Transport Patterns, Size Distributions, and Depolarization Characteristics of Dust Particles in East Asia in Spring 2018

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Abstract The long-range transport of Asian dust and its complex interaction with anthropogenic pollutants have a significant yet poorly quantified effect on the global climate. In this study, dust events were observed based on an optical particle counter with a polarization detection module at an urban site in North China. The temporal variations in the size distribution and morphological changes of dust particles were analyzed. The results demonstrate that the dust events were induced by a deep Siberian low-pressure anomaly and/or Mongolian cyclone in the low troposphere. The morphology of dust particles, as indicated by the depolarization ratio ($\delta = s/(s + p)$), the ratio of the parallel signal to the total backscattering signal, varied significantly at different transport heights. A decreasing tendency was found when dust was trapped within the boundary layer over polluted areas, particularly when the atmospheric loading of pollutants and the relative humidity were high. During a typical polluted dust case, the $\delta$ value of mineral dust ($D_p = 5 \mu m$) decreased by 39%, as the conditions were stagnant in East China, consistent with the Cloud-Aerosol LIDAR with Orthogonal Polarization results. The morphological variation was evaluated according to the aspect ratio, presuming that the dust particles were ellipsoidal. The interaction between dust particles and water vapor could lead to an increase of at least 6% in volume when the relative humidity increased up to 90%. An aerosol classification based on $\delta$ values and size information could be performed to estimate the contributions of different aerosol types to pollution days. The results should be helpful for evaluating satellite-borne and ground-based light detection and ranging data analyses.

1. Introduction

As major atmospheric compounds, mineral dust aerosols play a complex role in the terrestrial system. These compounds can affect the regional/global climate by disturbing atmospheric chemistry processes (Martin et al., 2003), biogeochemical cycles (Archer et al., 2000; Shao et al., 2013), cloud processes (Coopman et al., 2016; Liu et al., 2012; Sand et al., 2017; Tobo et al., 2010), and the radiation budget (Takemura, 2005; Yang et al., 2016). Dust aerosols are also a well-known human health hazard that can induce respiratory, cardiovascular, and dermatological disorders and infectious disease (de Longueville et al., 2013; Griffin, 2007; Goudie, 2014; Zhang et al., 2016). East Asia is the second-largest dust source region in the world. The annual dust emissions in East Asia are estimated to be 214 Tg yr$^{-1}$, dominating the atmospheric load over China and Mongolia (approximately 70%) (Tanaka & Chiba, 2006). Moreover, Asian dust aerosols can be transported long distances, causing significant impacts on a global scale (Grousset et al., 2003; Huang et al., 2015; Uno et al., 2009; Zhang et al., 2018).

To date, it remains challenging to accurately estimate the radiative effects of mineral dust since many uncertainties remain regarding the optical, mixing state, and hygroscopic properties of dust particles (Denjean et al., 2015, 2016; Kubilay et al., 2003). This is especially true in East Asia, a region characterized by both frequent dust processes and severe air pollution (Su et al., 2019; Xing et al., 2017; Zhang et al., 2019). Mixed dust plumes are reported to exhibit physical properties that are even more complex. For example, mineral dust...
mixed with air pollution can lead to brownish haze, which has a cooling effect on the Earth by absorbing and scattering solar radiation reaching the surface (Kaufman et al., 2002). However, once carbonaceous compo-
nents attach to the surface of the dust, the mixture of polluted dust can have a strong heating impact on the atmosphere (Seinfeld et al., 2004). In addition, dust particles act as active condensation nuclei, that is, cloud condensation nuclei and ice nuclei (Coopman et al., 2016; Kaufman et al., 2002; Koehler et al., 2009, 2010; Tobo et al., 2010), and the hydrophilic capacity of natural dust is limited (Tang et al., 2017). Nevertheless, when dust particles react with acid gas (e.g., sulfur dioxide or nitric acid) and/or undergo a coating process, these particles become more hygroscopic due to the presence of soluble matter on the surface (Liu et al., 2008; Sullivan et al., 2009; Wang et al., 2010). Dust particles can also provide reactive surfaces for the catalytic con-
version of reactive pollutants such as SO₂ (Li, Bei, et al., 2016; Nie et al., 2014; Zhang et al., 2015), which lar-
gely increases the complexity of heterogeneous chemical reactions in the atmosphere. These findings all emphasize the importance of characterizing the mixing states between mineral dust and other types of aerosols.

The depolarization of backscattered light can be applied to understand irregular changes in dust morphology and further distinguish the external/internal mixing states of dust particles in real time. In principle, the depolarization ratio of the scattering signal corresponding to spherical particles is almost zero because the particle-scattered light has the same polarization plane (p component) as the incident light; in contrast, the depolarization ratio of nonspherical particles is large because of the increase in the polarization compo-
nent in the perpendicular plane (s component). The depolarization ratio has been widely adopted by depo-
larization light detection and ranging (LIDAR) systems to analyze the spatiotemporal distribution of dust aerosols across large spatial scales (δa = s/p, at 532 nm). For example, in East Asia, a regional elastic scatter-
ing LIDAR network called the Asian Dust and Aerosol LIDAR Observation Network has been in operation since 2001 (Shimizu, 2004; Shimizu et al., 2016; Singh et al., 2008). In addition, in 2006, a space-based two-
wavelength polarization-sensitive backscatter LIDAR named Cloud-Aerosol LIDAR with Orthogonal Polarization (CALIOP) was launched onboard the Cloud-Aerosol LIDAR and Infrared Pathfinder Satellite Observations satellite, which broadened the view of ground-based observations. LIDAR has been widely used to analyze the vertical transects of aerosols and clouds at high temporal and spatial resolutions and the depolarization properties of dust particles during long-range transport (Huang et al., 2008; Itahashi et al., 2010; Uno et al., 2008, 2009; Zhang et al., 2018). However, it is still challenging to provide sufficient information on the external/internal mixing states of dust aerosols because the external mixing of dust and spherical particles may lead to optical characteristics that are similar to those of internally mixed pol-
luted dust (Pan et al., 2019; Sugimoto et al., 2015). Additionally, mixtures of dust and pollutants occur mostly in the boundary layer (Li, Shao, et al., 2016; Li et al., 2017; Zhang et al., 2005), while the “surface returns” received by space-borne LIDAR may somewhat mask aerosol information (Venkata & Reagan, 2016). In addition, ground-based LIDAR observations are blind to the information in the few hundreds of meters above the surface due to incomplete laser/receiver overlap (Xiang et al., 2018). To improve the information related to the depolarization ratio, single particle-based light scattering experiments have been conducted to further reveal the depolarization properties of the particles in the air. A summary of the instruments used to detect single particle-based depolarization properties is provided in Table 1. Glen and Brooks (2013) improved the Cloud Aerosol and Spectrometer (Baumgardner et al., 2001), which can measure the intensities of only forward and backward scattering light, and added a polarized module to detect the nonsphericity of particles. Baumgardner et al. (2014) further designed an open-path cloud probe onboard an aircraft to detect the polarization properties of cloud droplets and ice crystals.

