Anonymous Referee #1

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This manuscript presents observations of air quality and meteorological parameters including ground level data and vertical profiles. Those measurements were used to analyze emissions and atmospheric processes that had significant impacts on $PM_{2.5}$ air quality during the study period. The topic is relevant to ACP. The approach and applied methods are largely valid. The scientific contribution is good. However, the presentation has room for improvement. My comments and suggestions are listed below.

Response: We thank the referee for the comments which help us improve the quality of our manuscript. We address the reviewer's comments below. The original comments are in blue and our responses are in black.

1. Abstract. The last sentence is unclear, "Further vertical observations are needed to investigate the pollutants transport especially during the explosive increase pollution episode." 1) if you recommend that others should use vertical methods, it could read: Future studies may consider including vertical observations to aid investigation of pollutant transport especially during episodic events of rapidly increasing concentrations, or 2) if you believe that what you have done in this study is not enough, it could read: Vertical observations beyond those explored in this study may be necessary to investigate pollutant transport, especially during episodic events of rapidly increasing concentrations.

Response: This sentence is to recommend that others should use vertical methods. We have corrected as the reviewer suggested.

2. Section 3.1, "Period 1 (October 27th to November 2nd) and period 2 (November 3rd to November 12th) were defined to represent the periods before and during the APEC summit." Given that "Three pollution episodes were selected to discuss the pollution characteristics during the observation (Fig. S3): Episode 1 (October 27th to November 1st) represents the period before the emission control. Episode 2 (November 2nd to 5th) was the first pollution episode during the emission control plan. Episode 3 (November 6th to 11th) was the second pollution episode during the emission control plan." (pg 7, L10) and "Summit held in Beijing from November 5th to 11th, 2014. A strict air pollution control plan was carried out in the BTH Region to

improve air quality in Beijing from November 2nd to 11th for APEC" (pg 4, L3), the selection of Period 1 and Period 2 seem to be rather arbitrary and confusing. I suggest using "Episode 1 and Episodes 2 and 3 combined" and define the three episodes at the beginning of section 3.1.

Response: First, the control plan started from November 3rd instead of November 2nd. We apologize for this typo. To address the comment by the reviewer, we removed the definition of "period 1" and "period 2" from the manuscript and only discuss the characteristics changes before and after control. As suggested by the reviewer, we defined the three episodes at the beginning of section 3.1.

3. Pg 8, L13, "The high level of $PM_{2.5}$ is typical in Beijing during the autumn." Please provide the average $PM_{2.5}$ concentration during this episode.

Response: The average concentration of $PM_{2.5}$ reached to 140μg/m^3. It has been added in the article as the reviewer suggested.

4. Pg 9, L14, this paragraph seems to be less relevant; it could be better placed in the Supplemental Information.

Response: We agree. It has been placed in the Supplemental Information.

5. Pg 9, L23," For episode 1, the regional component accounted for 75%", did you mean, "For episode 1, the regional component accounted for 75% of $PM_{2.5}$ mass concentration observed in Beijing"?

Response: yes, to be more accurate, we mean the regional component accounted for 75% of $PM_{2.5}$ mass concentration observed at Liulihe site. We have corrected in the article.

6. Pg 9, L27, "After that vertical wind direction kept downward and promoted the pollutants accumulation, especially SNA." It is uncommon to have a prolonged period and/or a wide spread of downward winds that will result in great changes in atmospheric pressure. The authors may need to provide more data to support this claim or clarify where the downward winds were and for how long.

Response: Both the atmosphere pressure and wind speed decreased from October 30th to November 1st (as shown in Figure R1). This indicates that Liulihe site was probably in the rear of cold anticyclone. The steady weather conditions promoted the accumulation of air pollutants. Meanwhile, the vertical downward wind was unfavorable for pollutants dispersion. Weather Research & Forecasting Model (WRF) modeling results also show the whole region was under control of weak downward wind from late night on October 30th. (Figure R2, modeling parameters are provided in supplemental information). As a whole, the pollutants were easily accumulated. We agree that "After that vertical wind direction kept downward and promoted the pollutants accumulation, especially SNA" is a little arbitrary. So we have corrected this statement as well as the discussion in page 9, line 7. The atmosphere pressure figure and regional wind vertical speed figure have been added to the supplemental information.

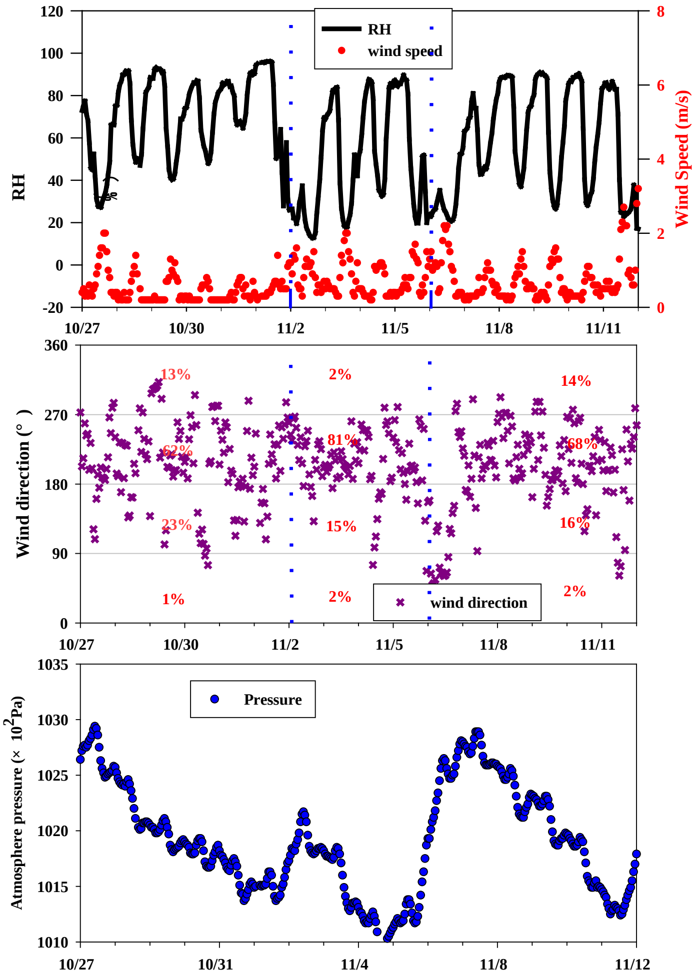

Figure R1 Meteorology conditions on the ground during the observation at Liulihe site

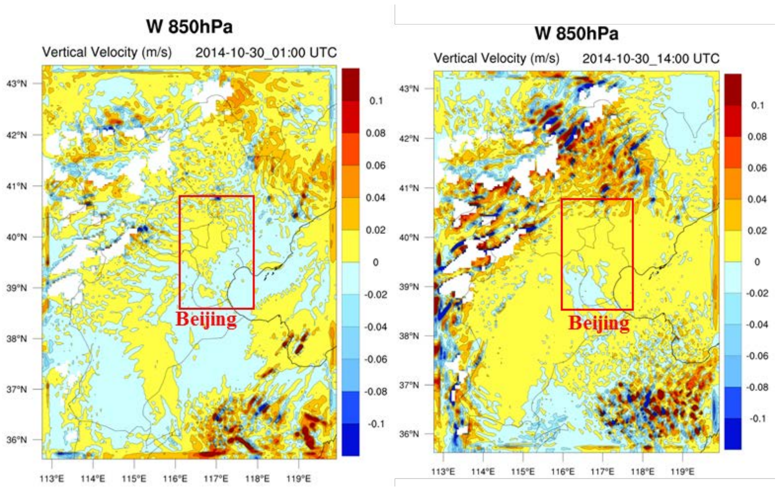

Figure R2 Regional wind vertical speed

7. Pg 11, L18, "Concentrations of BC, a marker of vehicular emission in urban settings, had two peaks every day. One was in the early morning and another was in the morning rush hour of 9:00am." The second peak in BC concentrations (blue line in Fig 11) seems to be near noon, please clarify.

Response: we check the data and confirm that the second peak is actually at 10:00-11:00am. The second peak might be resulted from vehicles from outside coming into Beijing. Vehicles not registered in Beijing are banned to come into Beijing in the rush hour (7:00 am to 9:00 am), which reduces the morning peaks and smoothes the traffic flow. The vehicles coming into Beijing reach a peak after morning rush hour (http://wenku.baidu.com/link?url=SjtPVT1tgo4ON0KDQ5py8ehw1ZAzUr3k0mSd74D3F-8lOQZPPvedZiro6E5-MOeFFuww7VZjy3XwRqfU-mHXkg0_8kSy5p9FGyokfrFZX0e). As a result, a second peak appeared in the late morning at Liulihe site where is close_the entrance from Hebei Province into Beijing. We have corrected the discussion as suggested.

8. References were missing at times, e.g. pg 6, L9, HYSPLIT.

Response: We check all the references in the article and corrected/added the following references.

Add missing reference: Tang, G., Zhu, X., Hu, B., Xin, J., Wang, L., Münkel, C., Mao, G. and Wang, Y. (2015) Impact of emission controls on air quality in Beijing during APEC 2014: lidar ceilometer observations. Atmospheric Chemistry and Physics 15(21), 12667-12680.

Adjust reference order: Ng, N.L., Herndon, S.C., Trimborn, A., Canagaratna, M.R., Croteau, P.L., Onasch, T.B., Sueper, D., Worsnop, D.R., Zhang, Q., Sun, Y.L. and Jayne, J.T. (2011) An Aerosol Chemical Speciation Monitor (ACSM) for Routine Monitoring of the Composition and Mass Concentrations of Ambient Aerosol. Aerosol Science and Technology 45(7), 780-794.

Correct the author name of citing reference (page 6, line 1) from "Fernald, 1984" to "Frederick, 1984".

The reference for HYSPLIT and Trajstat is "Wang, Y., Zhang, X. and Draxler, R.R. (2009) TrajStat: GIS-based software that uses various trajectory statistical analysis methods to identify potential sources from long-term air pollution measurement data. Environmental Modelling & Software 24(8), 938-939."

9. Figs 9e & 10e, legends seem to be missing.

Response: we have added the legends for the figures.

10. The use of English language is largely satisfactory. However, there are quite a few awkward sentences and word choices, some examples are listed below:

Response: We have carefully checked the article and polished the sentences. In addition, a copy-editing team will further help to improve the language after the manuscript is accepted for publication at ACP.

1) Pg 5, L25, it could read, "Vertical wind profiles indicate the transport direction. Vertical RH profiles reflect the strength of heterogeneous reaction at different layers."

Response: we have corrected as the suggested.

2) Pg 7, L30, "10-day observation", the entire observation seems to be either 16 days based on the three episodes (pg 7) or 17 days ("The field campaign was conducted from October 27th to November 12th, 2014." pg 5, L1).

Response: we have corrected the description of observation days.

3) Pg 10, L23, it could read: "Dry and clean air mass from the north arrived in…"

Response: we have corrected as the suggestions.

4) The word "plan" could be omitted in "control plan" after the Introduction.

Response: we have corrected as the suggestions.

5) The use of the word "pollution" is unconventional at times, e.g. "It was also noticed that the pollution occurred when the emission control plan just started," (pg 10, L31).

Large cities like Beijing would not be free from air pollution, however, the extent of air pollution may vary. Please clarify the meaning of "pollution".

Response: we check the article and clarify all the pollution might not be clear.

- Correct "large quantity of emissions has caused serious air pollution in China" (page 3, line 3) to "large quantity of emissions has caused serious particulate matter pollution in China."
- Correct "The significantly reduced local emissions led to reduced complexity of pollution process" (page 4, line 15) to "The significantly reduced local emissions led to reduced complexity of particulate matter pollution process".
- Correct "However, the general characteristics derived from ground-level observation are insufficient to identify the leading cause of air pollution, local emissions, regional transport, or both." (page 8, line 4) to "However, the general characteristics derived from ground-level observation are insufficient to identify the leading cause of particulate matter pollution, local emissions, regional transport, or both.".
- Replace the word "pollution" in the sentence "Rather than chemical reaction, aged aerosols settled down and had important contribution to the pollution in episode 2." (page 10, line 24) to "high $PM_{2.5}$ concentration".
- Correct the sentence "Even when local emission control was conducted effectively, the uncontrolled regional emission still led to severe pollution in Beijing." (page 10, line 29) to "Even when local emission control was conducted effectively, the uncontrolled regional emission still led to severe particulate matter pollution in Beijing.".
- Correct the sentence "This study indicates that the meteorology condition on the ground sometime couldn't explain the pollution process, especially the pollutions impacted by transport significantly." (page 11, line 27) to "This

study indicates that the meteorology condition on the ground sometime couldn’t explain the air pollution process, especially the air pollution episodes significantly impacted by regional transport of air pollutants”.

