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Abstract

Heterogeneous processes on dust particles are important for understanding the chemistry and radiative balance of the atmosphere. This paper investigates an intense Asian dust storm episode observed at Mount Heng (1250 m a.s.l.) in Southern China on 24–26 April 2009. A set of aerosol and trace gas data collected during the study was analyzed to investigate their chemical evolution and heterogeneous photochemistry as the dust traveled to Southern China. Results show that the mineral dust arriving at Mt. Heng experienced significant modifications during transport, with large enrichments in secondary species (sulfate, nitrate, and ammonium) compared with the dust composition collected at an upwind mountain top site (Mount Hua). A photochemical age “clock” ($-\log_{10}(\text{NO}_x/\text{NO}_y)$) was employed to quantify the atmospheric processing time. The result indicates an obvious increase in the abundance of secondary water-soluble ions in dust particles with the air mass’ photochemical age. Based on the observations, a 4-stage evolution process is proposed for carbonate-rich Asian dust, starting from fresh dust to particles coated with hydrophilic and acidic materials. Daytime-enhanced nitrite formation on the dust particles was also observed, which indicates the recent laboratory result of the TiO_2 photocatalysis of NO_2 as a potential source of nitrite and nitrous acid.

1 Introduction

Mineral dust is injected into the atmosphere under specific meteorological conditions in deserts or semiarid areas, generating dust storms. In East Asia, the Taklimakan Deserts in Western China and the Gobi Deserts in Mongolia and Northern China are the two major source regions of dust, with annual emissions between 100 Mtyr^{-1} and 460 Mtyr^{-1} (Laurent et al., 2006). Upon entering the atmosphere, especially when transported over heavily polluted regions, the chemical and surface nature of mineral dust particles undergo significant changes, in turn influencing a number of atmospheric

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processes (Zhuang et al., 1992; Usher et al., 2003; Formenti et al., 2011, and the references therein).

In the past few decades, several large field campaigns (e.g., TRACP, ACE-Asia and INTEX-B) have studied the atmospheric processes of Asian dust (Jacob et al., 2003; Arimoto et al., 2006; Singh et al., 2009). The evolutionary processes of dust particles during their eastward transport to the Western Pacific have been already investigated in Northeast Asia. These studies have significantly improved understanding of the heterogeneous reactions between Asian pollution and mineral dust. Sullivan et al. (2007a) found that nitrate tended to accumulate on calcium-rich dust, while sulfate tended to accumulate on aluminosilicate-rich dust. Li and Shao (2009) demonstrated nitrate being coated onto dust particles with transmission electron microscopy. The direct uptake of chlorine on dust particles by heterogeneous reaction with HCl was clearly verified by Sullivan et al. (2007b). Sullivan and Prather (2007) observed internally mixed organic dicarboxylic acid (DCA) with dust particles and proposed a mechanism of heterogeneous oxidation of DCA on the surface of Asian dust. However, to our knowledge, there have been very few studies of the chemical evolution of Asian dust during its transport to Southern China, where the dust is subject to higher temperature, higher humidity, and possibly different emissions compared with eastward transport.

Heterogeneous reactions on dust particles have been extensively investigated, and most previous studies have focused on nighttime chemistry (Usher et al., 2003). Photo-induced/enhanced reactions of some reactive gases on dust particles have recently been demonstrated by laboratory studies (Cwiertny et al., 2008 and the references therein). Nicolas et al. (2009) observed photo-enhanced O₃ decomposition on mimic mineral dust particles of TiO₂/SiO₂ mixtures; while Ndour et al. (2008) detected the photo-enhanced uptake of NO₂ on both TiO₂/SiO₂ mixed particles and real Saharan and Arizona dust particles. The latter reaction received great attention because it may be a missing source of daytime nitrous acid (HONO); to our knowledge, however, the extent of that reaction has not been observed in the real atmosphere.

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As part of China's National Basic Research Project on acid rain, a comprehensive field campaign was conducted from March to May 2009 at Mt. Heng in Hunan province, Southern China (Fig. 1), during which an intense dust storm was observed on 24–26 April. The aim of the specific work is to investigate the impact of this Asian dust storm on the atmospheric chemistry at rural and remote mountain regions in Southern China. Special attention is given to the chemical evolution of the dust by comparison with the aerosol composition concurrently observed at another mountain site near the dust source region (Wang et al., 2011), as well as the role of heterogeneous photochemistry in the nitrite production. The general features and transport pathway of this dust storm are described first. The chemical evolution of the dust particles during transport from Mt. Hua to Mt. Heng is then investigated. A photochemical age “clock” ($-\log_{10}(\text{NO}_x/\text{NO}_y)$) is employed to examine the changes in the major secondary water-soluble ions with age. The evidence of the daytime production of nitrite on dust particles is also presented, and its potential contribution to the gas-phase HONO is discussed.

2 Experimental methodologies

2.1 Site description

Mount Heng is situated in Hunan Province, Southern China, and is approximately 500 km and 900 km from the South China Sea and the East China Sea, respectively (Figs. 1a and 2). The field campaign was conducted at a meteorological station at the summit of Mount Heng ($27^{\circ}18' \text{ N}$, $112^{\circ}42' \text{ E}$, 1269 m a.s.l.). There are few local emission sources around the measurement site (Sun et al., 2010). The closest cities are Heng Yang and Xiang Tan, which are approximately 50 km to the south and 70 km to the north, respectively.

2.2 Field campaign, sample collection and measurement techniques

The field campaign covered a 3-month period from March to May 2009. A comprehensive suite of trace gases, aerosols, and rain and cloud water samples was measured/collected. The present study is focused on the dust storms observed during 20–29 April 2009 (Fig. 3). The dataset employed in this paper includes continuous data of NO_x , NO_y , $\text{PM}_{2.5}$, PM_{10} and black carbon (BC); filter-based element data of Si, Al, Fe, Ca and Ti in $\text{PM}_{2.5}$ and Al in TSP; and size-resolved water-soluble ions (SO_4^{2-} , NO_3^- , NO_2^- , NH_4^+ , Ca^{2+}). Missing data points were due to instrument maintenance or malfunction.