In this study, we used a bench-top optical particle counter equipped with a depolarization module (polariza-
tion optical particle counter [POPC]) to detect the size-resolved polarization of individual particles at the sur-
face (Kobayashi et al., 2014). This instrument provides information on the size and depolarization ratio of a particle, which can be used to analyze the temporal variation in the morphology of individual dust particles (Pan et al., 2015, 2017; Wang et al., 2017). The individual aerosol particle type was identified according to both the particle size and the depolarization property. The POPC is capable of distinguishing four types of aerosols, as shown in Figure 1: (1) anthropogenic pollution particles with Dp < 1 μm and δ < 0.2 or 1 μm < Dp < 2.5 μm and δ < 0.1; (2) coarse-mode mineral dust with Dp > 2.5 μm and δ = 0.3 ± 0.1 and fine-mode dust with Dp < 1 μm and δ > 0.2 or 1 μm < Dp < 2.5 μm and δ > 0.1; (3) polluted dust with Dp > 2.5 μm and δ < 0.2; and (4) sea salt particles with 1 μm < Dp < 2.5 μm and δ < 0.1 (Kobayashi...
et al., 2014; Pan et al., 2015, 2016; Tian et al., 2018, 2019). All external and internal mixing processes of dust events can be captured by the POPC. We focused on two typical large-scale outflows of Asian dust that occurred in the spring of 2018. The meteorological conditions and transport patterns were discussed, and the mixing state of the polluted dust was estimated. In the final section, the s component versus p component pattern of bulk aerosols on typical pollution days was described, and the contributions of anthropogenic pollutants, dust, and polluted dust to the detected signal were discussed. The final purpose of this study was to quantitatively investigate the morphological evolution of dust particles and comprehensively determine reasonable key factors in the formation of spherical dust particles.

2. Methods

2.1. Ground-Based Observations

Field measurements were carried out in the spring (March, April, and May) of 2018 at an urban site in Hengshui (37.7°N, 115.6°E) in the North China Plain (NCP). The POPC employs a linearly polarized laser to provide a collimated incident beam of light at a wavelength of 780 nm and can measure light scattering from particles over a diameter range from 0.5 to 10 μm. The scattered light was collected from two separate angles (acceptance angle of 45°), namely, forward (60°) and backward (120°), to detect the size (water equivalent optical diameter) and nonsphericity of the particles, respectively. Light scattered in the backward direction passes through a polarizer, which splits light into perpendicular (s polarized) and parallel (p polarized) signals. In this study, the polarization ratio δ was defined as $\delta = s / (s + p)$. The variation in the δ value provides information about the shape and size of the particles. For irregular particles, the polarization state is no longer identical to that of the incoming light. The uncertainties of δ values depend on various particle properties, that is, the aerosol water loss and the refractive index. We estimate the uncertainty of the δ value to be <13%. During the observation, the inlet flow rate was set to 80 ccm (cubic centimeters per minute), and the diluted air flow rate was set to 920 ccm to avoid the coincidence error of several particles being simultaneously illuminated by the laser. The residence time of the sample air mass in the tube was estimated to be ~3.8 s. During measurement, the relative humidity (RH) inside the measuring chamber was stable at 29.5 ± 0.9%.

The mass concentrations of the ambient particles derived from the POPC were comparable to those of a nearby state-controlled environmental monitoring station (0.9 km southeast of the POPC observation site), with correlation coefficients of 0.93 and 0.91 for PM$_{2.5}$ and PM$_{10}$, respectively (supporting information Figure S1). The particle mass from the POPC was estimated based on the

### Table 1

| Instrument/basic function | Detection angle ($\theta$, °) | Incident wavelength (λ, nm) | Aerosol sample | Reference |
|---------------------------|-------------------------------|-----------------------------|----------------|----------|
| HOLOGraphic Instrument for Microscopic Objects | 178 | 532 | Ice crystals (2–118 μm) | Amsler et al. (2009) |
| Laser scattering and depolarization instrument (SIMONE) | 178.2 | 488 | Small ice crystals (1–15 μm) | Schnaiter et al. (2012) |
| Cloud and Aerosol Spectrometer with Polarization | 168–176 | 680 | Dust particles (0.6–50 μm) | Glen and Brooks (2013) |
| Cloud Particle Spectrometer with Polarization Detection | 146–172 | 685 | Cloud particles (liquid and ice) (2–50 μm) | Baumgardner et al. (2001) |
| Polarization-resolved angular optical scattering detection instrument$^b$ | 150–180 | 650 and 680 | Bioaerosol particles (500 nm spheres; 500 × 1,000 nm cylinder-line particles) | Redding et al. (2014) |
| Polarization optical particle counter$^c$ | 120 | 780 | Mineral dust, sea salt, and anthropogenic pollutants (0.5–10 μm) | Kobayashi et al. (2014) |

$^a$The scattering angle to detect the depolarization properties. $^b$The authors did not give the instrument a precise name. $^c$Instrument used in this study.
hypothesis of a spherical shape with the density varying linearly from 1.77 to 2.4 g cm$^{-3}$ for fine- (0.5 μm) and coarse-mode (10 μm) aerosols.

Meteorological data, including the surface wind speed and direction, and visibility data were collected from the China Meteorological Administration. Air quality and meteorology observations were taken every 1 hr. The large-scale circulation analysis was based on the daily data set of the European Centre for Medium-Range Weather Forecasts global atmospheric reanalysis (ERA-Interim) (website: https://apps.ecmwf.int/datasets/data/interim-full-daily/) (Berrisford et al., 2009, 2011).

2.2. Space-Borne LIDAR Observations
During each of these events, the CALIOP and Moderate Resolution Imaging Spectroradiometer (MODIS) satellites sampled the dust plumes multiple times. The MODIS Level 3 Deep Blue aerosol optical depth (AOD) product (Version 6.1) (website: https://giovanni.gsfc.nasa.gov/giovanni/) reflects the optical information of all aerosols for the entire atmospheric column (Butt et al., 2017; Ichoku, 2002; Levy et al., 2013). This product was used to analyze the spatial distributions of the dust and pollution plumes in this study. The vertically resolved physical properties of aerosols are also crucial for understanding the dust transport process. We used Version 4.2 of the Level 2 aerosol profile products and vertical feature mask (VFM) products, which are both derived from layer products and interpolated onto a vertical grid. The Level 2 aerosol profile products provide vertically resolved optical properties (e.g., extinction coefficients and depolarization ratios) for each atmospheric column, while the information in the VFM allows aerosols to be classified as clean marine, dust, polluted continental, clean continental, polluted dust, and smoke (website: https://eosweb.larc.nasa.gov/project/calipso) (Winker et al., 2009).