6) Throughout the manuscript, the word “kept” could be replaced by, for example, “continuously”, or “was retained”.

Response: we check the word in the article and replace some of them.

- We replace the “kept” in the sentence “it still kept in the southwest above 500m, indicating significant influence of regional transport.” (page 9, line 5) to “was retained”.
- We replace the “kept” in the sentence “RH was high, wind speed kept low and wind direction was dominated by southwest in the surface.” (page 9, line 22) to “was continuously”.
- We replace the “kept” in the sentence “In episode 2, pollutants left from episode 1 kept in the boundary layer in the region.” (page 12, line 4) to “was retained”.

The manuscript by Hua et al. investigated the role of regional transport on the formation of PM pollution in Beijing by combining ground and vertical measurements during a unique period (APEC). The results showed that regional transport played a major role in PM pollution but varied substantially among different episodes. In addition, the impact of emission control on PM reduction was also discussed. This study provide new insights into the formation of PM pollution from the view of vertical measurements, and the conclusions showed that vertical measurements are of critical to explain the ground observations. This manuscript falls within the scope of ACP. I recommend it for publication after addressing the following comments.

We thank the referee for supporting the publication. We will put more efforts to improve the quality of our manuscript. We address the reviewer's comments below. The original comments are in blue and our responses are in black.

1. Abstract: it is better to mention the exact location for the ground and vertical measurements.

Response: the location has been added in the abstract.

2. Page 2, line 1: suggest using "aerosol optical properties, winds, relative humidity, and temperature"

Response: we have corrected as suggested.

3. Page 5, line 19: change "semi volatility" to "semi volatile"

Response: we have corrected as suggested.

4. Page 6, line 3: no blind area for CFL-03?

Response: There are 300m blind area for CFL-03. We have added it to the article.

5. Page 6, line 15: Cite Jia et al. (2008) where this technique was developed, not in this study. In addition, the approach in Jia et al. (2008) might have large uncertainties in determining the baseline (the lowest points) because of multiple influences from local emissions, regional transport, and secondary processes, I suggest the authors adding several sentences to discuss the uncertainties in quantification of the contributions of regional transport.

Response: we had cited in page 6, line 19 in the original manuscript. We have cited in line 15 as suggested.

The uncertainties discussion is also added to the manuscript. The uncertainty evaluation mainly includes systematic errors, random errors and sensitivities. The major systematic errors depend on the calibration of instruments for $PM_{2.5}$ concentration measurement. Minor systematic errors might be from the judging the location and height of the daily minima and the sensitivities analysis suggests these errors are less than 10%. Random errors include data measurement and quantification step, such as identifying the daily minima properly, dealing with days without less-obvious afternoon minima and using linear interpolation between the daily minima. All these errors are evaluate by Jia et al. (2008). As a whole, this technique has an uncertainty of 40%-50% for results of daily regional transport.

6. Page 7, line 19: It is not appropriate to call "PM1 chemical components" because BC was not included. Either use non-refractory PM1 or adding BC in Figure 1.

Response: we correct the "PM_1 chemical components" to "non-refractory PM_1 chemical components".

7. Please mark the three episodes in Figures 2 and 3, or explain the vertical dash lines in the captions.

Response: we have corrected the figures as suggested.

8. Please try to combine Figures 4, 9 and 10 in one page for easy reading. Also, add the units for the color bars.

Response: we have corrected the figures as suggested.

9. The colors of chemical species in the figures should be synchronized, e.g., Figure 1 vs. Figure 2, otherwise, it is very confusing.

Response: we have corrected the figures as suggested.

10. Figure 4: change "wind vertical direction and wind speed" to "wind vertical speed". The wind direction of "up" and "down" was already mentioned in the notes. Same as in Figure 5 and Figure 9.

Response: we have corrected the figures as suggested.

11. Suggest combining Figure 5 and Figure 8 together for easy reading and comparisons.

Response: we have corrected the figures as suggested.

MANUSCRIPT

Investigating the impact of regional transport on $PM_{2.5}$ formation using vertical observation during APEC 2014 Summit in Beijing

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ABSTRACT

During APEC (Asia-Pacific Economic Cooperation) Economic Leaders' 2014 Summit in Beijing, 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 of vertical observation methods to investigate the influence of regional transport on $PM_{2.5}$ pollution in Beijing before and during the APEC Summit. Vertical profiles of extinction coefficient, wind, 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), 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 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 crucial role in the heavy $PM_{2.5}$ pollution. In episode 3 (November 6th to 11th), emission from large point sources had been controlled for several days while primary emissions from 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 (aerosol optical ~~profile~~properties, wind ~~profile~~s, relatively humidity ~~profile~~ and temperature ~~profile~~) 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.~~Further vertical observations are needed to investigate the pollutants transport especially during the explosive increase pollution episode.~~

1. Introduction

With a rapid economic development and increases in energy consumption, large quantity of emissions has caused serious particulate matterair pollution in China. Monitoring data show that Beijing-Tianjin-Hebei (BTH) region is one of the most polluted region in China (Zhao et al., 2013; Wang et al., 2014). The region was home to eight out of the top 10 most polluted Chinese cities in 2014 (MEP-Ministry of Environment Protection, 2015). In 2014, the annual average $PM_{2.5}$ (particulate matter with aerodynamic diameter less than 2.5 μm) concentration reached 95 μg/m^3 in the BTH region. With 21.5 million residents and 5.3 million vehicles, Beijing has been burdened with severe pollution episodes frequently in recent years (Beijing Municipal Bureau of Statistics, 2014). The capital is surrounded by mountains in three directions (north, west and east). The top three most polluted cities in China (Baoding, Xingtai and Shijiazhuang) are located in the south to Beijing. Polluted air mass from the south contributes to $PM_{2.5}$ pollution in Beijing (Wang et al., 2015). Source apportionment by Beijing Environmental Protection Bureau indicates regional transport contributed 28%-36% to $PM_{2.5}$ 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 few researches have studied the causes of heavy polluted episodes in BTH region and show regional transport plays an important role in pollution formation. The sharp $PM_{2.5}$ build-up events in Beijing were unique while accumulation pollution process occurred at other cities in the region. This indicated that $PM_{2.5}$ was probably transported to Beijing from other cities (Zheng et al., 2015; Ji et al., 2014; Tao et al, 2014; Zhao et al., 2013). In the meanwhile, most severe pollutions are under stable synoptic meteorological conditions in Beijing (Sun et al., 2015; Zheng et al., 2015; Zhao et al., 2013). The low wind speed and stable synoptic meteorological condition at ground level cannot explain the reason that regional transport makes significant contribution to severe pollution. A previous study has shown the secondary aerosol in Beijing probably mainly formed over regional transport according to a vertical observation from the ground to 260m height. (Sun et al., 2015). Therefore, vertical profiles of meteorology and air quality might help us to understand the impacts of regional transport to heavy 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 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 5^{th} to 11^{th}, 2014. A strict air emission pollution control plan was carried out in the BTH Region to improve air quality in Beijing from November $3^{rd}2^{nd}$ to 11^{th} for APEC. According to a conservative estimate by MEP, production of 9,289 plants were paused and 3900 plants were running at reduced capacity in six provinces (Beijing, Tianjin, Hebei, Shanxi, Shandong and Inner Mongolia); and more than 40 thousand construction sites were shut down temporally (http://www.zhb.gov.cn/gkml/hbb/qt/201411/t20141115_291482.htm). Other measures include traffic control (50% of private passenger vehicles and 70% of buses were off-road) and frequent road 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 pollutant concentrations during the Summit (Wen et al., 2015; Tang et al., 2015; Han et al., 2015; Chen et al., 2015; Sun et al., 2016a). The significantly reduced local emissions led to reduced complexity of particulate matter pollution process, thus providing a unique opportunity to investigate the influence of transport events on $PM_{2.5}$ levels in Beijing.

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) in southwest Beijing before and during the control period of the APEC 2014 Summit. Vertical profiles of temperature, RH (relative humidity), wind speed and direction, and extinction coefficient were observed as well as pollutants concentration and meteorological parameters on the ground. The characteristics of three $PM_{2.5}$ pollution episodes were analysed. Findings of this study will help explore vertical observation methods for in-depth analysis of the meteorological and transport influence. Furthermore, it can aid the development of future air quality management strategies in BTH and other regions around the globe, including emission control and air surveillance.

2. Field observation and analysis methods

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-level and vertical observations. Detailed information of instruments at Liulihe site is provided in Table S1. Ground-level observations included meteorological parameters, mass concentration of $PM_{2.5}$/PM_{10}, SO_2, NO_x and O_3 as well as physical and chemistry properties of PM. $PM_{2.5}$/PM_{10} mass concentration was determined by the TEOM method. Particle size distribution from 3nm to 10μm were measured by a spectrometer assembled in-house including one Nano scanning mobility particle sizers (NSMPS), one scanning mobility particle sizers (SMPS), and one aerodynamic particle sizer (APS) (Liu et al., 2014).

ACSM (Aerosol Chemical Speciation Monitor), a low-maintenance aerosol mass spectrometer, was used to measure non-refractory (NR) particulate matter with aerodynamic diameters smaller than 1μm (PM_1) (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) based on the monitoring site condition (see supporting information) was used in this study. The NR-PM_1 concentration measured by ACSM tracks well with $PM_{2.5}$ measured by the TEOM (R^2=0.91) and 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 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 were distinguished (Fig. S3), i.e., HOA (hydrocarbon-like organic aerosol), SVOOA (semi volatile~~volatility~~ oxygenated organic aerosol) and LVOOA (low volatile~~volatility~~ oxygenated organic aerosol).

Beyond ground-level concentrations of routinely monitored air pollutants and meteorological parameters, the assessment was aided by vertical observations including vertical extinction coefficient profile, as well as vertical wind, RH and temperature profiles. The vertical extinction coefficient profiles depict the distribution of PM, which could be used to infer mixing process of particles transported in from high evaluations and those near the ground. Vertical wind profile indicate ~~can help figure out~~ the transport direction. Vertical RH profiles reflect the strength of heterogeneous reaction at different layers.~~Vertical RH profile can provide the RH information at transport layers, thus helping investigate heterogeneous reaction at the layers.~~ Vertical temperature profiles provide information on the stability of and mixing in the boundary layer. Lidar was used to observe the vertical optical properties of atmospheric aerosols at Liulihe site. The lidar consists of three parts, including emitting system, receiving system and signal analogue system (Chen et al., 2015). The laser source emitted pulse at 355/532nm. The pulse energy is 30MJ at 355nm and 20MJ at 532nm. The pulse repetition is 20Hz.

The telescope for receiving system is based on a Cassegrain design. Diameter of the telescope is 200mm with a vertical resolution of 7.5m. The particle backscatter coefficient and extinction coefficient was retrieved by Fernald method (Frederick et al., 1984Fernald 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).

2.2 Back trajectory analysis

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

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):

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

(1)

Where N is the total number of hourly $PM_{2.5}$ concentrations in a specific time period, C_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):

$$R = \frac{A_r}{A_t} \times 100\% \quad (2)$$

The uncertainty evaluation mainly includes systematic errors, random errors and sensitivities. The major systematic errors depend on the calibration of instruments for $PM_{2.5}$ concentration measurement. Minor systematic errors might be from the judging the location and height of the daily minima and the sensitivities analysis suggests these errors are less than 10%. Random errors include data measurement and quantification step, such as identifying the daily minima properly, dealing with days without less-obvious afternoon minima and using linear interpolation between the daily minima. All these errors are evaluate by Jia et al. (2008). As a whole, this technique has an uncertainty of 40%-50% for results of daily regional transport.

3. Results and discussion

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 3rd to November 12th) compared to the period before control (October 27th to November 2nd) as shown in Fig. S4 (a).Period 1 (October 27th to November 2nd) and period 2 (November 3rd to November 12th) were defined to represent the periods before and during the APEC summit. Concentrations of $PM_{2.5}$, SO_2 and NO_2 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_2 (37%) and SO_2 (36%), while the reduction in $PM_{2.5}$ was smaller (21%) but still significant (Fig. S4 (b)).

