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Chemical composition and mass size distribution of $PM_{1.0}$ at an elevated site in central east China

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Abstract

Size-resolved aerosol chemical compositions were measured continuously for one and half years with an aerosol mass spectrometer (AMS) to characterize the mass and size distributions (MSDs) of each component in bulk, fresh and aged submicron particles (approximately PM$_{1.0}$) at Mountain Tai, an elevated site in Central East China (CEC) from June 2010 to January 2012. The majority of the regionally-dispersed aerosols were found to be contributed from short distance mixed aerosol, mostly from its south with organics and sulfate as the major components. The annual mean mass concentrations of organics, sulfate, nitrate, ammonium and chloride were 11.2, 9.2, 7.2, 5.8 and 0.95 μg m^{-3}, respectively, which are much lower for organics and sulfate, and slightly lower for nitrate, ammonium and chloride than those at the nearby surface rural sites. High organics were observed for all four seasons, and the relatively fresh organic aerosol (OA) containing high proportion of less-photo chemically OA, were found from long-range transported aerosol from northwest. Semi-volatile and low-volatile oxidized OAs together contributed approximately 49 %, 55 % in spring and 72 % and 51 % in winter of total OA, showing at least 50 % of OA can be attributable to SOA. Seasonally, the chemical components at the elevated site showed a "winter high and autumn low" pattern, with organics, sulfate and ammonium peaking in summer. Though no obvious differences of MSDs were seen for various chemical components in the planetary boundary layer (PBL) and free troposphere (FT), the concentrations were a factor of 5–7 higher in PBL than in FT. The averaged MSDs of particles between 30–1000 nm for organics, sulfate, nitrate, and ammonium are approximately log-normal with similar mass median diameters (MMDs) of 539, 585, 542, and 545 nm, respectively, which were slightly larger than those in ground sites within North China Plain (NCP). Obvious differences in MMDs were found between fresh and aged aerosols for sulfate and ammonium, with smaller increased size-factors for the Mt. Tai aerosols than those in less polluted areas. All these exhibit the relative aged and well-mixed aerosol observed.

1 Introduction

The influence of regionally dispersed fine aerosols on weather-climate becomes more important than that immediately influenced by local specific source and by coarser particles, whose chemical composition and size distribution are two critical parameters. The mass-size distributions (MSDs) of fresh and aged aerosol chemical components are also critical for improving the simulation and validation of the aerosol size distributions and their optical properties (Zhou et al., 2012), especially for the aerosol in elevated sites. The size distributions of the principal fine particle constituents also have important impacts on visibility (Watson, 2002), aerosol radiative properties and cloud-nucleating ability of aerosol particles (Boucher et al., 2013).

As to the global scale, tropospheric aerosols are highly variable in time and space, in which the aerosol chemical components over China are found to be much higher than those in majority areas of the world, except for urban area in S. Asia (Zhang et al., 2012a). The sampling site in this study, Mountain Tai (Mt. Tai), is located in the Central East China with a peak elevation of ~ 1500 m (a.s.l.) optimal for investigating the regionally dispersed pollution, as the tropospheric aerosols over mountains are most often representative of regional- to large-scale mixed origins (Li et al., 2011; Wang et al., 2011). Although there are some recent studies that investigated particulate concentrations at Mt. Tai, they have been limited on the measurement of organic molecular compositions (Fu et al., 2008), gas-phase total peroxides (Ren et al., 2009), role of biogenic volatile organic compounds (Fu et al., 2010), the impact of open crop residual burning on O_3, CO, black carbon (BC or called EC) and organic carbon (OC) (Yamaji et al., 2010), EC-OC and inorganic ions in PM_{10} during spring time (Wang et al., 2011), individual particle analysis (Li et al., 2011), and they are only limited on one or two seasons with daily sampling resolutions. It is needed to have systematic measurements of MSDs of chemical components in fine aerosol particles, covering seasonal variations on a long-term basis, which is essential to accurately and objectively assess the characterization of various chemical components of fine aerosols in high atmospheric level in CEC.

This paper presents a data set covering one and half years of measurement of the mass concentrations and size distributions of selected chemical components in $PM_{1.0}$. The purpose was to assess the regionally representative concentration levels of different aerosol chemical components, and to obtain the seasonal variations. Through the identification of relative fresh (associated with new particle formation event) and aged aerosol, and aerosols in planet boundary layer (PBL) and free troposphere (FT) air mass, the MSDs of organic, sulfate, nitrate and ammonium under different conditions were estimated for better characterizing the aerosol chemical compositions of well-mixed aerosol in this area, and also for model verification. As the site is often influenced by regional-scale transported pollutants, the chemical and size properties of $PM_{1.0}$ from different air masses are also discussed. Finally, the secondary organic aerosol (SOA) fraction was identified by using a positive matrix factorization method (PMF) which was based on the time-resolved on-line measurements.

2 Experimental

2.1 Campaign description

As part of aerosol-cloud interaction campaign, supported by National Key Project of Basic Research, the aerosol chemical composition analyses were conducted at the summit of Mt. Tai (36.251° N, 117.101° E), located in Shandong Province of China with the highest elevation (1534 m a.s.l.) in CEC and settled along the pathway of Asian continental outflow. Tai'an is the nearest small city, 15 km away in the south with ~ 500 000 population (Fig. 1) while Jinan, the capital city of Shandong Province (population: 2.1 million) is 60 km away in the north. Because Mt. Tai station is situated close to the top of the planetary boundary layer, and the aerosol samples received are representative of the region rather than the immediate locality, it is a suitable site for investigating the regionally dispersed aerosol pollution over the heavily polluted CEC.

