Dear Dr. Barbara Ervens,

Thank you very much for your patience and help! Thank you for the opportunity to resubmit our manuscript (acp-2016-1183) to your journal. Based on the reviewers' and editor's comments, we have carefully revised our manuscript, including supplementing the reference, enhancing our discussion and supplementing the conclusion. We are confident that it is ready for publishing in a high quality journal. Detailed item-by-item responses to the comments are listed below.

Best regards,

Yours sincerely,

Jianhua Qi

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Response to comments

Comments by Reviewer #1, including some remarks by the editor.

1) Distinguishing the three categories

a) The reviewer still questions the representativeness of the three categories. As the main characteristics is the ammonia and nitrate content, evaporation of these compounds might bias the conclusion on which category the samples belong to.

Can you comment on this possible bias?

Response: As reviewer suggested, gas-particle interactions, particle-particle interactions, and dissociation of semi volatile species can lead to the sampling loss of ammonia and nitrate (Dougle and Ten Brink, 1996; Pathak et al., 2004; Wang and John, 1988), especially at high temperature (e.g., higher than 30℃) and low relative humidity (e.g. less than 40%)(Pathak et al., 2004). Pathak et al. (2004) studied the sampling loss of ammonium and nitrate in PM2.5 using a speciation sampler equipped with two denuders and a filter-pack system, and found that ammonium loss was low with a ratio less than 11% but the nitrate loss from the Teflon filter were significant. Chang et al. (2000) found that sampling loss is rather small for undenuded filter sampling and the higher loss observed for the denuded filter because the denuder removes all of the nitric acid, a condition that enhances volatilization. In addition, Wang and John (1988) found ammonium nitrate evaporative loss was less than 10% when the loaded mass is abundant (more than 2500 μg) in Teflon filters. Our samples were collected on quartz microfiber filters only for 4 hrs using high-volume air sampler without denuder at a flow rate of 1 m^3/min, corresponding to a high mass on filter (more than 2500 μg for all samples). The evaporation of ammonia and nitrate were very likely negligible under such samplings.

Pathak, R.. K., X. H., Yao, and C. K., Chan.: Sampling artifacts of acidity and ionic species in PM2.5, Environ. Sci. Technol., 38, 254-259, 2004.

Dougle, P. G., H. M., Ten Brink.: Evaporative losses of ammonium nitrate in nephelometry and impactor measurements, J. Aerosol Sci., 27(S1), S511-512, 1996.

Chang, M. C., C., Sioutas, S., Kim, H., Gong Jr., and Linn W. S.: Reduction of nitrate losses from filter and impactor samplers by means of concentration enrichment, Atmos. Environ., 34, 85-98, 2000.

Wang, H.C., and John, W.: Characteristics of the Berner Impactor for sampling inorganic ions. Aerosol. Sci. Tech., 8, 157-172, 1988.

b) The reviewer criticizes that the expression ‘adequate separation’ in l. 92 is too vague. Please clarify.

Response: The sentence has been revised into “The on-line data in high time-resolution can allow identifying two dust events accurately from the start to the end.” in l. 96-97 in the revised manuscript.

2) Assumptions on emissions

The reviewer asks why emissions for the year 2008 were used as they may not be representative for the following years. As you explain that data from the year 2006 were extrapolated to the year 2008, I (the editor) wonder if the same methodology could be applied in order to obtain

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emission scenarios for the subsequent years, too.

Response: Zhang et al. (2009) generated the emissions of air pollutants in 2006 including NOX and NH3 over East Asia, and they updated the emission inventory in 2008 for us being used in this study, using technology-based approach with detailed activity and technology information. Thus it is only available for emission of 2008 but not every year during 2008-2011. The annual variation of air pollutant emission was likely small at those periods, e.g., NOx shown in Fig. R1, especially the spatial distribution over China.

