Response to Referee #2

The manuscript by Ding et al. investigated the roles of emission control and meteorology in decreasing PM2.5 in eastern China using long-term measurements. The results showed that control of open biomass burning and fossil-fuel combustion are the two major factors in reducing PM2.5 in early summer and winter, respectively. Such long-term measurements are very limited in China, which makes this study be important to understand the impact of emission sources, chemical mechanisms and meteorology processes on the reductions of aerosol species. This manuscript is overall well written, and I recommend it for publication in ACP.

Major comment:

High concentrations of $PM_{2.5}$ in Yangtze River Delta were often associated with the transport from north China, particularly in winter season. Considering that the air quality in Beijing and Hebei province has been significantly improved during the last 6 years, I suggest the authors expanding some discussions how air quality improvement in eastern China was potentially associated with that in northern China.

Response: Thanks a lot for the suggestion. We added some additional results based on back-trajectory cluster analysis to evaluate potential reduction associated with emission reduction from different regions, including the northern China. Please see P7, Lines 20-35 and Figure 7.

Response to Referee #3

The manuscript by Ding et al., reported a long-term continuous trend of $PM_{2.5}$, chemical species, and the precursors at the SORPES station in Nanjing, which is defined as a regional background station in the YRD region. With application of LPDM and comprehensive analysis with other supporting data, the authors investigated the impacts of emissions from fossil fuel combustion and open biomass burning and of year-to-year meteorology on the trends of primary and secondary $PM_{2.5}$ in this region. The study revealed the effect of air pollution control measures over the YRD region in the past years. The paper is structured and written in a clear, thorough, and objective fashion that gives readers a clear understanding of the trends of $PM_{2.5}$, related compounds and the effect of air pollution control measures. I recommend the paper to be published after minor revisions. Detailed comments are as follows:

1. Page 4 Line23, Fig. 2 shows the trends of $PM_{2.5}$ mass concentration and the two key precursors (SO_2 and NO_2) since 2011, and the main $PM_{2.5}$ chemical components (BC, SO_4^{2-} and NO_3^-) since 2013. Please also give the trends of NH_4^+ and NO.

Response: Thanks for the suggestions. WE added the trends of NH_4^+ and NO in the revised version. Please see the updated Figure 2.

2. Page 4, Line 32-34, "Among the two precursors, SO2 showed an even more significant reduction with an annual decrease about 17

Response: Here we don't understand the point as the sentence is incomplete, but we rewrote this sentence to avoid any misunderstanding in the revise version. Please see P4, Line 35- P5, Line 1.

3. Page 5, Line 1-2, ". . .achieved a very big success of air pollution prevention from desulfurization in power plant factories in recent years", this effect is not only due to the desulfurization in power plants, but also the measures including "replacement of coal with natural gas or electricity, etc".

Response: Thanks. We included this point in the revised version.

4. Page 5 Line 13-14, "Here the results show that the efforts in reducing PM2.5 also co-benefited to the mitigation of global warming", please give more evidence or reference.

Response: Here our point is in the context of black carbon as a well-known forcer for global warming. We rewrote this sentence with references.

5. Page 5 Line 21-23, to examine the change before and after the "Ten measures", the authors separate the time period into 2011-2014 and 2015-2018. Since the "Ten measures" policy

was released in August, 2013; why did not the authors separate the time period into 2011-2013 and 2014-2018?

Response: Thanks for the suggestion. We separated the period into two periods, the first 3 and last 4 years, to investigate the overall trend in the past 7 years but not only the "Ten-Measures" alone. Considering year-to-year difference in meteorology, it is inappropriate to compare average for periods with 2 years versus 5 years. Multi-year average for similar lengths could minimize the year-to-year variation in meteorology.

6. Page 5 Line 3, please give the full name of the shortened "TRMM" when it is the first time to appear.

Response: We already defined it in the last paragraph of Session 2.2.

7. Page 5 Line 17, "Intensive emission from these activities could cause a secondary maximum of $PM_{2.5}$ in early summer", K^+ is a tracer of primary pollutants from biomass burning, therefore it should be primary instead of secondary.

Response: Here the "secondary maximum" is a typo. It is "second maximum, which means a peak following the maximum in seasonal (monthly) variation.

8. Page 6 Line 14 and Line 25, "dominate" should be "dominant".

Response: Thanks. We already corrected them.

9. Page 7 Line 1, "due to efficient control from large elevated coal burning sources, such as power plants" and coal replacement with natural gas or electricity.

Response: Thanks. We included this point in the revised version.

10. Page 7, Line 12-15, another reason is that the Ox concentrations, or the atmospheric oxidization capacity has been increasing in recently years, which will also enhance the formation of nitrate.

Response: Thanks. We already included this point in the revised version.

11. By only conducting the LPDM simulations based on the fixed MIX emission inventory for cold season (three months), could they quantify the influence of emission reduction and year-to-year change in meteorology?

Response: Here we only conduct simulations for the three winter months NDJ just because the following reasons:
1) The concentrations in the three months are dominantly high from the seasonal pattern. The winter emission reduction rate could reflect the overall annual results and show

stronger signal of reduction efforts.

2) Winter has less precipitation in this region. The LPDM simulations cannot well characterize wet-deposition and secondary formation of $PM_{2.5}$. So, this method has less uncertainty in winter.

We included these points in the revised version. Please see P8, Line 5-9.

12. Conclusions 2. an increased nitrate fraction in PM2.5 was observed because more NH3 were available for nitrate formation in the condition of reduced sulfate associated with a substantial reduction of SO2 and a moderate decrease of NOx. Another reason is the increase of Ox and atmospheric oxidization capacity.

Response: Thanks. We included this point in the revised version.

13. Figure 4, why did the authors separate by 2012-2014 and 2016-2018 instead of 2012-2013, 2014-2018?

Response: Same as the Response to the No.5 comment.

Significant reduction of PM$_{2.5}$ in eastern China due to regional-scale emission control: Evidences from the SORPES station, 2011-2018

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Abstract. Haze pollution caused by PM$_{2.5}$ is the largest air quality concern in China in recent years. Long-term measurements of PM$_{2.5}$ and the precursors and chemical speciation is crucially important for evaluating the efficiency of emission control, understanding formation and transport of PM$_{2.5}$ associated with the change of meteorology and for accessing the impact of human activities to regional climate change. Here we reported long-term continuous measurements of PM$_{2.5}$, chemical components, and their precursors at a regional background station, the Station for Observing Regional Processes of the Earth System (SORPES), in Nanjing eastern China since 2011. We found that PM$_{2.5}$ at the station has experienced a substantial decrease (-9.1%/yr), accompanied with even much significant reduction of SO$_2$ (-16.7%/yr), since the national "Ten measures" for air took action in 2013. Control of open biomass burning and fossil-fuel combustion are the two dominant factors that influence the PM$_{2.5}$ reduction in early summer and winter, respectively. In cold season (November-January), nitrate fraction was significantly increased, especially when air masses transport from north. More NH$_3$ available from a substantial reduction of SO$_2$ and increased oxidization capacity are the main factors for the enhanced nitrate formation. The changes of year-to-year meteorology contributed to 24% of the PM$_{2.5}$ decrease since 2013. This study highlights several important implications on air pollution control policy in China.