Three pollution episodes were selected to discuss the pollution characteristics during the observation (Fig. S5). Episode 1 (October 27th to November 1st) represents the period before the emission control. Episode 2 (November 2nd to 5th) was the first pollution episode during the emission control. Episode 3 (November 6th to 11th) was the second pollution episode during the emission control. $PM_{2.5}$ concentration at Miyun site (locate in northern Beijing, shown in Fig. S1, data source: Beijing EPB) is shown in Fig. S5 alongside Luilihe to demonstrate the synchronism of $PM_{2.5}$ levels at different sides in Beijing. Episode 1 (October 27th to November 1st) represents the period before the emission control. Episode 2 (November 2nd to 5th) was the first pollution episode during the emission control plan. Episode 3 (November 6th to 11th) was the second pollution episode during the emission control plan. At Luilihe, $PM_{2.5}$ concentration was the highest in episode 1 (140±70μg/m^3) before implementation of

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Formatted: Font: (Default) Times New Roman, (Asian) SimSun, 10 pt, English (United Kingdom) emission control, whereas the mean values were close during the last two episodes (91±75μg/m^3 and 89±61μg/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 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 17 days10-day observation. The frequency of southwest wind was above 60% during each of the three episodes, with the highest occurrence of 81% observed during episode 2.

The significant reduction in pollutant concentrations during APEC shown above implied that the emission control was effective. However, the general characteristics derived from ground-level observation are insufficient to identify the leading cause of particulate matterair pollution, local 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 conditions and local emission intensity suggest different transport or formation mechanisms during those two episodes. Therefore, vertical observations will be used to aid further investigation in each of the three episodes in the following section.

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

3.2.1 Pollution process in episode 1

Episode 1 (October 27th to November 1st) was before emission control. The average concentration of $PM_{2.5}$ reached to 140μg/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 PM_1 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 29th (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 Yongledian site (116°47′E, 39°43′N) near Liulihe site were used (Fig. 4(a)), because the optical lidar 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 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 downward vertical wind direction in Fig. 4 (b)) and mixed with aerosols on the ground (Fig. 4 (a)). The online particle size distribution also implied the transport process. During the same period (from 13:00 to 20:00 on October 28th), a new group of particles appeared and mixed with existing particles, indicating the arrival of aged aerosols (Fig. 4 (e)). As mentioned above, except secondary formation, other mechanisms might impact OM increase. The increase of OM might come from freshly-emitted 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 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~~kept~~ in the southwest above 500m, indicating significant influence of regional transport.

In the next two days (October 30th to 31st), vertical wind direction was downward which was unfavorable for the pollutants diffusion (Fig. 5(a))~~vertical wind direction was downward and pollutants were easily accumulated in the boundary layer (Fig. 5)~~. Weather Research & Forecasting Model (WRF) modeling results also show the whole region was under control of weak downward wind from late night on October 30th. (Fig. S8, modeling parameters are provided in supplemental information).What's more, both the atmosphere press and wind speed decreased at the same time (Fig. S6). This indicates the site was probably in the rear of cold anticyclone. The steady weather condition promote the pollutants accumulation. Meanwhile, high RH on the surface (Fig. S6) 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 0.26μg/m^3/h, 0.21μg/m^3/h, and 0.58μg/m^3/h, respectively. The peak of NH_4^+, SO_4^{2-} and NO_3^- concentrations was two days later than 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 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. 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 vertical temperature gradient decreased and vertical mixing became weak (wind vertical speed was very low). Consequently, $PM_{2.5}$ accumulated and had a sharp increase. Then clean and cold wind from north caused sharp increase of wind speed and decrease of atmosphere pressure. Based on the analysis above, pollution ended up at 18:00 when the week temperature ended and $PM_{2.5}$ decreased sharply (Fig. 6). The regional component is calculated based on the determination of baseline.

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 observed at Liulihe site, 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 was continuouslykept 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 the low wind speed, high RH can easily promoted the pollutants accumulation and downward vertical wind was unfavorable for pollutants diffusion.After that vertical wind direction kept downward and promoted the pollutants accumulation, especially SNA.

3.2.2 Pollution process in episode 2

Episode 2 (November 2nd to 5th) saw a lower mean $PM_{2.5}$ concentration (91±75μg/m^3) due to the implementation of emission control since November 2nd. Unlike the gradual accumulation of PM observed in episode 1, $PM_{2.5}$, OM and SNA had a sharp increase from November 4th to 5th. The concentrations of NH_4^+, SO_4^{2-} and NO_3^- increased at rates from the lowest to the highest of 0.88μg/m^3/h, 0.43μg/m^3/h, and 1.64μg/m^3/h, respectively, much faster than that in episode 1. OOA also increased much more significantly during this episode. The explosively increases of PM components mainly SA in such a short period of time is contrary to lower RH values in this episode leading to less heterogeneous reaction. Thus, such rapid increases in PM levels could be transport of aged aerosol from other regions, as hypothesized by previous studies where the transport process wasn't observed directly (Yue, et al., 2009; Massling., et al, 2009; Sun et al., 2014; Sun et al., 2016b).

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 1st, relatively high PM levels still resided at 1000m (from November 2nd to 3rd) as shown in the vertical extinction coefficient (Fig. 7). Furthermore, a band of high PM centered around 750 m were observed ((Fig. S8S9) on November 3rd at another site (Baoding site, 115°31′E, 38°52′N, shown in Fig. 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) 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. S9S10), consistent with the vertical wind profile observed at Liulihe (Fig.5 (b)Fig. 8 and Fig.9). On November 3rd and November 4th, the downward motion of air mass around 1000 m above ground intensified, bringing the aged aerosols down and mixing them with the aerosols on the ground. The well mixed boundary layer with regard to aerosol is evidence in Fig. 9 8 with a fairly uniform distribution from the ground to 900 m. Consequently, secondary chemical component concentrations of PM_1 (Fig. 2 and Fig. 3) started ascending with remarkably fast rates.

Dry and clean wind air mass from north directionthe north arrived in the early morning on November 5th. 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 high $PM_{2.5}$ concentrationpollution 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 high $PM_{2.5}$ level appearedpollution occurred when the emission control plan just started, which means this episode was partly caused by regional transport before control. Even when local emission control was conducted effectively, the uncontrolled regional emission still led to severe particulate matter pollution in Beijing.

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±61μg/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 (NH_4^+ and NO_3^-) or changed little (SO_4^{2-}). 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~~Fig. 8 and Fig. 10~~). 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. ~~S10~~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 ~~11~~ 10 depicts black carbon (BC) concentrations measured by Aethalometer and OM concentrations measured by ACSM. They tracked each other well during this episode. Concentrations of BC, a marker of vehicular emission in urban settings, had two peaks every day. One was in the early morning and another was after morning rush hour of 10:00-11:00 am.~~in the morning rush hour of 9:00am.~~ The first peak might result 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 large. The second peak might be resulted from vehicles from outside coming into Beijing. Vehicles not registered in Beijing are banned to come into Beijing in the rush hour (7:00 am to 9:00 am), which reduces the morning peaks and smoothes the traffic flow. The vehicles coming into Beijing reach a peak after morning rush hour (http://wenku.baidu.com/link?url=SjtPVT1tgo4ON0KDQ5py8ehw1ZAzUr3k0mSd74D3F-8lOQZPPvedZiro6E5-MOeFFuww7VZjy3XwRqfU-mHXkg0_8kSy5p9FGyokfrFZX0e). 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.

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 ~~significantly~~ 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.

Three episodes of different types under similar ground meteorological condition were discussed in this study. In episode 1, particle concentration accumulated under the unfavorable meteorological condition after transport occurred. The transport pollutants brought organic aerosol and SNA increased under high RH later. In episode 2, pollutants left from episode 1 ~~kept~~ was retained in the boundary layer in the region. When vertical wind direction changed to downward, the pollutants were settled down. As a result, OM and SNA increased much explosively. In episode 3, when the control ~~plan~~ had been conducted for several days, SNA and OA concentration increased much less while HOA and 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 equal efforts need to be paid to regional emission to ensure the clean air. What's more, diesel vehicle emission at night in Beijing might be an important pollution source and needs further investigation.

5~~.~~ Acknowledgements

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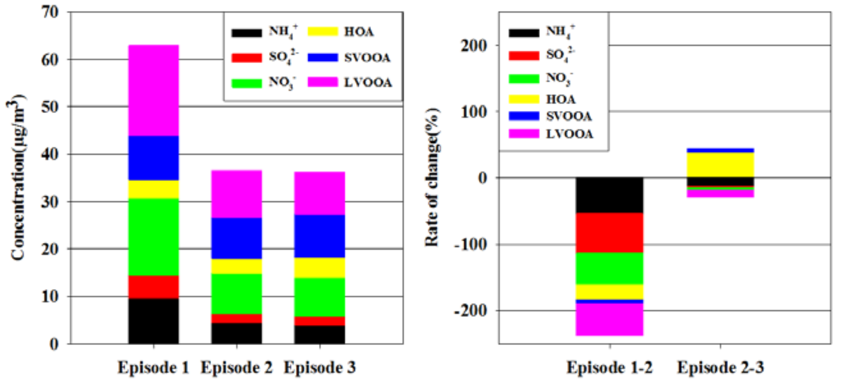
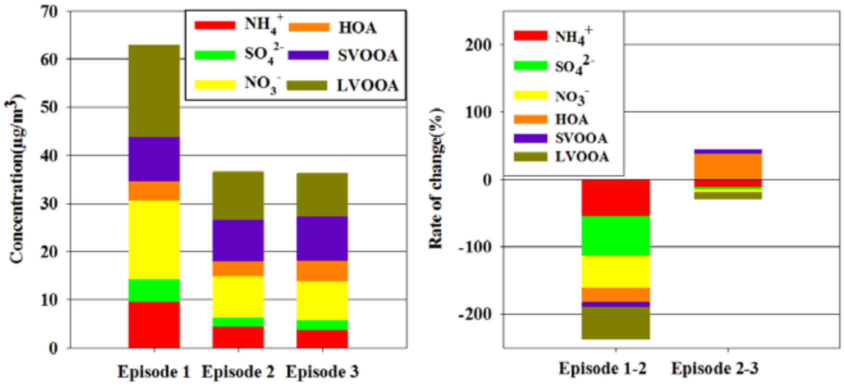
Figures



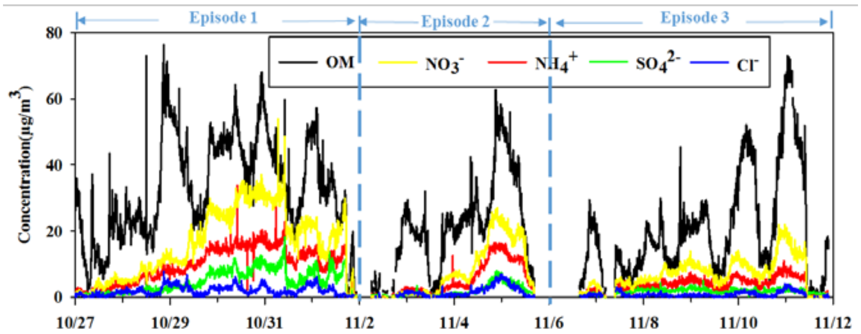


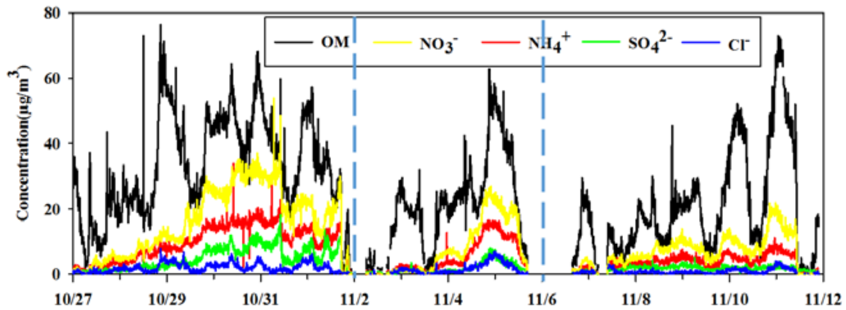
(a)