Measurements of trace gases: NO_y (defined as NO_x and its oxidation products and intermediates) was converted to NO with heated molybdenum oxide (MoO) and measured with a chemiluminescence analyzer (TEI model 42CY); NO_2 was converted to NO with a Blue Light converter (Meteorologie Consult Gubh) and measured with another NO analyzer (TEI model 42i). The NO_2 conversion efficiencies, determined by the method of gas-phase titration, were approximately 40% on average during the whole sampling period. The calibration methods followed those reported in Wang et al. (2001).

Sampling and analysis of aerosols: $\text{PM}_{2.5}$ and PM_{10} mass concentrations were continuously measured with a Tapered Element Oscillating Microbalance (TEOM, Thermo Electron Corporation, East Greenbush, NY, USA). BC was measured using an Aethalometer (Magee Scientific, Berkeley, California, USA, Model AE-21) at a wavelength of 880 nm. $\text{PM}_{2.5}$ filter samples were collected using a four-channel particle sampler (Thermo Andersen Chemical Speciation Monitor, RAAS2.5-400, Thermo Electron Corporation) with Teflon filters (Teflon™, 2 μm pore size and 47 mm diameter, Pall Inc.) at a flow rate of 16.7 Liters Per Minute (LPM) (Wu and Wang, 2007). Total suspended particles (TSP) samples were collected using medium-volume air samplers (TH-105C equipped with a PM10–100 inlet) at flow rate of 100 LPM on glass fiber filters (GFFs, 90 mm in diameter). The size-resolved particles were collected with a Micro-Orifice

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Uniform Deposit Impactor (MOUDI, MSP Company) with aluminum substrates (MSP Company) at a flow rate of 30 LPM (Nie et al., 2010). The elements in PM_{2.5} and TSP were measured using X-Ray Fluorescence (XRF) and inductively coupled plasma mass spectrometry (ICP-MS), respectively. The water-soluble ions were analyzed using ion chromatography (IC) (Dionex 90) (Wu and Wang, 2007). Further details can be found in our previous publications (Wang et al., 2001, 2010; Wu and Wang, 2007; Nie et al., 2010). During the dust storm of 24–26 April, we collected 4 sets of size-resolved aerosol samples using MOUDI: Sample-I, Sample-II, Sample-III and Sample-IV, with Sample-I and Sample-III collected during daytime and Sample-II and Sample-IV at night.

3 Results and discussion

3.1 General description of the dust episodes

Figure 2 shows the temporal variations of particle mass and chemical species observed at Mt. Heng during 20–29 April 2009, including PM₁₀, PM_{2.5}, and major mineral elements (Si, Al, Ca, Fe, Ti) in PM_{2.5}. During this period, a moderate dust storm was observed on 21–22 April, with hourly averaged PM₁₀ and PM_{2.5} concentrations of 153 $\mu\text{g m}^{-3}$ and 57 $\mu\text{g m}^{-3}$, respectively. A more intense dust storm attacked the sampling site on 24–26 April, as highlighted by the shadow in Fig. 2, with the hourly averaged PM₁₀ and PM_{2.5} concentrations reaching 562 $\mu\text{g m}^{-3}$ and 289 $\mu\text{g m}^{-3}$, respectively. This intense dust storm has also been captured at three other sites at Mt. Hua in the Shaanxi province of Central China (Fig. 1a), Mt. Tai in the Shandong province of Central-Eastern China (Wang et al., 2010) and the island of Taiwan. This dust event was also clearly shown in the MODIS true-color image for 24 and 25 April (Fig. 1b). These observations indicate a large geographical region of Southern and Eastern China being influenced by this dust storm. Due to the significant particle loading and

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more extensive transport, the specific analysis is focused on the second dust storm event (24–26 April).

In order to characterize the origin and transport pathway of the air masses, a NOAA HYSPLIT4 model (Hybrid Single-Particle Lagrangian Integrated Trajectory) was employed to calculate 72 h backward trajectories arriving at Mt. Hua and Mt. Heng during the dust event of 24–26 April. As illustrated in Fig. 3a, the dust originating in the Gobi deserts moved southward and passed over Central China (Shaanxi province), the North China Plain (Hebei province, Shanxi province and Henan province) and Southern China (Hubei province and Hunan province) and is expected to travel farther to the South China Sea.

3.2 Chemical evolution of mineral dust particles during transport

3.2.1 Comparison of size-resolved water-soluble ions of mineral dust particles between Mt. Hua and Mt. Heng

Figure 5 illustrates the size distributions of four major water-soluble ions, including sulfate, nitrate, ammonium and calcium, at Mt. Hua (sample of 24 April) and Mt. Heng (sample of 25 April). The size distribution patterns of sulfate, nitrate and calcium at both mountain sites were similar: sulfate had a bimodal size distribution, with one peak in the submicron range and another larger peak in the supermicron range; nitrate and calcium showed single peaks in the coarse mode. For ammonium, the size distribution exhibited different patterns at the two sites, being unimodal at Mt. Hua, but bimodal at Mt. Heng. Compared with Mt. Hua, all four ions in the supermicron particles at Mt. Heng showed significant enhancement (Fig. 1): sulfate increased moderately (approximately 56%), but the other three ions were enhanced up to 5–7 times.

The enrichment of water-soluble ions on dust particles during transport generally depends on the atmospheric processing time, transport pathway, and mixing processes (Formenti et al., 2011, and the references therein). Compared with Mt. Hua, the mineral dust particles arriving at Mt. Heng were subjected to the emissions in the polluted

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North China Plain (NCP, see Fig. 3). Thus, the more abundant water-soluble ions observed in the dust sample of Mt. Heng can be attributed to a more intensive interaction of dust with anthropogenic pollutants. The different degrees of enrichment for the different ions at Mt. Heng are influenced by the relative abundance of SO_2 , NO_x and NH_3 in the atmosphere and the factors affecting their uptake/conversion on dust particles. For example, due to a higher hygroscopicity of $\text{Ca}(\text{NO}_3)_2$ than CaSO_4 , the formation of $\text{Ca}(\text{NO}_3)_2$ would significantly enhance the uptake of water (and other soluble gases), resulting in a positive feedback until all the calcium transformed to calcium nitrate (Formenti et al., 2011). This process may offer an explanation for the much larger increase in nitrate than that of sulfate on the mineral particles during the transport from Mt. Hua to Mt. Heng (Fig. 4).

