Aerosol Vertical Mass Flux Measurements During Heavy Aerosol Pollution Episodes at a Rural Site and an Urban Site in the Beijing Area of the North China Plain

Renmin Yuan1, Xiaoye Zhang2,*, Hao Liu1, Yu Gui1, Bohao Shao1, Xiaping Tao5, Yaqiang Wang2, Junting Zhong2, Yubin Li3 and Zhiqiu Gao3

1School of Earth and Space Sciences, University of Science and Technology of China, Anhui, 230026, China
2State Key Laboratory of Severe Weather & Key Laboratory of Atmospheric Chemistry of CMA, Chinese Academy of Meteorological Sciences, Beijing 100081, China
3School of Geography and Remote Sensing, Nanjing University of Information Science and Technology, Nanjing 210044, China
4Center for Excellence in Regional Atmospheric Environment, IUE, CAS, Xiamen 361021, China.
5School of Physical Sciences, University of Science and Technology of China, Anhui, 230026, China

Correspondence: Renmin Yuan (rmyuan@ustc.edu.cn) and Xiaoye Zhang (xiaoye@cma.gov.cn)

Abstract:

Due to excessive anthropogenic emissions, heavy aerosol pollution episodes (HPEs) often occur during winter in the Beijing-Tianjin-Hebei (BTH) area of the North China Plain. Extensive observational studies have been carried out to understand the causes of HPEs; however, few measurements of aerosol vertical fluxes exist, despite them being the key to understanding vertical aerosol mixing, specifically during weak turbulence stages in HPEs. In the winter of 2016 and the spring of 2017, based on the light propagation theory and surface-layer similarity theory, aerosol vertical mass fluxes were measured by combining large aperture scintillometer (LAS) observations, surface PM$_{2.5}$ and PM$_{10}$ mass concentrations, and meteorological observations, including temperature, relative humidity (RH), and visibility, at a rural site in Gucheng (GC), Hebei Province, and an urban site at the Chinese Academy of Meteorological Sciences (CAMS) in Beijing located 100 km to the northeast. The near-ground aerosol mass flux was generally lower in winter than in spring and weaker in rural GC than in urban Beijing. This finding provides direct observational evidence from the perspective of vertical aerosol fluxes for a weakened turbulence intensity in winter and in polluted areas such as GC. The HPEs included a transport stage (TS), an accumulation stage (AS), and a removal stage (RS). During the HPEs from January 25, 2017 to January 31, 2017, in Beijing, the mean mass flux decreased by 51% from 0.0049 mg m$^{-2}$s$^{-1}$ in RSs to 0.0024 mg m$^{-2}$s$^{-1}$ in the TSs. During the ASs, the mean mass flux decreased further to 0.00087 mg m$^{-2}$s$^{-1}$, accounting for approximately 1/3 of the flux in the TSs. A similar reduction from the TSs to ASs was observed in the HPE from December 16, 2016 to December 22, 2016 in GC. The weakened mass flux indicates that the already weak turbulence would be further weakened by aerosol pollution to a certain extent, which would further facilitate aerosol accumulation.
1 Introduction

Recently, due to the country’s rapid development of industrialization and urbanization, China has experienced heavy aerosol pollution episodes (HPEs), particularly in the Beijing, Tianjin and Hebei (BTH) region, which is one of the most polluted areas in China (Zhang et al., 2012). The HPEs often last for a long duration in the BTH region and cover a wide area, particularly in winter; they also severely reduce near-ground visibility (Lei and Wuebbles, 2013) and can have detrimental effects on public health (He et al., 2018; Cao et al., 2012). This heavy pollution weather has received extensive attention in recent years, and many observational studies have been carried out (Zhong et al., 2018a; Sun et al., 2014; Wang et al., 2015; Guo et al., 2011; Zhang et al., 2009b; Huang et al., 2014). Modelling studies have also been performed to examine the regional transport of pollutants (Wang et al., 2014) and to study the important role of large-eddy convective turbulent mixing in the vertical transfer of pollutants from a field campaign in Beijing (Li et al., 2018). However, few studies on the turbulence contribution of the aerosol transport flux in the surface layer have been conducted.

Ground pollutant emissions are known as the main source of aerosols in the atmosphere. However, in previous studies, no measurements of ground emissions during heavy pollution events were collected. Surface emission data are currently required for model verification and pollution predictions, and these data are primarily obtained through emission inventories (Wu et al., 2012; Bond et al., 2004). The establishment of emission inventories is primarily based on emission activity and emission factor (EF) data (Akagi et al., 2011; Lu et al., 2011; Roden et al., 2006; Zhang and Tao, 2009). Emissions data are mainly obtained from statistical yearbooks (Zhang et al., 2009a). Some studies have used fixed EFs while others have implemented dynamic EFs (Bond et al., 2004; Zhang et al., 2009a). Many factors are considered in dynamic EFs, such as the size of a city, the degree of economic development, the type of fuel, the type of technology, the consumption of a product, the control technology, and so on, as well as estimates based on actual measured meteorological parameters and aerosol parameters (Chen et al., 2015; Karvosenoja et al., 2008; Shen et al., 2013). A numerical model has also been used to estimate average fleet emission factors in typical urban conditions (Ketzel et al., 2003; Krecl et al., 2018). The error in aerosol fluxes based on the use of emission inventories is very large (Liu et al., 2017; Zheng et al., 2017). Emission inventories constructed using the EF method provide only the total emission amount of atmospheric pollutants within a region. However, the emission data should be gridded to a suitable scale for air quality modelling and pollution predictions. Thus, near-surface aerosol emission data with a higher temporal and spatial resolution are urgently needed.

Many methods have been used to obtain aerosol flux data. For the upward transport of aerosols near the surface layer, the aerodynamic method was adopted in the early years, and the aerosol concentration gradient at different heights was measured and then calculated based on the similarity theory of the near-surface layer or calculated by the boundary layer box model, which can be based on meteorological data (Ceburnis et al., 2016; Hourdin et al., 2015; Zhang and Li, 2014). The emission rates of bioaerosols were also estimated from spore counts and molecular tracers (Elbert et al., 2007). The abundance of microbes and meteorological data were measured, and an estimate may be derived of the sea-air exchange of microorganisms (Mayol et al., 2014).