(b)

Figure 1. Non-refractory PM_1 chemical components at Liulihe site in the three episodes (a) average non-refractory PM_1 chemical components; (b) differences of chemical components among episodes

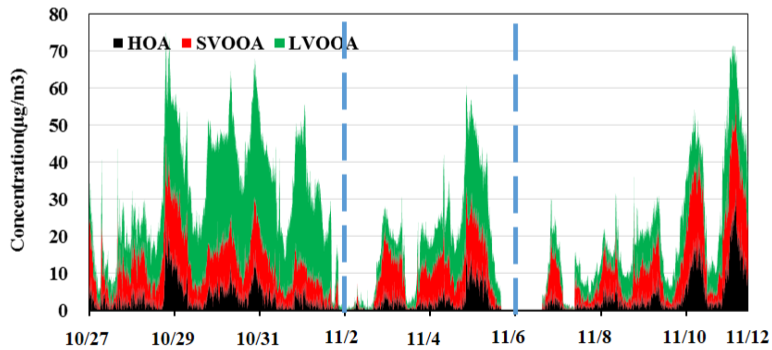
~~**Figure 1. Average PM_1 chemical components and the change rates during different episodes (a) average PM_1 chemical components; (b) change rates in chemical components**~~





<u>Figure 2. Temporal changes of non-refractory PM_1 chemical components at Liulihe site</u>

~~Figure 2. PM_1 chemical components during the observation at Liulihe site~~



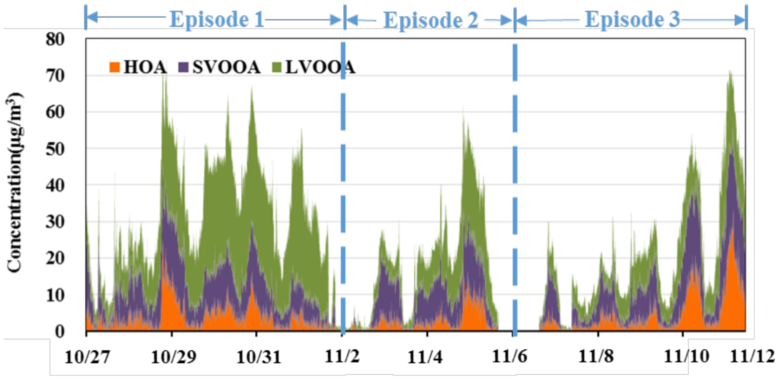
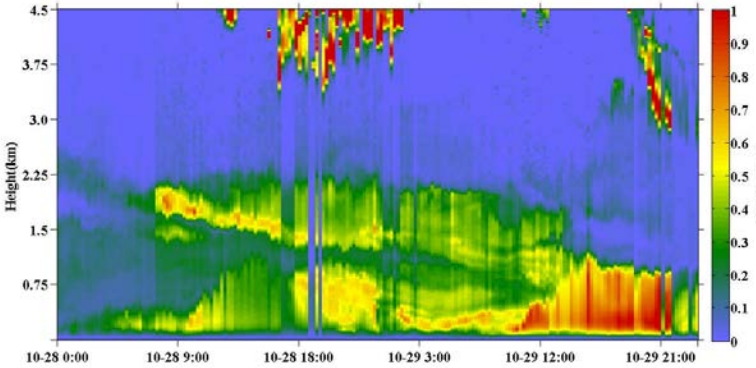


Figure 3. The temporal changes of organic components in PM_1 at Liulihe site~~PM_1 organic components during the observation at Liulihe site~~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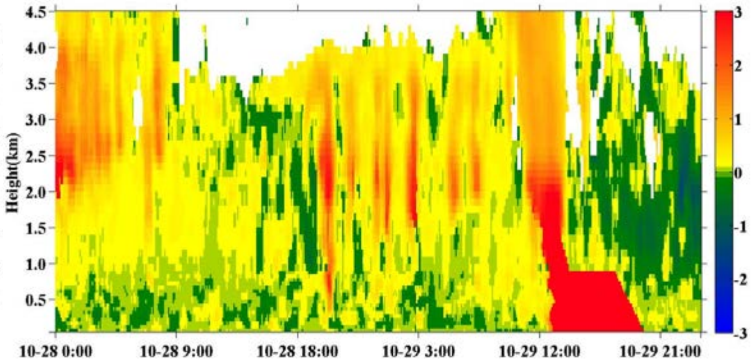
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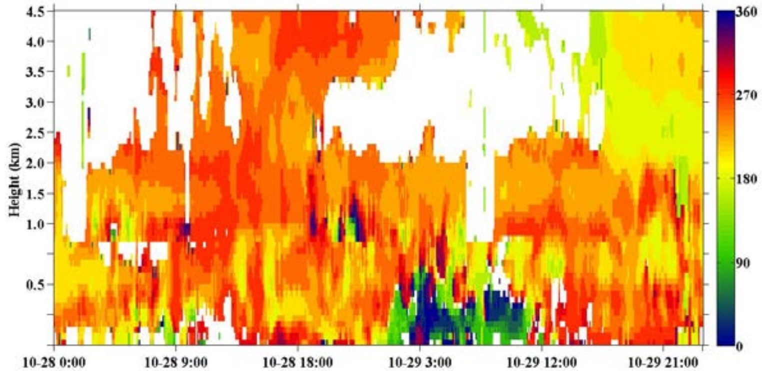
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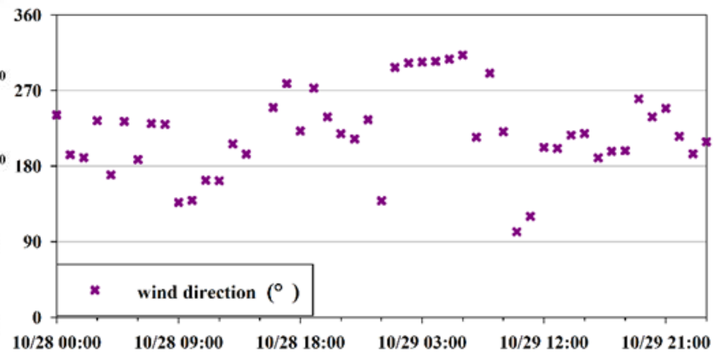
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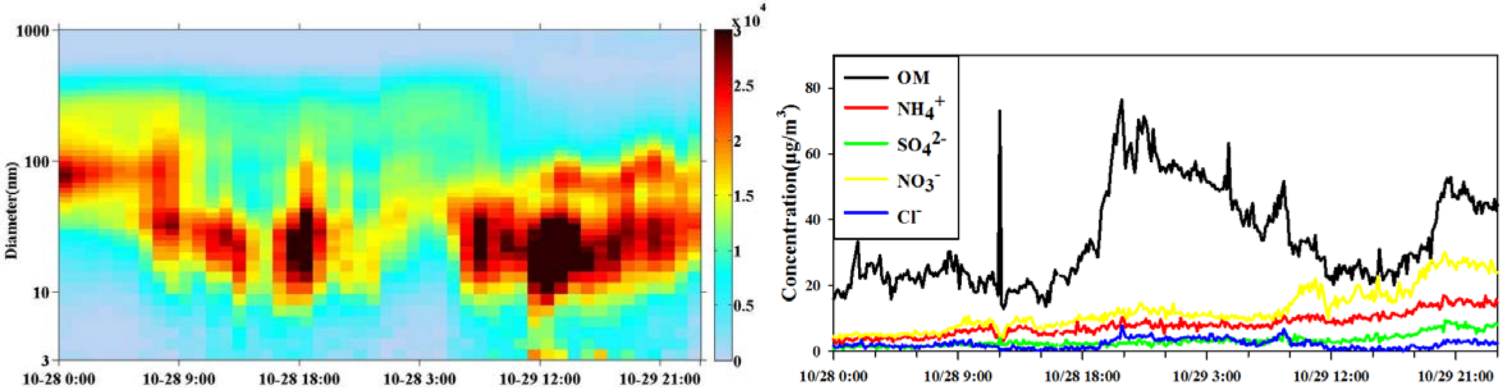
(c)

(d)

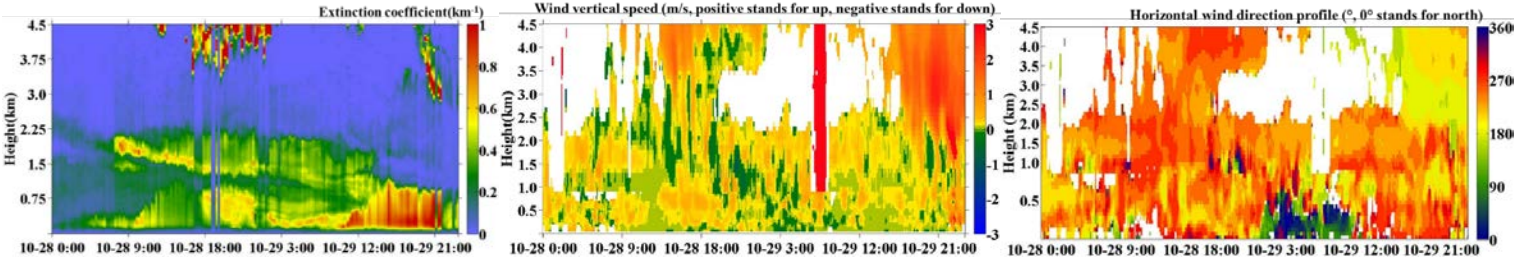


(e)

(f)

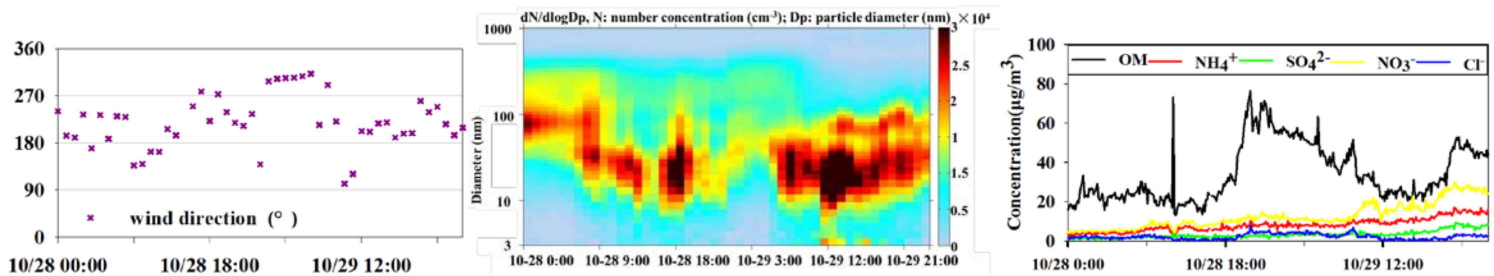
Figure 4. Parameters of particles and meteorology during episode 1

(a) Vertical profile of extinction coefficient (km^{-1}) (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^{-3}); Dp: particle diameter (nm)); (f) PM_1 chemical components.