3.2.2 Changes of mineral dust characteristics with photochemical age

In order to further investigate the evolution of the mineral dust, the atmospheric processing time of the air masses was calculated by the photochemical age ratio $-\log_{10}(\text{NO}_x/\text{NO}_y)$ (Kleinman et al., 2008), and then was related to the changes in water-soluble ions on the mineral dust particles. The photochemical age (Fig. 5) increased during the dust event (from 19:00, 24 April to 01:00, 27 April, local time). The ratios of the major secondary water-soluble ions (sulfate, nitrate and ammonium) to the PM mass in the supermicron range are shown in Figs. 6a–c. These results indicate overall increases in the relative abundances of all three ions with the photochemical age of the air parcels. During the time scale of the four samples, sulfate increased from 1.7 % to 2.4 %, nitrate from 2.5 % to 4.3 %, and ammonium from 0.2 % to 1.2 %. The increased accumulated secondary water-soluble ions would significantly enhance the solubility of mineral particles, thus affecting their lifetimes and Cloud Condensation Nuclei (CCN) or Ice Nuclei (IC) activity.

Acidity is another important factor that affects the chemical and physical properties of aerosols (Jang et al., 2002). For Asian dust particles, calcium is the major cation. Therefore, the ratios of the sum of major anions to calcium in equivalence in the su-

permicron range particles were calculated to investigate the changes of acidity of the mineral particles with the age of the air mass. As illustrated in Fig. 6d, evident acidification occurred with the aging of the air mass. The ratio increased from 0.5 in the first sample to 1.0 in the third sample and to 1.2 in the fourth sample.

3.2.3 Ammonium enrichment on dust and the aging process of carbonate-rich Asian dust

The formation of ammonium aerosol in the atmosphere would normally be suppressed by the invasion of a dust storm due to the dilution of the precursor (ammonia) and the uptake of acidic gases on dust particles (Song and Carmichael, 1999; Huang et al., 2010). However, during the dust event on 24–26 April, ammonium was observed at Mt. Heng, with bimodal peaks in both fine and coarse modes (Fig. 4e). In this section, discussions are focused on accumulated ammonium in the coarse mode. The ammonium abundance in the coarse-mode dust particles at Mt. Heng was relatively low and stable (2 % ~ 5 %) in the first three samples, but significantly increased to ~ 12 % in the fourth sample (Fig. 6c).

Previous field observations have indicated that for ammonium to form, mineral dust should first take up some acidic species that ammonia can partition to (Sullivan et al., 2007a). Carbonate-rich dust particles have a high alkalinity initially, and the uptake of secondary acids would first neutralize the alkaline carbonate and uptake of ammonia would follow. Thus some relationship between the amount of accumulated ammonium and the neutralized carbonate would be expected. In Asian dust, carbonates are mainly combined with calcium in the form of calcite (Matsuki et al., 2005). Therefore, the ratio of the sum of anions to calcium to was used to estimate the degree of neutralization of carbonate in the dust particles of Mt. Heng. In the first two samples, the alkaline carbonates were not fully neutralized, but some ammonium was still observed in the supermicron mode (Fig. 6c). This observation can be explained as: after the depletion of surface carbonate, the absorbed acidic species would uptake the gas-phase ammonia in parallel with neutralizing the carbonate in the inner core. Then, after the

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carbonate was fully consumed, ammonium was enhanced significantly in the fourth sample (see Fig. 6d).

Figure 7 presents a conceptual model summarizing the results of the particle evolution into a four-stage process for the carbonate-containing dust particles: freshly emitted dust (Stage 1), the uptake of secondary acidic species with little ammonium (Stage 2, as represented by the sample of Mt. Hua (see Fig. 4)), dust with ammonium accumulated on but without the depletion of carbonate (Stage 3, represented by Sample-I and Sample-II of Mt. Heng), and dust with all carbonate neutralized and significantly enhanced ammonium (Stage 4, represented by Sample-IV of Mt. Heng). This 4-stage evolution has important atmospheric implications. When the Asian dust particles have reached the third stage (i.e., with accumulated ammonium), the surface properties of the dust have changed from hydrophobic and alkaline to hydrophilic and acidic, thus significantly enhancing their CCN/IN capacity. At the fourth stage, the dust particles would be no different from the secondarily formed particles and could serve as favorable media to promote further reactions.

3.3 Photo-enhanced nitrite formation

High nitrite concentrations on dust particles were observed during the dust event of 24–26 April. Figure 8 shows the nitrite concentrations in the four samples following the temporal order during the dust episode of 24–26 April. The average nitrite concentration during this dust event was approximately $2.5 \mu\text{g m}^{-3}$, much higher than that during the non-dust days (approximately $0.3 \mu\text{g m}^{-3}$), indicating the promotion of nitrite formation by the dust particles. The enhanced nitrite formation during the dust event occurred only in the daytime samples (with the concentration up to $6 \mu\text{g m}^{-3}$ in Sample-III, see Fig. 8), suggesting a photochemical pathway yielding the observed nitrite.

Direct measurements of gas-phase HONO were unavailable during the Mt. Heng study. Nitrite is in the particle phase and can partition to the gas phase as HONO. A recent laboratory study (Ndour et al., 2008) observed a reduction of approximately 3 ppbv of NO_2 on irradiated real Sahara dust particles, with a concurrent production of

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5 pathways were not the major contributors to the observed nitrite formation. Despite the fact that aromatic compounds and humic acids were not measured, their concentrations were not presumed to increase much during the dust storm because mineral dust particles are not their sources. Considering that the daytime enhancement of nitrite production was only observed in the dust storm event, the third process should also be excluded as the key contributor.

