



- Investigating the impact of regional transport on PM<sub>2.5</sub>
   formation using vertical observation during APEC 2014
   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 20 the transport and formation mechanism of fine particulate matter ( $PM_{2.5}$ ). This study explores the use 21 of vertical observation methods to investigate the influence of regional transport on PM2.5 pollution in 22 Beijing before and during the APEC Summit. Vertical profiles of extinction coefficient, wind, 23 temperature and relative humidity were monitored. Three PM2.5 pollution episodes were analysed. In episode 1 (October 27th to November 1st), regional transport accompanied with the accumulation of 24 25 pollutants under unfavourable meteorological conditions led to the pollution. In episode 2 (November  $2^{nd}$  to  $5^{th}$ ), pollutants left from episode 1 were retained in the boundary layer for 2 days in the region 26 and then settled down to the surface, leading to an explosive increase of PM2.5. The regional transport 27 28 of aged aerosols played a crucial role in the heavy PM<sub>2.5</sub> pollution. In episode 3 (November 6<sup>th</sup> to 11<sup>th</sup>), emission from large point sources had been controlled for several days while primary emissions from 29 30 diesel vehicle might 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 31





- 1 (aerosol optical profile, wind profile, relatively humidity profile and temperature profile) improved
- 2 the understanding of regional transport influence on heavy pollution process. Further vertical
- 3 observations are needed to investigate the pollutants transport especially during the explosive increase
- 4 pollution episode.





## 1 1. Introduction

2	With a rapid economic development and increases in energy consumption, large quantity of emissions
3	has caused serious air pollution in China. Monitoring data show that Beijing-Tianjin-Hebei (BTH)
4	region is one of the most polluted region in China (Zhao et al., 2013; Wang et al., 2014). The region
5	was home to eight out of the top 10 most polluted Chinese cities in 2014 (MEP-Ministry of
6	Environment Protection, 2015). In 2014, the annual average $PM_{2.5}$ (particulate matter with
7	aerodynamic diameter less than 2.5 $\mu m)$ concentration reached 95 $\mu g/m^3$ 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 $\ensuremath{\text{PM}_{2.5}}$
14	in Beijing in 2012-2013. During some severe pollution periods, regional contribution was more than
15	50% (http://www.bjepb.gov.cn/bjepb/413526/331443/331937/333896/396191/index.html). Quite a
16	few researches have studied the causes of heavy polluted episodes in BTH region and show regional
17	transport plays an important role in pollution formation. The sharp $\text{PM}_{2.5}$ 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 investigate the impact of various factors influencing air quality. One of them was APEC (Asia-Pacific 2 3 Economic Cooperation) Economic Leaders' 2014 Summit held in Beijing from November 5th to 11th, 4 2014. A strict air pollution control plan was carried out in the BTH Region to improve air quality in Beijing from November 2<sup>nd</sup> to 11<sup>th</sup> for APEC. According to a conservative estimate by MEP, 5 6 production of 9,289 plants were paused and 3900 plants were running at reduced capacity in six 7 provinces (Beijing, Tianjin, Hebei, Shanxi, Shandong and Inner Mongolia); and more than 40 8 thousand construction down temporally sites were shut (http://www.zhb.gov.cn/gkml/hbb/qt/201411/t20141115\_291482.htm). Other measures include traffic 9 control (50% of private passenger vehicles and 70% of buses were off-road) and frequent road 10 11 sweeping and cleaning in Beijing. More detail emission control measures are supplied in the 12 supporting information. Studies have found that regional emission control effectively reduced air 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 pollution process, thus providing a unique opportunity to investigate the influence of 16 transport events on PM2.5 levels in Beijing.

17 The objective of the study is to investigate the impact of regional transport on  $PM_{2.5}$  in Beijing using 18 both ground-level and vertical observations. Field observation was conducted at a rural site (Liulihe) 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

28 Beijing is surrounded by mountains in the west, north and east directions, which blocks the pollutants

29 from spreading. The open air corridor in the south exposes the capital to air mass passing Hebei