(a) (b)

(c)



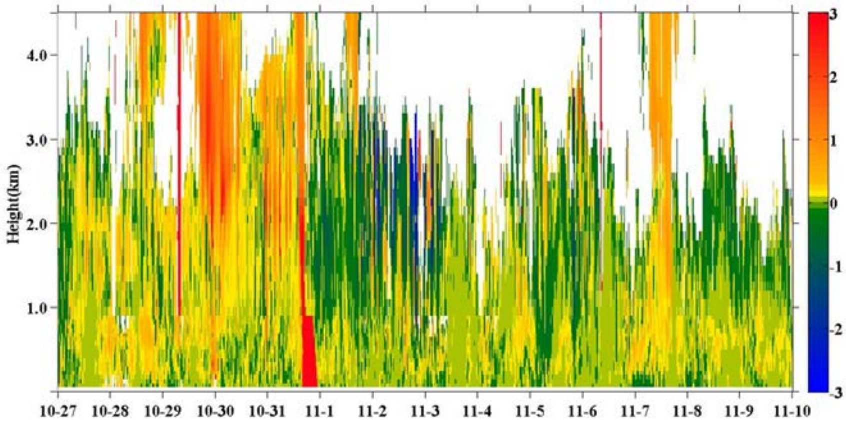
(d) (e)

(f)

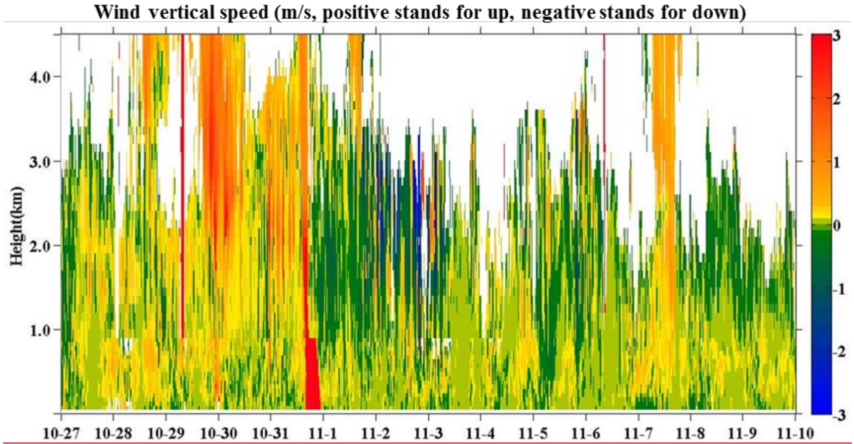
Figure 4. Characteristics of particulate matters and meteorological parameters during episode 1

(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-PM_1 chemical components

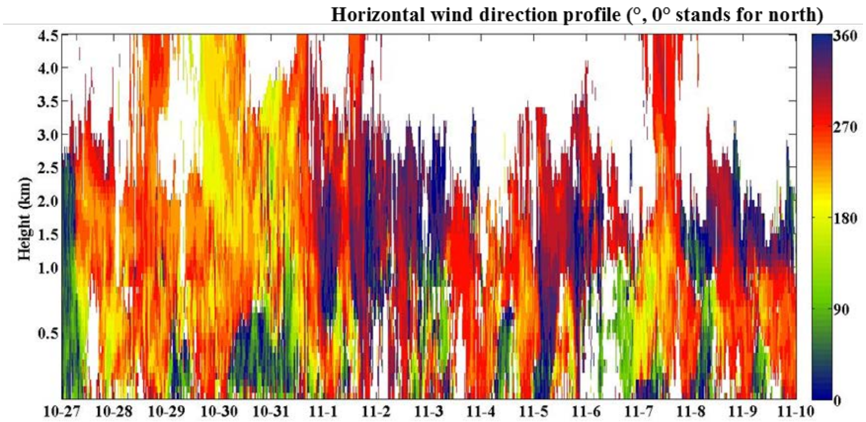
Formatted: Centered, Line spacing: 1.5 lines, Widow/Orphan control



~~**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**~~



(a)



(b)

Figure 5. Vertical profile of wind at Liulihe site

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

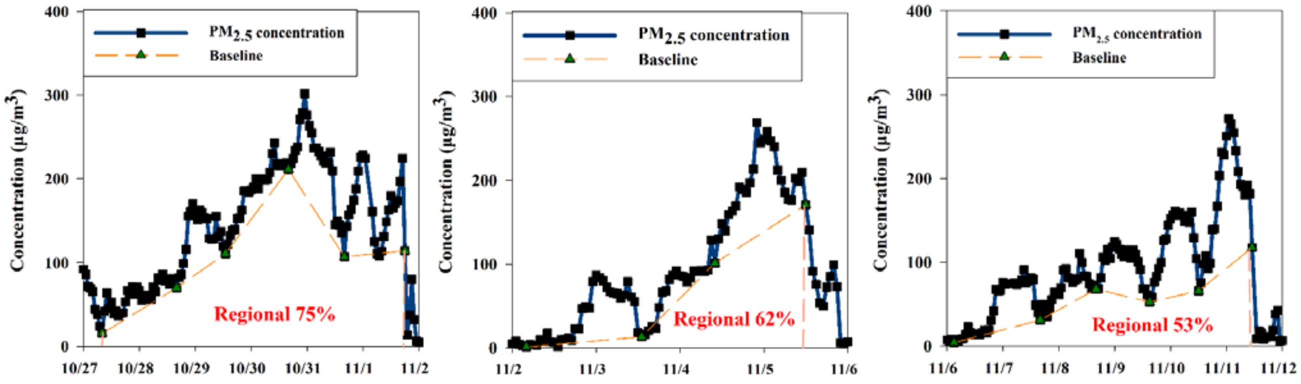


Figure 6. Regional and local components of the three episodes at Liulihe site

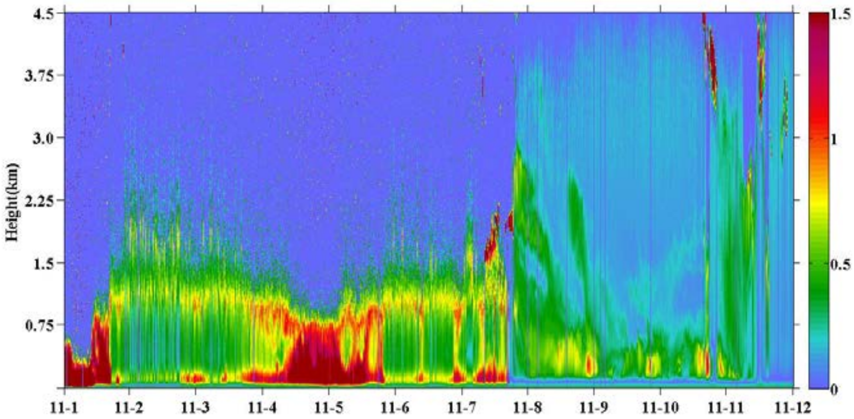


Figure 7. Vertical profile of extinction coefficient at Liulihe site

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

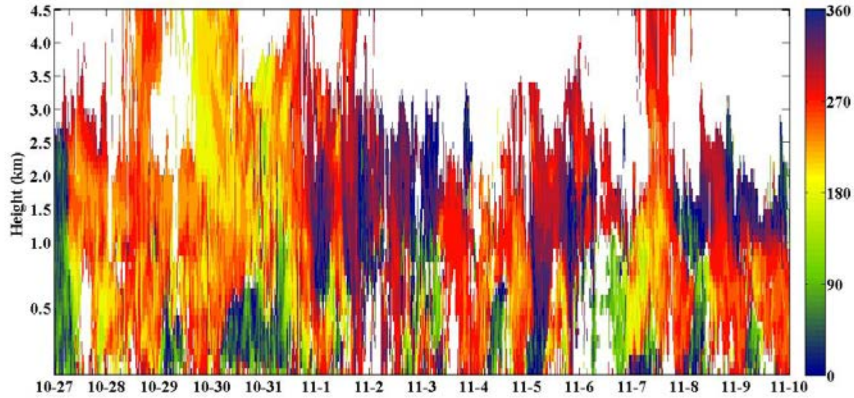
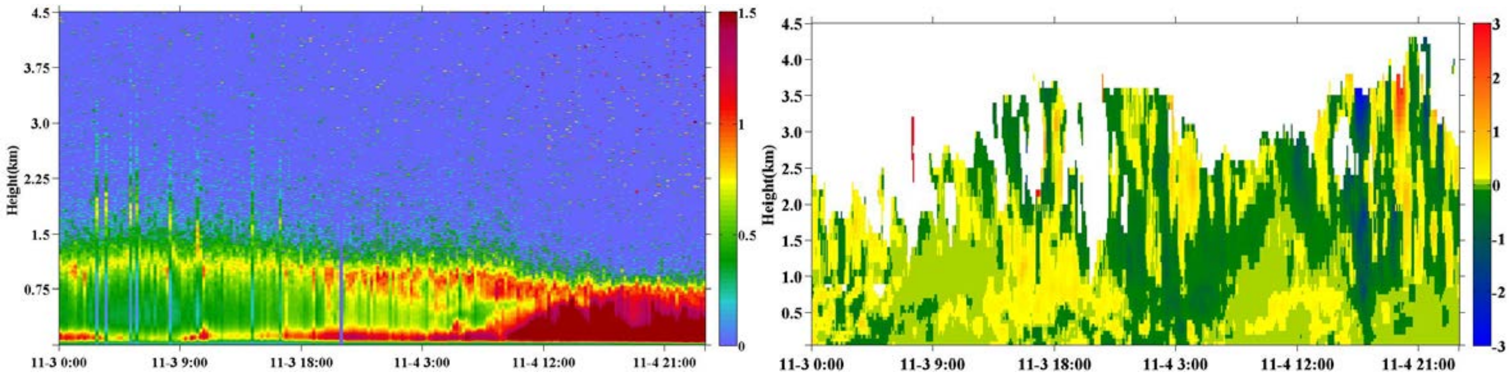


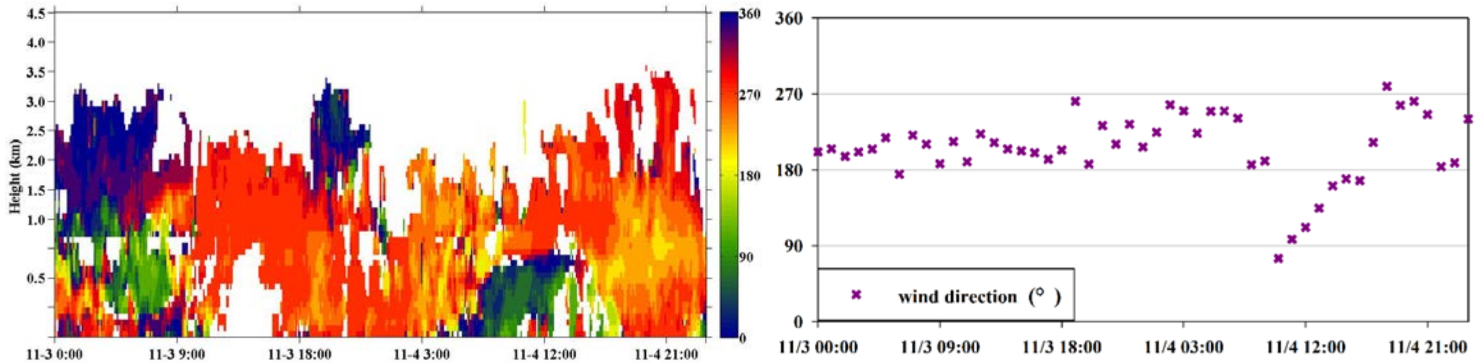
Figure 8. Vertical profile of wind horizontal direction during the observation at Liulihe site



Height(km)
11-3 0:00
11-3 9:00
11-3 18:00
11-4 3:00
11-4 12:00
11-4 21:00

~~(a)~~

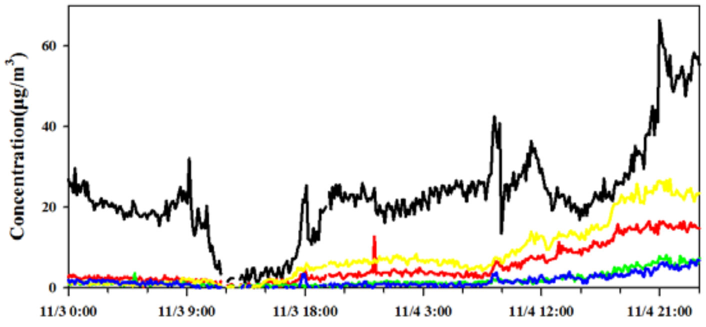
~~(b)~~



Height (km)
11-3 0:00
11-3 9:00
11-3 18:00
11-4 3:00
11-4 12:00
11-4 21:00
wind direction (°)
11/3 00:00
11/3 09:00
11/3 18:00
11/4 03:00
11/4 12:00
11/4 21:00

(c)

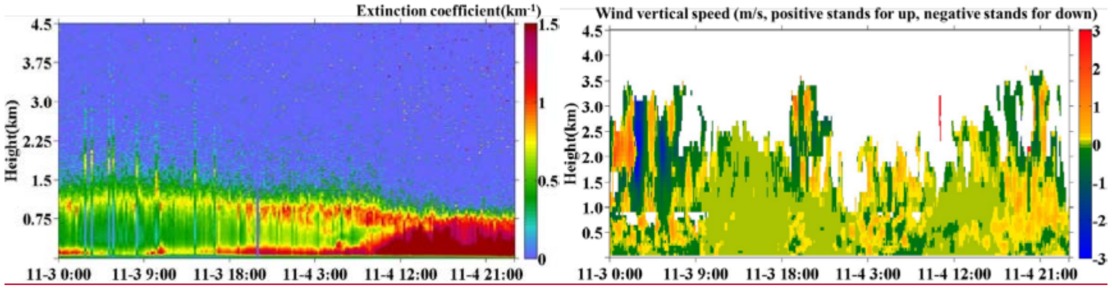
(d)