10 The fourth process of TiO_2 photocatalysis of NO_2 yielding HONO was recently demonstrated in the laboratory (Gustafsson et al., 2006; Ndour et al., 2008). The reaction mechanism can be simply described as nitrogen dioxides being reduced to nitrite ions by photo-produced electrons (the detailed mechanism was proposed in Ndour's study, 2008). In this study, the titanium concentrations indeed increased significantly during the dust storm of 24–26 April (see Fig. 2), implying that the surface TiO_2 photocatalysis may be responsible for the daytime nitrite formation. To further support this hypothesis, the correlations of daytime nitrite with titanium and with nitrate were also examined. Noting that the titanium concentration in the coarse mode particles ($\text{PM}_{>2.5}$) was unavailable, their values were calculated via the formula $\text{Ti}_{\text{coarse}} = \text{Ti}_{\text{fine}} \times (\text{Al}_{\text{coarse}}/\text{Al}_{\text{fine}})$ under the assumption that aluminum and titanium have the same size distributions. As tabulated in Table 3, for Sample-I, the ratio of $\text{NO}_2^-_{(2.5\text{--}10\mu\text{m})}/\text{NO}_2^-_{(0.45\text{--}2.5\mu\text{m})}$ was approximately 3.0, which was similar to that of titanium (approximately 2.8), but largely differed from that of nitrate (approximately 9.1). The result for Sample-III was similar. These results further indicate the role of TiO_2 photocatalysis on the observed daytime nitrite formation.

25 In summary, photo-enhanced nitrite formation on dust particles was observed, which indicated lab-demonstrated TiO_2 photocatalysis of NO_2 as an important source of daytime HONO under dust storm conditions. Given that there is increasing evidence for anthropogenic sources of photocatalyst-containing (including TiO_2) nanodusts emitted from industrial nanomaterials (Vicki, 2009), this process may play a potentially important role in atmospheric chemistry on a global scale. We therefore recommended more

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comprehensive studies to better evaluate the contribution of photoactive particles to gas-phase HONO and, in turn, OH radicals.

4 Conclusions

An intense Asian dust storm that originated from the Gobi desert was captured at a mountain site (Mount Heng) in Southern China. The mineral dust particles experienced significant modifications before arriving at Mt Heng, with large enrichments in secondary species, such as sulfate, nitrate and ammonium. The dust particles became more acidic and hydrophilic with the photochemical age of air parcels. The accumulation of ammonium on dust particles was verified, which is related to the neutralization of carbonate. Based on the observations, a four-stage conceptual model is proposed to describe the aging process of the carbonate-rich Asian dust. Photo-enhanced nitrite formation on the dust particles was also observed, indicating that the TiO_2 photocatalysis of NO_2 is a potentially important source of HONO (and nitrite) under dust storm conditions. In view of the potential for nanodust to become a global source of photocatalysts (TiO_2), this process may be an important daytime HONO source. More studies are needed in order to examine further the extent of photo-enhanced nitrite formation in the atmosphere and its implications for atmospheric chemistry.

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References

Arimoto, R., Kim, Y. J., Kim, Y. P., Quinn, P. K., Bates, T. S., Anderson, T. L., Gong, S., Uno, I., Chin, M., Huebert, B. J., Clarke, A. D., Shinozuka, Y., Weber, R. J., Anderson, J. R., Guaz-