From June 2010 to January 2012, an aerosol mass spectrometer (Q-AMS, Aerodyne Research Inc. Boston, MA, USA) was used to measure the mass concentrations and size distributions (30 nm–1 µm) of organics, sulfate, nitrate, ammonium and chloride with 5 min time resolution (Jayne et al., 2000; Zhang et al., 2011). The sampling periods for the AMS study were from 24 June–15 August, 22 September–11 October in 2010, 26 March–20 April, 5–30 June, 22 October–1 December in 2011 and 22 December 2011–13 January 2012. As a result of interruptions due to calibration, instrument failures and local burning events, 123 days of valid data were obtained. From July to December in 2010, a scanning mobility particle sizer (SMPS) (TSI 3936, TSI Inc.) was also used to monitor the number size distribution of aerosols over a size range of 12.6–552 nm at a time resolution of 5 min (2.5 min per scan, two scans) (Zhang et al., 2011). From 25 December 2010 until the end of the study, a tandem differential mobility particle sizer (TDMPS, IFT, Leibniz Institute for Tropospheric Research, Germany) and an aerodynamic particle sizer (APS, model 3321, TSI Inc., St Paul, USA) were used instead of the SMPS for particle number size distribution (PNSD) measurements; together these two instruments covered particles from 3 nm to 2.5 µm in diameter. To correct the concentrations for bounce at the vaporizer and the partial transmission of particles by the lens, a fixed particle-collection efficiency factor of 0.5 was used – this is the value that was obtained in studies conducted in Beijing (Sun et al., 2010; Zhang et al., 2011, 2012b), and a comparison of results presented below indicates that a reasonable correction was conducted.

Detection limits (DLs) for the AMS mass concentration were evaluated based on the mass spectra of particle-free ambient air, that is, air filtered with a HEPA filter. The DLs were defined as three times the standard deviation of each species signal in the particle-free air. During this study, the DLs for sulfate, nitrate, ammonium, organics and chloride for the 5 min averaged data were 0.03, 0.017, 0.063, 0.101, and 0.01 $\mu g\,m^{-3}$, respectively (Zhang et al., 2011).

The room temperature was controlled at 25 ± 3 °C, and the RH at 40–60 % and atmospheric air was sampled through a PM_{10} impactor, which was followed by a $PM_{2.5}$ cyclone (the flow rate was $16.7\,L\,min^{-1}$) and dried to a RH < 30 % with the use of an automatic aerosol dryer unit (Tuch et al., 2009).

2.2 Quality assurance of the data

To validate the assumed collection efficiency and demonstrate the data quality of our measurements, a plot of the NR-$PM_{1.0}$ mass concentration from the AMS against the reconstructed mass obtained with the SMPS is shown in Fig. 2. Here, the aerosol density used in SMPS mass calculations was assumed to be that of the average composition of $(NH_4)_2SO_4$, NH_4NO_3 and organics, whose densities are 1.77, 1.72, and $1.3\,g\,cm^{-3}$, respectively. As noted above, fixed collection efficiency (CE) of 0.5 was assumed for the entire campaign, which generally yielded a good correlation between the AMS and reconstructed SMPS mass data. The coefficient of determination (r^2) of AMS mass vs. SMPS mass was 0.744 with a slope of 0.984, demonstrating a good level of agreement in the masses obtained by the two methods.

2.3 Data separation of PBL, FT and different air-masses

Characterization of air masses sampled at the site from either PBL or FT is important to the subsequent analysis of the chemical data. In this study, the criteria developed by (Gallagher et al., 2011) were used to differentiate the data influenced by the PBL vs. the FT. Days without well-defined diurnal cycles of water vapour and number concentration were considered to have been influenced by the FT. The time of day with enhanced water vapour and high aerosol concentration at the mountaintop was considered to be a signal of PBL influence on the mountain.

To characterize the long range transport pathways, 72 h back trajectories were computed every 6 h (00:00, 6:00, 12:00 and 18:00 UTC) using the HYSPLIT-4.8 (Hybrid Single Partical Lagrangian Integrated Trajectories) model of the National Oceanic and Atmospheric Administration, USA (NOAA) (http://www.arl.noaa.gov/ready/hysplit4.html). The resolution of the terrain data in the HYSPLIT model is 1° × 1°, thus the real height of the mountain site is smoothed. So 1500 m above the model ground level was chosen as trajectory start height, which is about 840 hPa in the model and is a little lower than the pressure of the measurement site (845 hPa). TrajStat (Wang et al., 2009), a program that using trajectory statistical analysis data to identify potential source pathways and locations from long-term air pollution measurements, was used to construct clusters from the air-mass trajectories for long range transport pathways study. The Global Data Assimilation System (GDAS) meteorological data archives of the Air Resource Laboratory, NOAA, were used as the input.

3 Results and discussions

3.1 Relatively low concentration of chemical components in ~ 1500 m high atmosphere of Mt. Tai

The annual mean mass concentrations of organics, sulfate, nitrate, ammonium and chloride (OSNAC) in $PM_{1.0}$ were 11.2, 9.2, 7.2, 5.8 and 0.95 $\mu g\,m^{-3}$, respectively, totalling ~ 34.3 $\mu g\,m^{-3}$ (Table 1). This $PM_{1.0}$ concentration is roughly half of the ground-based values at urban Beijing (~ 500 km northwest of the Mt. Tai) (~ 76 $\mu g\,m^{-3}$) (Zhang et al., 2012b), lower than that in sub-urban Gucheng station (~ 52 $\mu g\,m^{-3}$), ~ 38 % of the sub-urban Tianjin level (~ 80 km southwest from Beijing), and lower than those at several urban/rural sites in the Pearl River Delta in China (Xiao et al., 2011). This $PM_{1.0}$ loading at Mt. Tai is higher than those in several European cities (10–30 $\mu g\,m^{-3}$) (Lanz et al., 2007) and several field campaigns conducted at various ground sites in urban areas, downwind of urban areas, and rural/remote locations in the mid-latitudes of the Northern Hemisphere (Zhang et al., 2007). It is much higher than springtime value (1.9 $\mu g\,m^{-3}$) at a high latitude site (Whistler Mountain with 2182 a.s.l.) (Sun et al., 2009), and comparable to that at Mexico City (Volkamer et al., 2006).