(e)

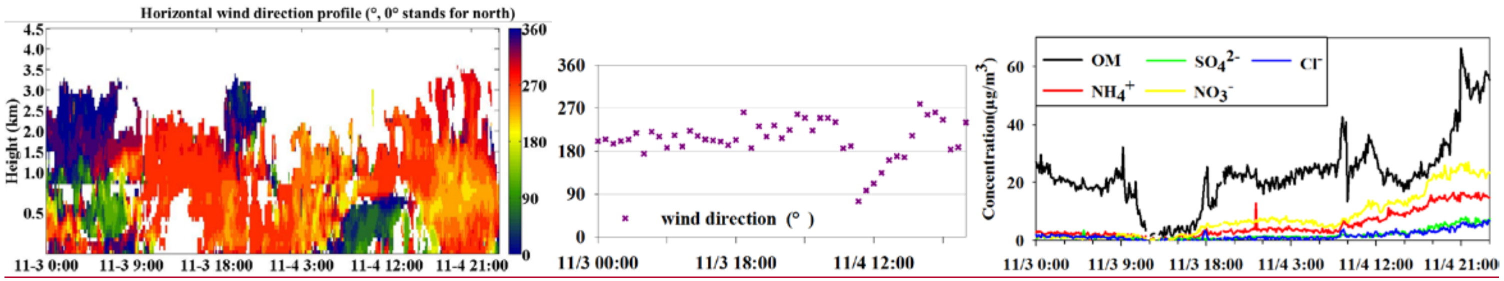
Figure 9. Parameters of particles and meteorology at Liulihe site during episode 2

(a) Vertical profile of extinction coefficient (km^{-1}); (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) PM_1 chemical components.



(a)

(b)

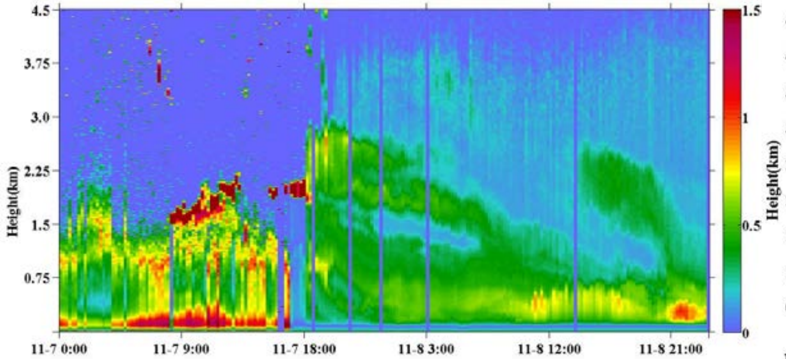


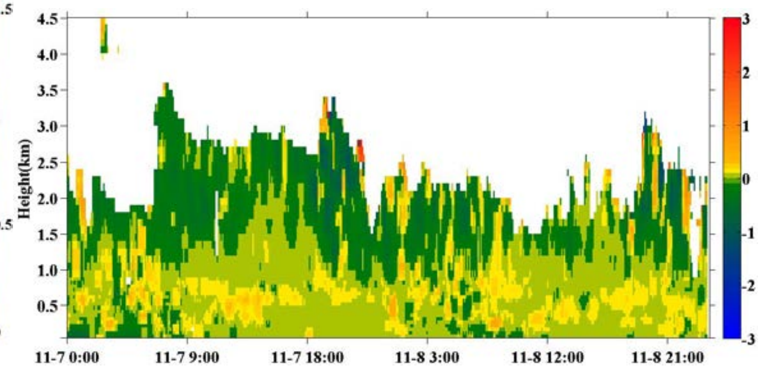
(c) (d)

(e)

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

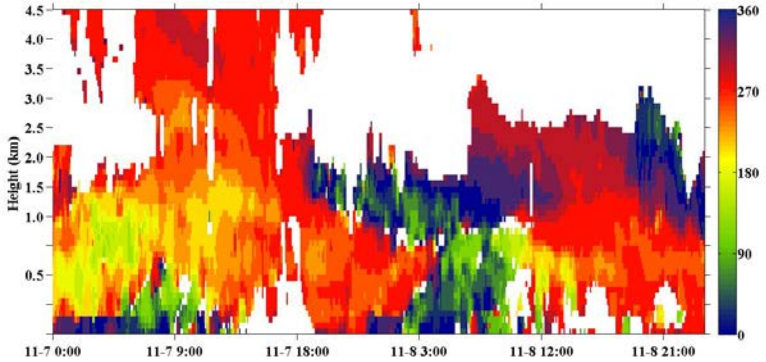
(a) Vertical profile of extinction coefficient;(b) Vertical profile of wind vertical direction and speed; (c) Horizontal wind direction profile; (d) wind direction on the ground; (e) NR-PM_1 chemical components

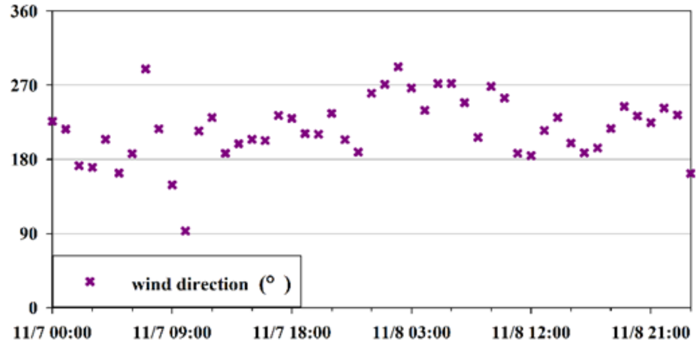




~~(a)~~

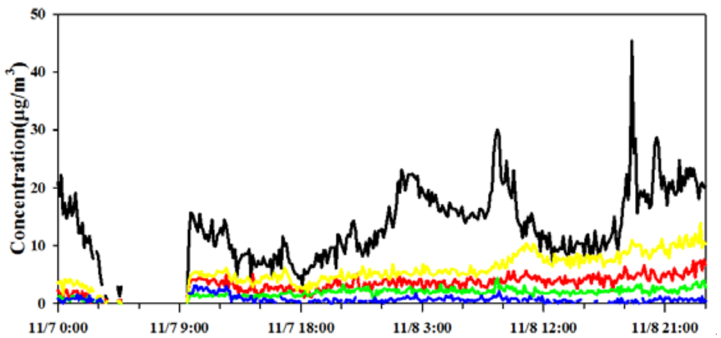
~~(b)~~


4.5
4.0
3.5
3.0
2.5
2.0
1.5
1.0
0.5
Height (km)
360
270
180
90
0
11-7 0:00
11-7 9:00
11-7 18:00
11-8 3:00
11-8 12:00
11-8 21:00


360
270
180
90
0
wind direction (°)
11/7 00:00
11/7 09:00
11/7 18:00
11/8 03:00
11/8 12:00
11/8 21:00

~~(c)~~

~~(d)~~

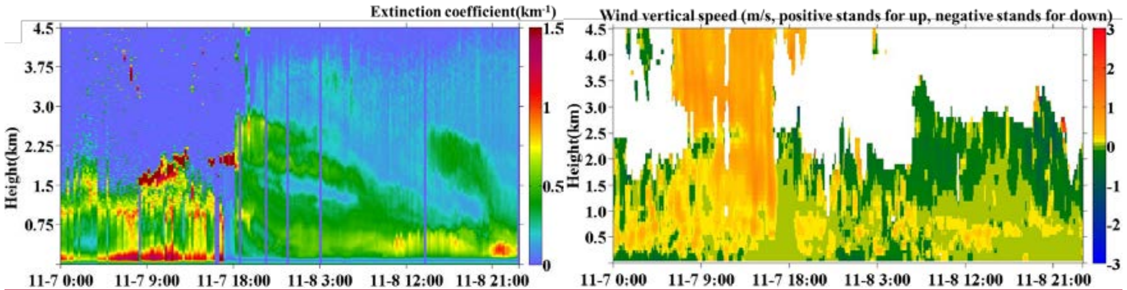

50
40
30
20
10
0
Concentration(μg/m3)
11/7 0:00
11/7 9:00
11/7 18:00
11/8 3:00
11/8 12:00
11/8 21:00

~~(e)~~

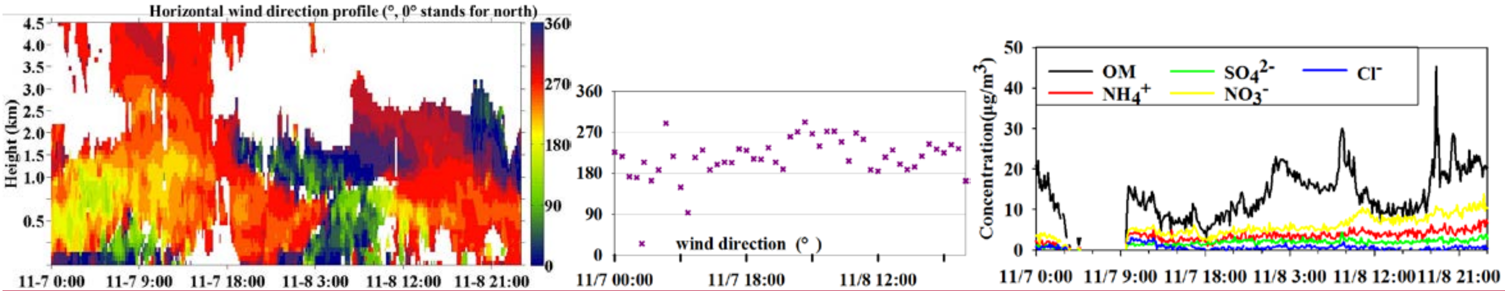
~~**Figure 10. Parameters of particles and meteorology at Liulihe site during episode 3**~~

~~**(a) Vertical profile of extinction coefficient (km^{-1}); (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) PM_1 chemical components.**~~



(a) **(b)**



(c) (d)

(e)

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-PM_1 chemical components

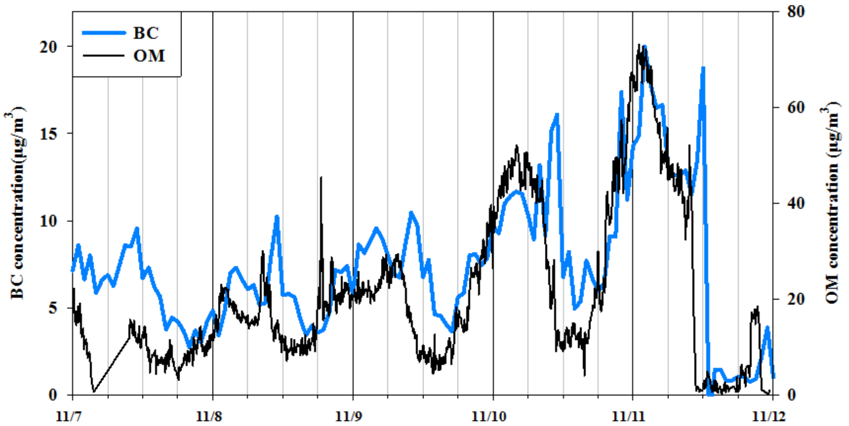


Figure 11. BC and OM concentrations of PM_1 at Liulihe site during episode 3

SUPPORTING INFORMATION

S1 Emission control measures during APEC

In Beijing, production of three largest thermal power plants and 141 industrial plants were restricted or stopped. Meanwhile, the number of private cars on road was reduced through odd/even license-plate rules and 70% of buses were off the road. Building constructions, municipal constructions and open burning were forbidden. Road sweeping and cleaning was conducted much more frequently to remove the road dust. The governmental staff had six days off from November 7th to November 12th to reduce the emissions from commuting. Moreover, ~~neighbouring~~neighboring provinces including Hebei, Tianjin, Shanxi, Inner Mongolia and Shandong implemented emission control plan. Furthermore, steady weather condition was forecast on November 4th and November 5th. Therefore, eighteen cities including Beijing, Tianjin, Langfang, Baoding and Shijiazhuang carried out emergency plans of emission control, to combat poor dispersion due to stable weather condition forecasted on November 4th and November 5th.)