With the use of instruments for measuring the number of aerosol particles during recent years (for example, the GP-WCPC3787 particle counter by TSI), the eddy covariance (EC) method has been applied, and measurements of the aerosol particle number flux have become possible. The
vertical transport flux of the aerosol particle number density \( F_p \) is denoted as a cross-correlation between the aerosol particle number concentration \( N \) and the vertical wind speed \( w' \) (Ripamonti et al., 2013). Based on this principle, the vertical velocity fluctuations and the fluctuations in the aerosol particle number density can be measured. As a result, the vertical transport flux of the aerosol particle number density has been measured in many cities, such as in Toronto, Canada (Gordon et al., 2011), Stockholm, Sweden (Vogt et al., 2011b), Helsinki, Finland (Ripamonti et al., 2013), London, UK (Harrison et al., 2012), the Blodgett Forest Observatory in the United States (Farmer et al., 2011), and measurements of sea salt aerosol fluxes in northern Europe (Brooks et al., 2009; Sproson et al., 2013). These results have shown the quantitative relationship among urban aerosol fluxes, urban vehicle emissions, and meteorological conditions (Jarvi et al., 2009) and have been used to determine sea salt aerosol transport characteristics and provide further knowledge of aerosol properties (Nemitz et al., 2009). These measurements have been mainly collected in cities because of their anthropogenic contributions to aerosol emissions. These data can be used as routine model inputs. Direct eddy covariance measurements of aerosol exchanges in tropical forests, where primary biological aerosol particles represent a substantial fraction of the airborne particulate matter (Graham et al., 2003), were also performed by Ahlm et al. (2010a and 2010b) and Whitehead et al. (2010), potentially giving a proxy for microbial emissions in tropical ecosystems.

Although measurements of urban aerosol particle number density fluxes have been collected, the current eddy correlation method only provides fluxes for the aerosol particle number density at a point. We know that the underlying surface of a city is very complex, and thus the aerosol particle flux is not homogeneous in the horizontal. For a complex underlying surface such as a city, these point measurements are not very representative. Therefore, it is of great importance to design an aerosol flux measurement system with an accurate spatial representation.

The use of eddy correlation principles to measure sensible heat fluxes has been widely performed (Lee, 2004). Current sensible heat fluxes can also be obtained using a large aperture scintillometer (LAS) based on the light propagation theory and similarity theory (Zeweldi et al., 2010). This configuration makes it possible to achieve aerosol mass flux measurements using the light propagation theory and similarity theory. Recently, we measured the imaginary part of the atmospheric equivalent refractive index structure parameter based on the light propagation theory (Yuan et al., 2015). The results showed that the imaginary part of the atmospheric equivalent refractive index structure parameter is related to turbulent transport and the spatial distribution characteristics of aerosols. Experiments also showed that there is a strong correlation between the imaginary part of the atmospheric equivalent refractive index and the mass concentration of aerosol particles (Yuan et al., 2016). Thus, similar to the temperature structure parameter reflecting the sensible heat flux, the structural parameter of the imaginary part of the atmospheric equivalent refractive index can reflect the mass flux of aerosol particles. This paper attempts to measure the aerosol mass flux in the BTH area.

To gain a deeper understanding of the interaction between atmospheric heavy pollution and weather in the BTH region, joint observations have been carried out in the BTH region since the winter of 2016 (Zhong et al., 2018b; Zhong et al., 2018a; Wang et al., 2018a; Shen et al., 2018). The observations reveal the large-scale and mesoscale transport processes of aerosols between heavy pollution episodes (HPEs) in the BTH region in the winter of 2016. Most HPEs in the BTH region are due to horizontal transport and unfavourable meteorological conditions (Zhong et al., 2018a). However, during HPEs, no research has been conducted in the BTH area on quantifying the
contribution of surface emissions to the concentration of pollutants. In this study, we focus on HPEs through field observations of aerosol transport based on the light propagation theory and surface similarity in the Beijing urban district and Gucheng suburban area. The second section of this paper introduces the theory of aerosol vertical transport flux measurements, the third section introduces the experiment, the fourth section gives the experimental results, and finally, the conclusion and discussion are presented in the fifth section.

2 Theory and methods

The theory for calculating the vertical flux of aerosol particles and the theory for calculating the friction velocity and characteristic temperature using the temperature and wind profiles is presented in the following subsections.

2.1 Calculation of the aerosol mass vertical flux

According to the micrometeorological principle (Stull, 1988), similar to the estimation method of the sensible heat flux, the aerosol flux $F_a$ can be obtained as follows:

$$ F_a = u_1 M_a $$

(1)

where $u_1$ is the friction velocity, which can be obtained from the temperature and wind speed profiles or directly from three-dimensional wind speed measurements; see Sec. 2.2. Prior experiments have shown that the motion characteristics of aerosol particles in the atmosphere approximate the motion of general scalars (Martensson et al. 2006; Vogt et al. 2011b). Therefore, aerosol particles can be approximated as scalars, and characteristic parameters $M_a$ similar to the scalars can be introduced, which can be regarded as the atmospheric aerosol mass concentration scale in the surface layer and deduced from surface layer similarity theory. This approximation is similar to the surface-layer temperature scale (Stull, 1988) as follows:

$$ \frac{C_a^2 (z-d)^{\nu_3}}{M_a^2} = \eta(\xi) $$

(2)

where $z$ is the measurement height, $d$ is the zero-displacement height (Evans and De Bruin, 2011; Hartogensis et al., 2003), $\xi = (z-d)/L$ is the nondimensional stability parameter, $L$ is the Monin-Obukhov (M-O) length and defined as $L = \frac{\bar{T} u_1^2}{\kappa g \bar{T}}$ (Stull, 1988), $\bar{T}$ is the average temperature, $T_s$ is the surface-layer characteristic temperature, $\kappa$ is the von Karman constant, which is 0.4, and $g$ is acceleration due to gravity. The stability function ($\eta(\xi)$) can be expressed as follows depending
on the stability condition (DeBruin et al., 1995):

\[ \eta(\xi) = a_1[1 - a_2^2 \xi]^{2/3} \]  

(3)

for unstable conditions ($\xi < 0$), and the following:

\[ \eta(\xi) = b_1[1 + b_2(\xi)^4] \]  

(4)

for stable conditions ($\xi \geq 0$) (Wyngaard et al., 1971).