These mean mass concentrations of chemical components are about factor of 2–3 lower (for organics and sulfate), and slightly lower (for nitrate, ammonium and chloride) than those at the near surface rural areas of China (Zhang et al., 2012a). This comparison is made on the basis of the fact that the secondary aerosols (sulfate, nitrate, ammonium and chloride, secondary organics), and high-temperature processes-emitted primary organics are mainly associated with fine aerosol particles with equal concentrations in PM_{10} and $PM_{1.0}$. For consistency, the mass of OC (Zhang et al., 2012a) has been converted to that of organics with conversion factor of 1.5.

Seasonally, minimum concentrations of all chemical components were found in autumn while sulfate, ammonium and organics peaked in summer and nitrate peaked in winter (Table 1). This is somewhat different from typical seasonal patterns of spring minimum in various aerosol components at ground-based measurement in China (Zhang et al., 2012a). Another unique feature of this site is the maximum concentration found in summer not only for sulfate, ammonium, but also for organics. This can be explained as the high secondary organics formed in summertime, due to plenty of VOCs, enhanced photochemistry, low air mass ventilation on a regional scale.

3.2 Relatively aged chemical components with summer maximum found at the elevated site

The MSDs of organics, sulfate, nitrate and ammonium in particles with diameters between 30 and 1000 nm were found to be approximately lognormal. The mass median diameters (MMDs) for bulk organics, sulfate, nitrate, and ammonium in entire observational period were quite similar, with 539, 585, 542 and 545 nm, respectively. The majorities of particle mass are with diameter larger than 100 nm (Table 1). Consequently, the relative larger MMDs were also found at this high elevated site than those at several ground sites in the NCP, including the sub-urban Gucheng station and sub-urban Tianjin, and urban Beijing (Table 1), showing the relative aged and regionally-dispersed fine aerosols received compared with those from ground-based measurement.

Information on the seasonality in the MSDs for the four major aerosol components is also presented in Table 1. The chemical components exhibited some similarities during the four seasons. Different from mass concentrations, seasonally, maximum MMDs for almost all chemical components were found in summer and secondary high values in autumn with minimum in spring, exhibiting the summer-half year is more conducive for aged fine aerosol in the higher atmosphere. The large sulfate, ammonium and organics in summer are consistent with the highest mass concentration observed in summer. In addition, the smallest MMDs were found in springtime, which was probably due to the transport of relatively fresh aerosols from the north.

The similar MMDs for each of the chemical components in the FT and PBL aerosols (Table 1) indicate the similarities of the characteristics of chemical components of aerosols in FT and PBL. However, the concentrations in PBL were a factor of 5–7 higher than those in FT, exhibiting that vast majority of aerosol was mixed within PBL.

3.3 MSDs of chemical components in fresh and aged episodes

As previously mentioned, the MSDs of organics, sulfate, nitrate and ammonium between 30 and 1000 nm particles were approximately lognormal. The standard deviations (σ_g) of the fitted MSDs are generally smaller than 2, showing that there were minor impacts of the nucleation mode or finer particle on the MSDs for each chemical component. Though the AMS cannot fully cover size range of ultra-fine particle, a fitting of mono mode log-normal distributions to the 30–1000 nm data provides a way to assess the relative importance of ultra-fine particles. Generally, a standard deviation of the fitted MSD near or above 2 was assumed to have relatively strong influences from ultra-fine particles, and relatively fresh aerosol particles. After new particle formation (NPF) event and local fresh air periods, which were identified based on the evolution of the size distributions and particle number concentrations following Kulmala's definitions of (Kulmala et al., 2004) and by the in-situ aerosol number size distribution and meteorological measurement, we can find the increased value of σ_g for the mono model fitting, and larger than 2 of σ_g for specific chemical components during relative fresh episodes (Table 2a). For comparisons, the MSDs of chemical components during aged pollution are also summarized in Table 2b.

The MMDs of organics, sulfate, nitrate and ammonium are typically smaller for fresh aerosols than for aged ones. This can be seen in the comparatively small MMDs (~ 373–459 nm) for organics during nucleation event or local fresh air at Mt. Tai and the ~ 30–70 % larger MMDs (~ 473–792 nm) during polluted episodes. The largest differences in MMDs between fresh and aged aerosols were found for nitrate and sulfate: the MMDs increased ~ 60–80 % and 50–80 %, respectively. Condensation is the possible process most responsible for transferring low-volatility vapours onto aerosol particles, and this is usually what is responsible for the particles' growth to larger sizes (Riipinen et al., 2011; Donahue et al., 2011). Coagulation is another possible important sink for ultrafine particles (Pierce and Adams, 2007), and the hygroscopicity of aerosol species also can contribute to their growth.

Compared with the Mt. Tai, the less MMD increasing factors for organics between fresh air and aged air have been reported for an urban ground site in Beijing (~ 38–61 %), a suburban site in Wuqing (~ 27–32 %), and a rural site in Gucheng (8–42 %). The increasing factors for sulfate, nitrate and ammonium in Beijing were all lower than that at Mt. Tai, suggesting again the relative aged aerosol observed in Mt. Tai. At Wuqing, the nitrate increasing factor was larger than other chemical components.