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- zotti, S. A., Sullivan, R. C., Sodeman, D. A., Prather, K. A., and Sokolik, I. N.: Characterization of Asian dust during ACE-Asia, *Global Planet. Change*, 52, 23–56, 2006.
- Aubin, D. G. and Abbatt, J. P. D.: Interaction of NO₂ with hydrocarbon soot: focus on HONO yield, surface modification, and mechanism, *J. Phys. Chem. A*, 111, 6263–6273, 2007.
- 5 Cwiertny, D. M., Young, M. A., and Grassian, V. H.: Chemistry and photochemistry of mineral dust aerosol, *Annu. Rev. Phys. Chem.*, 59, 27–51, 2008.
- Formenti, P., Schütz, L., Balkanski, Y., Desboeufs, K., Ebert, M., Kandler, K., Petzold, A., Scheuven, D., Weinbruch, S., and Zhang, D.: Recent progress in understanding physical and chemical properties of African and Asian mineral dust, *Atmos. Chem. Phys.*, 11, 8231–8256, doi:10.5194/acp-11-8231-2011, 2011.
- 10 George, C., Strekowski, R. S., Kleffmann, J., Stemmler, K., and Ammann, M.: Photoenhanced uptake of gaseous NO₂ on solid organic compounds: a photochemical source of HONO?, *Faraday Discuss.*, 130, 195–210, 2005.
- Gustafsson, R. J., Orlov, A., Griffiths, P. T., Cox, R. A., and Lambert, R. M.: Reduction of NO₂ to nitrous acid on illuminated titanium dioxide aerosol surfaces: implications for photocatalysis and atmospheric chemistry, *Chem. Commun.*, 42, 3936–3938, 2006.
- Huang, K., Zhuang, G., Li, J., Wang, Q., Sun, Y., Lin, Y., and Fu, J. S.: Mixing of Asian dust with pollution aerosol and the transformation of aerosol components during the dust storm over China in spring 2007, *J. Geophys. Res.-Atmos.*, 115, D00K13, doi:10.1029/2009jd013145, 2010.
- 20 Jacob, D. J., Crawford, J. H., Kleb, M. M., Connors, V. S., Bendura, R. J., Raper, J. L., Sachse, G. W., Gille, J. C., Emmons, L., and Heald, C. L.: Transport and chemical evolution over the Pacific (TRACE-P) aircraft mission: design, execution, and first results, *J. Geophys. Res.-Atmos.*, 108, 9000, doi:10.1029/2002jd003276, 2003.
- 25 Jang, M., Czoschke, N. M., Lee, S., and Kamens, R. M.: Heterogeneous atmospheric aerosol production by acid-catalyzed particle-phase reactions, *Science*, 298, 814–817, 2002.
- Kleffmann, J.: Daytime sources of nitrous acid (HONO) in the atmospheric boundary layer, *ChemPhysChem*, 8, 1137–1144, 2007.
- Kleinman, L. I., Springston, S. R., Daum, P. H., Lee, Y.-N., Nunnermacker, L. J., Senum, G. I., Wang, J., Weinstein-Lloyd, J., Alexander, M. L., Hubbe, J., Ortega, J., Canagaratna, M. R., and Jayne, J.: The time evolution of aerosol composition over the Mexico City plateau, *Atmos. Chem. Phys.*, 8, 1559–1575, doi:10.5194/acp-8-1559-2008, 2008.
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- Laurent, B., Marticorena, B., Bergametti, G., and Mei, F.: Modeling mineral dust emissions from Chinese and Mongolian deserts, *Global Planet. Change*, 52, 121–141, 2006.
- Li, W. J. and Shao, L. Y.: Observation of nitrate coatings on atmospheric mineral dust particles, *Atmos. Chem. Phys.*, 9, 1863–1871, doi:10.5194/acp-9-1863-2009, 2009.
- 5 Nicolas, M. L., Ndour, M., Ka, O., D'Anna, B., and George, C.: Photochemistry of atmospheric dust: ozone decomposition on illuminated titanium dioxide, *Environ. Sci. Technol.*, 43, 7437–7442, 2009.
- Matsuki, A., Iwasaka, Y., Shi, G. Y., Chen, H. B., Osada, K., Zhang, D., Kido, M., Inomata, Y., Kim, Y. S., Trochikine, D., Nishita, C., Yamada, M., Nagatani, T., Nagatani, M., and Nakata, H.: Heterogeneous sulfate formation on dust surface and its dependence on mineralogy: balloon-borne observations from balloon-borne measurements in the surface of Beijing, China, *Water Air Soil Poll.*, 5, 101–132, 2005.
- 10 Ndour, M., D'Anna, B., George, C., Ka, O., Balkanski, Y., Kleffmann, J., Stemmler, K., and Ammann, M.: Photoenhanced uptake of NO₂ on mineral dust: laboratory experiments and model simulations, *Geophys. Res. Lett.*, 35, L05812, doi:10.1029/2007gl032006, 2008.
- Nie, W., Wang, T., Gao, X., Pathak, R. K., Wang, X., Gao, R., Zhang, Q., Yang, L., and Wang, W.: Comparison among filter-based, impactor-based and continuous techniques for measuring atmospheric fine sulfate and nitrate, *Atmos. Environ.*, 44, 4396–4403, 2010.
- 15 Ramazan, K. A., Syomin, D., and Finlayson-Pitts, B. J.: The photochemical production of HONO during the heterogeneous hydrolysis of NO₂, *Phys. Chem. Chem. Phys.*, 6, 3836–3843, 2004.
- Singh, H. B., Brune, W. H., Crawford, J. H., Flocke, F., and Jacob, D. J.: Chemistry and transport of pollution over the Gulf of Mexico and the Pacific: spring 2006 INTEX-B campaign overview and first results, *Atmos. Chem. Phys.*, 9, 2301–2318, doi:10.5194/acp-9-2301-2009, 2009.
- 25 Song, C. H. and Carmichael, G. R.: The aging process of naturally emitted aerosol (sea-salt and mineral aerosol) during long range transport, *Atmos. Environ.*, 33, 2203–2218, 1999.
- Sörgel, M., Regelin, E., Bozem, H., Diesch, J.-M., Drewnick, F., Fischer, H., Harder, H., Held, A., Hosaynali-Beygi, Z., Martinez, M., and Zetzsch, C.: Quantification of the unknown HONO daytime source and its relation to NO₂, *Atmos. Chem. Phys.*, 11, 10433–10447, doi:10.5194/acp-11-10433-2011, 2011.
- 30 Stemmler, K., Ammann, M., Donders, C., Kleffmann, J., and George, C.: Photosensitized reduction of nitrogen dioxide on humic acid as a source of nitrous acid, *Nature*, 440, 195–198, 2006.

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Sullivan, R. C. and Prather, K. A.: Investigations of the diurnal cycle and mixing state of oxalic acid in individual particles in Asian aerosol outflow, *Environ. Sci. Technol.*, 41, 8062–8069, 2007.

Sullivan, R. C., Guazzotti, S. A., Sodeman, D. A., and Prather, K. A.: Direct observations of the atmospheric processing of Asian mineral dust, *Atmos. Chem. Phys.*, 7, 1213–1236, doi:10.5194/acp-7-1213-2007, 2007a.

Sullivan, R. C., Guazzotti, S. A., Sodeman, D. A., Tang, Y., Carmichael, G. R., and Prather, K. A.: Mineral dust is a sink for chlorine in the marine boundary layer, *Atmos. Environ.*, 41, 7166–7179, 2007b.

Sun, M., Wang, Y., Wang, T., Fan, S., Wang, W., Li, P., Guo, J., and Li, Y.: Cloud and the corresponding precipitation chemistry in South China: water-soluble components and pollution transport, *J. Geophys. Res.*, 115, D22303, doi:10.1029/2010jd014315, 2010.

Usher, C. R., Michel, A. E., and Grassian, V. H.: Reactions on mineral dust, *Chem. Rev.*, 103, 4883–4939, 2003.

Vicki H. G.: New directions: nanodust – a source of metals in the atmospheric environment?, *Atmos. Environ.*, 43, 4666–4667, 2009.

Wang, G., Li, J., Cheng, C., Hu, S., Xie, M., Gao, S., Zhou, B., Dai, W., Cao, J., and An, Z.: Observation of atmospheric aerosols at Mt. Hua and Mt. Tai in central and east China during spring 2009 – Part 1: EC, OC and inorganic ions, *Atmos. Chem. Phys.*, 11, 4221–4235, doi:10.5194/acp-11-4221-2011, 2011.