In Eqs. (3) and (4), $a_1$, $a_2$, $b_1$, $b_2$ and $e_1$ are constants, and different experiments have provided different values, although the differences between these results are small. Here, we take the parameters $a_1 = 4.9$, $a_2 = 9$, $b_1 = 5$, and $b_2 = 0$ (DeBruin et al., 1995).

$C^\nu_{M}$ in Eq. (2) is the aerosol mass concentration structure parameter. We assume that the aerosol particles in the atmosphere follow the movement of the air and satisfy the turbulent motion law. Previous studies have shown that the particle concentration fluctuation spectra follow a ‘5/3’ power law under unstable stratification conditions (Martensson et al., 2006; Vogt et al., 2011b), and the velocity-concentration co-spectra follows a ‘4/3’ power law (Martensson et al., 2006; Vogt et al., 2011a; Kaimal et al., 1972). Thus, the distribution of small particles can be considered as a conservative passive quantity analogous to the temperature. Then, at a separation ($r$) of the order in the inertial subrange in a locally isotropic field, the aerosol mass concentration (denoted as $M_a$) structure function ($D_{M_a}(r)$) follows a “2/3 law” (Wyngaard, 2010) and can be expressed as

\[ D_{M_a}(r) = [M_a(x) - M_a(x + r)]^2 = C^\nu_{M} r^{2/3} \],

where $x$ is the position vector, $r$ is the separation vector, and the overbar indicates the spatial average.

The following describes the method to deduce the aerosol mass concentration structure parameter $C^\nu_{M}$.

We assume that the aerosol particles are continuously dispersed in the air. The aerosol particles and gases in the atmosphere can be considered as an equivalent medium, and an atmospheric equivalent refractive index (AERI) $n_{eq}$ is introduced that contains the real part $n_r$ and the imaginary part $n_i$ of the equivalent refractive index. Thus, $n_{eq} = n_r + i n_i$. For visible light, the attenuation of light by gases in the atmosphere is very weak; the cause of the attenuation is the absorption and scattering due to aerosol particles. Therefore, the real part of the equivalent medium of aerosol particles and gases is determined by the gas composition of the air. The fluctuation of the real part is mainly determined by temperature fluctuations; the imaginary part is determined by the aerosol
particles, and the fluctuation of the imaginary part is determined by fluctuations in the aerosol concentration.

For visible light, there is a strong linear relationship between the variation of the real part of the AERI and the variation of the atmospheric temperature, i.e., \( R_n = \frac{\delta T}{\delta n_{Re}} \); thus, we have the following:

\[
R_n = -1.29 \times 10^4 \times (1 + \frac{7.52 \times 10^3}{\lambda}) \frac{\overline{T'}^n}{\overline{P}} \tag{5}
\]

which is based on the relationship between the real part of the AERI \( (n_{Re}) \) and atmospheric temperature (Tatarskii, 1961). Because the wavelength is deterministic, the ratio \( R_{TN} \) can be obtained by measuring the atmospheric temperature. The imaginary part of the AERI has a close correspondence with the extinction coefficient of the equivalent medium, and the extinction coefficient is inversely proportional to the visibility. Higher concentrations of aerosols in the atmosphere are related to lower visibility and vice versa; thus, the relationship between the imaginary part of the AERI and the atmospheric aerosol mass concentration can be established. The ratio of the atmospheric aerosol mass concentration to the imaginary part of the AERI \( R_{MN} \) can be defined as follows:

\[
R_{MN} = \frac{M}{n_{im}}. \tag{6}
\]

Theoretical analysis has revealed that \( R_{MN} \) is associated with the aerosol particle size distribution, mass density of the aerosol particles, and the aerosol particle refractive index. Because of the relatively small variations in particle size and aerosol refractive index (Dubovik et al., 2002), \( R_{MN} \) can be treated as a constant for surface-layer aerosols at a given location. \( M_n \) approximates the PM$_{10}$ value. The variable \( n_{im} \) can be calculated as follows (Yuan et al., 2016):

\[
n_{im} = \frac{0.55e^{-6} \times 3.912}{4\pi} \frac{L_v}{L'} \tag{7}
\]

where the unit of visibility \( (L_v) \) is m.

According to Eqs. (5) and (6), we have the following:

\[
C_n^2 = R_{TN}^2 C_{n,Re}^2 \tag{8}
\]

\[
C_{im}^2 = R_{MN}^2 C_{n,Im}^2 \tag{9}
\]

Thus, the temperature structure parameter \( C_n^2 \) and the aerosol mass concentration fluctuation structure parameter \( C_{im}^2 \) are converted into the measurement of the real and imaginary structure.
parameters of the AERI.

The measurement of relevant parameters is performed based on the light propagation theory. When light is transmitted in an equivalent medium, the AERI fluctuation will cause fluctuations in light intensity. Theoretical and experimental results have shown that the intensity fluctuation can be decomposed into high-frequency and low-frequency fluctuations. The high-frequency fluctuations are determined by fluctuations of the real part of the AERI, and the low-frequency fluctuations are determined by fluctuations of the imaginary part of the AERI. Thus, the real and imaginary structure parameters of the equivalent refractive index can be calculated by our developed LAS.

So far, we have completed the estimation of the aerosol mass transport flux.