- 30 Province (Fig. S1) a heavily polluted area in China. To investigate the impact of regional transport on
- 31 Beijing, a rural site (Liulihe site, 116°2'E, 39°36'N) was chosen in the southwest of Beijing. It was
- 32 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 4 PM<sub>2.5</sub>/PM<sub>10</sub>, SO<sub>2</sub>, NO<sub>x</sub> and O<sub>3</sub> as well as physical and chemistry properties of PM. PM<sub>2.5</sub>/PM<sub>10</sub> mass concentration was determined by the TEOM method. Particle size distribution from 3nm to 10µm 5 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 (PM<sub>1</sub>) (Ng et al., 2011). The ACSM data was calibrated with a collection efficiency (CE) value to compensate for the particle loss. The CE value of 0.45 recommended by Middlebrook et al. (2012) 12 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 (R<sup>2</sup>=0.91) and 15 the regression slope is 0.43 (Fig. S2). Positive matrix factorization (PMF) with the PMF2.exe algorithm was used to distinguish different components of OA measured by ACSM (Paatero and 16 17 Tapper, 1994). The PMF was performed and evaluated following the PMF analysis guide 18 (http://cires1.colorado.edu/jimenez-group/wiki/index.php/PMF-AMS\_Analysis\_Guide). Three factors 19 were distinguished (Fig. S3), i.e., HOA (hydrocarbon-like organic aerosol), SVOOA (semi volatility oxygenated organic aerosol) and LVOOA (low volatility oxygenated organic aerosol). 20

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 can help figure 26 out the transport direction. Vertical RH profile can provide the RH information at transport layers, 27 thus helping investigate heterogeneous reaction at the layers. Vertical temperature profiles provide 28 information on the stability of and mixing in the boundary layer. Lidar was used to observe the 29 vertical optical properties of atmospheric aerosols at Liulihe site. The lidar consists of three parts, 30 including emitting system, receiving system and signal analogue system (Chen et al., 2015). The laser 31 source emitted pulse at 355/532nm. The pulse energy is 30MJ at 355nm and 20MJ at 532nm. The 32 pulse repetition is 20Hz. The telescope for receiving system is based on a Cassegrain design. 33 Diameter of the telescope is 200mm with a vertical resolution of 7.5m. The particle backscatter





- 1 coefficient and extinction coefficient was retrieved by Fernald method (Fernald et al., 1984). CFL-03
- 2 phased array wind profile radar was used to monitor the vertical wind speed and direction with
- 3 resolutions of 50 m (0-1 km) and 100 m (1-5.5 km). Parameters of these instruments can be found in
- 4 another paper (Wang et al., 2013). Vertical profiles of atmospheric temperature and humidity were
- 5 derived by profiling radiometers. The channel centre frequencies were 22-32 GHz (K-Band) and 51-
- 6~~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 trajectory. 10 The model was run every 6 hours in a 24-hour mode back-trajectory mode at 1000 m above sea level 11 from Liulihe site to identify the origins and path way of air mass. The meteorology data used in the 12 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. The diurnal trend of PM<sub>2.5</sub> 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<sub>t</sub>), while its regional component is defined as the area under the baseline curve (A<sub>r</sub>). Both areas are approximated using trapezoid numerical integration as Eq. (1):

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

Where N is the total number of hourly  $PM_{2.5}$  concentrations in a specific time period, C<sub>i</sub> is total concentration (for A<sub>t</sub>) or baseline concentration (for A<sub>r</sub>) value at time t<sub>i</sub> (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):

$$27 \qquad \mathbf{R} = \frac{A_r}{A_t} \times 100\% \tag{2}$$





### 1 3. Results and discussion

## 2 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. Period 1 (October  $27^{th}$  to November  $2^{nd}$ ) and period 2 (November  $3^{rd}$  to November  $12^{th}$ ) were defined to represent the periods before and during the APEC summit. Concentrations of PM<sub>2.5</sub>, SO<sub>2</sub> and NO<sub>2</sub> decreased significantly during the emission control (Period 2) compared to before control (Period 1) as shown in Fig. S4 (a). The large rates of reduction were observed for NO<sub>2</sub> (37%) and SO<sub>2</sub> (36%), while the reduction in PM<sub>2.5</sub> was smaller (21%) but still significant (Fig. S4 (b)).

10 Three pollution episodes were selected to discuss the pollution characteristics during the observation 11 (Fig. S5). PM<sub>2.5</sub> concentration at Miyun site (locate in northern Beijing, shown in Fig. S1, data source: 12 Beijing EPB) is shown in Fig. S5 alongside Luilihe to demonstrate the synchronism of PM<sub>2.5</sub> levels at different sides in Beijing. Episode 1 (October 27th to November 1st) represents the period before the 13 14 emission control. Episode 2 (November 2<sup>nd</sup> to 5<sup>th</sup>) was the first pollution episode during the emission control plan. Episode 3 (November 6th to 11th) was the second pollution episode during the emission 15 control plan. At Luilihe, PM2.5 concentration was the highest in episode 1 (140±70µg/m3) before 16 17 implementation of emission control, whereas the mean values were close during the last two episodes 18  $(91\pm75\mu g/m^3 \text{ and } 89\pm61\mu g/m^3).$ 

The average concentration of online  $PM_1$  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  $NH_4^+$  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%.