2.3. Trajectory Model
The NOAA Air Resources Laboratory Lagrangian Trajectory Hybrid Single Particle Lagrangian Integrated Trajectory (HYSPLIT) model (website: https://ready.arl.noaa.gov/HYSPLIT.php/) was used for the complementary source analysis of dust aerosols observed over East China (Stein et al., 2009). Particles were released from the source location at an altitude of 50 m and were passively followed in the wind afterward. The NOAA Global Data Assimilation System (Chai et al., 2017) was used as the input for HYSPLIT. The concentration grid was set at a resolution of 0.05° with a vertical spacing of 2 km extending from the surface to 20 km.

3. Results and Discussion

3.1. Overview of the Dust and Pollution Events in the NCP in Spring 2018
The temporal variations in the volume concentration and depolarization ratio as a function of the particle size during the observation period are shown in Figure 2. The measurement was deployed in a typical spring period, during which the average mass concentrations of PM$_{2.5}$ and PM$_{10}$ were 57.7 ± 45.5 and 118.8 ± 77.9 μg/m$^3$, respectively. Approximately 86% of the hourly averaged PM$_{2.5}$ concentrations exceeded 75 μg/m$^3$, which is the Chinese Grade II guideline (CB 3095-2012). Meanwhile, the PM$_{10}$ concentration exceeded 150 μg/m$^3$ during 27.2% of the observation periods, indicating an influence of floating dust in the NCP. Two size modes dominated the volume size distribution in the atmosphere of East Asia during the springtime: Dust particles in the coarse mode (4–8 μm) generally had a clear nonspherical configuration (δ > 0.25), while anthropogenic pollutants in the fine mode (<2.5 μm) normally displayed a spherical shape with a δ value less than 0.1. Dust particles, especially those with relatively small diameters (2–3 μm), were occasionally present in a spherical configuration (δ < 0.1), possibly due to the complex mixing of natural dust and anthropogenic particles (Pan et al., 2015). The POPC captured a total of five dust events in the NCP (26–29 March, 30 March to 4 April, 7–13 April, 16–21 April, and 22–29 May). All the dust episodes were characterized by a pronounced peak of the volume size distribution in the coarse mode and a sharp increase in the δ value of fine-mode particles due to fine dust. Here, the two most intensive dust events occurred on 26–29 March (Episode I) and 7–13 April (Episode II) and were chosen for further study because both dust events affected nearly two thirds of China and part of the Northwest Pacific. The aerosols’ physical depolarization and hygroscopic properties as well as their transport patterns were explicitly analyzed.
3.2 Dispersion and Depolarization Properties of the Observed Dust Layers

Figures 3a–3h show an overview of the sources and spatial transport patterns of the atmospheric dust plumes in East Asia based on the MODIS AOD data set. During these two long-range dust transport events, the initial transport phases were both strongly zonal (confined between 30°N and 60°N) with a weak or absent meridional component. It is well known that the direct causes of long-range dust transport are weather disturbances and typical synoptic patterns. Moreover, continental- to planetary-scale circulations at middle and high latitudes can exert significant influences on weather conditions and affect dust plume activities (Gong et al., 2006; Mao et al., 2011; Zhao et al., 2013). To investigate the mechanisms of the dust cases in this study, the 500 and 1,000 hPa geopotential height anomaly patterns for the 3 day initial eastward transport stage of the events were composited from the ERA-Interim daily reanalysis data, as shown in Figures 3i–3l. To inspect the vertical structures of the two aerosol transport events, we applied the HYSPLIT backward trajectories and found the timing of the Cloud-Aerosol LIDAR and Infrared Pathfinder Satellite Observations cross section of the volume depolarization ratio ($\delta a = s/p$) at 532 nm and the aerosol types (VFM) that intersect the HYSPLIT path (Figures 4 and 5). In the HYSPLIT setup, short trajectories (3 days) were used to improve the reliability of the simulation (Stein et al., 2009, 2015).

3.2.1. Strong Siberian Low-Pressure System-Induced Dust Mass Outflow Process: Episode I

On 25 March, when persistent westerly warm advection became the carrier of dust, a cyclone system that split from a strong Siberian low-pressure system began to influence Northwest China (Figures 3i and 3j). Surface convergence around the low-pressure system provided ascending motion and lifted the dust mass into the high troposphere. A dense dust plume formed initially over the Lop Nur region at the eastern edge of the Taklimakan Desert on 25 March and moved quickly to the NCP within 2–3 days (Figures 3a–3c). Due to the limitations of satellite orbits, it was difficult to detect the dust layers near the source region of Lop Nur. The first CALIOP-observed pollution plume was detected at 19:04 UTC on 25 March over western Inner Mongolia (Figure 4a). The high $\delta a$ (0.25–0.5) implies that the aerosol layer was a dust layer with...
nonspheroidal coverage (Hu et al., 2007; Tian et al., 2015), and airborne dust appeared to be dispersed at high vertical heights of 2–10 km. On 27 March, yellow dust plumes masked northeastern and northern China, as these regions have a high AOD according to satellite observations (Figure 3c). The aerosol layers appear as thick elongated features ranging in thickness from 1 to 10 km with a length of 1,100–1,700 km. The altitude of the features decreased to <6 km on 28 March (Figure 4e) due to the deposition effect. The air quality over the NCP deteriorated on 28 March, and the maximum hourly averaged mass concentration of PM$_{2.5}$ reached 1,955 μg/m$^3$ in Beijing (39.98°N, 116.4°E), 706 μg/m$^3$ in Hengshui, and 484 μg/m$^3$ in Zhengzhou (34.75°N, 113.6°E) (Figure S2). Accordingly, signals of increased AOD caused by the dust mass were captured over the Korean Peninsula and migrated pervasively over the Yellow Sea of China (Figure 3d). In this case, the volume depolarization ratio (δv) of the dust mass was observed to be 0.2–0.5 and showed almost no difference between the dust source area and eastern China. This finding was because the dust plume was transported at a high altitude of >2 km until 28 March, when the obvious deposition of dust occurred on the Yellow Sea of China. The polluted dust (δv < 0.15) observed at a low altitude (<2 km) over Northeast China and at the Yangtze River Delta (Figure 4e) was likely due to local floating dust and pollutants.