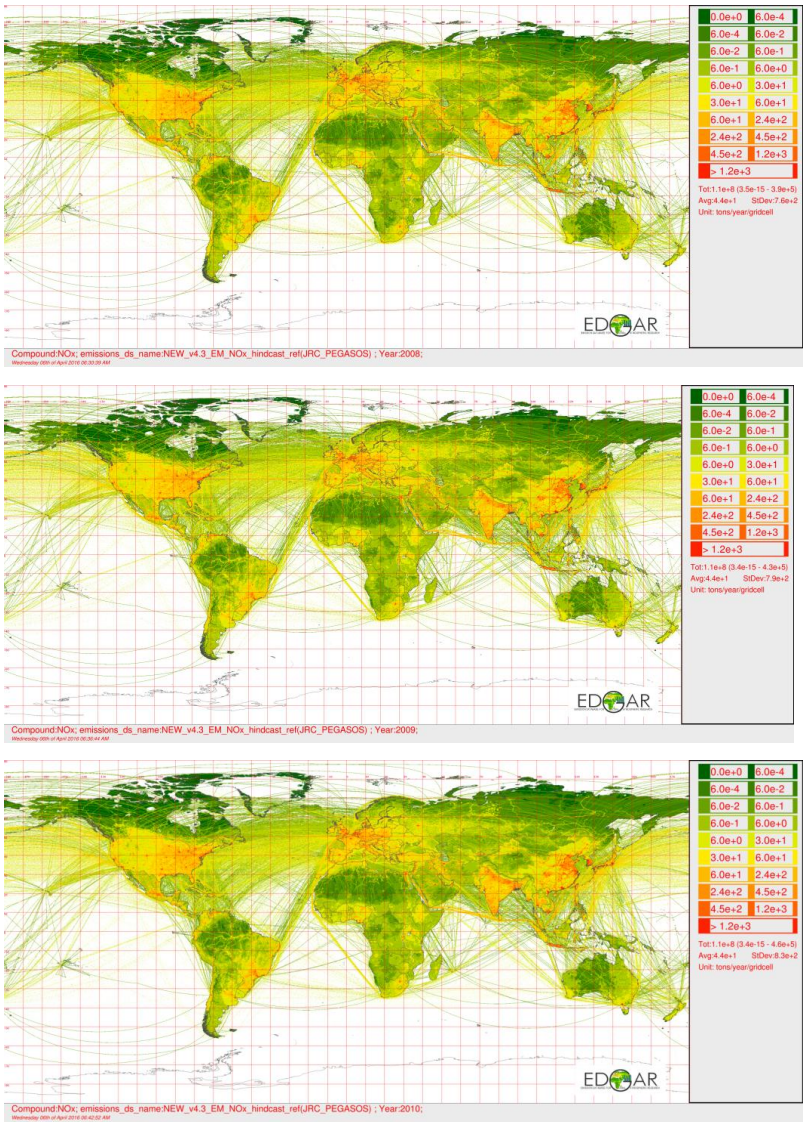

Figure R1. The EDGAR global anthropogenic emission inventory of NOx during 2008-2010 (From http://edgar.jrc.ec.europa.eu/overview.php?v=431) .

3) Importance of dust intensity

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The reviewer still questions the statement of the low importance of the dust intensity. Can you comment on the statement that Ca^{2+} should be dependent on the dust event intensity?

Response: According to Sand-dust weather almanac (CMA, 2009; 2010; 2012; 2013), half of the events were recorded as sand and dust storm (visibility less than 1000 m) and the rest was the severe sand and dust storm (visibility less than 500 m) in the source regions in Category 1 and 2, with exception of two events recorded as blowing dust (visibility reduce to 1000–10000 m) in Category 3. When the dust arrived at Qingdao after the long-range transport, we only observed floating dust (horizontal visibility less than 10000 m) in Qingdao. Therefore we didn't consider dust intensity as an important factor for $N_{NH4+NO3-}$ in dust events we studied in the last revised manuscript.

Just as Formenti et al. (2011) reviewed, studies on the mineralogical composition of unpolluted aerosol in dust source region in China are limited. The published references indicated the carbonate content and Dolomite ($CaMg(CO_3)_2$) generally can be used as a source tracer for Asia dust (Formenti et al. , 2011; Jong et al., 2008; Li et al., 2007; Shen et al., 2005). The carbonate content and Ca/Al ratio exhibits a geographical dependence with decreasing value from west to east (with exception of the Gurbantunggut desert), following the carbonate distribution in soil (Formenti et al., 2011). Thus, Ca concentration should highly depend on the dust source more than dust intensity.

CMA: Sand-dust weather almanac 2008, China Meteorological Press, Beijing, 10-64, 2009.

CMA: Sand-dust weather almanac 2009, China Meteorological Press, Beijing, 11-59, 2010.

CMA: Sand-dust weather almanac 2010, China Meteorological Press, Beijing, 11-79, 2012.

CMA: Sand-dust weather almanac 2011, China Meteorological Press, Beijing, 10-53, 2013.

Formenti, P., Sch ¨ utz, L., Balkanski, Y., Desboeufs, K., Ebert, M., Kandler, K., Petzold, A., Scheuvens, 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.

Jeong, G. Y.: Bulk and single-particle mineralogy of Asian dust and a comparison with its source soils, J. Geophys. Res., 113, D02208, doi:10.1029/2007jd008606, 2008.

Li, G., Chen, J., Chen, Y., Yang, J., Ji, J., and Liu, L.: Dolomite as a tracer for the source regions of Asian dust, J. Geophys. Res., 112, D17201, doi:10.1029/2007jd008676, 2007.

Shen, Z. X., Li, X., Cao, J., Caquineau, S., Wang, Y., and Zhang, X.: Characteristics of clay minerals in Asian dust and their environmental significance, China Particuology, 3, 260–264, 2005.

4) Category 3

I agree with the reviewer that Category 3 should be mentioned in the abstract. In addition, I am missing a discussion of Category 3 in Section 4.1.

Response: We are very sorry for the missing. We have supplemented the flux results of Category 3 in abstract in l.29-31. And we have supplemented the discussion of Category 3 in l. 310-319 in Section 4.1. However, the unique changes in NH_4^+ and NO_3^-, different from Category 1 and 2, need further investigation.

5) Inorganic nitrogen

a) The reviewer asks why the focus of the study was inorganic nitrogen. Can you estimate any possible contribution of organic nitrogen in the particles?

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Response: Inorganic nitrogen reportedly contributed to ~80% of the total water-soluble nitrogen (TDN) in atmospheric particles collected over the Yellow Sea and in Qingdao (Shi et al., 2010). In the region, the dry deposition flux of the inorganic nitrogen accounted for more than 75% for the TDN (Qi et al., 2013). When deposited to the ocean via atmospheric dry deposition, inorganic nitrogen has great impact on marine productivity due to its bioavailability. To update and improve our knowledge on reactive nitrogen carried by dust particles, we focused on nitrate and ammonium by excluding nitrite because of its very low concentration.

We have supplemented the contribution of organic nitrogen in the particles in l. 65-69 in *Introduction* Section.