Overall, most meteorological parameters changed little during the three episodes except RH (Fig. S6)). The average ground-level RH (69%) in episode 1 was higher compared with those in episode 2 (50%) and in episode 3 (58%). Wind speed remained low during the entire observation. The average wind speed was 0.5m/s, 0.8m/s and 0.7m/s in episode 1, episode 2 and episode 3, respectively. The dominant wind direction was southwest during the 10-day observation. The frequency of southwest





- 1 wind was above 60% during each of the three episodes, with the highest occurrence of 81% observed
- 2 during episode 2.

3 The significant reduction in pollutant concentrations during APEC shown above implied that the 4 emission control was effective. However, the general characteristics derived from ground-level 5 observation are insufficient to identify the leading cause of air pollution, local emissions, regional 6 transport, or both. Furthermore, the significant differences of particle chemical components changes 7 from episode 2 to episode 3 under similar ground-level meteorological conditions and local emission 8 intensity suggest different transport or formation mechanisms during those two episodes. Therefore, 9 vertical observations will be used to aid further investigation in each of the three episodes in the 10 following section.

### 11 3.2 Characteristics of heavy PM<sub>2.5</sub> pollution episodes and contribution of regional transport

#### 12 3.2.1 Pollution process in episode 1

Episode 1 (October  $27^{th}$  to November  $1^{st}$ ) was before emission control. The high level of PM<sub>2.5</sub> is typical in Beijing during the autumn. There were two unique features in this episode. One is the continued increases of PM<sub>2.5</sub> mass and PM<sub>1</sub> 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.

19 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 20 21 transport and pollutants accumulation later. Vertical extinction coefficient data observed at 22 Yongledian site (116°47′E, 39°43′N) near Liulihe site were used (Fig. 4(a)), because the optical lidar 23 at Liulihe didn't work in October. High level of PM appeared at approximately 2 km above ground (Fig. 4 (a)) and retained there for 1 day. The air mass came from the southwest where emissions were 24 25 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 26 27 downward vertical wind direction in Fig. 4 (b)) and mixed with aerosols on the ground (Fig. 4 (a)). 28 The online particle size distribution also implied the transport process. During the same period (from 29 13:00 to 20:00 on October 28th), a new group of particles appeared and mixed with existing particles, 30 indicating the arrival of aged aerosols (Fig. 4 (e)). As mentioned above, except secondary formation, 31 other mechanisms might impact OM increase. The increase of OM might come from freshly-emitted





1 organic particles and transported to the site instead of aged particles. One evidence is that both HOA

2 and OOA increased significantly. Another is that the OM peak appeared after the transport occurrence,

3 much earlier than SNA. It is noticed, even wind direction on the ground changed to north in the early

4 morning on October 29<sup>th</sup>, it still kept in the southwest above 500m, indicating significant influence of

5 regional transport.

6 In the next two days (October 30th to 31st), vertical wind direction was downward and pollutants were 7 easily accumulated in the boundary layer (Fig. 5). Meanwhile, high RH on the surface (Fig. S6) 8 enhanced the formation of SA (secondary aerosol) as pointed out by Pathak et al. (2009). Under this condition, NH<sub>4</sub><sup>+</sup>, SO<sub>4</sub><sup>2-</sup> and NO<sub>3</sub><sup>-</sup> concentrations increased at rates of 0.26µg/m<sup>3</sup>/h, 0.21µg/m<sup>3</sup>/h, and 9 10  $0.58 \mu g/m^3/h$ , respectively. The peak of NH<sub>4</sub><sup>+</sup>, SO<sub>4</sub><sup>2-</sup> and NO<sub>3</sub><sup>-</sup> concentrations was two days later than 11 OM. This also proved the organic particles were transported to Beijing and reached to the peak on 12 October 29th and secondary formation became severe later, both of which promoted the pollution 13 occurrence.