Figure 3. (a–h) Wind field at 10 m above the ground and distribution of the mean AOD at 550 nm derived from the MODIS Terra Deep Blue product in East Asia. (i–l) The 500 and 1,000 hPa geopotential height anomaly patterns for the 3 day initial eastward transport stage of the events on 26–28 March (Episode I) and 7–9 April (Episode II). The composite anomaly was calculated with respect to the 1979–2018 climatology based on ERA-Interim daily reanalysis data.
A high AOD was observed over the Shandong Peninsula beginning on 25 March, and the pollution mass strengthened during transport to the NCP on 26 March. Simultaneously, CALIOP observed polluted dust at a low altitude (below 2 km) over this region. In typical cities such as Zhengzhou and Hengshui, the PM$_{2.5}$/PM$_{10}$ ratio was found to be 0.5–0.7. The PM$_{2.5}$ concentration in Beijing reached 110.5 ± 57.5 μg/m$^3$, and the PM$_{2.5}$/PM$_{10}$ ratio was nearly 1. The high AOD was likely attributable primarily to pollutants and partially to floating dust. The POPC observations also reflected the occurrence of anthropogenic pollution; a detailed discussion will be presented in section 3.3.

3.2.2. Low-Altitude Transport of Dust Plumes Caused by a Shallow Low-Pressure System in Mongolia: Episode II

In this case, the strong dust plume was first observed in the Taklimakan Desert on 3 April. After 6 April, the hourly PM$_{10}$ concentration observed at the Kizilsu Kirghiz Autonomous Prefecture (39.72°N, 76.19°E) site inside the Taklimakan Desert exceeded 2,000 μg/m$^3$ for more than 95% of the observations over the next 3 days; this value is over 5 times higher than the average concentration (357.3 ± 599.4 μg/m$^3$) observed in the springtime (Figure S2). Due to the influence of a weak high-pressure system centered at North Sinkiang, the dust mass was confined within the Tarim Basin during the first few days until 7 April (Figure 3e), when a distinct thick dust layer at approximately 1–11 km above ground level was observed (Figure 5a). The subsequent export process was triggered by an enhanced midlatitude cyclone that first formed over Mongolia on 8 April due to the heating effect of warm advection in the lower troposphere (Figures 3k and 3l). This Mongolian low-pressure system was shallow with an upper limit of 850 hPa. The plume traveled more than 10° of longitude in 1 day, when it was channeled through the Hexi Corridor.
and the Alxa Desert and it affected much of northern and eastern China (Figures 3f–3h). The broad horizontal extent of the dust plume spanned nearly 50° of longitude. In this episode, the dust mass was suppressed by vertical diffusion (2–5 km) due to the insufficient lifting force (Figures 5b–5e). The air quality in central China deteriorated at noon on 9 April, as the maximum hourly PM$_{10}$ concentration was over 1,000 μg/m$^3$ in most cities in Inner Mongolia, including Alxa (38.85°N, 105.72°E), Baotou (40.65°N, 109.83°E), and Hohhot (40.76°N, 111.65°E). On 10 April, the dust plume severely influenced the NCP, as the PM$_{10}$ concentrations reached 815 and 690 μg/m$^3$ in Hengshui and Zhengzhou, respectively. Influenced by the east-west convergence zone near the ground at ~33°N in eastern China, on 11 April, the transport direction of the plume shifted from south to east toward the Yangtze River Delta, with hourly PM$_{10}$ concentrations reaching 400 μg/m$^3$ in Nanjing. The main part of the dust plume then returned to the NCP, and the dust episode ended on 14 April. During the eastward transport process, the volume $δ_a$ of the dust air mass at high altitudes (>2 km) remained at approximately 0.35 ± 0.1. On 9 April, when the dust cloud first arrived at the NCP, dust deposition in the planetary boundary layer (PBL) was clearly detected from vertical profiles of the aerosol types over East China, while the volume $δ_a$ of the dust layer at low altitudes (<2 km) was 0.05–0.2 as a result of the generated polluted dust (Figures 5c–5e).

From the above analyses, we confirm that a large-scale low-pressure system favored the 2–3 day zonal transport of dust plumes. The lifting mechanism and eastward outflow pattern of Asian dust were associated with two important outflow patterns. The first prominent climatological feature, represented by Episode I, was a strong low-pressure anomaly over Europe and western Siberia. The dust layer was lifted to altitudes of 0.5–12 km in the free troposphere and transported rapidly. The second pattern is represented by Episode II, in which the outbreak of a dust episode was correlated with a Mongolian cyclone anomaly in the low troposphere. Similar meteorological conditions were found by Zhu et al. (2008) and Tan and Wang (2014). Zhu

Figure 5. The same as Figure 4 but with the backward trajectories beginning at 00:00 UTC on 10 April (red hollow dot) and 00:00 UTC on 12 April (blue hollow dot).
et al. (2008) reported that the air temperature trend around Lake Baikal could affect the activity of Mongolian cyclones. The lifting effect and vertical diffusion of dust were comparably suppressed for the second pattern with dust plumes migrating <6 km during transport. The depolarization properties of the dust particles were more likely to change during low-altitude transport, especially at near-ground altitudes where pollution was severe. We further checked the atmospheric circulation patterns of the other three cases that occurred in spring 2018 and found that the two cases on 31 March to 2 April and 15–17 April corresponded to the first pattern, while another case (21–28 May) was similar to the second pattern (Figure S3).

3.3. Morphological Changes in the Dust Aerosols Over the Polluted Atmosphere in East China

For both cases, the variations in the volume depolarization ratio of dust particles were different during their eastward movement. The degree of variation in the morphology of dust particles could vary greatly under different air conditions. For example, the production of polluted dust may be inefficient during transport in the troposphere (Wu et al., 2016), while the generation of polluted dust may be more efficient within the PBL in a polluted area (Li & Shao, 2009; Pan et al., 2017). The POPC could provide much more accurate signatures of internally mixed Asian dust particles by detecting the single-particle depolarization ratio (δ). In Episode II, the dust air mass was stagnant over East China for approximately 5 days, enabling continuous observations of the depolarization properties of the same dust plume throughout the event.