Shi, J. H., Gao, H. W., Zhang, J., Tan, S. C., Ren, J. L., Liu, C. G., Liu, Y., and Yao, X. H.: Examination of causative link between a spring bloom and dry/wet deposition of Asian dust in the Yellow Sea, China, J. Geophys. Res-Atmos., 117, 127-135, 2012.

Qi, J. H., Shi, J. H., Gao, H. W., and Sun, Z.: Atmospheric dry and wet deposition of nitrogen species and its implication for primary productivity in coastal region of the Yellow Sea, China, Atmos. Environ., 81, 600-608, 2013.

b) I think 'inorganic nitrogen' is a too broad term. Only at one place in the manuscript it is mentioned that nitrite is excluded. I suggest being explicit and replacing 'inorganic nitrogen' by 'NH_4^+ and NO_3^-' throughout the manuscript.

Response: We have replaced "inorganic nitrogen" by "NH_4^+ and NO_3^-" throughout the manuscript.

6) Median vs Mean
Please correct the contradiction of l. 205 and Table S1 (cf reviewer comment)

Response: We are sorry for the confusion. We have supplemented the average concentration in Table S1. And we have revised the "median" into "mean" (Now l. 212) in the revised manuscript.

Additional editor comments
l. 22: Do you mean 'externally mixed', i.e. in separate particles?

Response: "externally mixed" has been revised into "existing separately" throughout the manuscript.

l. 26: What does < 3 refer to here?

Response: It referred to "Our modeled results satisfied the reasonable fit criteria, i.e. 90% of the scaled residuals were located between the range −3 and +3 for each species.", which has been moved to Section 2.3 in l. 158-160.

l. 57/8: This sentence is not clear. Please reword.

Response: We have reworded the sentence to "However, Zhang et al. (2010a) reported an interesting result, i.e., the concentrations of NO_3^- and NH_4^+ were lower during strong dust storm

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events than weak dust events. A high uncertainty appeared to exist for carrying amount of reactive nitrogen by dust particles." in l. 56-58.

l. 113: remove 'the'

Response: Revised.

l. 173: Are the emissions modeled, i.e. predicted based on assumptions of sources or are they an input to the model?

Response: We are sorry for the confusion. Zhang et al. (2009) generated the emissions of air pollutants in 2006 including NO_X and NH_3 over East Asia and they updated the emission inventory in 2008 for us being used in this study. And the sentence was supplemented in l. 180-182.

l. 204/5: This is ambiguous. As it is written, the text suggests that each individual sample pair exhibited a net increase of 82-1303%. Is this true? Or was this large range the range that was determined based on all samples?

Response: Yes, it's true. The sentence has been revised into "In each individual pair of dust day sample against reference sample, a net increase in the mass concentration of TSPs was observed. The percentages varied from 82 to 1,303% on basis of events, with a mean value of 403% (Table S1)" in l.210-212.

l. 239: This sentence needs to be improved. Do you mean 'the absolute increase…'? I don't understand what is meant by 'complex for the interactions'. It is very vague and grammatically wrong.

Response: The sentence has been revised into "Since air pollutant emissions, meteorological conditions, chemical reactions, and others can affect the concentrations of NH_4^+ and NO_3^- in atmospheric particles collected in dust days, the observed increase or decrease in the mass concentration of nitrate and ammonium in different dust samples against the reference implied the combined effect of those factors." in l. 245-248.

l. 259 and throughout the manuscript: What is meant by 'exterior sample'? Do you mean an outlier? How was this determined?

Response: We have revised "exterior sample" into "except for…" throughout the manuscript.

l. 262: 'It was commonly believed' should be changed here. What evidence was this assumption based on? Could it be concluded based on more than one study?

Response: We have rewritten the sentence into "Anthropogenic ammonium nitrate and ammonium sulfate were thought to be produced by gas, aqueous phase reaction and thermodynamic equilibrium processes and they usually internally mixed (Seinfeld and Pandis, 1998)." in l. 269-271.

Seinfeld, J. H., and Pandis, S. N.: Atmospheric Chemistry and Physics: From Air Pollution to Climate Change, 2nd Edition, Wiley, New York, 1191 pp., 1998.

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l. 267: Do you mean 'may be externally mixed'?

Response: "may externally exist with dust aerosols" has been revised into "very likely existed separately" in l. 273.

l. 270/1: I don't understand this. How does the dilution effect affect particle composition and what chemical reaction(s) is/are referred to here?

Response: We are very sorry for the confusion. According to the reference, Huang et al. reported that the higher concentration of SO_4^{2-} was observed in dust samples due to the heterogeneous reaction on the alkaline dust during dust storms, while the concentrations of NO_3^- and NH_4^+ decreased due to the dilution of the local pollution by a strong wind associated with the invaded dust. Because the low temperature, relative humidity, strong wind and the low pollution gases did not favor the chemical conversion, and the NOR (nitrogen oxidation ratio) was low and even less than 1% during the dust storm of Beijing (Yuan et al., 2008). We have revised the sentence to into "The observed NO_3^- and NH_4^+ in Asia dust samples were argued due to physically mixing two types of particles rather than the heterogeneous formation of nitrate and ammonium (Huang et al., 2010)." in l.276-278 to clarify this question.

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.

Yuan, H., Zhuang, G., Li, J., Wang, Z. and Li, J.: Mixing of mineral with pollution aerosols in dust season in Beijing: Revealed by source apportionment study, Atmos. Environ., 42, 2141–2157, 2008.

l. 275 and throughout the manuscript: Is there any evidence in previous studies that metal ions form stable salts in particles? References? Are these all salts or would also metal-sulfato-complexes be possible?