1 Introduction

Fine particulate matter, with aerodynamic diameter smaller than 2.5 µm (PM$_{2.5}$), which impacts negatively on human health and visibility (Cao et al., 2012; Zhang et al., 2017; Malm et al., 2004), has been considered as one of the main air pollutants in China (He et al., 2001; Yao et al.,
5 2002; Sun et al., 2006; Pathak et al., 2009; Zhang et al., 2015; Wang et al., 2017). To tackle this great challenge, China recently has implemented the Action Plan for Air Pollution Prevention and Control (i.e. the so-called national "Ten Measures" for air) in several developed critical regions since 2013 (Sheehan et al., 2014; Wang et al., 2017; Liu et al., 2018; Zheng et al., 2018; Liu et al., 2019). Measurement data, mainly from the ambient air quality monitoring network,
10 showed some evidences of improved haze pollution in many cities in recent years (Wang et al., 2017; Lang et al., 2017; Zhang et al., 2019). However, to give a robust and quantitative assessment of the change due to specific emission reduction, high quality long-term continuous measurements of PM$_{2.5}$ and its chemical composition and precursors together with comprehensive data analysis and model simulations are needed because PM$_{2.5}$ has complex
15 chemical compositions, and sources and formation mechanisms (Pathak et al., 2009; G. Wang et al., 2016; Cheng et al., 2016; Wen et al., 2018), and strong dependence on year-to-year meteorology (Zheng et al., 2015; Zhang et al., 2016; Zhang et al., 2019).

The Yangtze River Delta (YRD) is one of the developed and highly populated regions in China (Ding et al., 2013a; H. Wang et al., 2016). Under unfavorable meteorological conditions,
20 PM$_{2.5}$ in this region could reach to very high concentration in winter and early summer, contributed mainly by fossil fuel combustion and agricultural straw burning, respectively (Ding et al., 2013ab; Cheng et al., 2014; Huang et al., 2016; Ding et al., 2016a; H. Wang et al., 2016; Wang et al., 2018). Some recent studies reported the change of PM$_{2.5}$ and its chemical components in some YRD cities in a short period, e.g. 2-3 years (Wang H. et al., 2016; Wang et
25 al., 2017; Sun et al., 2018), however, so far there is a lack of long-term observational study with comprehensive measurements covering the entire period of national "Ten measures" period, i.e. 2013-2017, in this region.

In this study, we report the long-term continuous ground-based measurements of PM$_{2.5}$ and its chemical compositions as well as main precursors at the Station for Observing Regional
30 Processes of the Earth System (SORPES) in Nanjing, western YRD for the period of 2011-2018. Based on Lagrangian dispersion modeling and comprehensive analysis with other supporting data, we investigate the impacts of emissions from fossil fuel combustion and open biomass burning (BB) and of year-to-year meteorology on the trend of primary and secondary PM$_{2.5}$ in this region.

2

2 Data and Methods

2.1 Brief introduction to the SORPES station and instrumentations

The SORPES station (118°57′10" E, 32°07′14" N, 62 m a.s.l.) is a cross-disciplinary research and experiment platform established in 2011 to understand the impact of human activities in the rapidly urbanized and industrialized eastern China region (Ding et al., 2013a; 2016b). Because of the unique geographical location, i.e. downwind of the North China Plain (NCP) and the YRD city cluster but upwind of Nanjing downtown (with distance about 20 km), this site can be considered as a regional background station for air quality studies in eastern China (Ding et al. 2016b). Regional anthropogenic plumes from the NCP to YRD city clusters and the early-summer open BB smoke in eastern China (Fig. 1) can influence this site under a complex multi-scale transport associated with Asian monsoon (Ding et al., 2013a; Ding et al. 2016b).

Continuous measurement of $PM_{2.5}$ mass concentration and its precursors, e.g. sulfur dioxide (SO_2) and nitrogen oxides (NO_x) (nitric oxide (NO) + nitrogen dioxide (NO_2)) etc., started in August 2011. More species, such as $PM_{2.5}$ chemical compositions, including black carbon (BC) and water-soluble ions (e.g. sulfate (SO_4^{2-}), nitrate (NO_3^-), ammonium (NH_4^+), potassium(K^+), and calcium (Ca^{2-}), etc.) has been measured since 2013 (Xie et al., 2015; Sun et al., 2018; Wang et al., 2018; Shen et al., 2018). Details of the data, including instrumentation, measurement period, and data coverage, used in this study are given in Table S1 in the Supporting Information (SI). Briefly, instruments/analyzers for chemical compositions measurement are housed in a 2-floor building on the top of a small hill about 42 m above ground level. $PM_{2.5}$ mass concentration is measured by the online analyzer based on the light scattering and beta ray absorption method (Thermo Fisher Scientific, 5030SHARP, USA). The water-soluble inorganic ions, including SO_4^{2-}, NO_3^-, NH_4^+, and K^+ etc., are detected by the instrument for Measuring AeRosols and GAses (MARGA, Metrohm, Switzerland) (Xie et al, 2015; Sun et al, 2018). BC was measured using a 7-wavelenth Aethalometer (AE-31, Magee Scientific) and the data at a wavelength of 880 nm was used in this study (Shen et al., 2018; Virkkula et al., 2015). SO_2 and NO_x are measured by the online analyzers with a time resolution of 1–5 min (Thermo Fisher Scientific, 43i and 42i, respectively). All the instruments are routinely calibrated for different durations. During the continuous measurement period, most of the instruments have a data coverage over 80% (Table S1).

2.2 Lagrangian dispersion modeling and other data sources

In order to help quantitatively understand the influence of year-to-year change in meteorology on air pollutant transport and dispersion, we conducted backward Lagrangian Particle Dispersion Modeling (LPDM) using the Hybrid Single-Particle Lagrangian Integrated Trajectory

(HYSPLIT) model (Stein et al., 2015). We used the model to calculate single-particle backward trajectories and to conduct cluster analysis. We also estimated the hourly PM$_{2.5}$ concentration based on the particle dispersion simulations following a method developed by Ding et al. (2013a; 2013c). Briefly, for each hour during the study period, the model was run 2 days and 7 days

5   backwardly with 3,000 particles released every hour at the altitude of 100 m over the SORPES station. The model calculated the position of particles by mean wind and a turbulence transport component, and the spatiotemporal distribution of these particles were further used to calculate the potential source contribution by using footprint "retroplume" (i.e., the residence time at the altitude of 100 m above the ground surface) and an emission inventory (Ding et al., 2013a;

10   2013c). Global Data Assimilation System (GDAS) data were used to drive the model and the MIX emission inventory database for the year of 2010 (Fig. 1a) (M. Li et al., 2017) was used for a quantitative estimation of PM$_{2.5}$ concentration.