The increasing factors of MMD from fresh to aged aerosol at Mt. Tai are smaller compared with those in less polluted areas. At an urban site in Manchester, the particle sizes for the aerosol species in aged air were 110–270 % higher than fresh air. At a regional site in Pittsburgh, USA, the increasing factors for organics, sulfate, nitrate and ammonium from a nucleation event to aged air were 165 %, 200 %, 29 %, 160 %, respectively. Therefore, the evolution of MMDs at a certain degree indicates the pollution level and particle aging.

3.4 Transport pathways and associated chemical component changes

Seven main back-trajectory clusters were identified by using TrajStat and the HYSPLIT-4.8 model (Fig. 1). Cluster-1 (~ 17 % of the total) represents the shortest transport pathway, which was from the south. These trajectories passed over Xuzhou in Anhui Province, an area that suffers from serious pollution, mainly due to biomass burning (Wang et al., 2002; Woo et al., 2003; Suthawaree et al., 2010). Tai' an, a prefecture-level city, located on the southern flank of Mt. Tai, is a likely source for pollution when the airflow followed this pathway. Trajectories in Cluster-2 (~ 16 %) started over the Yellow Sea and passed over the eastern side of the Shandong Peninsula. Air masses following this relatively short path would bring both marine aerosol and ground-level air pollutants to Mt. Tai. Cluster-3 (another 25 %) shows a third short pathway; the back trajectories for these air masses were from the north/northeast of Mt. Tai, starting at Chengde in Hebei Province and then passing through Bohai sea and Shandong Peninsula. The three short distance clusters account for ~ 60 % of all the air masses, showing the majority of regionally-dispersed aerosol received at Mt. Tai was from short distance mixed aerosol, mostly from its south.

Cluster-4 (~ 7 %) shows the airflow from north of China to the NCP, including Tianjin and Hebei Province. These trajectories are representative of regional-scale transport path. Trajectories in Cluster-5 (20 %) originate over remote areas in north/north western China with a transport path above 3000 m high. These air masses swept through Hebei province to the site at high wind speed, which has assumed delivering most of aerosol in FT. Cluster-6 (~ 11 %) represents regional transport from the west of Mt. Tai. Cluster-7 (4 %) represents another group of trajectories from the further northwest with longer distances. The air masses in these four clusters account for another ~ 40 % of all transport, and bring less polluted aerosol from north. These can also be supported by the sums of the mass concentrations of the OSNAC of 51, 59 and 48 $\mu g\,m^{-3}$ for the first three transport path aerosols (in Fig. 1), and 36, 16, 34 and 14 $\mu g\,m^{-3}$ for the rest path aerosols, respectively. The mass loadings for Cluster 6 and 7 were the lowest of seven clusters. Even though they are still much higher than those at Whistler Mt. (1.2 µg m^{-3}), they are close to a similar level to what has been observed at Korea and Japan (11–13 µg m^{-3}) (Zhang et al., 2007). The relative smaller MMDs were also found for chemical components associated with long-distance transported aerosols (cluster 4–7), suggesting the relative fresh and unaltered aerosols (Table 3).

Organics and sulfate were the two largest fractions, accounting for ~ 30 % each for the OSNAC in the airflow associated with the first three short-distant paths (cluster 1, 2 and 3) from the south (Table 3), exhibiting again the more pollutants from Pan-Yangtze river delta area. The nitrate was about 20 % with mean concentrations of 9.0, 11.2, 10.9 µg m^{-3} for cluster 1, 2 and 3 respectively. The ammonium and chloride were the two smallest fractions with ~ 17 % and 2 %, respectively. The much higher organics were found for aerosol travelled long-distance from north relative to the short-distance moving aerosols from south. About 40 % organic were found in aerosols associated with transport cluster 4, 5, 6 and 7.

3.5 Composition of OA

To further investigate the secondary OA, PMF modelling of OA spectra (Aiken et al., 2009) was used to identify presumptive sources for the organic aerosol during each of four seasons at Mt. Tai. Different types of organics were resolved based on specific mass-spectral profiles, and the mass concentrations are presented in Table 4. The mass spectra for Hydrocarbon-like OA (HOA) is characterized by hydrocarbon ions of the general form C_nH_{2n+1} and C_nH_{2n-1}, including $C_3H_7^+$ (m/z 43), $C_4H_7^+$ (m/z 55), and $C_4H_9^+$ (m/z 57). A signal from m/z 60, a tracer ion for biomass burning organic aerosol (BBOA) (Alfarra et al., 2006; Aiken et al., 2009), which can be attributable to POA (Jimenez et al., 2009), was resolved in the mass spectra from spring and summer. In winter, another factor with high m/z 43 and m/z 60 was identified as the coal-combustion organic aerosol (CCOA) (Sun et al., 2013). About 34 % of the organics were from BBOA in summer. In winter, CCOA accounted for 34 % of the organics, showing the coal combustion influence (Table 4). HOA, BBOA and CCOA can be considered as primary organic matter (POA) directly emitted into the atmosphere, accounting for 51 %, 44 %, 28 %, 49 % of the total measured organics in spring, summer, fall and winter, respectively; For HOA, the smallest percent occurred in summer, but larger in spring and fall.