S2 ACSM data analysis

Although default collection efficiency (CE) of 0.5 is widely used, it varies based on aerosol composition, RH and aerosol acidity (Middlebrook et al., 2012). Considering aerosol was dried before ACSM sampling, the influence of RH can be ignored. What's more, NR-PM_1 chemical components measured in this study showed no acidity (cation/anion = 1.2). As a result, aerosol composition impact was considered in this study. CE= max (0.45, 0.0833 + 0.9167 × ANMF) was used (Middlebrook et al., 2012). ANMF is characterized by the ~~amomium~~ ammonium nitrated mass fraction (ANMF). CE was calculated to be 0.45. This value was also used in the previous study in Beijing (Sun et al., 2013).

S3 baseline definition for transport component calculation

In the morning on 1st November (episode 1), air mass from the north above 1000 m arrived Beijing. The vertical temperature gradient decreased and vertical mixing became weak (wind vertical speed was very low). Consequently, $PM_{2.5}$ accumulated and had a sharp increase. Then clean and cold wind from north caused sharp increase of wind speed and decrease of atmosphere pressure. Based on the analysis above, pollution ended up at 18:00 when the week temperature ended and $PM_{2.5}$ decreased sharply (Fig. 6).

S4 Weather Research & Forecasting Model (WRF) modeling analysis

WRF version 3.7 is utilized to generate the regional meteorological fields. The parameters have been introduced in our previous studies (Wang et al., 2015).

Table S1. Instruments information at Liulihe site.

Measurement index	Instruments	Time resolution
$PM_{2.5}$/PM_{10}	TEOM1405/1400a (Thermo Scientific, USA)	1hour
SO_2	API100E (Teledyne, USA)	1hour
NO_2	API200E (Teledyne, USA)	1hour
O_3	API400E (Teledyne, USA)	1hour
Off-line $PM_{2.5}$	Partisol 2300 (Thermo Scientific, USA)	23.5 hour
NR-PM_1 chemical composition (SO_4^{2-}, NO_3^-, NH_4^+, Cl^-, Organic Matter)	ACSM (Aerodyne Research Inc. USA)	8min
Particle size distribution	Nano SMPS&SMPS&APS 3321 (TSI Inc, USA)	5min
Absorption coefficient/black carbon	Aethalometer AE42 (Margee Scientific, USA)	1 min
Meteorological data (RH, wind speed/direction, temperature, atmospheric press)	WXT520 (VAISALA, Finland)	1hour
Wind profile	CFL-03 (23rd Institute of China Aerospace Science and Industry Corporation)	6min
Temperature and humidity profile	QFW-6000 (22nd Institute of China Electronic Technology Group Corporation)	2min

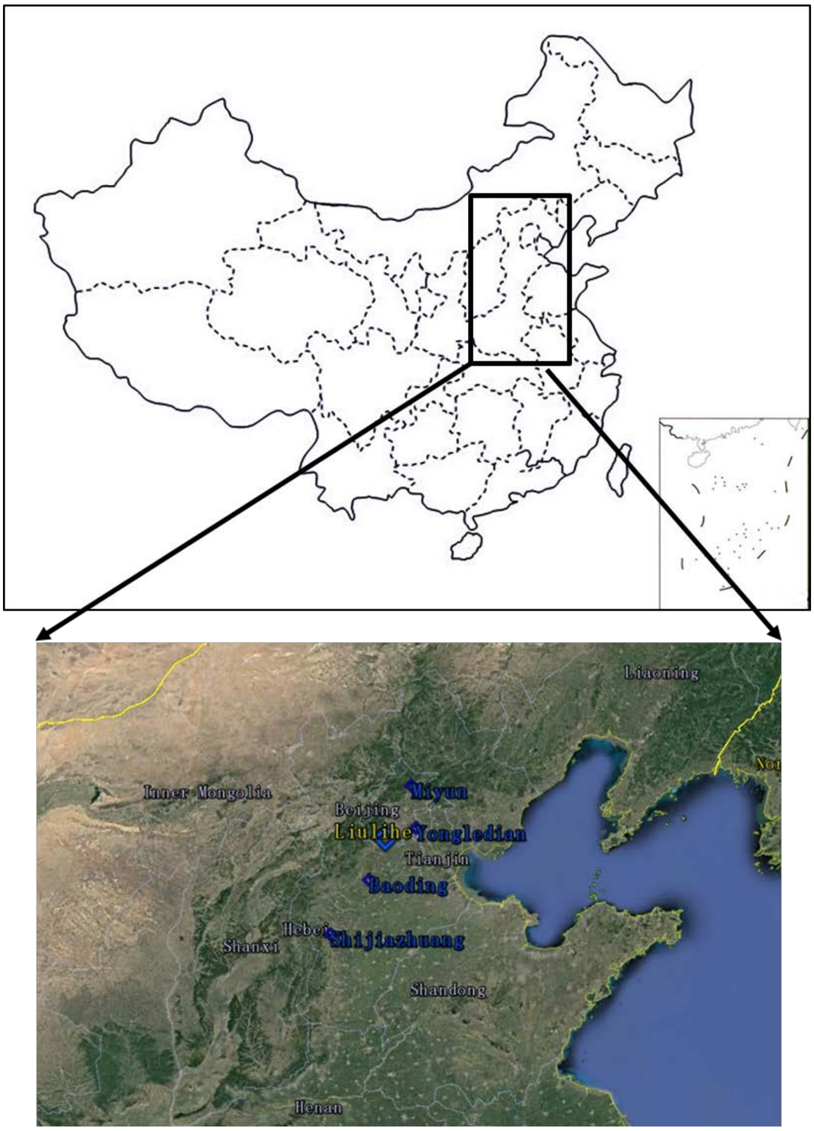


Figure S1. Field observation site location

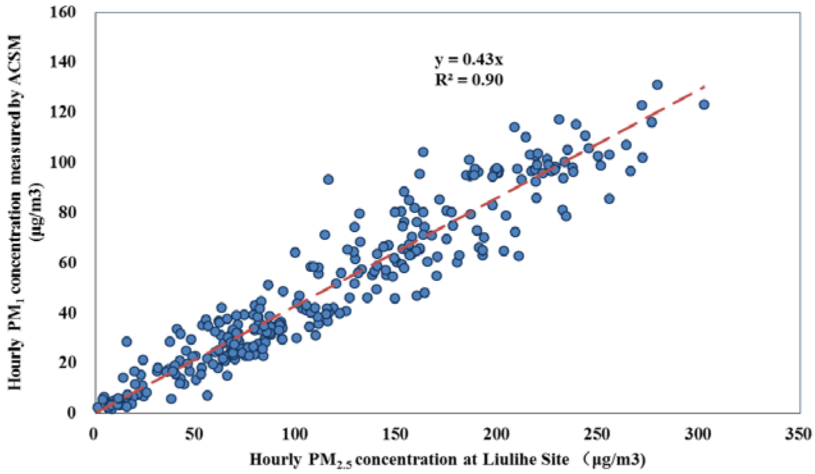


Figure S2. Correlation between NR-PM$_1$ (= Organic matter + SO_4^{2-} + NO_3^- + NH_4^+ + Cl^-) measured by the ACSM and PM$_{2.5}$ by the TEOM

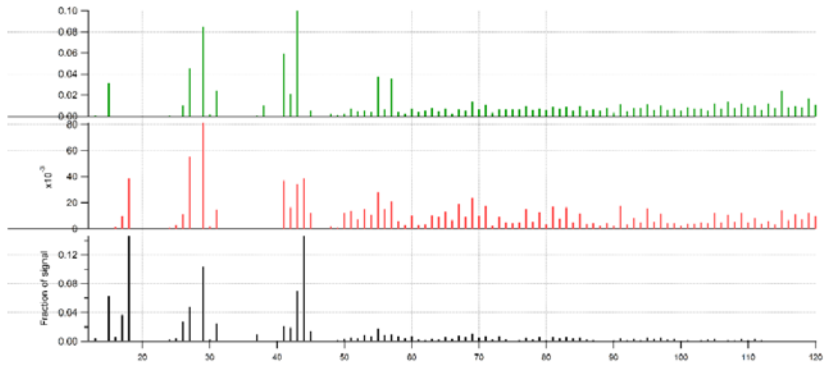
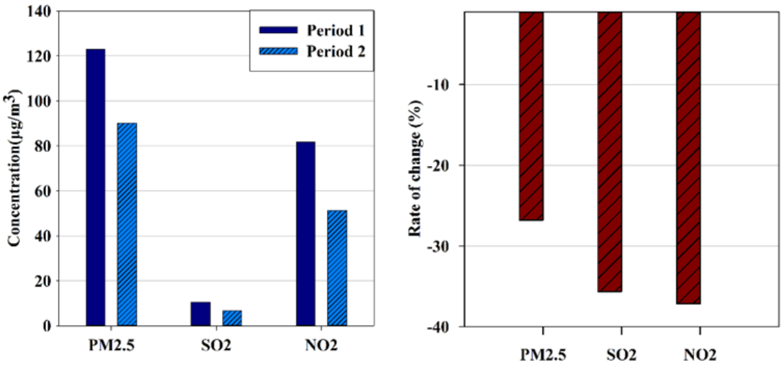


Figure S3. Factor profile performed by PMF



(a) (b)

Figure S4. Average concentration and change rate of pollutants during the observation. (a) Average concentration of pollutants; (b) Change rate of pollutants

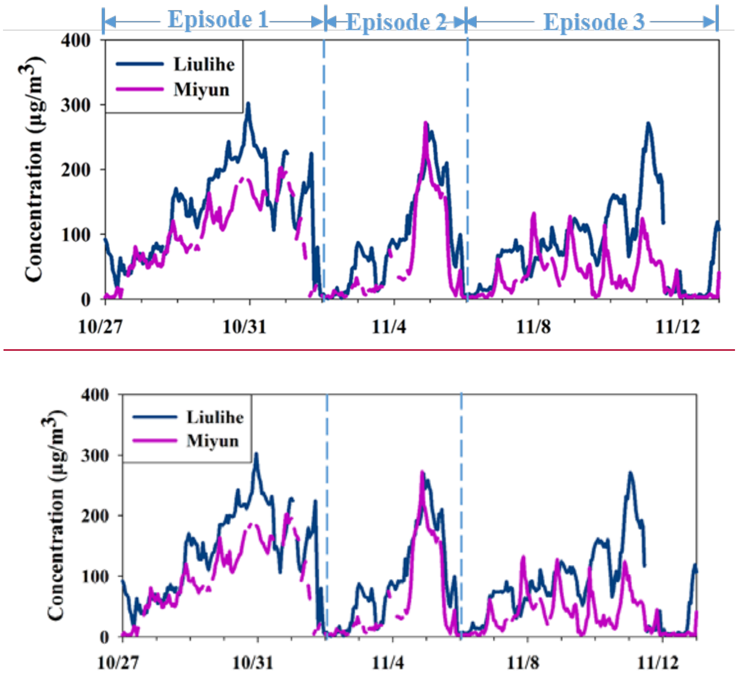
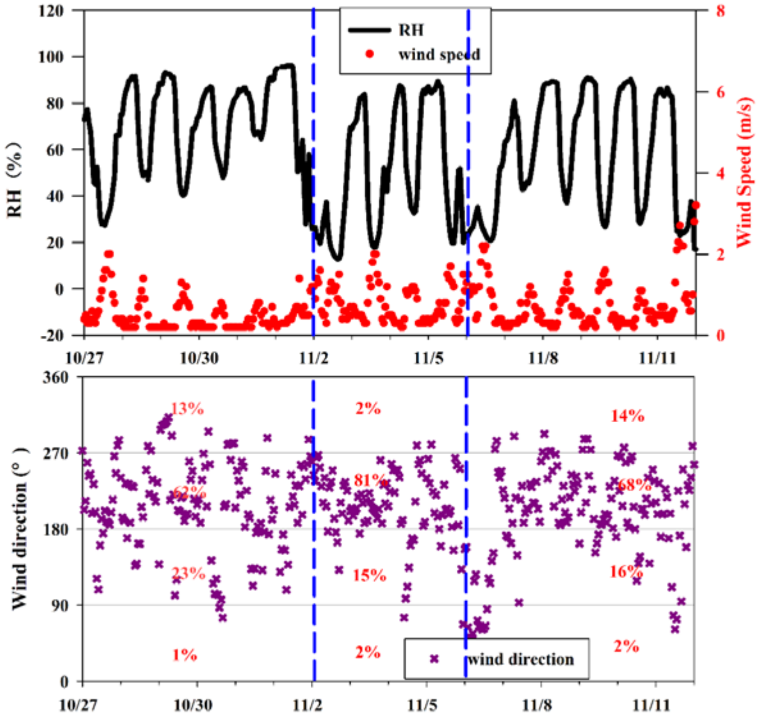