Besides the observations at the SORPES station and data used in the LPDM simulations, various data are used to support the data analysis and discussions. To identify the impact of

15   agricultural straw burning, we used BB emission inventory from the Global Fire Emissions Database, Version 4.1 (with small fires) (GFED4s) (Giglio et al., 2013) and MODIS (Moderate Resolution Imaging Spectroradiometer) Thermal Anomalies/Fire daily L3 Global Product (MOD/MYD14A1) (Boschetti et al., 2009). The ERA5 reanalysis data (https://software.ecmwf.int/wiki/display/CKB/ERA5+data+documentation) from European

20   Centre for Medium-Range Weather Forecasts (ECMWF) together with the Tropical Rainfall Measuring Mission (TRMM) satellite observed precipitation (Huffman et al., 2007) are also use to investigate the year-to-year difference of meteorology that may influence the PM$_{2.5}$ concentration.

3 Results and Discussion

25   Based on continuous measurement at the SORPES station, Fig. 2 shows the trends of PM$_{2.5}$ mass concentration and key precursors (SO$_2$ and NO$_x$ (NO+NO$_2$)) since 2011, and the main PM$_{2.5}$ chemical components (BC, SO$_4^{2-}$, NO$_3^-$, and NH$_4^+$) since 2013. Considering the difference in the observation duration and the specific emission control in east China associated with the national "Ten Measures" for air since 2013 (Sheehan et al., 2014; Wang et al., 2017; Liu et al., 2018), we

30   conducted linear regression for the two periods: August 2011-July 2018 and August 2013- July 2018, respectively. It can be found that PM$_{2.5}$ concentration and the mixing ratio of two precursors show an overall decreasing trend during the past seven years (-6.4%/yr, -12.1%/yr, -4.6%/yr, and -11.1%/yr for PM$_{2.5}$, SO$_2$, NO$_2$ and NO, respectively), but more remarkable decreasing trends (-9.1%/yr, -16.7%/yr, -5.2%/yr, and -14.1%/yr for PM$_{2.5}$, SO$_2$, NO$_2$ and NO,

35   respectively) since 2013. For SO$_2$, the 5-yr reduction almost reached 70-80% and showed

significantly higher reduction rate in comparison with NOx. It demonstrates that the YRD region, as one of the main industry bases with a huge consumption of coal, achieved a very big success of air pollution prevention from desulfurization in power plants and factories and from replacement of coal with natural gases and electricity in recent years. In fact, a national wide significant reduction of SO_2 in the past few years has been also reported by ground and satellite measurements and emission estimations (C. Li et al., 2017; Liu et al., 2018; Zheng et al., 2018).

Fig. 2d shows that NO_2, another precursor of nitrate, shows a decreasing trend (-5.2% /yr) since 2013, which is much smaller than that of SO_2. Accordingly, the two secondary inorganic water-soluble ions, SO_4^{2-} and NO_3^- showed different trends, with the former a more significant reduction (-10.6%/yr vs. -5.8% /yr). For BC, an important particle mainly from primary emission of incomplete combustion but with significant impact to climate and aerosol-boundary layer feedback (Bond et al., 2013; Ding et al., 2016a; Wang et al., 2018; Huang et al., 2018), it showed a decreasing trend (about -8.4%) between those of sulfate and nitrate. Although BC is a short-lived climate forcer contributing to global warming (Bond et al., 2013; IPCC, 2013; Ding et al., 2016a), so far there are no specific reduction policy focus on it. Here the results show that the efforts in reducing $PM_{2.5}$ also caused BC reduction, which co-benefited to the mitigation of global warming (Bond et al. 2013, IPCC, 2013).

In eastern China, agricultural straw burning is particularly strong in early summer, i.e. from middle-May to middle-June, after the harvest of wheat (Ding et al., 2013ab; Cheng 2013; Huang et al., 2016; Chen et al., 2017). Intensive emission from these activities could cause a second maximum of $PM_{2.5}$ in early summer (Ding et al., 2013a), and also severe haze events with high concentration of $PM_{2.5}$ and other pollutants. For example, the SORPES station recorded an hourly concentration of $PM_{2.5}$ over 400 $\mu g\ m^{-3}$ in 10 June 2012 (Ding et al., 2013a; Xie et al., 2015; Nie et al., 2015). To examine the change before and after the "Ten measures" took action, Fig. 3a presents the seasonal variation of $PM_{2.5}$ mass concentration averaged for the periods of 2011-2014 and 2015-2018, respectively. It clearly shows that the secondary maximum of $PM_{2.5}$ mass concentration in early summer was missing in recent years, instead of a relative flat change (Fig. 3a).

Since the "Ten measures" took action in 2013, Chinese government has conducted very strict emission control of agricultural straw burning by using real-time satellite as a tool to monitor these activities (Chen et al., 2017; Wang et al., 2017). The MODIS satellite fire counts data did demonstrate the significant reduction in BB activities in this region since 2013 (Fig. 3b). To further investigate the potential impacts of other factors, i.e. the change of air pollutant transport associated with circulation patterns, we conducted LPDM simulations for the BB seasons (15 May – 20 June) during the two three-year periods. Fig. 4 shows that the averaged transport

pathways of air masses didn't change much for the two periods, but the MODIS satellite fire counts showed a distinct difference in both total amount and spatial distribution. These BB emitted smoke could be transported to the SORPES station in two days when the north wind prevailed.

5 Fig. 3b also shows the averaged TRMM precipitation in the same area as the fire counts data (i.e. dashed square in Fig. 1b) during 15 May-20 June, 2011-2018. It is indicative of a certain reverse correlation of precipitation with the total amount fire counts. However, the relationship between precipitation and BB is complicated. On one hand, the harvest season is generally before the "Meiyu" season in this region (Kitoh & Takao, 2006). Farmers usually choose

10 continuous sunny day to harvest and to dry the wheat, while the straws are also easy to be burned in a dry condition (Feng et al., 2019). On the other hand, precipitation can influence wet deposition of smoke (Uematsu et al., 2010). Meanwhile, BB smoke has been demonstrated to modify rainfall. For example, Ding et al. (2013b) and Huang et al. (2016) reported a case of suppressed rainfall and changed rainfall pattern by BB smoke in this region based on the

15 SORPES observations and numerical modeling. Although more quantitative studies are still needed, here the year-to-year variation of precipitation shouldn't be the dominant factor influencing the substantial decreasing the $PM_{2.5}$, especially for the period between 2013-2018 when the precipitation didn't show a significant trend.

To confirm the impact of BB on the $PM_{2.5}$ reduction, we further investigate the measured BB

20 tracer, fine particulate K^+, as previous studies (Ding et al., 2013b; Xie et al., 2015; Nie et al., 2015; Zhou et al., 2017) suggested that it is a good tracer for agricultural straw burning in this region. The scatter plot of K^+ as a function of $PM_{2.5}$ mass concentration, color-coded with time, given in Fig. 3b clearly shows a dramatic decrease in the $K^+/PM_{2.5}$ proportion at the SORPES station, especially after 2015. Using the concentration of K^+ as a threshold, we identified BB (K^+

25 higher than the 75% percentile) and non_BB cases (K^+ lower than 25% percentile) and showed the scattering plots of sulfate and nitrate as a function of $PM_{2.5}$ in Fig. 5. It confirms that very efficient control of open BB emission is the dominant factor that influenced the early-summer $PM_{2.5}$ reduction in this region.