According to the previous derivation and analysis, there are two calculation schemes for determining the aerosol mass flux as follows:

\[
F_{d1} = \left( \frac{C_{n,\text{Im}}^2}{C_{n,\text{Re}}^2} \right)^{1/2} \frac{R_{MN}}{R_{PN}} u[T]\]  
\[
F_{d2} = u_x \sqrt{\frac{C_M 2^{2/3}}{\eta(x)}} = u_x R_{MN} \sqrt{\frac{C_{n,\text{Im}}^2 2^{2/3}}{\eta(x)}}
\]

When the free convection approximation (\(-\xi \gg 1\)) is assumed, based on the definition of the M-O length, and the similarity theory (Wyngaard et al., 1971), the following can be obtained:

\[
F_{d3} = a \left( \frac{\xi}{U} \right)^{1/2} R_{TN}^{1/2} (C_{n,\text{Re}}) \frac{1}{4} R_{MN} \left( C_{n,\text{Im}}^2 \right)^{1/2} (z - d)
\]

where the coefficient \(a = \frac{3}{4} d_1^{1/4} \frac{1}{4} K^{1/2}\) can be taken as 0.567 (DeBruin et al., 1995; Lagouarde et al., 2006). Eqs. (10)-(12) are the theoretical basis for the aerosol mass flux measurements.

According to Eqs. (10)-(12), the vertical transport flux of aerosol particles is related to the strength of turbulent fluctuations and aerosol mass concentration fluctuations.

### 2.2 Calculation of the friction velocity and surface-layer characteristic temperature

To calculate the aerosol vertical transport flux, according to Eq. (10), the values of the friction velocity \(u^*\) and the characteristic temperature \(T^*\) are required. These can be obtained via wind speed and temperature profile data. From the near-surface similarity theory, the temperature and wind speed data measured at two heights of \(z_1\) and \(z_2\) can be used in the expressions of the friction velocity \(u^*\) and the characteristic temperature \(T^*\) (Stull, 1988) as follows:

\[
u^* = \frac{k[U(z_2) - U(z_1)]}{\ln \frac{z_2}{z_1} - \Psi_v(\xi_1) + \Psi_v(\xi_2)}
\]
\[ T' = \frac{a(T(z_2) - T(z_1))}{0.74[\ln(\frac{z_2}{\xi}) - \Psi_u(\xi) + \Psi_T(\xi)]} \]  

where \( T(z_1) \) and \( T(z_2) \) are the measured velocities at heights \( z_1 \) and \( z_2 \), respectively, \( T(z_1) \) and \( T(z_2) \) are the measured temperatures at heights \( z_1 \) and \( z_2 \), respectively, \( \xi_1 \) and \( \xi_2 \) are the stabilities at heights \( z_1 \) and \( z_2 \), respectively, and \( \Psi_u \) and \( \Psi_T \) are the correction terms for the velocity and temperature profiles under the condition of stability \( L \). Under unstable conditions (Stull, 1988), we have the following:

\[ \Psi_u(\xi) = \ln[(1 + x^2)[1 + x^2/2] - 2\arctan(x) + \frac{\pi}{2}], \quad x = (1 - 15\xi)^{1/4} \]  

\[ \Psi_T(\xi) = \ln[(1 + y^2)/2], \quad y = (1 - 9\xi)^{1/2} \]

Under stable conditions (Cheng and Brutsaert, 2005), we have the following:

\[ \Psi_u(\xi) = -a\ln[\xi + (1 + \xi^3)^{1/3}], \quad a=6.1, \quad b=2.5. \]

\[ \Psi_T(\xi) = -c\ln[\xi + (1 + \xi^3)^{1/3}], \quad c=5.3, \quad d=1.1. \]

Based on Eqs. (13)-(18), the friction velocity \( u^* \) and characteristic temperature \( T^* \) can be determined.

3 Measurements and data processing

3.1 Introduction of Experiments

Observations were collected at two locations (two rectangles in Fig. 1a) from December 2016 to March 2017: a rural site in Gucheng (GC site), Hebei Province and an urban site at the Chinese Academy of Meteorological Sciences (CAMS site) in Beijing. The distance between the two locations is approximately 100 km. According to the theoretical methods defined in the preceding section, to estimate the aerosol transport flux, the ratio of the aerosol mass to the imaginary part of the AERI, the ratio of the temperature to the real part of the AERI, the real and imaginary parts of the atmospheric equivalent refractive index structure parameter (AERISP), the friction speed, and the characteristic temperature must all be obtained. If the free convection condition is satisfied, fewer parameters are required, including the real and imaginary parts of the AERISP, the ratio of the aerosol mass to the imaginary part of the AERI, the ratio of the temperature to the real part of the AERI, and the atmospheric temperature.

Two sets of LASs developed by our research group were installed at the top of the building of the Beijing Institute of Meteorological Sciences (point A in Fig. 1b) and at the top of a two-story building in the farm of the Central Meteorological Bureau of Gucheng Town, Baoding City (point D in Fig. 1c). The light intensity sampling frequency of the receiving end was 500 Hz, and a file was recorded every 20 minutes. Then, the real and imaginary parts of the AERISP were calculated.

In the CAMS site, the transmitter end of the LAS was placed on the roof of a building on the east side of the Institute of Meteorological Sciences, and the receiver end was placed at the top of the Chinese Academy of Meteorological Sciences. The propagation path was along an east-west direction. The distance between the two ends was 550 metres as shown in Fig. 1d. The light beam
passed over urban buildings, residential areas and urban roads. The beam height was 43 metres. The average height of the building below the beam was 24 metres, thus, the zero-displacement was 18 metres \((24 \times 0.67 = 18)\) (Leclerc and Foken, 2014), and the effective height of the beam was 25 metres. At the Beijing observation point, the temperature and wind speed of the near-surface atmosphere were measured simultaneously, and the measurement heights were 1.5 m and 10 m, respectively. To calculate the aerosol flux, it is necessary to obtain the ratio of the aerosol mass to the imaginary part of AERI and to measure the aerosol mass concentration and visibility. In Haidian District, there is a site to measure the visibility of the near-surface layer (point B in Fig. 1b), and the PM\(_{10}\) mass concentration measurements were collected at Guanyuan Station (see point C in Fig. 1B). The sampling interval for the visibility and PM\(_{10}\) mass concentration measurements was 1 h. The measurement height of points B and C in Fig. 1b was approximately 20 metres. The ratio of the aerosol mass PM\(_{10}\) to the imaginary part of the AERI was calculated based on the data. The measurements were collected at the GC site from January 15, 2017 to March 20, 2017.