Response: We have supplemented the reference to support our hypothesis, i.e. metal ions can form stable salts in particles. Cu existed in form of salts and organic complexes (Scheinhardt et al., 2013; Wang et al., 2016; Zhang et al., 2015). Sulfate can exist in many forms of metal salts in atmospheric particles, such as Na_2SO_4, K_2SO_4, $K_2Ca(SO_4)_2{\cdot}H_2O$, $Na_2Ca(SO_4)_2$, $Na_2Mg(SO_4)_2{\cdot}4H_2O$, $(NH_4)_2Mg(SO_4)_2{\cdot}6H_2O$, $Na_3(NO_3)(SO_4){\cdot}\ H_2O$ (Chabas and Lefèvre, 2000; Sobanska et al., 2012; Xie et al., 2005). in l. 283-293.

Chabas, A., and Lefèvre, R. A.: Chemistry and microscopy of atmospheric particulates at Delos (Cyclades–Greece), Atmos. Environ., 34, 225–238, 2000.

Scheinhardt, S., Müller, K., Spindler, G., and Herrmann, H.: Complexation of trace metals in size-segregated aerosol particles at nine sites in Germany, Atmos. Environ.,74,102-109, 2013.

Sobanska, S., Hwang, H., Choël, M., Jung, H., Eom, H., Kim, H., Barbillat, J., and Ro C.: Investigation of the Chemical Mixing State of Individual Asian Dust Particles by the Combined Use of Electron Probe X-ray Microanalysis and Raman Microspectrometry, Anal. Chem., 84 (7), 3145–3154, 2012.

Wang, H., An, J., Shen, L., Zhu, B., Xia, L., Duan, Q., and Zou, J.: Mixing state of ambient aerosols in Nanjing city by single particle mass spectrometry, Atmos. Environ., 132, 123-132, 2016.

Xie, R. K., Seip, H. M., Leinum, J. R., Winje, T., and Xiao, J. S.: Chemical characterization of, individual particles

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(PM10) from ambient air in Guiyang City, Sci. Total. Environ., 343(1-3).261-271, 2005.

Zhang, G., Han, B., Bi, X., Dai, S., Huang, W., Chen, D., Wang, X., Sheng, G., Fu, J., and Zhou, Z.: Characteristics of individual particles in the atmosphere of Guangzhou by single particle mass spectrometry, Atmos. Res., 153, 286-295, 2015.

Section 4.3: I got a bit lost in this Section? Which part is based on measurements and which based on model results? Please clarify.

Response: The emission of NO_x and NH_3, concentration of PM10 and its major components NO_3^- and NH_4^+ over East Asia were model results. We have clarified the modeled results in this section. In addition, the definition and method for transport distance over the sea, air temperature, RH, and average mixed layer for samples were shown in Section 2.4.

l. 351: 'totally off' is very colloquial. Is there any explanation for this discrepancy?

Response: The sentence has been revised into "For reference samples, simulated NH_4^+ concentrations sometimes can well reproduce the observational values, but the simulation was sometimes severely deviated from the observation. The deviation could be related to many factors which were out of scope of this study." in l.372-374.

l. 373: I cannot follow here. Why is ammonium excluded in Category 3? Isn't that a contradiction as you mention in the following sentence that you discuss here $N(NH_4^+ + NO_3^-)$?

Response: We are sorry for the confusion. The sentence has been revised into "The dry deposition fluxes of particulate $N_{NH4+NO3-}$ decreased by 50%, on average, in Categories 2 and 3, although the fluxes of ammonium of two samples in Category 3 increased." in l. 397-399.

l. 374: 'A larger decrease' than what? Please clarify.

Response: The sentence has been revised into "A larger decrease against the reference in the flux of nitrate was present in Categories 2 and 3, i.e., decreases of 73% and 46%, respectively." in l. 399-400.

l. 378 ff: Again, it is not clear whether the following text is based on observations or measurements. Please clarify.

Response: In the beginning of this section, we have stated that we calculated the dry deposition fluxes of aerosols particles, $N_{NH4+NO3-}$ and metal elements during dust and reference periods using the measured component concentrations and modeled dry deposition velocities (l. 385-387). And we have clarified the calculated values in this section.

Final comment: What are the main conclusions of your study? They should be summarized in a separate conclusion section after Section 5.

Response: We have summarized a separate conclusion in Section 5.

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The concentration, source and deposition flux of ammonium and nitrate~~inorganic nitrogen~~ in atmospheric particles during dust events at a coastal site in northern China

Jianhua Qi[1], Xiaohuan Liu[1], Xiaohong Yao[1], Ruifeng Zhang[1], Xiaojing Chen[1], Xuehui Lin[2], Huiwang Gao[1], Ruhai Liu[1]

[1]Key Laboratory of Marine Environment and Ecology, Ministry of Education, Ocean University of China, Qingdao, 266100, China

[2]Qingdao Institute of Marine Geology, Qingdao, 266100, China

Correspondence to: Jianhua Qi (qjianhua@ouc.edu.cn)