Besides the early summer, months in the cold season, e.g. November, December and January

30 (NDJ), experienced strong $PM_{2.5}$ reduction in the past few years (Fig. 3a). In order to further explore the inter-relationship among precursors and secondary and primary particles, we show the scatter plots of different species color-coded with time in Fig. 6. For the SO_2-NO_x plot, it changed from a bifurcation pattern in the earlier years to a latest monotonous one (Fig. 6a). The bifurcated pattern of SO_2 versus NO_x indicates the impact of emissions from elevated coal

35 burning point sources (with high SO_2/NO_x ratio) and scattered vehicle sources (Wang et al.,

6

2002). The pattern change in recent years further confirms that the reduction of SO_2 was mainly due to efficient control from large elevated coal burning sources, such as power plants, and coal replacement with natural gas or electricity (Wang et al., 2002. Liu et al., 2018).

For the scatter plot of SNA versus BC (Fig. 6b), a less-changed SNA/BC slope from 2014 to 2018 somehow suggests the co-benefited reduction of short-lived climate forcers from mitigation of haze pollution in China. Since SNA are mainly secondary products due to oxidation of SO_2 and NO_x and neutralization of NH_3 (Pinder et al., 2007) and BC is a tracer of primary pollutants from combustion sources (Bond et al., 2013), the similar slope here also indicates that the overall proportion of secondary particles in $PM_{2.5}$ was less changed. However, the sulfate/$PM_{2.5}$ ratio was significantly reduced from 2014 to 2018 (Fig. 6c), linked to the remarkable reduction of its precursor SO_2 as shown in Fig. 2b. While for nitrate, an overall increased nitrate/$PM_{2.5}$ ratio could be clearly seen in recent years, especially for 2018, despite of a moderate decreasing of NO_2 (Fig. 2d). It has been well known that $PM_{2.5}$ has a nonlinear response to the reduction of sulfate because decreases in sulfate may increase aerosols when more nitric acid may enter the aerosol phase (West et al., 1999; Pinder et al., 2007; Liu et al., 2019). Here the regional scale sulfate reduction should have increased the availability of NH_3 for the formation of nitrate (Pathak et al., 2009; Huang et al., 2012; Wang et al., 2011; Liu et al., 2018; Liu et al., 2019). In addition, the increased atmospheric oxidization capacity will also enhance the formation of nitrate (Wen et al., 2018; Liu et al., 2019).

To further investigate the change in SNA and BC associated with air masses from main source regions in eastern China, we carried out back-trajectory cluster analysis for the months of NDJ during 2013-2018 (Fig. S1). In Fig. 7, we show the pie charts of SNA and BC in $PM_{2.5}$ at SORPES between 2013 and 2017, i.e. the beginning and end of the first 5-year "Ten measures", for air masses originated from NCP (Figs. 7a,d), Central-Eastern China plain (Figs. 7b,e), and the YRD (Figs. 7c,f), respectively. The results show a significantly higher reduction (-52.3%) in the air masses from NCP than the other two (~32%), indicating more strict and efficient control in North China (Liu et al., 2019; Li et al., 2019). For the fraction of chemical composition, sulfate show significant decrease of from NCP (from 23% to 19%) and YRD (from 18% to 14%), but nitrate show significant increase (from 24% to 29% for YRD air masses and to 32% for air masses from the two regions in the north). While ammonium shows 1% increase in all the three regions. These results confirmed the increased availability of NH_3 for the formation of SNA (Pathak et al., 2009; Liu et al., 2018, Liu et al., 2019; Li et al., 2019), and suggest a stricter NH_3 emission reduction as a potentially efficient means for further $PM_{2.5}$ mitigation in the eastern China.

Meteorological conditions, especially transport and dispersion, could significantly influence PM$_{2.5}$ concentration (Ding et al., 2013ab; Zhang et al., 2016). From the air quality management perspectives, it is very important to quantify the influence of emission reduction and year-to-year change in meteorology based on the observed trend of air pollutants. Because NDJ are the three

5   months with highest PM$_{2.5}$ concentration at SORPES (Fig. 3a), the trend during these months should also dominate the overall trend in annual average. In addition, the total precipitation during this period were relatively low (upper panel of Fig. 7), indicating a limited influence of wet deposition. Considering the limitation of LPDM in characterizing wet deposition and also secondary PM formation, we only chose NDJ to conduct the LPDM simulations based on the

10   fixed MIX emission inventory for 2010 (M. Li et al., 2017) to quantify the impacts from emission reduction and from the change in meteorology. To better characterize the influence of year-to-year differences in meteorology alone based on LPDM, we scaled the simulation results to make the modeled median values equal to observation for the first year, i.e. November 2011-January 2012. This procedure removes other systematic differences between the LPDM

15   simulation and the observation, e.g. uncertainty of emission inventory, parameterization of boundary layer etc. in the model. In fact, for the results with all years scaled using this method, the LPDM simulations could well reproduce the day-to-day variation of PM$_{2.5}$ (Fig. S2). The good agreement implies that the primary and secondary PM$_{2.5}$ had a consistent change with day-to-day weather, which controls the transport and dispersion of pollutants.

20   Fig. 8 shows the trends of observed PM$_{2.5}$ concentration at the SORPES station and the scaled LPDM simulation results to the first-year observation (i.e. 2011/2012) in NDJ from 2012 to 2018. The observations in the three months show a consistent trend with the annual average given in Fig. 1a. From the LPDM simulation results based on fixed emission inventory, we can find that the meteorology-influenced PM$_{2.5}$ had strong year-to-year variation, with a minimum

25   medium value as 84.8 µg m^{-3} in 2011/2012 to 108.4 µg m^{-3} in 2013/2014, i.e. with a year-to-year difference up to 28%. The difference in the two years were mainly due to different transport and dispersion patterns related to different large-scale circulations (Fig. 9). In the NDJ of 2013/2014, the eastern China were dominated by a stagnant High isolated from the continental High, causing more sub-regional influence of air masses in the eastern China (Fig. 9d) than the NDJ of

30   2011/2012. For the LPDM simulations with fixed emission inventory, an increasing trend (about 1.11 %/yr) existed for the entire 7 years but a moderate decreasing trend (-2.6%/yr) in the latest five years (Fig. 8). Here the distinctly different trends for the 5-year and 7-year periods indicate that the interannual variation could substantially influence the understanding of trend in air quality for a period less than one decade. For the period since 2013, i.e. when the "Ten

35   measures" took action, the observed decrease of PM$_{2.5}$ (-10.9%/yr) are mainly induced by emission reduction (76%) and the year-to-year change in mereology contributed to the left 24%,

i.e. about a quarter of the overall one. Here our estimation is consistent with that by Zhang et al. (2019), in which meteorology was estimated to contribute 20%-30% of the deceased trend in the YRD region during the period of 2013-2017.