In the GC site (point D in Fig. 1c, namely, the LAS position) in Gucheng, Baoding, Hebei, the transmitter of the LAS was placed on the roof of a two-story building with a height of 8 m, and the receiving end was located in a room in a three-story building on the west side of National Highway 107 at the same height as the transmitting end. The distance between the transmitting end and the receiving end was 1670 metres. The terrain between the transmitting end and the receiving end was flat, with farmland, a national road and sporadic trees below the beam, as seen in Fig. 1e. Near the light beam, there was a 30-metre-high meteorological observation tower, in which the temperature, relative humidity (RH), and wind speed were measured at 5 levels (1 m, 3 m, 8 m, 18 m, and 28 m). The friction speed and characteristic temperature were calculated according to the temperature wind speed profile. Visibility observations were made in Xushui District near the LAS position (see point E in Fig. 1c). The PM\(_{10}\) mass concentration was measured in Beishi District (see point F in Fig. 1c).

From Fig. 1c, the three observation points (points D, E and F in Fig. 1c) formed a nearly straight line and were distributed in a northeast-southwest direction. During the experimental observation period, a northeast-southwest wind prevailed; thus, the Xushui District visibility data and Beishi District PM\(_{10}\) data can approximate the situation of the scintillometer position. The measurements were collected at the GC site from November 17, 2016 to March 30, 2017.

### 3.2 Data quality control

There are two types of variables, e.g., mean variables and fluctuation variables. Mean variables include temperature, wind speed, wind direction, PM\(_{10}\), and visibility for averages of 30 minutes or 60 minutes. Data quality control for the mean variables was conducted by comparing the measured data at different heights or from personal experience. All the measured mean data were determined to be adequate. Fluctuation variables include the high-frequency intensity fluctuation data measured by the LAS, the real and imaginary parts of the AERISP, and the calculated aerosol flux. Quality control mainly includes the elimination of spike and supplementing missing data.

Peaks in the light intensity fluctuation data appear because the received signal quickly increases when the light signal is blocked, such as due to birds along the transmission path. This situation is automatically determined by the data processing program. When this happens, the current 20-minute period is rejected. For the real and imaginary parts of the AERISP and the aerosol flux data, (a) 3 times the standard deviation (SD) of the anomaly and (b) 3 times the SD of the adjacent difference
were determined. The method for judging 3 times the SD of the anomaly was applied to obtain a
trend of two-hour averages. Then, the difference between the measured value and the trend at each
moment was calculated, and the mean and SD of the difference were also calculated. The data with
differences from the trend exceeding 3 times the SD were considered as spikes. The method for
judging the difference of 3 times the SD of the adjacent differences was to first calculate the
difference between adjacent observations and then calculate the mean and SD of the difference. Any
data whose adjacent difference deviated from the mean of the adjacent difference by more than 3
the SD was considered an error. Less than 5% of the data were considered to contain spikes or errors.
The data determined to be errors were supplemented by the average of the nearby observations.
Of course, if data were missing over a long period, the missing gap could not be filled. For this
situation, further supplementation was not considered.

Other errors in the measurements using a LAS due to specific reasons (Moene et al., 2009); for
example, the impact of the uncertainty in the exact shape of the turbulence spectrum and the
intermittent variations in the characteristics of that spectrum on the LAS signal were not considered
in this study.

4 Results

First, the visibility and PM$_{10}$ aerosol mass concentration results at the CAMS site and the GC
site are given and compared. Then, the characteristics of aerosol transport in typical weather
conditions at the CAMS site and the GC site are discussed. Finally, the aerosol flux characteristics
under heavy pollution weather conditions are analysed.

4.1 Relationship between $n_{st}$ and PM$_{10}$

To obtain the ratio of the atmospheric aerosol mass concentration to the imaginary part of the
AERI ($n_{st}$) $R_{AV}$, PM$_{10}$ and visibility were measured.

The maximum PM$_{10}$ concentration in the Baoding area appeared at 1:00 on January 28, 2017
(up to 1071 µg m$^{-3}$), and the maximum PM$_{10}$ concentration in the Beijing area appeared at 2:00 on
January 28, 2017 (up to 917 µg m$^{-3}$). This heavy pollution event swept through Beijing and the
surrounding areas, reaching a maximum at almost the same time. The visibility at the corresponding
time was less than 500 metres. The imaginary part of the AERI can be calculated from the visibility
(Yuan et al., 2016). Fig. 2a shows a scatter diagram of the imaginary parts of the AERI and PM$_{10}$
data measured in the Beijing area; there is a strong correlation between the AERI and aerosol particle
mass concentration, with a linear correlation coefficient of 0.96. The fitted linear in Fig. 2a has a
slope of 3845 kg m$^{-3}$. Therefore, $R_{AV}$ was taken as 3845 kg m$^{-3}$ for the Beijing area to estimate the
aerosol vertical transportation flux. Similarly, Fig. 2b shows the results for the Baoding area, and
$R_{AV}$ was set to 3711 kg m$^{-3}$ for the Baoding area to estimate the aerosol vertical transportation flux.
The two ratio coefficients are relatively close. Figs. 2a and 2b also show that in the case of light
pollution, Beijing’s $R_{AV}$ is slightly larger.

Furthermore, Figs. 2a and 2b show that although there is a large scattering between PM$_{10}$ and
$n_{st}$ that may be attributed to a large separation between the two measurement locations for visibility
and PM$_{10}$, there is a strong linear correlation between the imaginary part of the AERI and PM$_{10}$. The
imaginary part of the AERI has a slightly stronger correlation with the PM$_{10}$ data obtained in the
Baoding area than in the Beijing area.
The following provides the results of the aerosol transport flux under typical weather conditions in Beijing and Baoding for the period from March 10, 2017 to March 17, 2017.

