures

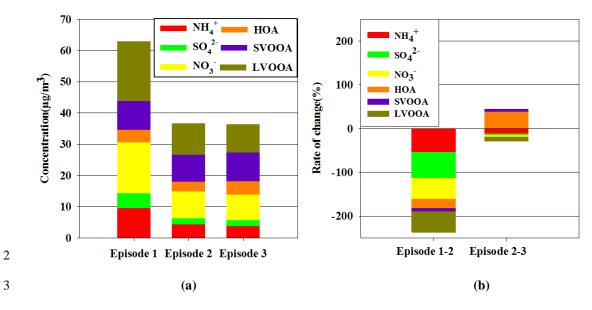


Figure 1. Non-refractory PM₁ chemical components at Liulihe site in the three episodes (a)
 average non-refractory PM₁ chemical components; (b) differences of chemical components

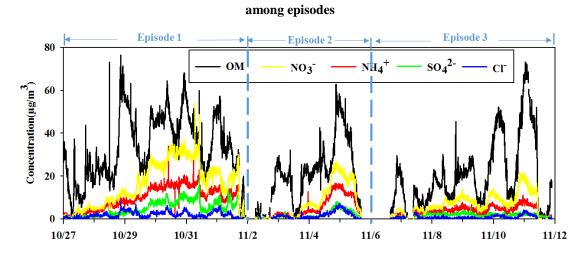


Figure 2. Temporal changes of non-refractory PM1 chemical components at Liulihe site

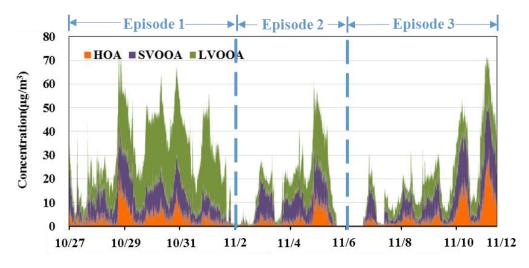


Figure 3. The temporal changes of organic components in PM1 at Liulihe site

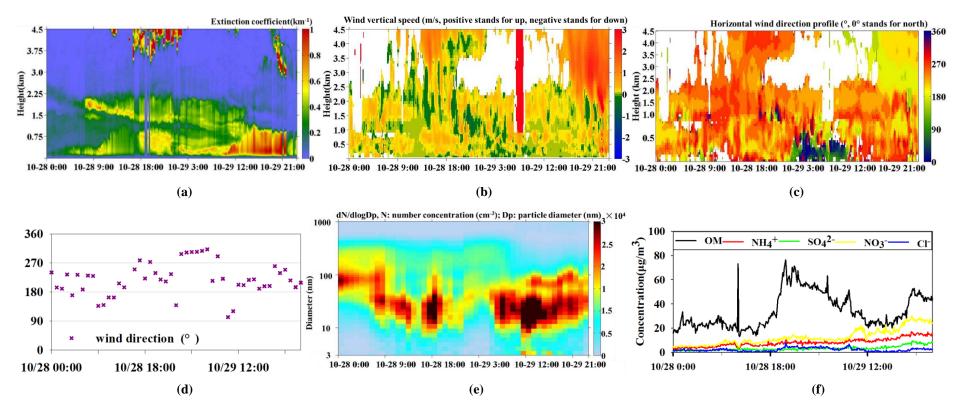
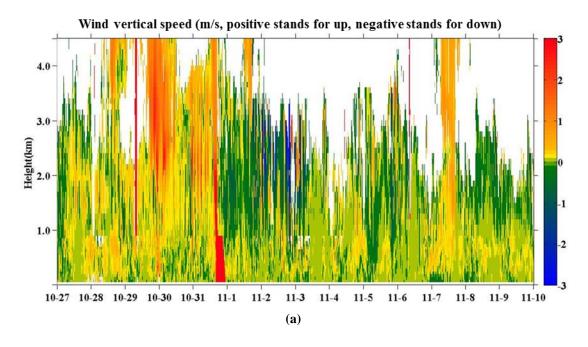
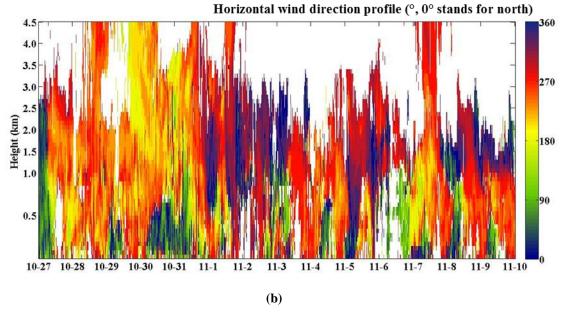


Figure 4. Characteristics of particulate matters and meteorological parameters during episode 1

(a) Vertical profile of extinction coefficient (Yongledian site); (b) Vertical profile of wind vertical direction and speed; (c) Horizontal wind direction profile; (d) wind

direction on the ground; (e) Particle size distribution; (f) NR-PM₁ chemical components