5 **4 Conclusions**

In this study, we make a comprehensive analysis based on long-term continuous high-quality measurements of $PM_{2.5}$, relevant chemical compositions and precursors at a regional background station SORPES in Nanjing, eastern China since 2011. By utilizing Lagrangian dispersion modeling and various data, we quantified the impact of changes in different emission sources 10 and in year-to-year meteorology on the observed trends, especially after the implementation of Action Plan for Air Pollution Prevention and Control (i.e. the national "Ten measures" for air) in 2013. Main conclusions are given below:

1) We found a substantial reduction of $PM_{2.5}$ together with stronger reduction of SO_2 at the SORPES station during 2011-2018, and much faster decreasing trends (-9.1%/yr in $PM_{2.5}$ 15 and -16.7%/yr in SO_2) associated with concurrent reduction in sulfate (-10.6%/yr), nitrate (-5.8%/yr) and BC (-8.4%/yr) during 2013-2018, i.e. after the "Ten measures" implemented.

2) The early-summer agricultural straw burning has been found to be significantly reduced since 2013 in the eastern China. It significantly reduced $PM_{2.5}$ concentration at the 20 SORPES station during May-June, resulting in a change in the seasonal pattern of $PM_{2.5}$. In cold season (November-January), the fraction of nitrate in $PM_{2.5}$ was significantly increased, especially when air masses came from north. Besides an increased oxidization capacity, more NH_3 were available for nitrate formation in the condition of reduced sulfate associated with a substantial reduction of SO_2 are the main reason causing the 25 enhanced nitrate formation. A stricter NH_3 emission reduction is suggested for the further $PM_{2.5}$ mitigation in the eastern China.

3) The year-to-year difference in meteorological conditions could cause strong change in wintertime $PM_{2.5}$ concentrations by mainly influencing the transport and dispersion of air pollutants. The change in meteorology contributed to 24% of the observed decrease in 30 $PM_{2.5}$ at the SORPES station in November-January during 2013-2018, with the left 76% caused mainly by emission reduction.

This study demonstrates the unique role of long-term high-quality continuous measurement at a regional background station in the understanding of the impact of emission sources, chemical mechanisms and meteorology processes on the change of atmospheric components.

Based on comprehensive and in-depth analysis of the data, the long-term measurement can provide a quantitative understanding about the large-scale air quality measures and can also raise some insights on the policy making for future air pollution prevention and control.

5   **Author contributions.** AD designed this study, carried out the data analysis, and wrote the paper with contributions from all co-authors. XH participated the data analysis, modeling and plotted some figures. WN, XC, ZX, LZ, ZNX, YX, XQ, YS, PS, JW, LW, WQ and XZ conducted the measurements at the SORPES station.

Competing interests. The authors declare that they have no conflict of interest.