4.2 Characteristics of aerosol flux in the Beijing region

To analyse the aerosol transport flux characteristics, we present the time series of the conventional meteorological parameters. The measurement site is the Beijing Meteorological Observatory, which is 20 metres above the ground. The time series of temperature, RH, wind speed, wind direction, PM$_{10}$, $C_{n,Rc}^{-2}$, $C_{n,Im}^{-2}$ and aerosol flux are shown in Figs. 3a-3h, respectively. The temperature has an obvious diurnal variation, indicating that this period had primarily sunny weather. The RH from March 10, 2017 to March 17, 2017, was less than 60%, and the RH for most of the time period was less than 30%. The wind speed was low; only during the period from March 11 to March 14 was the wind strong. At 6:00 on March 12, the maximum wind speed was 4.2 m s$^{-1}$. At that time, there was no dominant wind direction. Moreover, two light pollution events occurred (MEP, 2012) on March 11 and March 16, with PM$_{10}$ concentrations approaching 200 μgm$^{-3}$. From the data of $C_{n,Rc}^{-2}$ and $C_{n,Im}^{-2}$ in Figs. 3f and 3g, the real part of the AERISP $C_{n,Rc}^{-2}$ has obvious diurnal variations, i.e., smaller in the morning and at night and larger at noon. The imaginary part of the AERISP $C_{n,Im}^{-2}$ had no obvious diurnal variation. According to Fig. 3g, there are some peak values, i.e., some sudden increases and decreases, which may be related to sudden changes in wind direction, as shown in Fig. 3d.

Aerosol fluxes in Beijing are calculated using the assumption of free convection. Because there was no measurement of wind speed and temperature profiles near the LAS measurement location, the friction velocity and characteristic temperature could not be calculated. Because the height of the LAS instrument at the CAMS site was 43 m, the conditions assumed for free convection were easily satisfied. During the day, the surface layer is usually unstable. At night, for the city, even if there is an inversion at a higher altitude, due to the existence of the urban heat island, the surface layer is often weakly unstable. The stable stratification situation is rare (Li et al., 2007).

From the aerosol flux time series given in Fig. 3h, the aerosol flux is large at noon and small in the morning and at night, which is mainly because of the strong convection at noon. However, large aerosol fluxes also occurred on the nights of March 11 and March 12, which were related to high wind speeds. The mean aerosol flux measured at this observation point during this period was 0.0039 mgm$^2$s$^{-1}$.

4.3 Characteristics of aerosol flux at the GC site

Similarly, Figs. 4a-4d provide the time series of temperature, RH, wind speed and wind direction at 3 metres and 18 metres for the GC site, and Figs. 4e-4h show the PM$_{10}$, $C_{n,Rc}^{-2}$, $C_{n,Im}^{-2}$ and aerosol flux curves over time. According to Fig. 4a, the temperatures at both heights show obvious diurnal variations. The daytime is characterized by unstable stratification, and at night, stable stratification prevails. Moreover, in the morning and evening, there is a transition period between the stable and unstable stratification. Here, $\nu$, $T_*$ and MO length $L$ were calculated from the wind speed and temperature measured at 3 m and 18 m on a meteorological tower. Fig. 4b shows a plot of the two levels of RH over time, again with obvious diurnal variations. The RH of the GC site was lower at the CAMS site. Figs. 4c and 4d provide the time series of wind speeds and wind
directions at two levels. At 6:00 on March 12, the wind speed was relatively high, and the maximum
at 18 metres was 6.5 m s\(^{-1}\). At the same time, the maximum wind speed was reached in the Beijing
area, although the speed was lower in Beijing. The overall trend of wind direction at the GC site
was more consistent with the results of the CAMS site.

Figure 4e shows the PM\(_{10}\) trend of over time. There were two light pollution events on March
11 and March 16. The overall trend is the same as in Fig. 3(e) except that there is a slight difference.
Figs. 4f and 4g show the time series of the imaginary and real parts of the AERISP for the GC site.
The real part of the AERISP is large at noon, and the optical turbulence is strong. The real part of
the AERISP is small during the morning and evening, and the corresponding turbulence is weak.
The imaginary part of the AERISP given in Fig. 4g does not show an obvious diurnal variation, and
there may be some sharp peaks.

Figure 4h shows the aerosol mass vertical flux changes over time. The aerosol flux has a
significant diurnal variation characteristic associated with turbulent transport near the surface. The
mean aerosol flux measured at the GC site during this period was 0.0016 mg m\(^{-2}\) s\(^{-1}\). This value is
much smaller than the results for the CAMS site. Human activities contribute to increased water
vapor releases in urban areas compared to rural areas, as observed by Dou et al. (2014), and
especially for the night-time SBL in winter. During our observation period, the RH of the city was
lower than the rural area. However, human activities cause more aerosol particles in urban areas
than in rural areas.

4.4 Aerosol flux during heavy pollution periods

In the winter of 2016, there were several heavy pollution events. Generally, based on the PM\(_{2.5}\)
daily mean mass concentration limit in the primary standard of China’s national environmental
quality standards (EPD, 2012), a pollution episode is referred to as the period during which the
PM\(_{2.5}\) concentration exceeds 80 \(\mu\)g m\(^{-3}\) for 3 consecutive days between two clean periods, while a
period when the PM\(_{2.5}\) level is less than 35 \(\mu\)g m\(^{-3}\) is defined as a clean period. Pollution episodes
with peak PM\(_{2.5}\) values of more than 400 \(\mu\)g m\(^{-3}\) or less than 300 \(\mu\)g m\(^{-3}\) are termed heavy-pollution
episodes (HPEs) or light-pollution episodes (LPEs), respectively (Zhong et al., 2017).