Abstract. Asian dust has been reported to carry anthropogenic reactive nitrogen during transport from source areas to the oceans. In this study, we attempted to characterize NH_4^+ and NO_3^- in atmospheric particles collected at a coastal site in northern China during spring dust events from 2008 to 2011. Based on the mass concentrations of NH_4^+ and NO_3^- in each total suspended particle (TSP) sample, the samples can be classified into increasing or decreasing types. In Category 1, the concentrations of NH_4^+ and NO_3^- were 20%-440% higher in dust day samples relative to samples collected immediately before or after a dust event. These concentrations decreased by 10-75% in the dust day samples in Categories 2 and 3. Back trajectory analysis suggested that multiple factors such as the transport distance prior to the reception site, the mixing layer depth on the transport route and the residence time across highly polluted regions, might affect the concentrations of NH_4^+ and NO_3^-. NH_4^+ in the dust day samples was likely either in the form of ammonium salts existing separately ~~externally~~ with dust aerosols or as the residual of incomplete reactions between ammonium salts and carbonate salts. NO_3^- in the dust day samples was attributed to various formation processes during the long-range transport. The positive matrix factorization (PMF) receptor model results showed that the contribution of soil dust increased from 23% to 36% ~~(90% of the scaled residuals located between the reasonable range −3 and +3~~~~90% of the residuals<3.0 and, r^2=0.97)~~ on dust days with decreasing contributions from local anthropogenic inputs and associated secondary aerosols. The estimated deposition flux of ~~inorganic nitrogen~~$N_{NH4+ + NO3-}$ varied greatly from event to event, e.g., the dry deposition flux of

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$N_{NH4+ +NO3-}$ ~~particulate inorganic nitrogen~~ increased by 9-285% in Category 1, but decreased by 46%-73% in Category 2~~ and 11-48% in Categories 3~~. In Categories 3, the average dry deposition fluxes of particulate nitrate and ammonium decreased by 46% and increased by 10%, respectively, leading to ~~while the average ammonium deposition flux increased by 10%.~~11-48% decrease in the fluxes of $N_{NH4+ + NO3-}$.

Keywords: aerosols, nitrogen, dust, source apportionment, dry deposition flux

1 Introduction

Reactive nitrogen carried in dust particles can be transported over a long distance, and the atmospheric nitrogen deposition in oceans has been recognized as an important external source of the nitrogen supporting phytoplankton growth (Duce et al., 2008; Zhang et al., 2010b). This hypothesis has been evaluated through incubation experiments, in situ experiments, and the use of satellite observational data (Banerjee and Kumar 2014; Guo et al., 2012; Liu et al., 2013; Shi et al., 2012; Tan and Wang, 2014). However, the process is dynamic due to the worldwide changing emissions of NO_x and NH_3 in the last few decades. For example, China and most of the developing countries in Asia experienced a large increase in emissions of NH_3 and NO_x while a substantial decrease in emissions occurred in Europe over the last three decades (Grice et al., 2009; Liu et al., 2017; Ohara et al., 2007; Skjøth and Hertel, 2013). The change would affect the nitrogen carried by dust particles to some extent, and updated studies are thereby essential.

Asian dust is one of three largest dust sources on earth. Asian dust has been reported to not only frequently cross over the mainland and the China Seas, but also to occasionally reach the remote northern Pacific Ocean or North America (Creamean 2013; Tan and Wang, 2014; Van Curen and Cahill, 2002; Zhang and Gao, 2007). In an extreme case, Asian dust was found to be transported more than one full circuit around the globe in approximately 13 days (Uno et al 2009). During the long-range transport, dust particles may mix with anthropogenic air pollutants and consequently undergo complicated chemical reactions (Cui et al., 2009; Li et al., 2014; Ma et al., 2012; Wang et al., 2011; Wang et al., 2016b; Wang et al., 2017a; Xu et al., 2014; Yang et al., 2002). For example, a few studies have shown that the concentrations of atmospheric particulate NO_3^- and NH_4^+ on dust storm days were 2-5 times larger than those prior to the events in Beijing (Liu et al., 2014; Liu and Bei, 2016).

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Fitzgerald et al. (2015) found that almost all Asian dust events observed in Korea contained considerable amounts of nitrate. However, Zhang et al. (2010a) reported an interesting result, i.e., the concentrations of ~~the~~ NO_3^- and NH_4^+ ~~showed~~were lower ~~concentrations~~ during strong dust storm events than weak dust events~~stronger dust storms corresponded to the smaller increases in these ions~~. A high uncertainty appeared to exist for c~~This raises the uncertainty~~complex for~~about~~ carrying amount of reactive nitrogen by dust particles.

A few contradictory results were also reported in the literature, which made the scientific issue even more complicated. For example, the concentration of NO_3^- in atmospheric aerosols on dust days was significantly lower in comparison to the concentration measured immediately before or after the event at a rural site in Yulin near the Asian dust source region (Wang et al., 2016b). The phenomenon was also observed in Shanghai, a mega city at a few thousands of kilometers from dust source zones in China, and more downwind sites (Kang et al., 2013; Li et al., 2014; Wang et al., 2013).

Inorganic nitrogen reportedly contributed to ~80%~~is a major component~~ of the total water-soluble nitrogen (TDN) in atmospheric particles~~ulates, and is reported to contribute ~80% of the TDN~~ collected over the Yellow Sea and in Qingdao (Shi et al., 2012). In the region, t~~T~~he dry deposition flux of the inorganic nitrogen accounted for more than 75% for the TDN (Qi et al., 2013). When deposited to the ocean via atmospheric dry deposition, inorganic nitrogen has great impact on marine productivity due to its bioavailability. To update and improve our knowledge on reactive nitrogen carried by dust particles, we collected atmospheric aerosol particles during and prior to (or post, but only when no sample was collected prior to dust events) at a coastal site adjacent to the Yellow Sea in each spring of 2008-2011. The concentrations of NO_3^- and NH_4^+ ~~(We later refer to dissolved inorganic nitrogen (DIN) as the sum of focused on nitrate and ammonium by excluding nitrite because of its very low concentration.~~

~~)inorganic nitrogen~~ and other components were determined for analysis. In this study, we focused on nitrate and ammonium by excluding nitrite because of its very low concentration. W~~w~~e first characterized the concentrations of NH_4^+ ~~+~~ and NO_3^- ~~inorganic nitrogen~~ in dust samples by comparing them with the values in atmospheric particles measured either prior to or post the event. We then conducted source apportionment to quantify their sources. Finally, we calculated and discussed the deposition flux of atmospheric particulate NH_4^+ ~~+~~ and NO_3^- ~~inorganic nitrogen~~ during dust events.