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Supporting Information

Figs. S1-2 and Table S1

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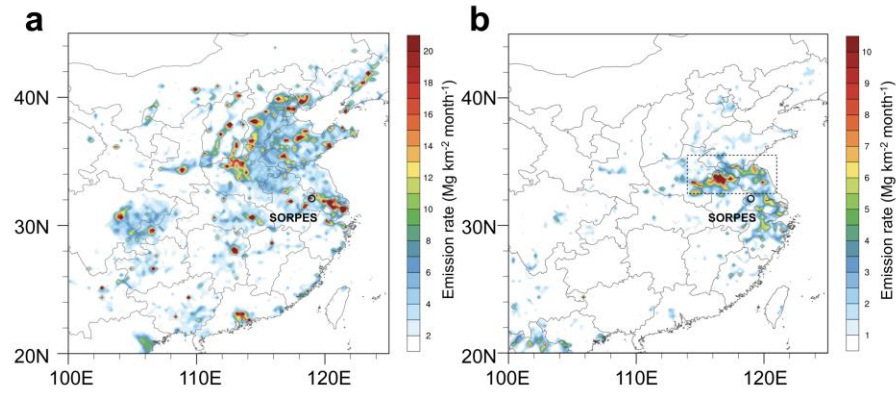
Figure 1. Spatial distributions of **(a)** anthropogenic emission of primary PM$_{2.5}$ in 2010 (Data from the MIX inventory available in MEIC database of Tsinghua University) and **(b)** averaged carbon emission from open biomass burning in May-June during 2012-2017 (Data from GFEDv4 available at https://daac.ornl.gov/cgi-bin/dsviewer.pl?ds_id=1293).
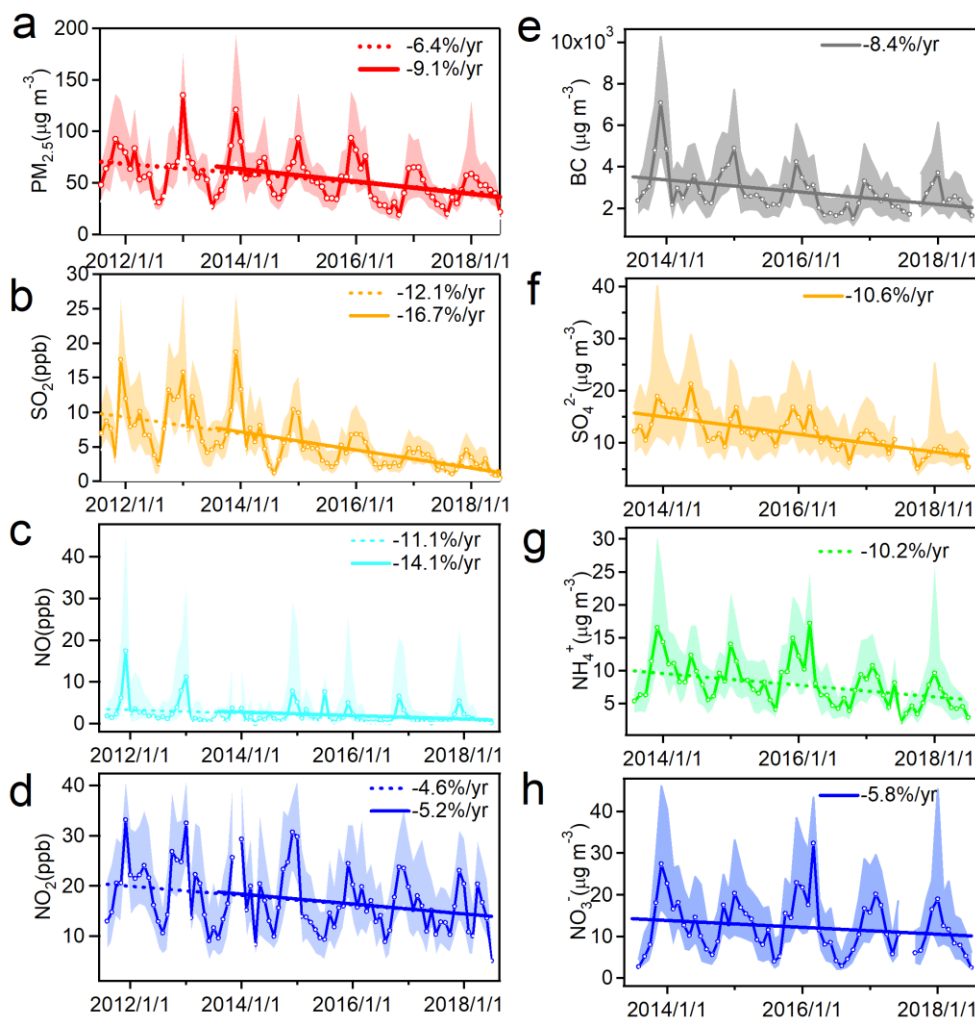
Figure 2. Monthly statistics and trends for **(a)** $PM_{2.5}$, **(b)** SO_2, **(c)** NO, **(d)** NO_2, **(e)** BC, **(f)** SO_4^{2-}, **(g)** NH_4^+, and **(h)** NO_3^- observed at the SORPES station. Note: For BC, SO_4^{2-}, NH_4^+ and NO_3^-, only data during August 2013-July 2018 are shown here. The solid lines marked with open cycles represent the monthly medium value and shaded areas mark the data from 25th to 75th percentiles. Dashed and solid lines show the linear regression fitting for data during 2011-2018 and 2013-2018, respectively.
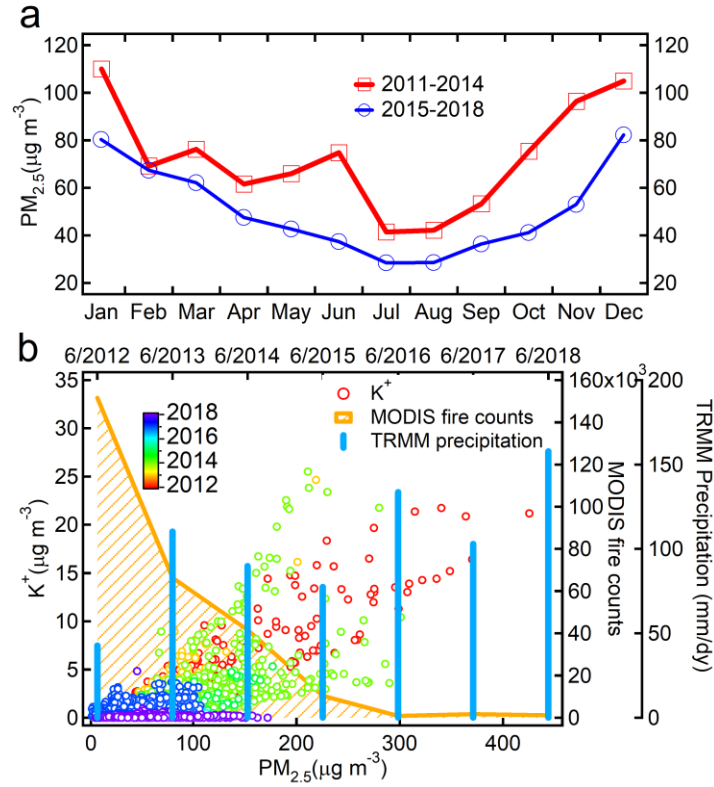
Figure 3. (a) Seasonal variation of PM$_{2.5}$ mass concentration measured at the SORPES station during 2011-2014 and 2015-2018, and **(b)** Scatter plots of K$^+$ as a function of PM$_{2.5}$ concentration measured at the SORPES station and the sum of MODIS fire counts and of TRMM precipitation in the BB domain (square in Fig. 1b) during 15 May-20 June, 2012-2018.
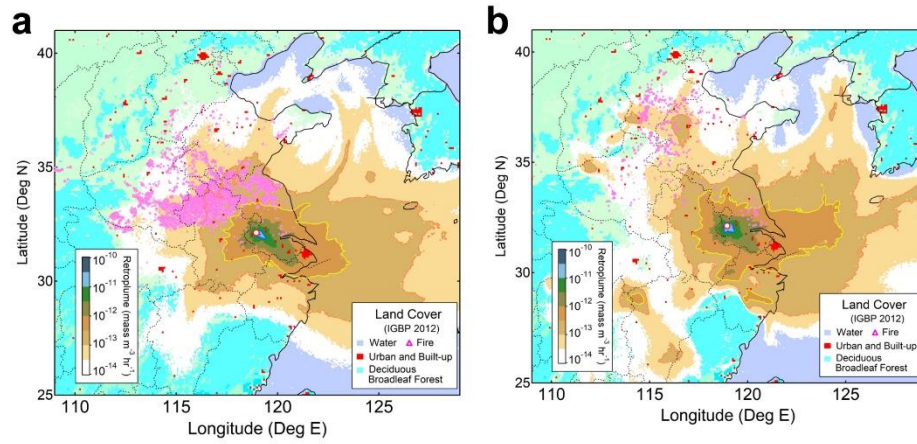
Figure 4. Averaged retroplume from 2-day backward Lagrangian dispersion modeling and MODIS fire counts for the period of 15 May- 20 June during **(a)** 2012-2014 and **(b)** 2016-2018.

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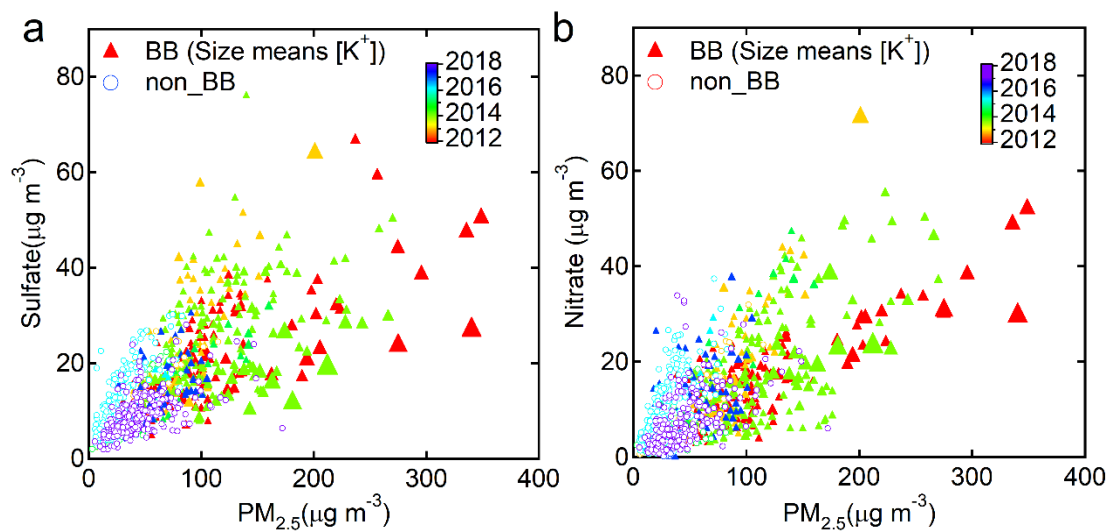
Figure 5. Scatter plot of **(a)** sulfate and **(b)** nitrate as a function of $PM_{2.5}$ for biomass burning (with K^+ higher than 75% percentile) and non-biomass burning (with K^+ lower than 25% percentile) cases for the period of 15 May- 20 June during 2012-2018.