A heavy pollution event began on December 1, 2016 and ended on January 10, 2017. Relevant
observational experiments were performed in the Beijing and Baoding areas, including observations
of meteorological parameters and aerosol parameters, to understand the causes of the heavy
pollution. The observations show that the beginning of the HPE was characterized by pollutant
transport under southerly conditions, i.e., the formation of heavy pollution was mainly caused by
the pollutants transported from southern Beijing, which we call the transport stage (TS). Usually,
after the TS stage, the explosive growth of PM\(_{10}\) and the rapid accumulation of pollutants occur,
which is called the accumulative stage (AS). During the other period within the HPEs, aerosol
particles are usually removed from the atmosphere, which is called the removal stage (RS). During
heavy pollution events, there is a lower boundary layer depth, low wind speeds and high RH, and
the PM\(_{10}\) concentration increases rapidly and reaches a very high value (Zhong et al., 2017). There
were 7 TS stages in the 2016 winter heavy pollution event, and the AS stage appeared immediately
after 4 TS stages. These included 00:00 on December 1 to 03:20 on December 4, 18:40 on December
15 to 00:00 on December 22, 00:00 on December 29 to January 2, and 00:00 and 8:40 on January 2
to 00:00 on January 5.
During this period, we used a LAS to conduct an observational study of the aerosol vertical flux in the GC site, which was from 00:00 on December 1, 2016, to 00:00 on December 22, 2016. No corresponding observations were made at the Beijing site during this period. Here, we first discuss the observation results of the GC site, Baoding City, as shown in Fig. 5. Fig. 5a shows the time series of the aerosol vertical transport flux. Figs. 5b-5g show the time series for the real and imaginary parts of the AERISP, the temperature and RH at 18 metres, and the wind speed and direction. Purple curves indicate the TS stages, red curves indicate the AS stages, and grey curves indicate the RS stages.

According to Fig. 5a, in the TS stages and the RS stages, the aerosol flux exhibited diurnal variations, while the AS stage did not show a diurnal variation. There were some peaks in the TS stage. The average aerosol flux of the TS stages was 0.00065 mgm$^{-2}$s$^{-1}$, the average value of the AS stages was 0.00025 mgm$^{-2}$s$^{-1}$, and the average value of the RS stages was 0.00063 mgm$^{-2}$s$^{-1}$. The aerosol transport fluxes in the TS and RS stages were similar, while the aerosol transport flux in the AS stage was much smaller than the TS and RS stages.

According to Fig. 5b-5c, the imaginary structure parameters and the real structure parameters of the refractive index in the TS and RS stages exhibited diurnal variations, while the AS stage did not exhibit a diurnal variation. Fig. 5d shows that except for the second AS event (22:00 on December 19, 2016 to 00:00, December 22, 2016), the temperature showed a diurnal variation. During the AS stage, the RH (see Fig. 5e) was close to 100%, while the RH during the TS and RS stages was lower. Moreover, Fig. 5f shows that during this time, the wind speed was relatively weak, although the wind speed was slightly stronger on December 5. As shown in Fig. 5g, during the TS and AS stages, southerly winds prevailed, while during the RS period, northerly winds prevailed.

The high wind speed and convection in the TS and RS stages contributed to the upward transport of aerosol particles, whereas the low wind speed and stable stratification in the AS stage were not conducive to the upward transport of aerosol particles.

During the heavy pollution period from December 1, 2016 to January 10, 2017, we did not conduct surface aerosol flux observations at the CAMS site. From January 25 to January 31, the pollution in the Beijing area also reached the level of heavy pollution. During this heavy pollution period, a measurement of surface aerosol fluxes at the CAMS site was conducted. Figure 6 shows the results of the meteorological and pollutant observations for 6 days from 00:00 on January 25, 2017 to 00:00 on January 31, 2017. According to Fig. 6, northerly winds prevailed after 12:00 on January 26, when the concentration of PM$_{10}$ dropped rapidly from 254 μg m$^{-3}$ at 12:00 to 5 μg m$^{-3}$ at 15:00. During the period 12:00-24:00 on January 26, the average wind speed was 2.6 ms$^{-1}$. On January 27, southerly winds prevailed, the average wind speed was only 0.8 ms$^{-1}$, and the aerosol concentration (PM$_{10}$) increased slowly; the increase began at 6:30 before increasing rapidly at 17:50, reaching more than 300 μg m$^{-3}$ at 23:00 and 917 μg m$^{-3}$ at 2:00 am on January 28, which was the maximum aerosol concentration over the 6 day period. Then, the aerosol concentration decreased gradually. The average wind speed on January 27 was 0.6 ms$^{-1}$, southerly winds prevailed, and the mean PM$_{10}$ concentration was 440 μg m$^{-3}$, which constitutes a serious pollution level. The average PM$_{10}$ concentration during the period from 00:00 on January 25 to 00:00 on January 31 was 170 μg m$^{-3}$.

According to the previous characteristics for the TS and AS stages, a period of southerly winds can be determined as the TS stage. Thus, January 27 can be designated as the TS stage, January 28 can be determined as the AS stage, and January 29 can be determined as the RS stage. During
Beijing's heavy pollution event in January 2017 (20170125-20170131), the mean aerosol vertical flux in the TS stage was 0.0024 mg m\(^{-2}\) s\(^{-1}\), the average value during the AS stage was 0.00087 mg m\(^{-2}\) s\(^{-1}\). 2.5 and the RS stage was 0.0049 mg m\(^{-2}\) s\(^{-1}\). The overall average value was 0.0032 mg m\(^{-2}\) s\(^{-1}\).

Even during heavy pollution events, the RH in Beijing was lower than in the outer suburbs. According to Fig. 6e, the RH exceeded 60% in the period from 3:00 to 6:00 on January 26, where the maximum value was 63%, and the RH was less than 60% in the remaining periods. In urban areas, when the RH is low, heavy pollution incidents can occur. In Beijing, during the AS stage, the vertical flux of aerosol was less than during the TS and RS stages.