At Mt. Tai, one can still find other large fraction (Table 4) of oxygenated organic aerosol (OOA) that is considered to be secondary generally (Herndon et al., 2008; Volkamer et al., 2006), in which two subtypes of OOA can be distinguished by large fraction of CO_2^+ (m/z 44) for low-volatility oxidized organic aerosol (LV-OOA) and by $C_3H_7^+$ (m/z 43), CO_2^+ (m/z 44), $C_3H_3O^+$ (m/z 55) and $C_3H_5O^+$ (m/z 57) for semi-volatile oxidized organic aerosol (SV-OOA). According to (Jimenez et al., 2009), LV-OOA is strongly correlated with non-volatile secondary species such as sulfate and has a high O : C, mainly attributable to regional, heavily aged OA; SV-OOA has a higher correlation with semi-volatile species such as ammonium nitrate and ammonium chloride and has a lower O : C, consistent with less–photo chemically fresh OA. SV-OOA and LV-OOA together contributed 49, 55, 72 and 51 % from spring to winter, showing at least 50 % of OA can be attributable to SOA in regionally-dispersed aerosol in the CEC. Extra high proportions of SOA found in fall, probably exhibiting the influence of high biomass burning season in October in China (Zhang et al., 2012a). From the trajectories analysis, we find ∼ 16 % and 8 % air-mass associated with long-distance transport cluster (6/7) in fall. This identification about relative contributions of POA and SOA is similar to the report from 2 yr filter measurement from 16 sites in various regions of China, in which report the secondary organic carbon contributed ∼ 55 % and 60 % for urban and rural aerosol, respectively (Zhang et al., 2012a).

For different transport trajectories, the LV-OOA usually accounts for > 40 % of the total organics, exhibiting the regional, heavily aged OA observed at this high mountain site. Of the total organics associated with the two most long-distance cluster 5 and 7, one can find the singular high proportion SV-OOA, indicating that the less–photo chemically fresh OA with semi-volatile characteristics is associated mostly with long-distance air-mass from north.

4 Summary

The mass concentrations and size distributions of chemical components in $PM_{1.0}$ were characterized in situ with AMS at the summit of Mt. Tai (∼ 1500 m a.s.l.) from June 2010 to January 2012.

The mass concentration of organics, sulfate, nitrate, ammonium and chloride (OSNAC) in $PM_{1.0}$ at Mt. Tai was lower than those at urban Beijing, but higher than those at several cities and mountain sites in the USA and Europe. The mean mass concentrations observed in the near surface rural sites of China are about a factor of 2–3 higher for organics and sulfate and slightly higher for nitrate, ammonium and chloride than those in this high elevated Mt. Tai. This probably indicates that the formation of sulfate and organics in the near surface is faster than that of nitrate, leading to more organics and sulfate found in the near surface and close to their emission sources where the relative longer formation time for nitrate probably is beneficial to have more nitrate mixed in large scope and in the elevated site.

The seasonal pattern at the Mt. Tai site is slightly different from typical seasonal patterns in various aerosol components at ground-based measurement in China ("spring minimum"). Seasonally, high concentrations of all chemical components in ∼ 1500 m high level were found in winter with minimum in fall except for organics, sulfate and ammonium which had another peak in summer. The MSDs of chemical components exhibited some similarities during the four seasons. Different from the seasonality of mass concentrations, maximum MMDs for almost all chemical components were found in summer and secondary high values in autumn with minimum in spring, indicating the summer-half of the year is more conducive for aged fine aerosol in high atmosphere. In addition, the smallest MMDs were found in springtime, and this was probably due to the transport of relatively fresh aerosols from the north.

The MSDs for organics, sulfate, nitrate, and ammonium are approximately lognormal with more than 99 % mass fraction attributable to the particle with diameter larger than 100 nm by the AMS measurement. The larger MMDs were found at Mt. Tai than those in ground sites, showing the relative aged and well-mixed aerosol observed. The smaller MMDs (∼ 373–459 nm) were found for organics in relative fresh aerosols comparing to aged aerosols with MMDs of ∼ 473–792 nm. The largest differences in MMDs between fresh and aged aerosols were found for nitrate and sulfate. The MMDs increased by ∼ 60–80 % and 50–80 %, respectively. Compared with the Mt. Tai, the less MMD increasing factors for organics between fresh air and aged air have been reported for an urban ground site in Beijing (∼ 38–61 %), a suburban site in Wuqing (∼ 27–32 %), and a rural site in Gucheng (8–42 %). The increasing factors for sulfate, nitrate and ammonium in Beijing were all lower than that of Mt. Tai, suggesting again the relative aged aerosol observed in Mt. Tai.

At Mt. Tai, one can still find the large fraction (normally > 50 %) of total OA can be attributable to oxygenated organic aerosol (OOA) that is considered to be secondary generally. Extra high proportions of SOA found in fall, probably exhibiting the influence of high biomass burning season in October of China. Regionally, heavily aged OA are observed with > 40 % low-volatility oxidized organic aerosol (LV-OOA) in the total OA. Hydrocarbon-like OA (HOA), biomass burning organic aerosol (BBOA) and coal-combustion organic aerosol (CCOA) considered as primary organic matter (POA) directly emitted into the atmosphere, also accounted together for 51, 44, 28 and 49 % of the total measured organics in spring, summer, fall and winter, respectively. Of the total organics associated with the two most long-distance cluster 5 and 7, one can find the singular high proportion semi-volatile oxidized organic aerosol (SV-OOA), indicating that the less–photo chemically fresh OA with semi volatile characterises is associated mostly with long-distance air-mass from north.

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Table 1. MMDs and mass concentrations for chemical components in this study and comparison with other studies in China and other countries.