14 To quantify the impacts of regional transport, the transport component is calculated with the method 15 introduced in section 2.2. The baseline needs to be defined first especially for pollution end timing. 16 Here the vertical observation and ground observation were combined to discuss when the pollution ended. In the morning on 1st November, air mass from the north above 1000 m arrived Beijing. The 17 18 vertical temperature gradient decreased and vertical mixing became weak (wind vertical speed was 19 very low). Consequently, PM2.5 accumulated and had a sharp increase. Then clean and cold wind from 20 north caused sharp increase of wind speed and decrease of atmosphere pressure. Based on the analysis 21 above, pollution ended up at 18:00 when the week temperature ended and PM<sub>2.5</sub> decreased sharply 22 (Fig. 6). The regional component is calculated based on the determination of baseline.

For episode 1, the regional component accounted for 75%, indicating the important influence of regional transport on the pollution. It can be seen that episode 1 was a pollution episode influenced by transport process in Beijing. RH was high, wind speed kept low and wind direction was dominated by southwest in the surface. Vertical observation showed pollutants transported from southwest settled down. OM concentration increased significantly when the transport PM was observed. After that vertical wind direction kept downward and promoted the pollutants accumulation, especially SNA.

### 29 **3.2.2 Pollution process in episode 2**

Episode 2 (November  $2^{nd}$  to  $5^{th}$ ) saw a lower mean PM<sub>2.5</sub> concentration (91±75µg/m<sup>3</sup>) due to the implementation of emission control since November  $2^{nd}$ . 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 1 concentrations of NH4+, SO42- and NO3- increased at rates from the lowest to the highest of 2 3 0.88µg/m3/h, 0.43µg/m3/h, and 1.64µg/m3/h, respectively, much faster than that in episode 1. OOA 4 also increased much more significantly during this episode. The explosively increases of PM 5 components mainly SA in such a short period of time is contrary to lower RH values in this episode 6 leading to less heterogeneous reaction. Thus, such rapid increases in PM levels could be transport of 7 aged aerosol from other regions, as hypothesized by previous studies where the transport process 8 wasn't observed directly (Yue, et al., 2009; Massling., et al, 2009; Sun et al., 2014; Sun et al., 2016b).

9 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 10 1st, relatively high PM levels still resided at 1000m (from November 2nd to 3rd) as shown in the 11 12 vertical extinction coefficient (Fig. 7). Furthermore, a band of high PM centered around 750 m were observed ((Fig. S8) on November 3rd at another site (Baoding site, 115°31'E, 38°52'N, shown in Fig. 13 14 S1) in the BTH region, suggesting a wide-spread PM aloft in the region. During the next two days, the 15 pollutants were transported in the region and the slow winds (average speed of 4.8m/s at 1000 m) allowed aerosols ample time to age in their journey. Back trajectories showed transport of air mass 16 17 from the southwest at the night of November 3<sup>rd</sup> (Fig. S9), consistent with the vertical wind profile 18 observed at Liulihe (Fig. 8 and Fig.9). On November 4th, the downward motion of air mass around 19 1000 m above ground intensified, bringing the aged aerosols down and mixing them with the aerosols 20 on the ground. The well mixed boundary layer with regard to aerosol is evidence in Fig. 9 with a 21 fairly uniform distribution from the ground to 900 m. Consequently, secondary chemical component 22 concentrations of PM1 (Fig. 2 and Fig. 3) started ascending with remarkably fast rates.

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

Rather than chemical reaction, aged aerosols settled down and had important contribution to the pollution in episode 2. Vertical observations found that the aged aerosol settled down and caused the explosive increase of SNA in such a short time, which can't be explained by the ground-level observations. It was also noticed that the pollution occurred when the emission control plan just started, which means this episode was partly caused by regional transport before control. Even when





- 1 local emission control was conducted effectively, the uncontrolled regional emission still led to severe
- 2 pollution in Beijing.

### 3 3.2.3 Pollution process in episode 3

4 During episode 3 (November 6<sup>th</sup> to 11<sup>th</sup>), Luilihe site recorded a relatively high average  $PM_{2.5}$ 5 concentration of  $89\pm61\mu$ g/m<sup>3</sup>. Furthermore, this episode is characterized by much more and faster 6 increases in OM concentrations than SNA (Fig. 2 and Fig. 3). Specifically, concentrations of aerosol 7 related with fuel combustion (HOA) increased significantly. While SNA increased slowly (NH<sub>4</sub><sup>+</sup> and 8 NO<sub>3</sub><sup>-</sup>) or changed little (SO<sub>4</sub><sup>2-</sup>). All of these indicate primary emission rather than the formation of SA 9 was the dominant cause.