5 Discussions and conclusions

During the winter of 2016 and the spring of 2017, HPEs occurred frequently in the BTH area. This study investigated the aerosol vertical mass flux and compared its magnitude during different stages of HPEs, including RSs, TSs, and ASs, in two representative urban and rural sites, including the CAMS site in Beijing and the GC site in Hebei Province. Based on the light propagation theory and surface-layer similarity theory, the aerosol vertical mass flux was obtained by combining LAS observations, surface PM\(_{2.5}\) and PM\(_{10}\) mass concentrations, and meteorological observations, including air temperature and RH. We found that under favourable meteorological conditions for pollution dispersion, i.e., from March 10, 2017 to March 17, 2017, the vertical aerosol mass flux exhibited striking diurnal variations, with the mass fluxes reaching peak values at noon and lowering in the morning and evening. During the HPEs from January 25, 2017 to January 31, 2017 in Beijing, the vertical aerosol mass flux varied substantially during the different stages. Specifically, the mean mass flux decreased by 51% from 0.0049 mg m\(^{-2}\) s\(^{-1}\) in the RSs to 0.0024 mg m\(^{-2}\) s\(^{-1}\) in the TSs, which was partly due to the wind speed reduction from strong northerly winds in the RSs to southerly winds in the TSs. During the ASs, the mean mass flux decreased further to 0.00087 mg m\(^{-2}\) s\(^{-1}\), which accounted for approximately 1/3 of the flux during the TSs. Due to the cooling effect of elevated aerosols in the ASs, the near-ground temperature decreased and caused or reinforced the inversion, which suppressed the turbulence diffusion. The weakened mass flux would further facilitate aerosol accumulation. During the HPE from December 01, 2016, to December 22, 2016, in Gucheng, the mean mass flux was similar in the RSs and TSs, ranging from 0.00063 mg m\(^{-2}\) s\(^{-1}\) to 0.00065 mg m\(^{-2}\) s\(^{-1}\). This is partly because Gucheng was less affected by strong northerly winds than Beijing. Thus, the wind speed varied slightly from the RSs to TSs. However, the mass flux decreased substantially to 0.00025 mg m\(^{-2}\) s\(^{-1}\) in the ASs, which was merely 1/3 of the mean flux in the TSs.

In this study, the aerosol emission flux was also estimated in these two rural and urban sites. Generally, compared with the emissions in spring, we found that in winter, the near-ground emissions were weaker in suburban areas and were similar in urban areas. In suburban areas, although the aerosol concentrations were relatively high (Shen et al., 2018), the upward emitted aerosol flux was smaller than in urban areas. During the ASs of the HPEs, the aerosol emission flux from the ground was weaker than for the RSs and TSs at both the CAMS and GC sites, which indicates that surface pollutant emissions are not the major cause of explosive PM\(_{2.5}\) growth. During the ASs with weak solar radiation, the factors most associated with aerosol concentration changes were horizontal transport and BL height variations, which might be the main causes of increased PM\(_{2.5}\) (Zhong et al., 2018a; Zhong et al., 2018a; Zhang et al., 2018).

Due to the lack of necessary experimental conditions, such as meteorological towers and EC systems, current experimental results cannot be compared with EC methods. According to the
literature data, the two methods have been compared indirectly, and the estimated aerosol flux under different measurement conditions is consistent in magnitude (Yuan et al., 2016). However, a direct comparison of the two methods is in development.

Compared with the EC method, the aerosol flux has high spatial representativeness based on the principle of light propagation, and there is no need to install a high tower. However, the estimation of aerosol fluxes using the LAS method still has theoretical and practical deficiencies. At present, the LAS method for the aerosol flux regards the aerosol particles as ordinary scalar molecules. At the same time, based on the assumption of the equivalent medium, the imaginary part of the AERI is taken for granted as proportional to the aerosol mass concentration. This is often not the case. The actual turbulence spectrum shape often deviates from the “-5/3” law, and turbulence intermittent and scintillation saturation can also occur (Moene et al., 2009). The applicability of the near-surface layer similarity theory to the aerosol particle motion under stable layer conditions also has many problems. The formation of new particles and changes in aerosol particle size distribution also affect the scintillation in light propagation. There are also practical problems such as untimely maintenance, rainfall and low visibility, and platform vibrations required for observation. All these problems will cause errors in final estimates, so more theoretical and experimental research is needed.

**Author contributions.** Renmin Yuan and Xiaoye Zhang designed experiments and wrote the manuscript; Renmin Yuan, Hao Liu, Yu Gui, Bohao Shao, Yaqi Wang, Junting Zhong and Xiaoping Tao carried out experiments; Renmin Yuan analyzed experimental results. Yubin Li and Zhiqiu Gao designed experiments and discussed the results.

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Figure 1. Photographs of the measurement site. (a) Map of the experiment area in the Beijing urban area and suburban area and (b) expanded view of the Beijing experiment area, which is marked as the shaded rectangle in (a). (c) Expanded view of the Baoding experiment area, which is marked as the shaded rectangle in (a). (d) Satellite image of the CAMS site and (e) the satellite image of the GC site. Figs. 1a, b, c, and d © Google.

Figure 2. Scatterplots of aerosol mass concentration $M_a$ vs. the imaginary part of the AERI for (a) the Beijing area and (b) the Baoding area.
Figure 3. Temporal variations in (a) air temperature, (b) RH, (c) wind speed, (d) wind direction, (e) PM$_{10}$, (f) real part of the AERISP, (g) imaginary part of the AERISP and (h) aerosol mass flux in the Beijing area from March 10, 2017 to March 17, 2017.
Figure 4. Temporal variations in (a) air temperature, (b) RH, (c) wind speed, (d) wind direction, (e) PM$_{10}$, (f) real part of the AERISP, (g) imaginary part of the AERISP and (h) aerosol mass flux in the Baoding area from March 10, 2017 to March 17, 2017.
Figure 5. Temporal variations in (a) aerosol flux, (b) imaginary part of the AERISP, (c) real part of the AERISP, (d) air temperature, (e) RH, (f) wind speed, and (g) wind direction in the Baoding area during a heavy pollution period, i.e., December 1, 2016 to December 22, 2016.
Figure 6. Temporal variations in (a) aerosol flux, (b) imaginary part of the AERISP, (c) real part of the AERISP (d) air temperature, (e) RH, (f) wind speed, and (g) wind direction in the Beijing area during a heavy pollution period, i.e., January 25, 2017 to January 31, 2017.