Location	Site type	period	Lognormal Distribution between 30 to 1000 nm							
				Organics		Mass[b] (µg m^{-3})		Sulfate		Mass (µg m^{-3})
			MMD	σ_g	% [a]		MMD	σ_g	%	
Mt. Tai, N. China Plain	Regional	Annual (2011)	538	1.7	99	11.2	585	1.7	99.0	9.2
		Spring (2011)	457	1.8	99	8.6	517	1.9	100	7.3
		Summer (2010, 2011)	549	1.7	100	16.4	661	1.8	100	20.1
		Autumn (2010, 2011)	531	1.7	100	5.7	582	1.7	100	5.7
		Winter (2011)	556	1.7	100	11.6	574	1.8	100	8.7
		FT[c]	339–514	1.5–1.9	93–99	3.7–12.8	367–457	1.6–1.9	99.0	1.3–8.8
		PBL[d]	462–522	1.7–1.9	99	13.5–18.2	566–590	1.6–1.9	99.0	11.9–21.7
Gucheng, N. China Plain	Rural	Winter (2006)	514	0.8	99	41.4	506	0.8	99	9.9
		Spring (2006)	394	1.0	99	9.7	426	0.9	99	5.7
Wuqing, Tianjin, N. China Plain	Suburban	Summer (2009)	457	1.9	99	12.1	563	1.7	100	13.3
		Fall (2009)	475	1.9	99	50.8	566	1.8	100	23.5
Beijing, N. China Plain	Urban	Annual (2008)	413	1.9	99	33.5	503	1.7	100	15.0
Whistler mountain, Canada	High altitude	Spring (2006)	313	2.0	93	1.1	461	1.6	83	0.6
Instituto Mexicano del Petroleo, Mexico	Urban	Spring (2006)	305	2.0	86	29.2	434	1.7	81	6.7
Vienna, Austria	Urban	Winter (2004)					480	2.2		1.5
Ljubljana, Slovenia	Urban	Winter (2004)	550	1.9		29.2	440	2.1		6.7
Fukue, Japan	Rural/remote	Spring (2003)	394	1.6	87	5.0	440	1.5	94	4.8
Tokoy, Japan	Urban	Summer (2003)	291	2.8	84	5.6	508	1.8	75	3.2
		Fall (2003)	286	2.4	83	15.4	439	1.9	80	19.5
		Winter (2003)	252	2.6	87	10.3	439	1.6	84	15.3

Table 1. Continued.

Location		Nitrate		Mass (µg m^{-3})		Ammonium		Mass (µg m^{-3})	
	MMD	σ_g	%		MMD	σ_g	%		References
Mt. Tai, N. China Plain	540	1.74	99	7.2	541	1.7	99	5.8	This study
	489	1.82	100	8.8	495	1.9	99	5.6	
	633	1.63	100	8.3	614	1.7	100	11.0	
	565	1.69	100	3.8	551	1.7	100	2.9	
	501	1.82	100	9.6	506	1.8	100	6.8	
	365–550	1.5–1.8	97–99	0.2–14	351–502	1.5–1.8	96–99	0.4–8	
	558–561	1.6–1.8	99	7.7–14.9	544–561	1.7–1.8	99	7.1–13.2	
Gucheng, N. China Plain	483	0.80	99	10.7	439	0.8	100	9.1	Zhang (2011)
	527	0.75	99	4.6	434	0.9	100	3.7	
Wuqing, Tianjin, N. China Plain	585	1.61	100	6.2	535	1.6	99	7.3	
	521	1.85	100	17.5	489	1.8	100	17.9	
Beijing, N. China Plain	492	1.69	100	14.4	460	1.7	99	8.7	Zhang et al. (2012)
Whistler mountain, Canada					536	1.8	70	0.2	Sun et al. (2009)
Instituto Mexicano del Petroleo, Mexico	371	1.7	87	4.5	394	1.8	83	1.8	Aiken et al. (2009)
Vienna, Austria									Hitzenberger et al. (2006)
Ljubljana, Slovenia	420	1.8		4.5	600	2.3		1.8	
Fukue, Japan	427	1.5	99	0.6				1.6	Takami et al. (2005)
Tokoy,Japan	523	1.6	33	1.0	527	1.6	76	3.2	Takagawa et al. (2006)
	430	2.0	80	20.0	451	2.4	77	23.2	
	333	1.9	90	9.4	358	1.8	85	14.4	

[a] percentage of the measured mass for 100–1000 nm particles in the 30 to 1000 nm size fraction, [b] mass concentration; [c] free troposphere average; [d] planetary boundary layer average.

Table 2a. Characteristics of the lognormal mass-size distributions of chemical components for relative fresh aerosols.