Wang, T., Cheung, V. T. F., Anson, M., and Li, Y. S.: Ozone and related gaseous pollutants in the boundary layer of Eastern China: overview of the recent measurements at a rural site, *Geophys. Res. Lett.*, 28, 2373–2376, 2001.

Wang, T., Nie, W., Gao, J., Xue, L. K., Gao, X. M., Wang, X. F., Qiu, J., Poon, C. N., Meinardi, S., Blake, D., Wang, S. L., Ding, A. J., Chai, F. H., Zhang, Q. Z., and Wang, W. X.: Air quality during the 2008 Beijing Olympics: secondary pollutants and regional impact, *Atmos. Chem. Phys.*, 10, 7603–7615, doi:10.5194/acp-10-7603-2010, 2010.

Wang, Y., Sun, M., Li, P., Li, Y., Xue, L., and Wang, W.: Variation of low molecular weight organic acids in precipitation and cloudwater at high elevation in South China, *Atmos. Environ.*, 45, 6518–6525, 2011.

Wu, W. S. and Wang, T.: On the performance of a semi-continuous PM_{2.5} sulphate and nitrate instrument under high loadings of particulate and sulphur dioxide, *Atmos. Environ.*, 41, 5442–5451, 2007.

Zhou, X., Gao, H., He, Y., Huang, G., Bertman, S. B., Civerolo, K., and Schwab, J.: Nitric acid photolysis on surfaces in low-NO_x environments: significant atmospheric implications, *Geophys. Res. Lett.*, 30, 2217, doi:10.1029/2003gl018620, 2003.

5 Zhuang, G., Yi, Z., Duce, R. A., and Brown, P. R.: Link between iron and sulphur cycles suggested by detection of Fe(n) in remote marine aerosols, *Nature*, 355, 537–539, 1992.

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Table 1. Concentrations of nitrite, nitrate and titanium in both fine particle mode (PM_{2.5}) and coarse particle mode (PM_{>2.5}) in the two daytime samples during the dust storm of 24–26 April 2009.

Sample	Fine particles ($\mu\text{g m}^{-3}$)			Coarse particles ($\mu\text{g m}^{-3}$)		
	NO ₂ ⁻	Ti	NO ₃ ⁻	NO ₂ ⁻	Ti*	NO ₃ ⁻
0425 Daytime	0.68	0.58	2.0	2.02	1.61	18.2
0426 Daytime	1.5	0.42	1.0	4.49	1.12	10.7

* Ti in coarse particles was calculated by $Ti_{\text{coarse}} = Ti_{\text{fine}} \times (Al_{\text{coarse}}/Al_{\text{fine}})$.

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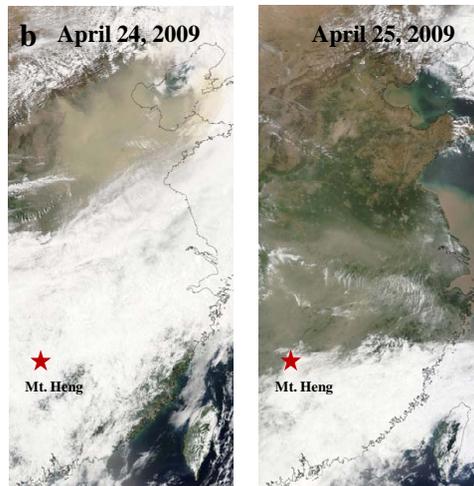
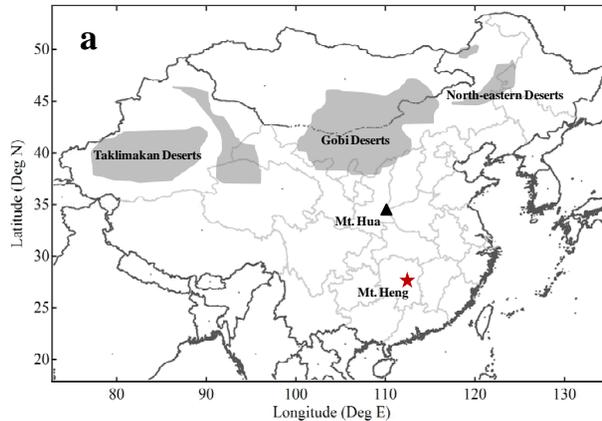



Fig. 1. (a) Map showing Mt. Heng and Mt. Hua. The major deserts in East Asia are marked by the shadows in the map. **(b)** MODIS true-color imagery of Eastern China on 24 and 25 April 2009.

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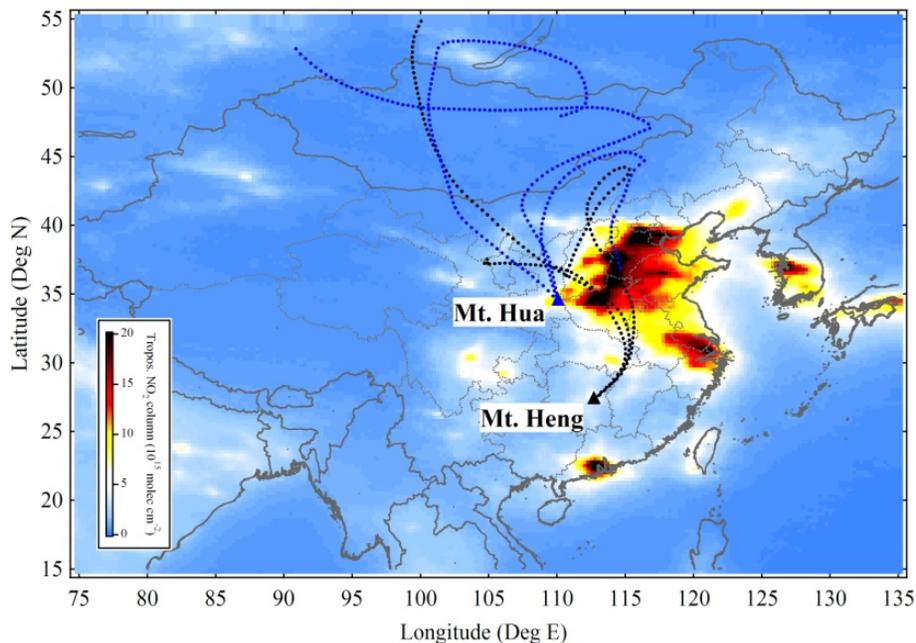


Fig. 2. The 72-h backward trajectories for the dust event at Mt. Hua and Mt. Heng. The NO_2 column data from GOME and SCIAMMACHY (averaged over 1996–2007, Wang et al., 2009) are shown in the shaded areas.