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2 Experimental methods

2.1 Sampling

Fig. 1 shows the sampling site, which is situated at the top of a coastal hill (Baguanshan) in Qingdao in northern China (36 ° 6' N, 120 ° 19' E, 77 m above sea level) and is approximately 1.0 km from the Yellow Sea to the east. A high-volume air sampler (Model KC-1000, Qingdao Laoshan Electronic Instrument Complex Co., Ltd., China) was set up on the roof of a two-story office building to collect total suspended particle (TSP) samples on quartz microfiber filters (Whatman QM-A) at a flow rate of 1 m^3/min. Prior to sampling, the filters were heated at 450 ℃ for 4.5 hrs to remove organic compounds. Our sample collection strategy involved collecting dust samples representing long-range transported particles. We followed the definition of dust events adopted in the regulations of surface meteorological observations of China (CMA, 2004; Wang et al., 2008) and identified dust events based on the meteorological records (Weather Phenomenon) of Qingdao from the Meteorological Information Comprehensive Analysis and Process System (MICAPS) of the China Meteorological Administration. Due to no dust events lasting over 12 hrs (Lee et al., 2015; Su et al., 2017; Zhang et al., 2007), we collected one dust sample with a 4-hr duration in a day. The sampling for dust particles started only when the measured PM_{10} mass concentration in Qingdao (http://www.qepb.gov.cn/m2/) and the forecasted dust mass over Asia (http://www-cfors.nies.go.jp/~cfors/) had greatly increased.

On March 20-21, 2010, two dust events subsequently swept Qingdao. ~~The 4-hr dust samples with IDs of 20100320 and 20100321 may not capture the entirety of the two events. However, t~~The on-line data in high time-resolution can allow identifying two dust events accurately from the start to the end. The data confirmed that the 4 hr dust samples with IDs of 20100320 and 20100321 were well separated from each other for the two events, although they may not capture the entirety of the two events, ~~allow adequate separation of the two dust event samples.~~ The same was true for the dust samples with IDs of 20110501, 20110502. Table 1 lists the sampling information. Based on the forecast, we also collected aerosol particle samples immediately before, which were regarded as the reference samples. These reference samples were further classified into sunny day samples and cloudy day samples. For those events missing sampling prior to dust events, we collected post-dust samples under clear and sunny weather conditions as early as possible.

Asian dust events were mostly observed in the spring at the sampling site. Our intensive samplings

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were concentrated in the period of March to May in 2008-2011, when a smaller outbreak for Asian dust events was observed in northern China (Fig. S3). Overall, a total of 14 sets of dust samples and 8 sets of comparison samples were available for analysis in this study.

To facilitate the coastal sampling data analysis, sand samples were collected at the remote site of Zhurihe (42 °22'N, 112 °58'E) in the Hunshandake Desert, one of the main Chinese sand deserts, in April 2012. Sand samples were packed in clean plastic sample bags and were stored below -20 ℃ before the transfer. An ice-box was used to store the samples during transport to the lab for chemical analysis.

2.2 Analysis

The aerosol samples were weighted according to the standard protocol. The sample membranes were then cut into several portions for analysis. One portion of each aerosol sample was ultrasonically extracted with ultra-pure water in an ice water bath for determining inorganic water-soluble ions using ICS-3000 ion chromatography (Qi et al., 2011). The sand samples collected at the Zhurihe site were analyzed using the same procedure. ~~We later refer to dissolved inorganic nitrogen (DIN) as the sum of nitrate and ammonium by excluding nitrite because of its very low concentration.~~

One portion of ~~the~~ each aerosol filter was cut into 60 cm^2 pieces and digested with HNO_3+$HClO_4$+HF (5:2:2 by volume) at 160 ℃ using an electric heating plate. The concentrations of Cu, Zn, Cr, Sc and Pb were measured using inductively coupled plasma mass spectrometry (Thermo X Series 2), while the concentrations of Al, Ca, Fe, Na and Mg were measured using inductively coupled plasma atomic emission spectroscopy (IRIS Intrepid II XSP). Field blank membranes were also analyzed for correction.

One portion of aerosol sample was digested with an HNO_3 solution (10% HNO_3, 1.6 M) at 160 ℃ for 20 min in a microwave digestion system (CEM, U.S.). The Hg and As in sample extracts were analyzed following the U.S. Environmental Protection Agency method 1631E (U.S. EPA, 2002) using cold vapor atomic fluorescence spectrometry (CVAFS). The detection limits, precisions and recoveries of water-soluble ions and metal elements are listed in Table 2.

2.3 Computational modeling

The enrichment factor of metal elements was given by

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$$EF_i = \frac{(X_i / X_{Re})_{aerosols}}{(X_i / X_{Re})_{crust}} \quad (1)$$

where subscripts *i* and *Re* refer to the studied metal and the reference metal, respectively; $(X_i/X_{Re})_{aerosols}$ is the concentration ratio of metal *i* to metal *Re* in the aerosol samples; and $(X_i/X_{Re})_{crust}$ is the ratio of metal *i* to metal *Re* in the Earth's crust. For the calculation of the enrichment factor of the metal elements, scandium was used as the reference element (Han et al., 2012), and the abundance of elements in the Earth's crust given by Taylor (1964) was adopted.