Figures 6a–6d plot the temporal variations in the PM$_{2.5}$ and PM$_{10}$ mass concentrations, PM$_{2.5}$/PM$_{10}$ ratio, RH, and δ values of the ambient particles during each episode. For comparison, we selected coarse-mode particles at an optical size of D$_{p}$ = 5 μm to represent the dust particles and fine-mode particles at...
Dp = 1 μm for anthropogenic pollutants. The size-dependent depolarization ratio and volume concentration of the aerosols in each episode are shown in Figures 6e and 6g. Previous studies indicated that dust particles could also undergo hygroscopic processes due to the continuous absorption of moisture, which could alter their morphology (Krueger et al., 2003). Here, the δ values of the dust particles as a function of the RH were investigated (Figures 6f and 6h). Additionally, to further analyze the nonsphericity and mixing states of the dust particles, scatter plots between the particle size and the δ values are presented in Figure 7.

3.3.1. Typical Externally Mixed Dust Process: Episode I
Beginning on 24 March, a southerly wind (2–5 m/s) prevailed in the NCP. The PM$_{2.5}$/PM$_{10}$ ratio slightly fluctuated near 0.7 ± 0.09 (Figure 6a), indicating a negligible contribution of coarse-mode dust to the PM$_{10}$ concentration compared with that of fine-mode pollutants. The POPC results confirm that during this non-dust period, the observation site was dominated by spherical particles concentrated at <1 and ~2 μm (Figure 7a). When the dust plume first arrived on the afternoon of 28 March, strong northerly winds (4–6 m/s) dominated the NCP, and the volume size distribution had a pronounced peak at 4–5 μm. As expected, the hourly averaged δ value of 1 μm particles sharply increased to twice the reference value (0.1 ± 0.02) under the influence of irregularly shaped fine dust (Figure 6b). The observed δ values of the particles markedly increased

![Figure 7. Variation in the dependence of the depolarization ratio (δ) on the particle size. The colors and contour lines represent the dV/dlogDp and volume fraction of the particles, respectively. For better comparison, the volume concentration was normalized to a maximum value of 1 using the following formula: Standard value = (truth value-minimum)/(maximum-minimum).](image-url)
with size when the diameter was <4 μm; however, the δ values of the 4–10 μm dust particles remained almost unchanged (Figure 6e). This result suggests that the δ value was sensitive mainly to the shape irregularity and size of the particles. At this time, the atmospheric aerosols were an external mixture of dust and spherical aerosol particles (Figure 7c). The δ value of the uncontaminated dust particles (Dp = 5 μm) was found to be 0.28 ± 0.05, almost irrespective of the RH (Figure 6f), demonstrating the weak hydrophobic nature of dust. On 29 March, the PM2.5 mass concentration decreased by 58% because of rapid gravitational deposition. A volume loss of spherical particles also occurred in the fine mode (Figure 6d) due to dispersion on a windy day. Throughout this episode, the RH exhibited a significant diurnal variation with an increasing trend at night (70–85% at midnight) and a decreasing trend in the daytime (<40% at noon). The high humidity at night usually corresponded to a slight downward fluctuation in the δ value of the dust particles. Significant internal mixing between the dust and pollutants was not observed in this case.

### 3.3.2. High RH and High Proportion of Secondary Pollutants in Atmospheric Particles Leading to the Significant Production of Polluted Dust: Episode II

In this case, the dust mass had a layered structure, as detected by CALIOP; substantial amounts of polluted dust were present at low altitudes within the PBL, while pure dust was found to be transported at higher altitudes (2–6 km) (Figures 5c–5e). During the onset of the dust plume in East China (10 April), the total volume of coarse-mode particles (Dp > 2.5 μm, with a median diameter of 4.2 μm) increased. The δ values for the dust particles (5 μm) and fine particles were found to be 0.28 ± 0.05 and 0.21 ± 0.01, respectively (Figure 6d). Over the next 3 days (11–13 April), the dust air mass was slowly transported and hovered over North and East China (Figures S4a–S4h). Strong moisture advection (850 hPa) from the Northwest Pacific beginning on 12 April (Figures S4i–S4l) led to a significant increase in the surface RH (95 ± 5%). The PM2.5-10 mass concentration decreased due to gravity settlement and dilution, whereas the PM2.5 mass concentration was maintained at a high level of 70 ± 5 μg/m³ and the PM2.5/PM10 ratio gradually increased to >0.9 on 13 April because of secondary pollutant generation (Figure 6c). Simultaneously, the δ value of the dust particles (5 μm) decreased by ~39% to 0.17 ± 0.06 due to continuous internal mixing with pollutants (Figure 7k). The δ value of the particles at 1 μm also decreased to 0.06 ± 0.01, 40% lower than the reference value. Calibration of the POPC showed that the δ values for purely spherical particles at 5, 7, and 10 μm were 0.08, 0.09, and 0.1, respectively, and were almost zero for the particles at Dp = 1, 2, and 3 μm. This result means that particles of all sizes on 13 April were altered to be more spherical because of the absorption of moisture. The ability of particles to uptake water depends mostly on their chemical composition (Martin, 2000). In this case, δ5μm was negatively correlated with the RH: The value was 0.26 ± 0.02 when RH < 60% and decreased to 0.21 ± 0.02 when RH > 80% (Figure 6h). This result suggests that the polluted dust particles were related to hygroscopic aerosols.

The scattering of light by a particle is affected by various factors, such as the incident light properties (i.e., wavelength), the physical-chemical properties of the particle (i.e., size range, morphology, refractive index, and water content), and the orientation of the particle relative to the incident light (Zenker et al., 2017). To assess the potential response of the POPC to nonspherical particles, Pan et al. (2017) took a simplistic approach using prolate spheroids at an equivalent optical diameter of 5 μm to represent dust particles with an aspect ratio range of 1.0–1.9. The dust particles were characterized by the ellipsoid model because, in previous field observation studies, the typical dust particles collected from natural sources were mostly in the shapes of blocks, sharp diamonds, and elongated rectangles, which can be easily depicted by the ellipsoid model (Chou et al., 2008; Haywood et al., 2008; Kandler et al., 2017; Okada et al., 2001; Rocha-Lima et al., 2018; Wagner et al., 2012). In Episode II, the observed reference aspect ratio for the uncoated dust particles (δ value = 0.28 ± 0.05) was 1.69 ± 0.01 and decreased by ~1.2% when the RH varied from 10% to 70% (Figure S5). A significant decrease in δ occurred when the RH was above 70%, such that the aspect ratio decreased by 4.8% to 1.59 ± 0.01. This decrease corresponded to an increase of at least ~6.3% in the total volume of the matter coating the dust surface. Pure dust particles from natural sources usually have poor hygroscopicity (Gibson et al., 2006). Ma et al. (2012) observed that at 90% RH, fresh CaCO3 particles contain only 0.1% water. Bohr et al. (2010) found that the water adsorbed onto the surface of a single CaCO3 crystal remained constant in thickness (1.55 ± 0.1 nm) when the RH varied from <4% to 90%. However, HNO3, NO2, and N2O5 uptake can lead to the formation of nitrate, which has much higher hygroscopicity, on dust particles (Nie et al., 2014; Seisel et al., 2005; Tang et al., 2012; Underwood et al., 2001). This result may explain the significant response of dust to the RH in Episode II.
We further inspected the depolarization properties of the aerosols in the three other dust events in the spring of 2018 and confirmed that the hygroscopicity of the dust particles increased on 15–22 April and 22–29 May (Figures S6 and S7). A common feature among these three cases was that the air mass first moved south to ~35°N in East China and then returned to the NCP. The high PM2.5–10 concentration observed at the observation site was combined with the prevailing south wind. In both cases, pollutants were generated at the end, and the PM2.5/PM10 ratio reached 0.8. We conclude that the internal mixing process of mineral dust particles generally occurred during the later period of the dust process, and a necessary condition was a high proportion of pollutants in the atmosphere. Weather conditions, especially the wind direction and RH, also played a key role in the decreasing δ values.