	Parameter[a]	Mt. Tai Nucleation event	Gucheng Nucleation event	Fresh air	Wuqing Nucleation event	Primary emission	Beijing Nucleation event	Clean Period	Manchester Fresh air	Pittsburgh Nucleation event	Clean Period
Organics	MMD	373–459	293–391	247–438	319–392	314–349	214–411	231–397	171	196	354
	σ_g	1.6–2.5	2.0–2.3	1.8–4.2	1.6–1.7	2.1–2.3	1.8–3.4	1.7–3.4	3	2.9	1.7
	%	98–99	98–99	96–99	96–98	99	94–99	96–99	90		82
	Mass	2.1–8.9	2.4–18	2.3–4.3	1.9–5.4	21.5–70.6	5.7–22.4	3.6–12.8	2.9		
Sulfate	MMD	329–485	249–423	143–418	404–517	263–361	247–567	271–442	306	192	415
	σ_g	1.6–2.0	1.7–2.2	1.4–1.8	1.3–1.7	1.9–2.1	1.3–2.6	1.6–2.3	1.8	2.8	1.6
	%	98–99	98–100	95–99	98–100	99	95–100	98–99	94		81
	Mass	1.1–6.4	0.5–5.8	0.8–1.2	0.5–1.2	4.0–5.9	1.0–18	0.4–5.6	2.5		
Nitrate	MMD	296–500	229–451	371–466	228–309	204–310	213–518	221–459	236	437	363
	σ_g	1.6–2.3	1.8–3.0	2–19.6	1.2–2.0	1.9–1.9	1.3–39.2	1.6–2.9	2.2	1.1	1.5
	%	99	98–100	98–99	96–98	97–99	93–99	96–99	92		91
	Mass	0.9–11.6	0.2–9.1	0.2–0.5	0.2–1.2	2.6–10.7	0.3–6.0	0.2–3.8	2.2		
Ammonium	MMD	340–441	234–309	134–362	326–379	200–277	217–526	216–618		215	401
	σ_g	1.6–2.1	1.5–2.3	1.4–1.7	1.3–1.7	1.8–2.0	1.3–2.3	1.2–2.0		2.4	1.6
	%	97–99	95–99	94–97	93–96	97–99	93–99	93–99			79
	Mass	0.9–6.5	0.3–4.7	0.5	0.4–1.1	4.2–9.8	0.6–7.3	0.2–2.5			
Reference		This study	Zhang (2011)				Zhang et al. (2011)		Allan et al. (2003)	Zhang et al. (2005)	

[a] Mass median diameter (MMD, nm), standard deviation (σ_g), percentage of the measured mass for 100–1000 nm particles in the 30 to 1000 nm size fraction, and mass concentration (Mass, µg m^{-3}).

Table 2b. Characteristics of the lognormal mass-size distributions of chemical components for relatively aged aerosols.

	Parameter[a]	Mt. Tai	Gucheng	Wuqing	Beijing	Manchester		Pittsburgh		Mt.Whistler	
		Aged air	Aged air	Aged air	Aged air	High local wind	Aged air	Polluted period	Polluted with sulfate	Dust event	Organic event
Organics	MMD	473–792	373–517	347–559	345–567	347	619	234	520	660	267
	σ_g	1.5–2.9	1.7–1.9	1.6–2.5	1.4–3.0	1.3		2.3	1.4	1.1	1.9
	%	99–100	99–100	97–100	99	91		91	91.2	0	82
	Mass	7.9–62.8	9.9–60.3	6.3–73.3	9.1–66.3	0.3		4.2		0.1	3.3
Sulfate	MMD	501–825	391–527	348–717	319–612	404	657	332	578	585	445
	σ_g	1.6–2.1	1.7–2.3	1.5–2.7	1.3–2.1	1.2		1.6	1.3	1.6	1.7
	%	99–100	99–100	98–100	99–100	76		94	77.1	70	80
	Mass	7.8–44	9.4–15.6	1.9–42.7	4.7–41.3	0.7		5.4		2	0.6
Nitrate	MMD	482–774	443–591	304–702	316–585	391	697	286	564		231
	σ_g	1.5–1.9	1.5–1.9	1.5–2.3	1.3–2.0	1.3		1.3	1.1		1.7
	%	99–100	99–100	98–100	99–100	81			70.1		99
	Mass	5.9–43	3.3–14.3	0.7–37	3.0–36.7	0					0.1
Ammonium	MMD	473–697	378–494	297–625	299–568			336	557	591	700
	σ_g	1.5–1.9	1.5–2.2	1.5–1.9	1.3–1.9			1.5	1.3	1.6	1
	%	99–100	99	96–100	99–100			92	90	69	85
	Mass	5–28	5.4–11.7	1.0–33.9	5.1–22.1			7.6		0.4	0.3
Reference		This study	Zhang (2011)		Zhang et al. (2011)	Allan et al. (2003)		Zhang et al. (2005)		Sun et al. (2009)	

[a] Abbreviations as in Table 2a.

Table 3. Mass median diameters (MMD, nm, standard deviations (σ_g) and mass concentration (Mass, µg m^{-3}) for chemical components in samples grouped by air-mass trajectory cluster.

Air mass	Organics			Sulfate			Nitrate			Ammonium			Chloride
	MMD	σ_g	Mass	MMD	σ_g	Mass	MMD	σ_g	Mass	MMD	σ_g	mass	Mass
Cluster-1	545	1.7	16.8	570	1.7	15.6	582	1.7	9.0	542	1.7	8.7	0.9
Cluster-2	533	1.6	18.2	584	1.7	17.6	581	1.6	11.2	553	1.7	10.2	1.0
Cluster-3	511	1.7	16.4	528	1.8	11.4	514	1.7	10.9	497	1.8	8.1	1.2
Cluster-4	464	1.7	12.8	517	1.7	9.1	470	1.7	7.4	478	1.7	5.8	0.8
Cluster-5	458	1.6	6.8	515	2.0	3.7	462	2.1	2.9	440	2.0	2.5	0.5
Cluster-6	417	1.8	12.7	464	2.0	6.8	435	1.8	8.0	437	1.9	5.4	1.0
Cluster-7	426	2.0	5.2	459	1.9	3.4	357	2.3	2.5	395	2.2	2.1	0.4

Table 4. Seasonal averaged mass concentrations (unit: μg m^{-3}) and standard deviation with percentage (in parentheses) of specific types of organic aerosols from Positive matrix factorization analysis.