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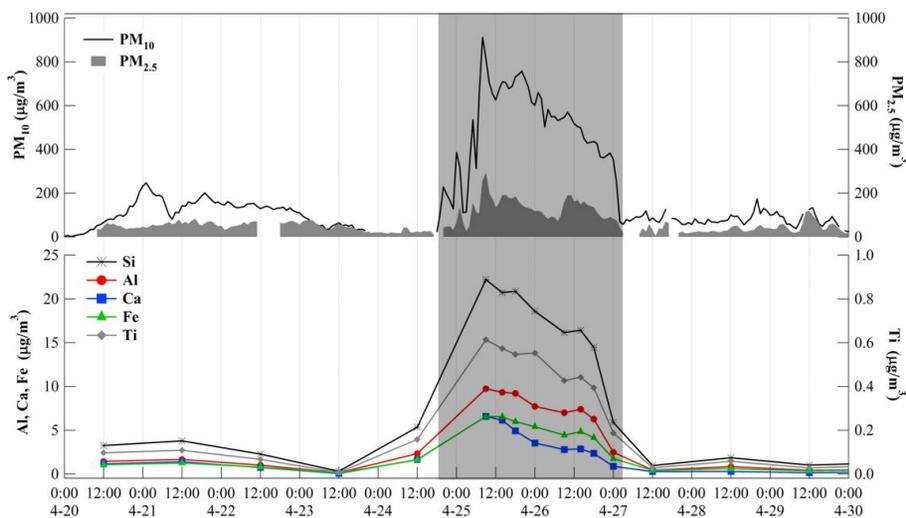


Fig. 3. Temporal variations of PM_{10} , $PM_{2.5}$ and mineral elements (Si, Al, Ca, Fe, Ti) in $PM_{2.5}$ at Mt. Heng during 20 to 29 April 2009.

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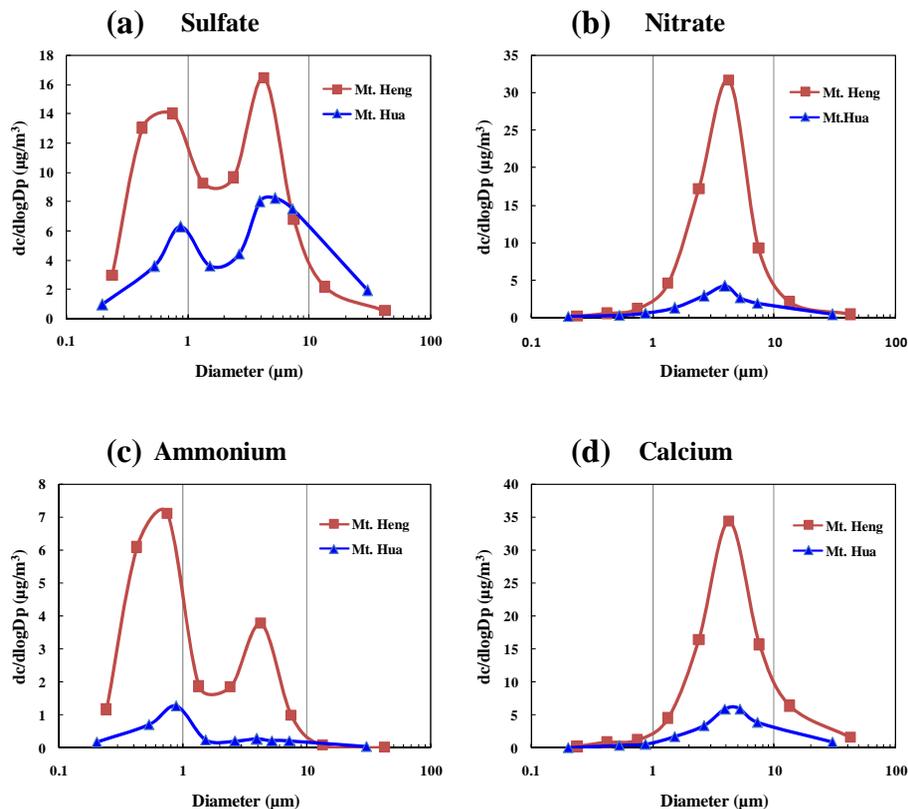


Fig. 4. Size distributions of (a) sulfate, (b) nitrate, (c) ammonium and (d) calcium at Mt. Hua and Mt. Heng during the dust event on 24–26 April.

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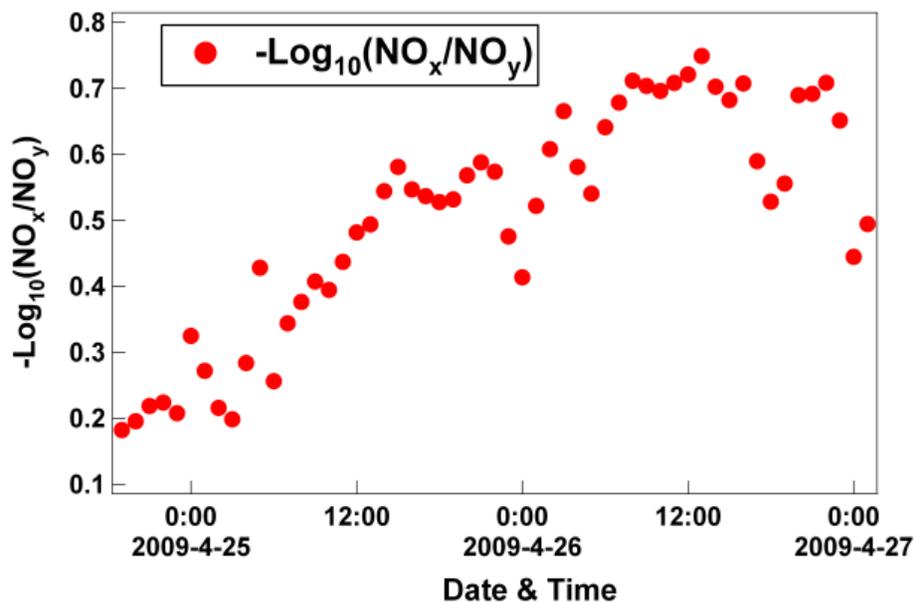