3.4. Depolarization Properties of Bulk Aerosols

As mentioned, LIDAR measures the volume depolarization properties of bulk atmospheric aerosols and determines the aerosol type according to the optical properties of particles within the grid. For example, CALIOP applies a systematic aerosol type selection rule with grid precision based on the estimated
particulate depolarization ratio, the 532 nm integrated attenuated backscatter ($\gamma'$), the layer top ($Z_{\text{top}}$) and base ($Z_{\text{base}}$) altitudes, and the land cover types (Winker et al., 2009), while the POPC identifies the optical properties of an air mass based on particle-by-particle detection. Hence, the POPC cannot provide vertical information on an aerosol mass; however, the instrument can still help further understand and investigate LIDAR data.

Figure 8 shows a scatter plot between the s component and p component and the contribution of each aerosol type to the total detected signals on typical pollution days: 26 March, which was dominated by anthropogenic pollutants; 7 April, which was dominated by transported natural dust; and 12 April, when a large amount of polluted dust was present. The scatter plot of anthropogenic pollutant particles is concentrated at approximately −0.07 of the log (s component) and 0.04 of the log (p component). On the typical day of anthropogenic pollution, the air pollution particles were dominated by the p component in the backscattering signals (81.1%), while dust particles contributed more than 60% to the s component because of the irregular morphology (Figure 8a). Figures 8b and 8c show the optical properties of the Asian dust and mixed dust that were first to arrive. The signal of dust dominated the sum of the s component, which was 91.2% and 92%, whereas dust accounted for approximately half of the p component. The proportion of polluted dust that accounted for the s component was maintained at 6–7%, while the proportion in the p component doubled, increasing by a factor of 1.3 compared to the value on 7 April. This finding may be explained by the more complex interaction between dust and water vapor. The scattering signal of coarse-mode particles (Dp > 2.5 μm) was fitted, as the black line shows, and the slope of the distribution is indicated by $s/p$. Apparently, the slope was lower when a substantial amount of polluted dust was present (Figure 8c), as the slope was determined by the proportion of internally mixed dust.

This estimation method based on the POPC-detected single-particle depolarization ratio and optical size is not completely accurate; for example, it cannot identify the proportion of fine dust involved in the internal mixing process, and the contribution of air pollutants may be overestimated. However, the result of this approach may still contribute to the in-depth analysis of LIDAR-detected data and improve the accuracy of classification results, especially in the PBL. Of note, the results were not exactly comparable because the aerosol scattering angle measured with LIDAR was different.

4. Conclusions

Asian dust is often mixed with air-polluting aerosols during transport. The mixing states of dust and other aerosols are an important concern in the assessment of aerosol effects on climate. In this study, the springtime outflow and depolarization properties of Asian dust processes were investigated via synergetic analysis with POPC and space-borne LIDAR observations.

We identified two prominent outflow patterns. Pattern I was characterized by a strong Siberian low-pressure anomaly spinning from 40°E to 110°E with a horizontal longitude scale of 60–90° at 500 hPa. Pattern II was induced by a shallow Mongolian low-pressure system. The atmospheric stratification was significantly different between the different circulation patterns. Pattern I had a strong uplifting flow, and the elevated dust layer tended to be transported at higher altitudes (2–10 km) and mostly stayed uncontaminated. In Pattern II, dust was transported in the low troposphere (<6 km), and a tendency toward a significant decrease in the volume depolarization ratio and the production of polluted dust was found in the PBL. Among the five cases of large-scale dust outbreaks in the spring of 2018, three cases belonged to Pattern I, while the other two cases belonged to Pattern II.

In the PBL of North China, fine-mode spherical particles (pollutants) almost always existed in the background, generally leading to $\delta$ values of −0.1 ± 0.02. Once influenced by dust air masses, irregular dust in the fine mode will lead to a sharp increase in the $\delta$ value, reaching twice the reference value.

Polluted Asian dust was observed at the ends of the dust events when most of the coarse-mode particles were deposited. High RH and a high proportion of secondary pollutants in the atmosphere are necessary for the formation of polluted dust. Under extremely humid conditions (RH > 90%), an increase of ~6.3% in the volume of polluted dust particles at 5 μm was estimated due to the continuous coating process and absorption of moisture. We also estimated the contribution of each aerosol type to the s component and p
component signals of bulk aerosols on different pollution days. The results may contribute to the analysis of LIDAR detection data.

This study demonstrated the essential role that the POPC instrument plays as a supplement for LIDAR polarization observations, especially in high industrial pollution. The comparison of different dust episodes provides in-depth insights into key meteorological factors that affect changes in the mixing states of mineral dust and other aerosols. This study also provides clues and plays an essential role in assessing the contributions of the interaction between dust and anthropogenic aerosols to direct and indirect climate effects.

Data Availability Statement

The data used in this study are available online (http://doi.org/10.5281/zenodo.3856127).

Acknowledgments

This work was supported by the National Natural Science Foundation of China (Grant 41675128) and the National Key Research and Development Project (Grant 2016YFC0207904). We thank NASA and the French National Centre for Space Studies for the CALIOP data, the NOAA ARL for the HYSPLIT transport and dispersion model, and the ECMWF for the ERA-Interim data.