Sub-type organics	Spring	Summer	Fall	Winter
Primary OA				
HOA	2.3 ± 1.2 (27 %)	1.6 ± 1.8 (10 %)	1.6 ± 0.7 (28 %)	2.2 ± 0.8 (15 %)
BBOA	2.1 ± 1.1 (24 %)	5.1 ± 10.7 (34 %)		
CCOA				5.2 ± 3.7 (34 %)
Secondary OA				
SV-OOA		3.1 ± 4.2 (21 %)	2.1 ± 1.3 (36 %)	
LV-OOA	4.2 ± 2.3 (49 %)	5.3 ± 6.4 (35 %)	2.1 ± 1.1 (36 %)	7.7 ± 6.1 (51 %)

[a] Abbreviations: HOA = hydrocarbon-like organic aerosol, BBOA = biomass-burning organic aerosol, CCOA= coal combustion organic aerosol, semi-volatile oxidized organic aerosol = SV-OOA, and low-volatility oxidized organic aerosol = LV-OOA.

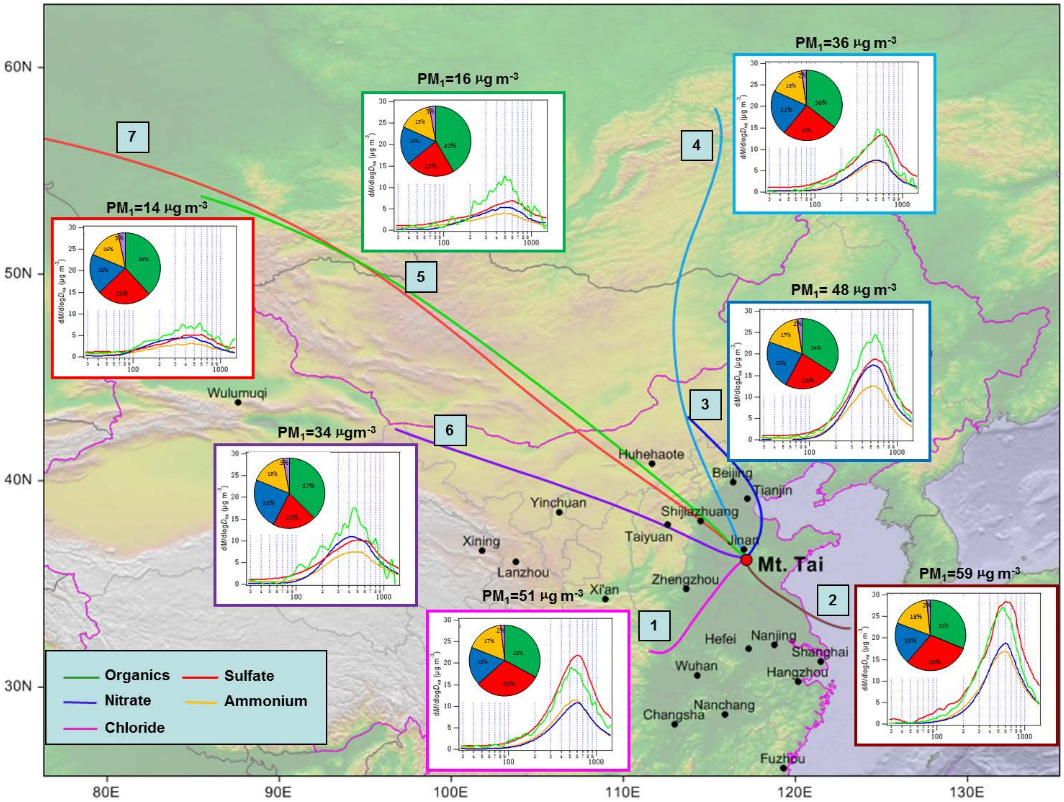

Figure 1. Mt. Tai station and averaged mass concentrations, mass-size distribution and percentage of chemical components associating with different air-mass trajectory clusters.

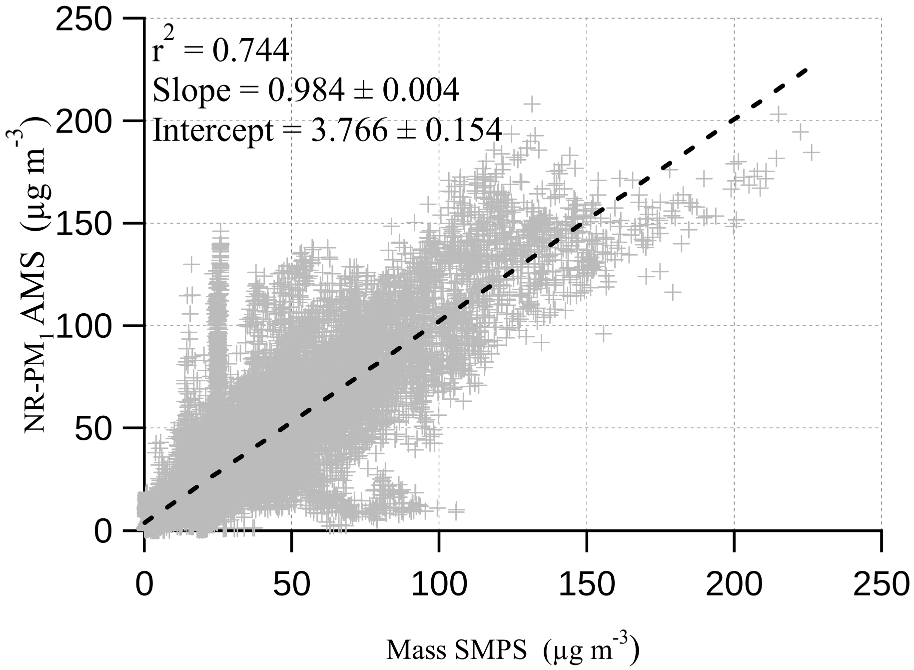

Figure 2. Correlation between AMS and (reconstructed) SMPS mass concentrations.