(a) Wind vertical speed; (b) Wind horizontal direction profile

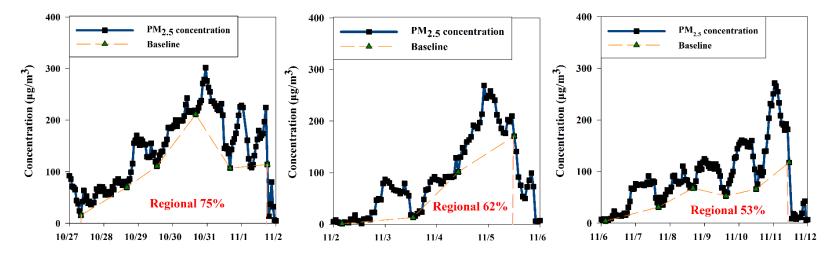


Figure 6. Regional and local components of the three episodes at Liulihe site

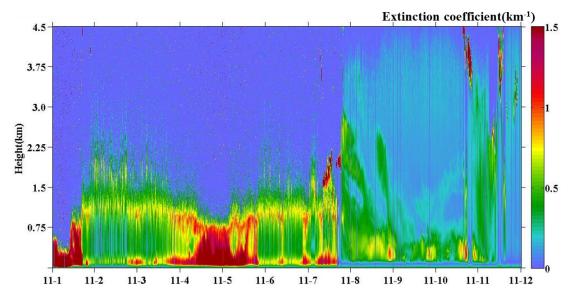


Figure 7. Vertical profile of extinction coefficient at Liulihe site

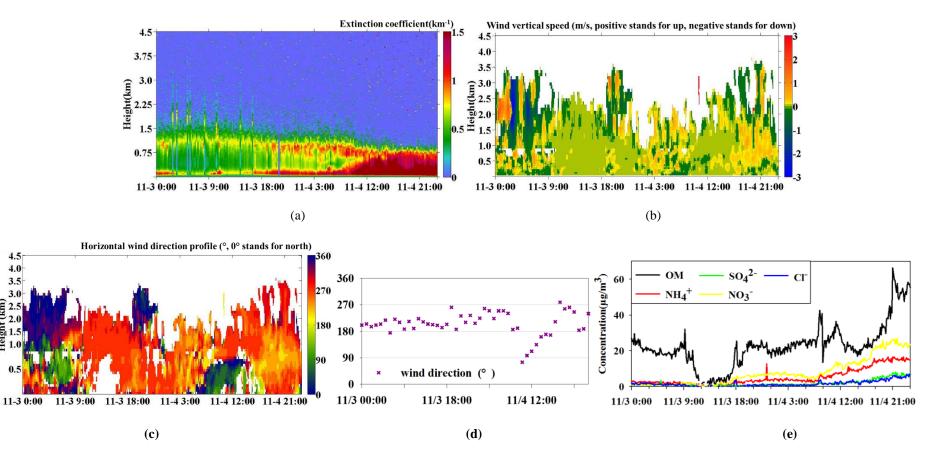


Figure 8. Characteristics of particulate matters and meteorological parameters during episode 2

Height (

(a) Vertical profile of extinction coefficient; (b) Vertical profile of wind vertical direction and speed; (c) Horizontal wind direction profile; (d) wind direction on the

ground; (e) NR-PM₁ chemical components

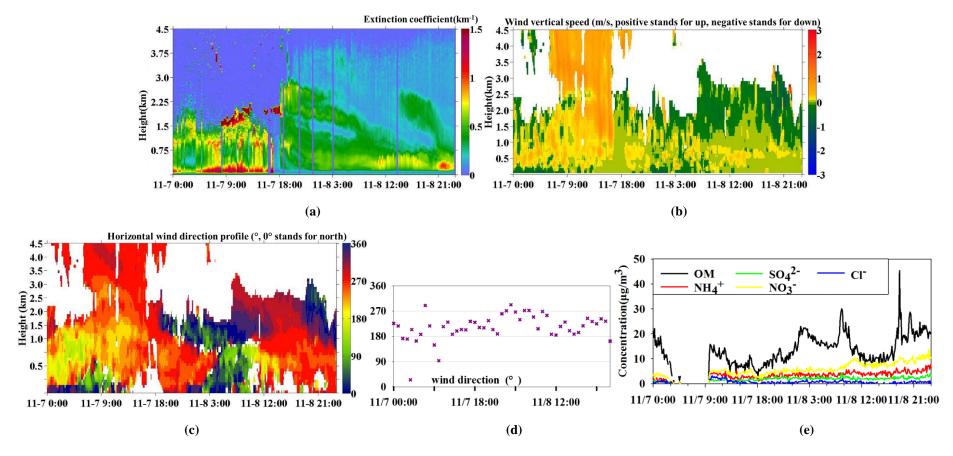


Figure 9. Characteristics of particulate matters and meteorological parameters at Liulihe site during episode 3

(a) Vertical profile of extinction coefficient; (b) Vertical profile of wind vertical direction and speed; (c) Horizontal wind direction profile; (d) wind direction on the ground; (e) NR-PM₁ chemical components

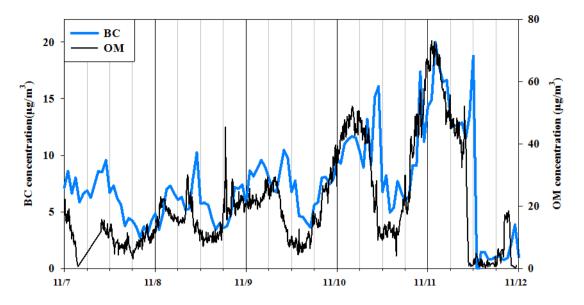


Figure 11. BC and OM concentrations of PM_1 at Liulihe site during episode 3