The 72-h air mass back trajectories were calculated for each TSP sample using TrajStat software (Wang et al., 2009) and National Oceanic and Atmospheric Administration (NOAA) GDAS (Global Data Assimilation System) archive data (http:// www.arl.noaa.gov/ready/hysplit4.html). The air mass back trajectories were calculated at an altitude of 1500 m to identify the dust origin. In addition, the distance over sea of the air mass for each sample was measured from the trajectory using TrajStat software (Wang et al., 2009).

The positive matrix factorization (PMF) is a commonly used receptor modeling method. This model can quantify the contribution of sources to samples based on the composition or fingerprints of the sources (Paatero and Tapper, 1993; Paatero, 1997). The measured composition data can be represented by a matrix X of *i* by *j* dimensions, in which *i* number of samples and *j* chemical species were measured, with uncertainty *u*. *X* can be factorized as a source profile matrix (*F*) with the number of source factors (*p*) and a contribution matrix (*G*) of each source factor to each individual sample, as shown in Equation 2.

$$X_{ij} = \sum_{k=1}^{p} G_{ik} F_{kj} + E_{ij} \quad (2)$$

where E_{ij} is the residual for species *j* of the i-th sample.

The aim of the model is to minimize the objective function *Q*, which was calculated from the residual and uncertainty of all samples (Equation 3), to obtain the most optimal factor contributions and profiles.

$$Q = \sum_{i=1}^{n} \sum_{j=1}^{m} (E_{ij}/u_{ij})^2 \quad (3)$$

The EPA PMF 3.0 model was used to obtain the source apportionment of atmospheric particulates on dust and comparison days. Our modeled results satisfied the reasonable fit criteria, i.e. 90% of the scaled residuals were located between the range −3 and +3 for each species. The correlation coefficient between the predicted and observed concentrations was 0.97.

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Dry deposition velocities were obtained using Williams' model (Williams, 1982) by accounting for particle growth (Qi et al., 2005). Williams' model is a two-layer model used to calculate the dry velocity of size-segregated particles over the water. In an upper layer below a reference height (10 m), the deposition of aerosol particles is governed by turbulent transfer and gravitational settling. In the deposition layer, the gravitational settling of particles is affected by particle growth due to high relative humidity. To obtain the deposition velocity of different particle sizes, Williams' model needs many input parameters, such as the wind speed at 10-m height (U_{10}), air/water temperature, and relative humidity. Relative humidity, air temperature and U_{10} from the National Centers for Environmental Prediction (NCEP) were used in this study. Surface seawater temperature data was collected from the European Centre for Medium-Range Weather Forecasts (ECMWF). The meteorological and seawater temperature data had a six-hour resolution. According to a previously reported method (Qi et al., 2013), the dry deposition fluxes of the particles and the nitrogen species were calculated for dust and comparison days.

The CMAQ model (v5.0.2) was applied over the East Asia area to simulate the concentrations of PM_{10}, NO_x and NH_3 for 14 samples collected during 11 dust events. The simulated domain contains 164×97 grid cells with a 36-km spatial resolution, and the centered point was 110 °E, 34 °N. The vertical resolution includes 14 layers from the surface to the tropopause, with the first model layer at a height of 36 m above the ground level. The meteorological fields were generated by the Weather Research and Forecasting (WRF) Model (v3.7). Considering that the simulated area is connected to the Yellow Sea, the CB05Cl chemical mechanism was chosen to simulate the gas-phase chemistry. Zhang et al. (2009) generated t~~T~~he emissions of ~~–~~air pollutants in 2006 including NO_X and NH_3 over East Asia~~ for each dust event were also modeled using the CMAQ model according to the emission inventory in 2008, which~~ and they updated the emission inventory ~~was generated by extrapolating the 2006 activity data~~ in 2008 for us being used in this study~~to the year 2008 using the method described by Zhang et al. (2009)~~. Initial conditions (ICONs) and boundary conditions were generated from a global chemistry model of GEOS-CHEM. All the dust events simulations are performed separately, each with a 1-week spin-up period to minimize the influence of the ICONs. The validation of the application of the CMAQ model in China has been reported by Liu et al. (2010a, b).

2.4 Other data sources and statistical analysis

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Meteorological data were obtained from the Qingdao Meteorological Administration (http://qdqx.qingdao.gov.cn/zdz/ystj.aspx) and the MICAPS of the Meteorological Administration of China. Different weather characteristics, such as sunny days, cloudy days and dust days, were defined according to information from the MICAPS and Qingdao Meteorological Administration. According to the altitude, longitude and latitude of the 72-hr air mass back trajectory of each dust sample, the pressure level, temperature and relative humidity (RH) data along the path of the air mass were derived from the NCEP/NCAR re-analysis system (http://www.esrl.noaa.gov/psd/data/gridded/data.ncep.reanalysis.html) for each sample. The mixed layer depth during the air mass transport of dust samples was obtained from the HYSPLIT Trajectory Model (http://ready.arl.noaa.gov/hypub-bin/trajasrc.pl) using the same method. Then the average mixing layer, transport altitude, air temperature and RH were calculated as an average of all points on the air mass back trajectory of each sample. Spearman correlation analysis was applied to examine the relationships of nitrate and ammonium with transport parameters, and P values of <0.05 were considered to be statistically significant.