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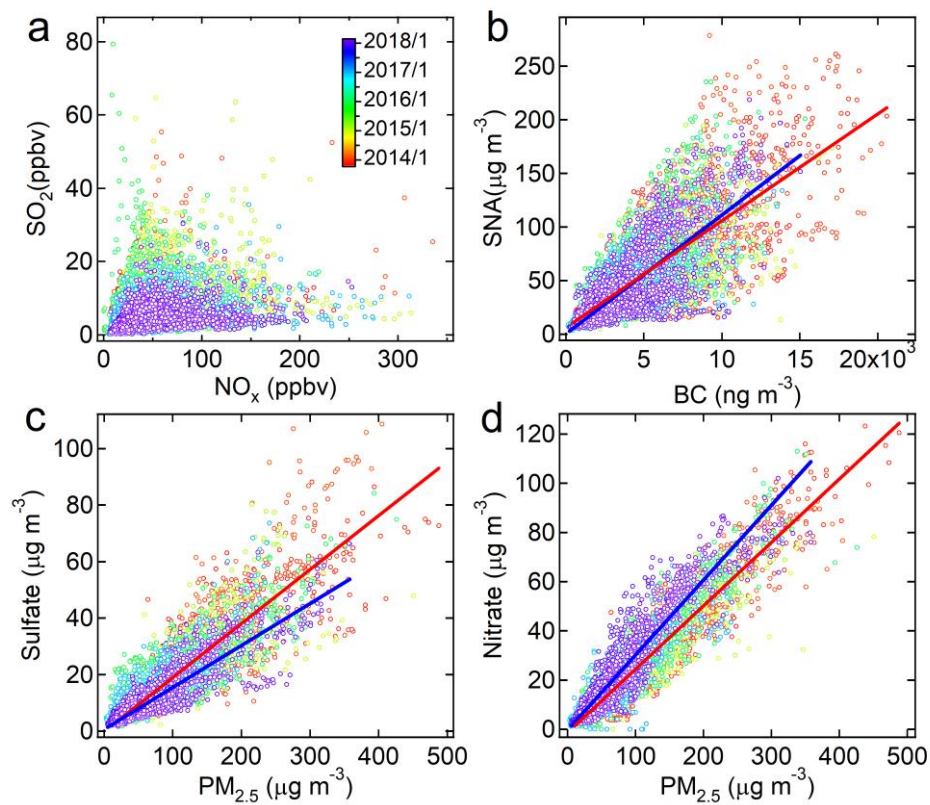
Figure 6. Scatter plots of **(a)** SO_2 versus NO_x, **(b)** SNA(i.e. the sum of sulfate, nitrate and ammonium versus BC), **(c)** SO_4^{2-} versus $PM_{2.5}$, and **(d)** NO_3^- versus $PM_{2.5}$ in November, December and January during 2013-2018.
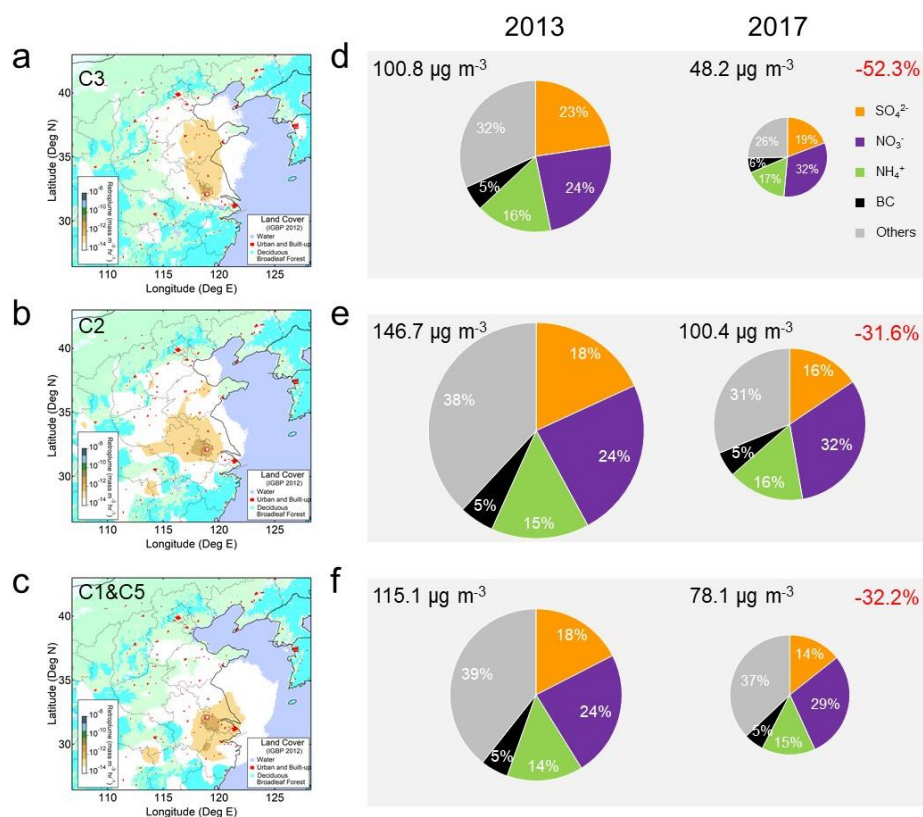
Figure 7. Averaged retroplume for air masses from **(a)** NCP (C3 in Fig. S1), **(b)** Central-Eastern Plain (C2 in Fig. S1), and **(c)** YRD (C1 & C5 in Fig.S1), and **(d-f)** a comparison of pie charts of SNA and BC in $PM_{2.5}$ for the three types of air masses in NDJ of 2013 (November and December 2013 and January 2014) and 2017 (November and December 2017 and January 2018), corresponding to the three transport pathways of different air masses given in (a-c). Note: Averaged $PM_{2.5}$ concentrations for the corresponding air masses and periods are shown in the top-left of each pie chart and also indicated by the size of the pie charts. The reduction rates between the two periods are shown in red color in the top left of right panel (in percentage).
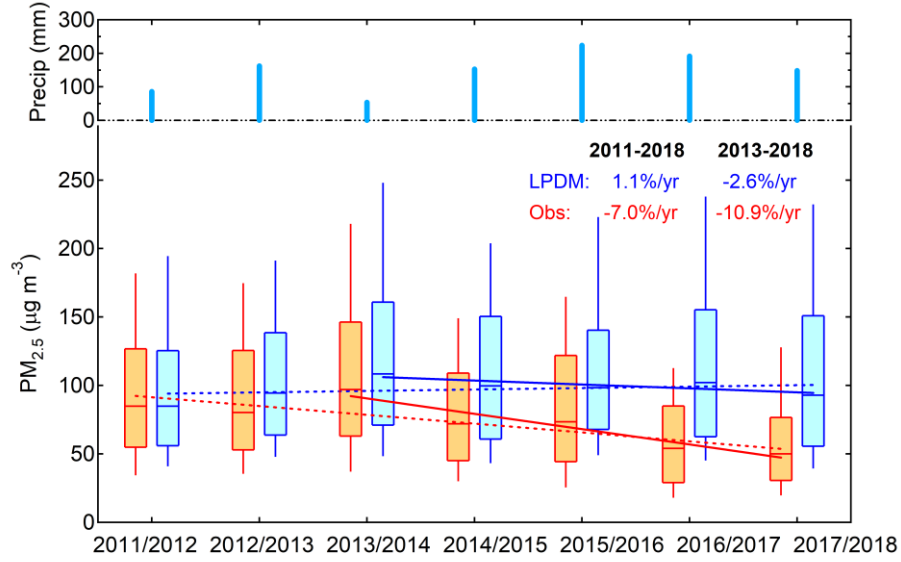
Figure 8. Statistics of PM$_{2.5}$ concentrations from the SORPES observation and the scaled LPDM simulations to the first-year observation (lower panel) and a sum of 3-month TRMM precipitation averaged in a 2°×2° grid (118° E-120° E, 31° N -33° N) around the SORPES station (upper panel) for November-December-January during 2011-2018.
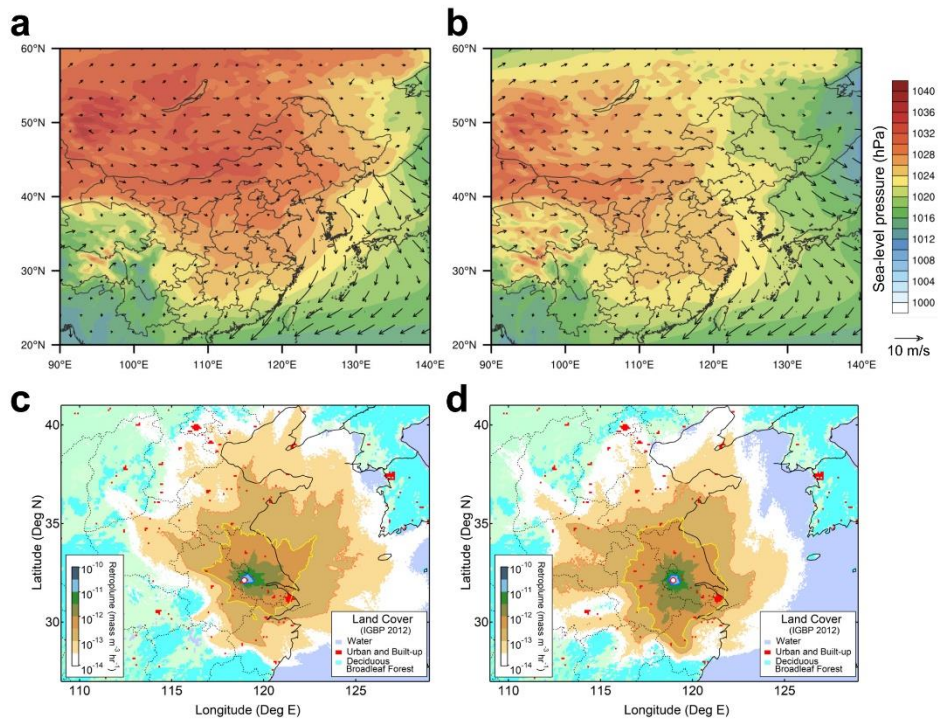
Figure 9. Averaged sea-level pressure and wind flows for **(a)** November 2011-January 2012 and **(b)** November 2013-January 2014, and **(c)** and **(d)** averaged retroplume from 2-day backward Lagrangian dispersion modeling for the corresponding period in **(a)** and **(b)**, respectively.