References

Amsler, P., Stetser, O., Schnaiter, M., Hesse, E., Benza, S., Moehler, O., & Lohmann, U. (2009). Ice crystal habits from cloud chamber studies obtained by in-line holographic microscopy related to depolarization measurements. Applied Optics, 48(30), 5811–5822. https://doi.org/10.1364/AO.48.058511

Archer, D., Winguth, A., Lea, D., & Mahowald, N. (2000). What caused the glacial/interglacial atmospheric pCO2 cycles? Reviews of Geophysics, 38(2), 159–189. https://doi.org/10.1029/1999RG000066

Baumgardner, D., Jonsson, H., Dawson, W., O’Connor, D., & Newton, R. (2001). The cloud, aerosol and precipitation spectrometer: A new instrument for cloud studies. Atmospheric Research, 59-60, 251–264. https://doi.org/10.1016/s0169-8095(01)00119-3

Grousset, F. E., Ginoux, P., Bory, A., & Biscaye, P. E. (2003). Case study of a Chinese dust plume reaching the French Alps. Geophysical Research Letters, 30(6), 1277. https://doi.org/10.1029/2002GL016833

Haywood, J. M., Pelon, J., Formenti, P., Bharhal, N., Brooks, M., Capes, G., et al. (2008). Overview of the dust and biomass-burning experiment and African monsoon multidisciplinary analysis special observing period 0. Journal of Geophysical Research, 113, D00C17. https://doi.org/10.1029/2008JD001077

Hu, Y., Vaughan, M., Liu, Z., Lin, B., Yang, P., Flitner, D., et al. (2007). The depolarization-attenuated backscatter relation: CALIPSO lidar measurements vs. theory. Optics Express, 15(9), 5327–5332. https://doi.org/10.1364/oe.15.055327
Huang, J., Minnis, P., Chen, B., Huang, Z., Liu, Z., Zhao, Q., et al. (2008). Long-range transport and vertical structure of Asian dust from CALIPSO and surface measurements during PACDEX. *Journal of Geophysical Research, 113*, D23212. https://doi.org/10.1029/2008JD010620

Huang, Z. W., Huang, J. P., Hayasaka, T., Wang, S. S., Zhou, T., & Jin, H. C. (2015). Short-cut transport path for Asian dust directly to the Arctic: A case study. *Environmental Research Letters, 10*(11), 114018. https://doi.org/10.1088/1748-9326/10/11/114018

Ichoku, C. (2002). A spatio-temporal approach for global validation and analysis of MODIS aerosol products. *Geophysical Research Letters, 29*(12). https://doi.org/10.1029/2001GL013206

Itahashi, S., Yumimoto, K., Uno, I., Eguchi, K., Takemura, T., Hara, Y., et al. (2010). Structure of dust and air pollutant outflow over East Asia in the spring. *Geophysical Research Letters, 37*, L20806. https://doi.org/10.1029/2010GL044776

Kandler, K., Schulte, L., Deutscher, C., Ebert, M., Hofmann, H., Jäckel, S., et al. (2017). Size distribution, mass concentration, chemical and mineralogical composition and derived optical parameters of the boundary layer aerosol at Tinfou, Morocco, during SAMUM 2006. *Tellus B: Chemical and Physical Meteorology, 69*(1), 32–50. https://doi.org/10.1111/j.1600-0898.2006.00385.x

Kaufman, Y. J., Tanre, D., & Boucher, O. (2002). A satellite view of aerosols in the climate system. *Nature, 419*(6903), 215–223. https://doi.org/10.1038/nature01091

Kobayashi, H., Hayashii, M., Shiraishi, K., Nakura, Y., Enomoto, T., Miura, K., et al. (2014). Development of a polarization optical particle counter capable of aerosol type classification. *Atmospheric Environment, 97*, 486–492. https://doi.org/10.1016/j.atmosenv.2014.05.006

Koehler, K. A., Kreidenweis, S. M., DeMott, P. J., Petters, M. D., Prenni, A. J., & Mohler, O. (2010). Laboratory investigations of the impact of mineral dust aerosol on cold cloud formation. *Atmospheric Chemistry and Physics, 10*(23), 11955–11968. https://doi.org/10.5194/acp-10-11955-2010

Krupke, B. J., Grassian, V. H., Idema, M. J., Cowin, J. P., & Laskin, A. (2003). Probing heterogeneous chemistry of individual atmospheric particles using scanning electron microscopy and energy-dispersive X-ray analysis. *Analytical Chemistry, 75*(19), 5170–5179. https://doi.org/10.1021/ac034455t

Kubilay, N., Cokacar, T., & Oguz, T. (2003). Optical properties of mineral dust outbreaks over the northeastern Mediterranean. *Journal of Geophysical Research, 108*(D21), 4666. https://doi.org/10.1029/2003JD003798

Levy, R. C., Mattoo, S., Munchak, L. A., Remer, L. A., Sayer, A. M., Patadia, F., & Hsu, N. C. (2013). The collection 6 MODIS aerosol products over land and ocean. *Atmospheric Measurement Techniques, 6*(11), 2989–3034. https://doi.org/10.5194/amt-6-2989-2013

Li, G., Bei, N., Cao, J., Huang, R., Wu, J., Feng, T., et al. (2016). A possible pathway for rapid growth of sulfate during haze days in China. *Science Reports, 6*, 3034. https://doi.org/10.1038/srep06634

Liu, X., Shi, X., Zhang, K., Jensen, E. J., Gettelman, A., Barahona, D., et al. (2012). Sensitivity studies of dust ice nuclei effect on cirrus clouds with the Community Atmosphere Model CAM5. *Atmospheric Chemistry and Physics, 12*(24), 12,061–12,079. https://doi.org/10.5194/acp-12-12061-2012

Liu, Y. J., Zhu, T., Zhao, D. F., & Zhang, Z. F. (2008). Investigation of the hygroscopic properties of Ca(NO3)2 and internally mixed Ca(NO3)2/Na2CO3 particles by micro-Raman spectrometry. *Atmospheric Chemistry and Physics, 8*(23), 7205–7215. https://doi.org/10.5194/acp-8-7205-2008

Ma, Q., Liu, Y., Liu, C., & He, H. (2012). Heterogeneous reaction of acetic acid on Mg0.8Al0.2O3 and CaCO3 and the effect on the hygroscopic behaviour of these particles. *Physical Chemistry Chemical Physics, 14*(23), 8403–8409. https://doi.org/10.1039/c2cp40530e

Mao, R., Ho, C.-H., Shao, Y., Gong, D.-Y., & Kim, J. (2011). Influence of Arctic Oscillation on dust activity over northeast Asia. *Atmospheric Environment, 45*(2), 326–337. https://doi.org/10.1016/j.atmosenv.2010.10.020

Martin, S. T. (2000). Phase Transitions of Aqueous Atmospheric Particles. *Chemical Reviews, 100*(9), 3403–3454.