Fig. 5. Photochemical age (defined as $-\log_{10}(\text{NO}_x/\text{NO}_y)$) during the dust event on 24–26 April at Mt. Heng.

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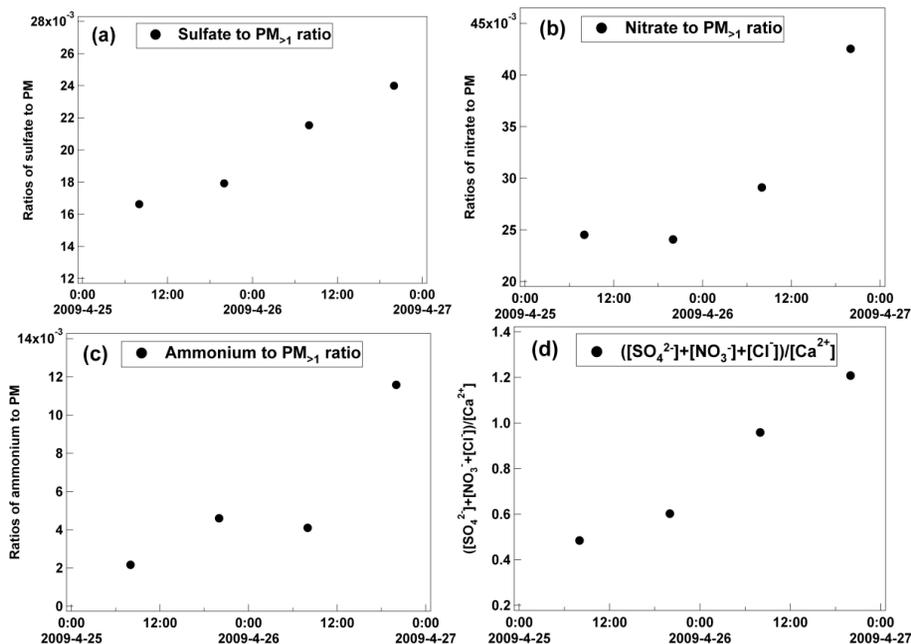


Fig. 6. Concentration ratios of (a) sulfate to PM mass, (b) nitrate to PM mass, (c) ammonium to PM mass and (d) sum of major anions to calcium in equivalence, in the supermicron range particles collected during the dust event on 24–26 April at Mt. Heng.

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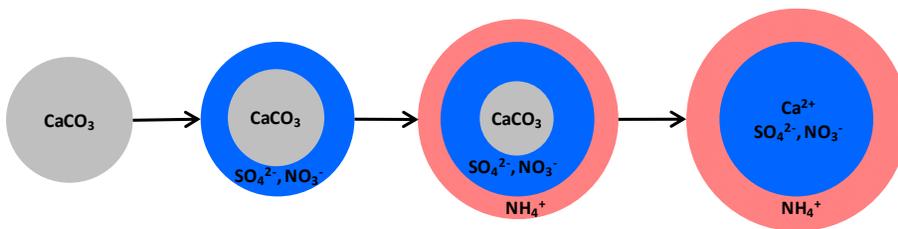



Fig. 7. Conceptual model showing the four aging stages for the carbonate-rich dust particles.

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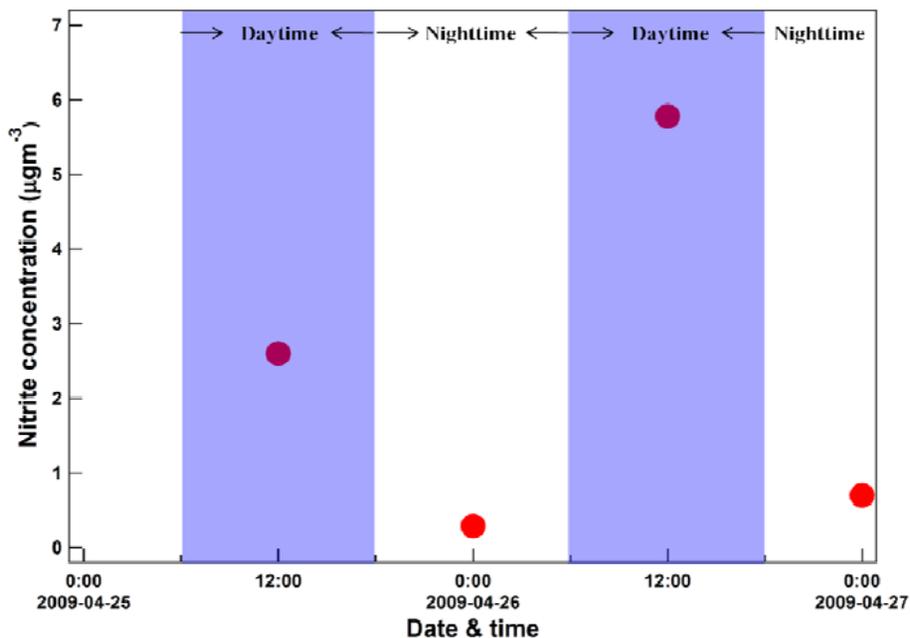


Fig. 8. Nitrite concentrations in total suspended particulates collected during the dust event on 24–26 April at Mt. Heng.

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