Supplementary Information

Table S1. Instrumentation, observational period and data coverage for main parameters measured at the SORPES station.

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Parameter	Instruments	Resolution	Period	Data coverage
$PM_{2.5}$	Thermo Fisher Scientific SHARP 5030	5 min	August 2011-July 2018	97.8%
SO_2	Thermo Fisher Scientific TEI-43i	5 min	August 2011-July 2018	98.3%
NO_2, NO_x	Thermo Fisher Scientific TEI-42i	5min	August 2011-July 2018	93.9%
BC	Magee Scientific AE31	5min	May 2013-July 2018	91.9%
SO_4^{2-}, NO_3^-, NH_4^+, K^+ etc.	Metrohm, MARGA	1 hr	May-June 2012 May 2013-July 2018	83.7%
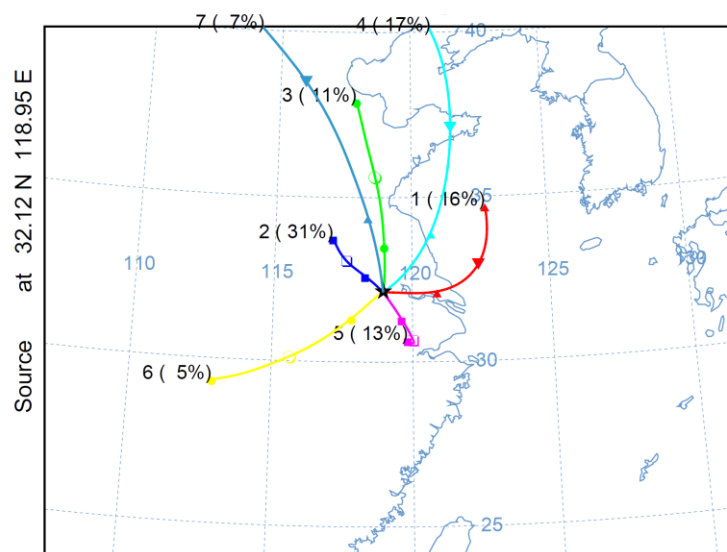
Figure S1. Cluster analysis for back-trajectories at SORPES station calculated using the HYSPLIT model for NDJ (November, December January) from November 2013 to January 2018. Note: 36-hr backward trajectories are used here. Total 7 clusters are identified and the percentage of each cluster is shown in the averaged trajectories. All trajectories were calculated for the altitude of 100 m above the SORPES station and GDAS data was used to drive the model.
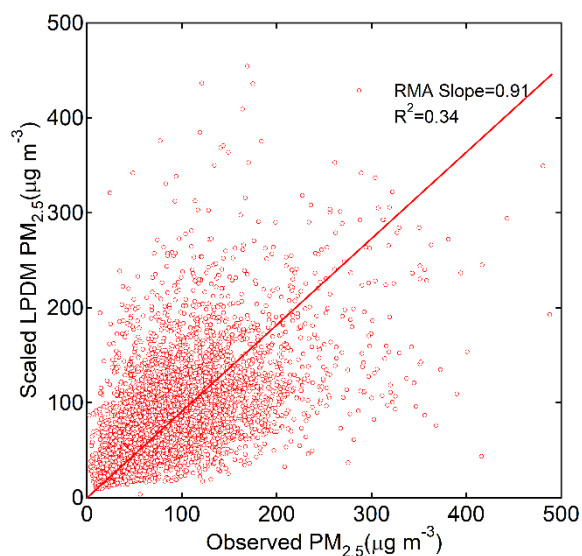
Figure S2. Scatter plot of scaled LPDM simulated PM$_{2.5}$ versus observations at the SORPES station for the period of November-January during 2013-2018. Note: Here the "scaled" means for each month the simulation result was adjusted to have the medium value matched with observation.

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