10 Vertical extinction coefficient shows pollutants appeared at 2000-2500m on November 7<sup>th</sup>. The air 11 mass came from the northwest and the vertical convection bringing them down on November 7<sup>th</sup> and 12 8<sup>th</sup> (Fig. 7, Fig. 8 and Fig. 10). Air mass trajectories at 1000 m also show air mass arrived in Beijing 13 from the south on November 7<sup>th</sup> but changing to the northwest on November 8<sup>th</sup> (Fig. S10). Because 14 the northwest was less polluted and the effective emission control in BJH region during the APEC, 15 the regional transport of PM was weakened. This is supported by an estimated regional contribution 16 of 53% to PM<sub>2.5</sub> in Beijing, much lower than in episode 1 (75%) and episode 2 (63%).

17 Figure 11 depicts black carbon (BC) concentrations measured by Aethalometer and OM concentrations measured by ACSM. They tracked each other well during this episode. Concentrations 18 19 of BC, a marker of vehicular emission in urban settings, had two peaks every day. One was in the 20 early morning and another was in the morning rush hour of 9:00am. The first peak might result from 21 diesel vehicle emissions (Westerdahl, et al., 2009). This is because transportation of goods to Beijing 22 via heavy-duty diesel vehicles has been permitted at night only, and the number of trucks was large. 23 When the regional emission control was conducted effectively and air mass was from relatively clean 24 areas, traffic emissions in and around the city became the dominant source.

## 25 4. Conclusion

This study indicates that the meteorology condition on the ground sometime couldn't explain the pollution process, especially the pollutions impacted by transport significantly. 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.





1 Three episodes of different types under similar ground meteorological condition were discussed in 2 this study. In episode 1, particle concentration accumulated under the unfavorable meteorological 3 condition after transport occurred. The transport pollutants brought organic aerosol and SNA 4 increased under high RH later. In episode 2, pollutants left from episode 1 kept in the boundary layer 5 in the region. When vertical wind direction changed to downward, the pollutants were settled down. 6 As a result, OM and SNA increased much explosively. In episode 3, when control plan had been 7 conducted for several days, SNA and OA concentration increased much less while HOA and 8 increased significantly. The pollution might be caused by the primary emission from diesel vehicles.

9 Our research suggests regional transport of air pollutants has significant contribution (up to 70%) to 10 severe secondary particle pollution, even when local emission was controlled effectively (53%, such 11 as in APEC summit). Although lots of efforts were paid to air quality management in Beijing, the 12 equal efforts need to be paid to regional emission to ensure the clean air. What's more, diesel vehicle 13 emission at night in Beijing might be an important pollution source and needs further investigation.

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1 Figures



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average  $PM_1$  chemical components; (b) change rates in chemical components





Figure 2. PM1 chemical components during the observation at Liulihe site

















(a) Vertical profile of extinction coefficient (km<sup>-1</sup>) (Yongledian site); (b) Vertical profile of wind vertical direction and speed (m/s, positive stands for up, negative stands for down);(c) Horizontal wind direction profile (°, 0° stands for north); (d) wind direction on the ground; (e) Particle size distribution (dN/dlogDp, N: number concentration (cm<sup>-3</sup>); Dp: particle diameter (nm)); (f) PM1 chemical components.







Figure 5. Vertical profile of wind vertical direction and speed (m/s, positive stands for up and negative stands for down) during the observation time at Liulihe site









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Figure 7. Vertical profile of extinction coefficient during the observation at Liulihe site



Figure 8. Vertical profile of wind horizontal direction during the observation at Liulihe site







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Figure 9. Parameters of particles and meteorology at Liulihe site during episode 2

(a) Vertical profile of extinction coefficient (km<sup>-1</sup>); (b) Vertical profile of wind vertical direction and speed (m/s, positive stands for up, negative stands for down);(c)

Horizontal wind direction profile ( °, 0 ° stands for north); (d) wind direction on the ground; (e) PM1 chemical components.

















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(a) Vertical profile of extinction coefficient (km<sup>-1</sup>); (b) Vertical profile of wind vertical direction and speed (m/s, positive stands for up, negative stands for down);(c)

Horizontal wind direction profile (  $, 0 \,^\circ$  stands for north); (d) wind direction on the ground; (e) PM1 chemical components.



Figure 11. BC and OM concentrations of PM1 at Liulihe site during episode 3