Figure S5. Hourly $PM_{2.5}$ concentrations at Liulihe and Miyun during the observation



RH
wind speed
RH (%)
Wind Speed (m/s)
10/27
10/30
11/2
11/5
11/8
11/11
Wind direction (°)
13%
62%
23%
1%
2%
81%
15%
2%
14%
68%
16%
2%
wind direction

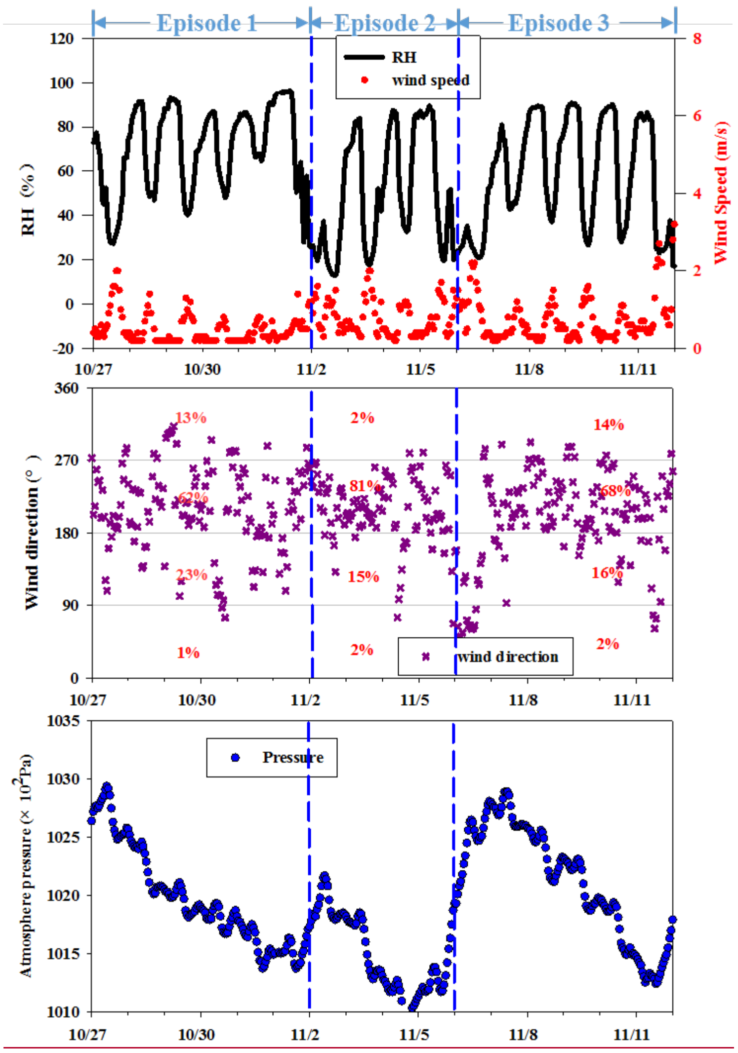


Figure S6. Meteorology conditions on the ground during the observation at Liulihe site

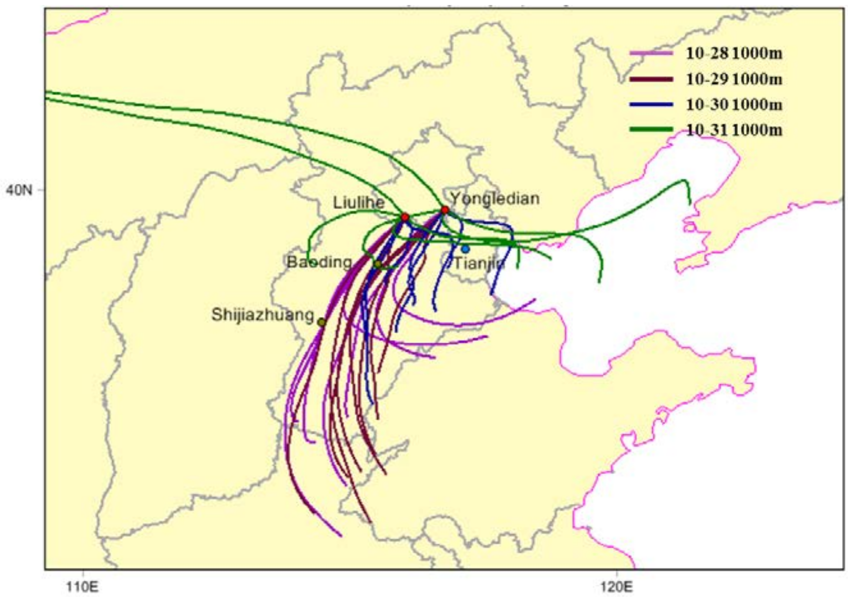


Figure S7. Air mass trajectory analysis during episode 1

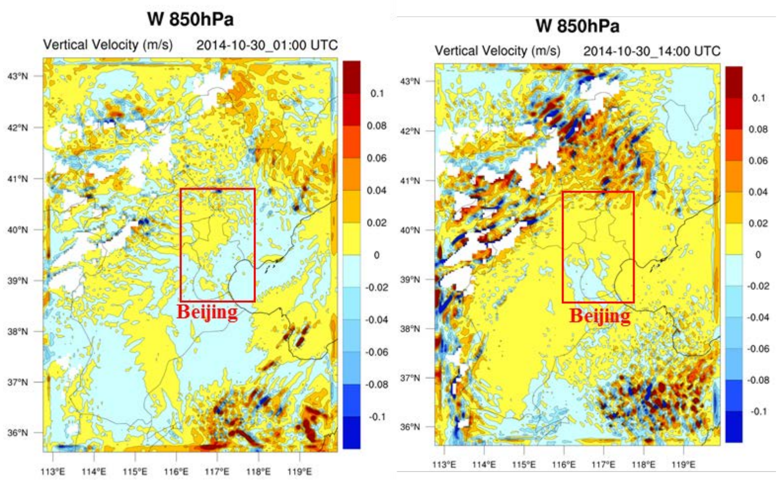


Figure S8. Regional wind vertical speed generated by WRF

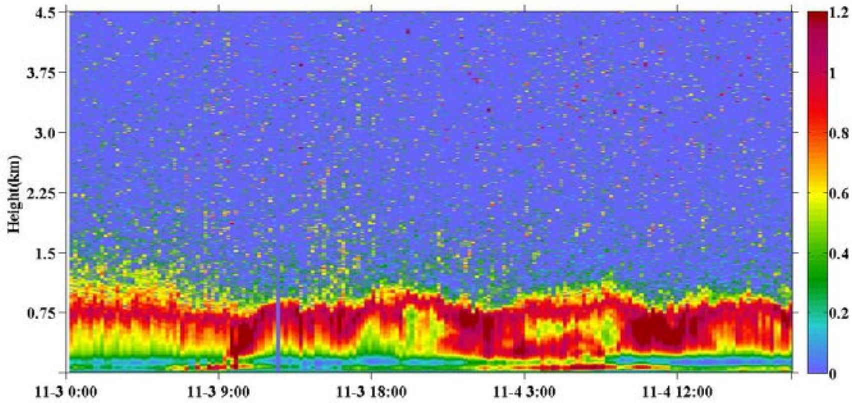


Figure ~~S8~~S9. Vertical profile of extinction coefficient at Baoding site during episode 2 (km^{-1})

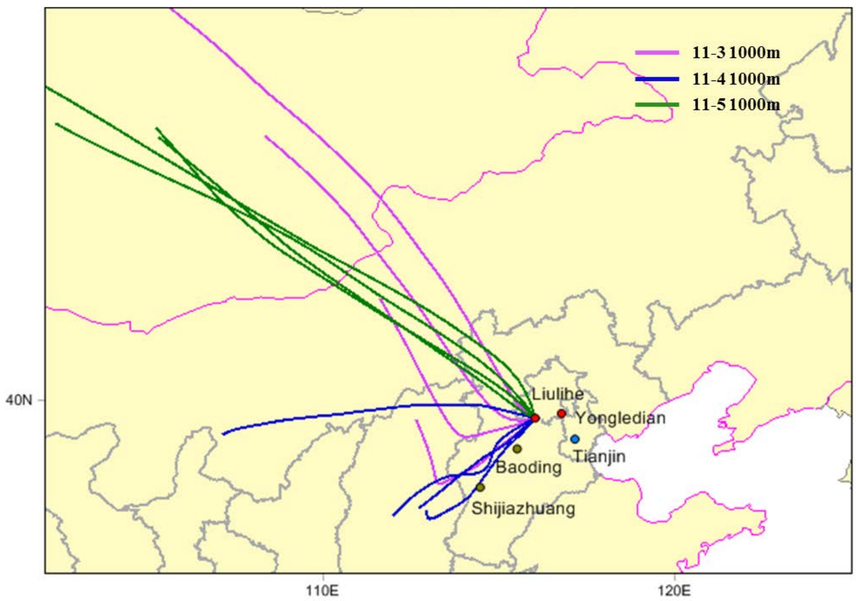


Figure ~~S9~~S10. Air mass trajectory analysis during episode 2

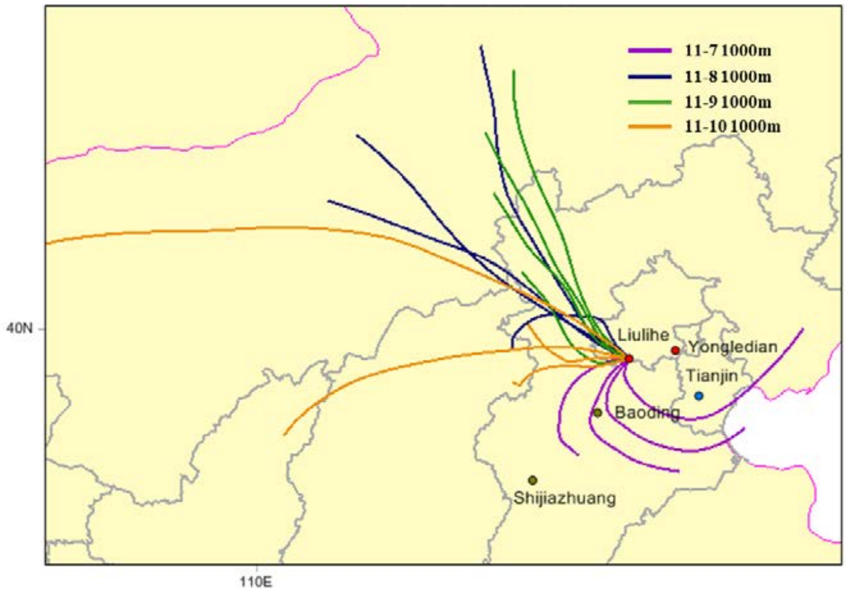


Figure ~~S10~~S11. Air mass trajectory analysis during episode 3

References

Middlebrook, A.M., Bahreini, R., Jimenez, J.L. and Canagaratna, M.R. (2012) Evaluation of composition-dependent collection efficiencies for the aerodyne aerosol mass spectrometer using field data. Aerosol Science and Technology 46(3), 258-271.

Sun, Y.L., Wang, Z.F., Fu, P.Q., Yang, T., Jiang, Q., Dong, H.B., Li, J. and Jia, J.J. (2013) Aerosol composition, sources and processes during wintertime in Beijing, China. Atmospheric Chemistry and Physics 13(9), 4577-4592.

Wang, J., Wang, S., Voorhees, A.S., Zhao, B., Jang, C., Jiang, J., Fu, J.S., Ding, D., Zhu, Y. and Hao, J. (2015) Assessment of short-term PM 2.5-related mortality due to different emission sources in the Yangtze River Delta, China. Atmospheric Environment 123, 440-448.