Martin, R. V., Jacob, D. J., Yantosca, R. M., Chin, M., & Ginoux, P. (2003). Global and regional decreases in tropospheric oxidants from photochemical effects of aerosols. *Journal of Geophysical Research, 108*(D3), 4097–4097. https://doi.org/10.1029/2002JD002622

Nie, W., Ding, A., Wang, T., Kerminen, V. M., George, C., Xue, L., et al. (2014). Polluted dust promotes new particle formation and growth. *Scientific Reports, 4*, 6634. https://doi.org/10.1038/srep06634

Okada, K., Heintzenberg, J., Kai, K., & Qin, Y. (2001). Shape of atmospheric mineral particles collected in three Chinese arid-regions. *Geophysical Research Letters, 28*(16), 3123–3126. https://doi.org/10.1029/2000GL012798

Pan, X., Uno, I., Wang, Z., Nishizawa, T., Sugimoto, N., Yamamoto, S., et al. (2017). Real-time observational evidence of changing Asian dust morphology with the mixing of heavy anthropogenic pollution. *Scientific Reports, 7*(1), 335. https://doi.org/10.1038/s41598-017-00444-w

Pan, X. L., Ge, B., Wang, Z., Tian, Y., Liu, H., Wei, L., et al. (2019). Synergistic effect of water-soluble species and relative humidity on morphological changes in aerosol particles in the Beijing megacity during severe pollution episodes. *Atmospheric Chemistry and Physics, 19*(1), 219–232. https://doi.org/10.5194/acp-19-219-2019

Pan, X. L., Uno, I., Hara, Y., Kuribayashi, M., Kobayashi, H., Sugimoto, N., et al. (2015). Observation of the simultaneous transport of Asian mineral dust aerosols with anthropogenic pollutants using a POPC during a long-lasting dust event in late spring 2014. *Geophysical Research Letters, 42*, 1593–1598. https://doi.org/10.1002/2014GL062491

Pan, X. L., Uno, I., Hara, Y., Okada, K., Yamamoto, S., Wang, Z., et al. (2016). Polarization properties of aerosol particles over western Japan: Classification, seasonal variation, and implications for air quality. *Atmospheric Chemistry and Physics, 16*(15), 9863–9873. https://doi.org/10.5194/acp-16-9863-2016

Redding, B., Pab, Y., Wang, C., Videen, G., & Caoa, H. (2014). Polarization resolved angular optical scattering of aerosol particles. https://doi.org/10.1117/12.2050022

Rocha-Lima, A., Martins, J. V., Remer, L. A., Todd, M., Marshall, J. H., Engelstaedter, S., et al. (2018). A detailed characterization of the Saharan dust collected during the Fennec campaign in 2011: In situ ground-based and laboratory measurements. *Atmospheric Chemistry and Physics, 18*(2), 1023–1043. https://doi.org/10.5194/acp-18-1023-2018
Wu, F., Zhang, D., Cao, J., Guo, X., Xia, Y., Zhang, T., et al. (2016). Limited production of sulfate and nitrate on front-associated dust storm particles moving from desert to distant populated areas in northwestern China. *Atmospheric Chemistry and Physics Discussions, 17*(23), 14473. https://doi.org/10.5194/acp-2016-853

Xiang, Y., Liu, J. G., Zhang, T. S., Lv, Y. H., & Fu, Y. B. (2018). Uncertainty factors of aerosol optical properties inversion by lidar. *Laser & Optoelectronics Progress, 55*(9), 092801. https://doi.org/10.3788/LOP55.092801

Xing, C., Liu, C., Wang, S., Chan, K. L., Gao, Y., Huang, X., et al. (2017). Observations of the vertical distributions of summertime atmospheric pollutants and the corresponding ozone production in Shanghai, China. *Atmospheric Chemistry and Physics, 17*(23), 14,275–14,289. https://doi.org/10.5194/acp-2017-14275-2017

Yang, X., Zhao, C., Zhou, L., Wang, Y., & Liu, X. (2016). Distinct impact of different types of aerosols on surface solar radiation in China. *Journal of Geophysical Research: Atmospheres, 121*, 6459–6471. https://doi.org/10.1002/2016JD024938

Zenker, J., Collier, K. N., Xu, G., Yang, P., Levin, E. J. T., Suski, K. J., et al. (2017). Using depolarization to quantify ice nucleating particle concentrations: A new method. *Atmospheric Measurement Techniques Discussions, 1–42*. https://doi.org/10.5194/amt-2017-166

Zhang, C., Liu, C., Hu, Q., Cai, Z., Su, W., Xia, C., et al. (2019). Satellite UV-Vis spectroscopy: Implications for air quality trends and their driving forces in China during 2005-2017. *Light: Science & Applications, 8*(1), 100. https://doi.org/10.1038/s41377-019-0210-6

Zhang, D., Iwasaka, Y., Shi, G., Zang, J., Hu, M., & Li, C. (2005). Separated status of the natural dust plume and polluted air masses in an Asian dust storm event at coastal areas of China. *Journal of Geophysical Research, 110*, D06302. https://doi.org/10.1029/2004JD005305

Zhang, R., Wang, G., Guo, S., Zamora, M. L., Ying, Q., Lin, Y., et al. (2015). Formation of urban fine particulate matter. *Chemical Reviews, 115*(10), 3803–3855. https://doi.org/10.1021/acs.chemrev.5b00067

Zhang, X., Zhao, L., Tong, D., Wu, G., Dan, M., & Teng, B. (2016). A systematic review of global desert dust and associated human health effects. *Atmosphere*, 7(12), 158. https://doi.org/10.3390/atmos7120158

Zhang, X.-X., Sharratt, B., Liu, L. Y., Wang, Z. F., Pan, X. L., Lei, J. Q., et al. (2018). East Asian dust storm in May 2017: Observations, modelling, and its influence on the Asia-Pacific region. *Atmospheric Chemistry and Physics, 18*(11), 8353–8371. https://doi.org/10.5194/acp-18-8353-2018

Zhao, Y., Huang, A., Zhu, X., Zhou, Y., & Huang, Y. (2013). The impact of the winter North Atlantic Oscillation on the frequency of spring dust storms over Tarim Basin in northwest China in the past half-century. *Environmental Research Letters, 8*(2), 024026. https://doi.org/10.1088/1748-9326/8/2/024026

Zhu, C., Wang, B., & Qian, W. (2008). Why do dust storms decrease in northern China concurrently with the recent global warming?. *Geophysical Research Letters, 35*, L18702. https://doi.org/10.1029/2008GL034886