3 Results

3.1 Characterization of aerosol samples collected during dust events

We first examined the mass concentrations of TSP samples and the concentrations of crustal and anthropogenic metals therein through a comparison with the ~~reference~~ samples collected on dust days and reference samples on immediately before or after days, providing the background information for our target species analyzed later. The comparative results are highlighted below. For these reference samples, the TSP mass concentrations ranged from 94 to 275 $\mu g \cdot m^{-3}$, with an average of 201 $\mu g \cdot m^{-3}$ (Fig. 2, Table S1). The TSP mass concentration increased substantially to 410-3857 $\mu g \cdot m^{-3}$ in dust day samples, with an average of 1140~~.3~~ $\mu g \cdot m^{-3}$. In each individual pair of dust day sample against reference sample, ~~the~~ a net increase in the mass concentration of TSPs was observed. The percentages varied from 82 to~~-~~ 1,303% on basis of events, with a ~~median~~ mean value of 403% (Table S1). A similar increase was present in the crustal elements in each pair of samples. For example, the mean concentrations of Sc, Al, Fe, Mg and nss-Ca (usually used as a typical dust index) increased by more than a factor of two. On the other hand, the enrichment factors (EF) of Al, Fe, Ca, and Mg were less

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than three in dust day samples with values less than 14 in the reference samples (Table 3). Lower values are indicative of elements from a primarily crustal origin. The average mass concentrations of anthropogenic elements, such as Cu, Pb, Zn, Cr, Hg and As, in dust day samples increased by 107% to 722% against those in the reference sample; however, the EF of the anthropogenic metal elements decreased in the former. This indicates that dust particles likely carried more anthropogenic elements, although their relative contribution to the total mass was lower than that in the reference sample. Note that Sample 20110415 was excluded for further analysis. It was judged as a local blowing dust event because no corresponding dust event existed upwind.

3.2 Concentrations of NH_4^+ and NO_3^- ~~inorganic nitrogen~~ in dust day samples

When the mass concentrations of NH_4^+ and NO_3^- in each pair of TSP samples were compared, the concentrations of NH_4^+ increased by 8%-473% in some dust day samples (20080301, 20080315, 20090316, 20100315, 20100320, 20100321, 20110418 and 20110502), but decreased by 28-84% in other dust day samples (Fig. 3, Column NH_4^+ and NO_3^- in Table S1). The same was generally true for the measured concentrations of NO_3^-.

Considering the relative values of NH_4^+ and NO_3^- in dust day samples relative to the reference samples, we classified the dust day samples into three categories (Table 4). In Category 1, the mass concentrations of NH_4^+ and NO_3^- were larger in dust day samples against the reference samples. In Category 2, the reverse was true. In Category 3, the mass concentrations of NO_3^- were lower in the dust samples than in the reference samples, whereas the concentrations of NH_4^+ were close to the reference. As reported, the Yellow Sea encountered dust storms mainly derived from the Hunshandake Desert (Zhang and Gao, 2007). We thereby compared our observations with the sand particles collected from this desert (Table 5). The ratios of mass concentrations of nitrate and ammonium to the total mass of sand particles were very low, i.e., less than 81 μg/g, which are approximately three orders of magnitude less than the corresponding values in our dust samples. The values obtained from atmospheric aerosols at the urban sites of Duolun (Cui, 2009) and Alxa Right Banner (Niu and Zhang, 2000), which are closer to the desert, increased on dust days, but were still over one order of magnitude lower than the corresponding values in this study (Table 5). The mixing and chemical interaction between anthropogenic air pollutants and dust particles during transport from the source zone to the reception site likely played an important role in increasing the ratios, leading to extremely larger ratio values at

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this site relative to those in source dust and in upwind atmospheric particles (Cui et al., 2009; Wang et al., 2011; Wu et al., 2016). ~~However, the increase or decrease in the mass concentration absolute of nitrate and ammonium in different dust samples against the reference implied the complex combined result of many factors, such as~~ Since air pollutant emissions, meteorological conditions, chemical reactions, and other ~~factor~~s can affect the concentrations ~~abundance of~~ NH_4^+ and NO_3^- in atmospheric particles collected in dust days, the observed increase or decrease in the mass concentration of nitrate and ammonium in different dust samples against the reference implied the combined effect of those factors. ~~for the interactions.~~

4. Discussion ~~and conclusion~~

4.1 Theoretical analysis of the three categories

Ammonium salts are common in atmospheric particles with diameters of less than 2 μm (Yao et al., 2003; Yao and Zhang, 2012). Many modeling studies have shown that the gas-aerosol thermodynamic equilibrium is assumed to be fully attained for inorganic ions, including ammonium salts in $PM_{2.5}$ (Dentener et al., 1996; Underwood et al., 2001; Wang et al., 2017a; Zhang et al., 1994; Zhang and Carmichael, 1999). Reasonably good agreements between ammonium salt modeling results and observations reported in the literature support the validity of this assumption (Chen et al., 2016; Penrodet al., 2014; Walker et al., 2012). Supposing that a thermodynamic equilibrium had been attained by the ammonium salts in Category 1, the reactions between carbonate salts and ammonium salts, such as 1) $(NH_4)_2SO_4$+ $CaCO_3 \Rightarrow$ $CaSO_4$ + NH_3 (gas) +CO_2 (gas) +H_2O and 2) $2NH_4NO_3$+ $CaCO_3 \Rightarrow Ca(NO_3)_2$ + $2NH_3$ (gas) +CO_2 (gas) +H_2O, will release NH_3 (gas) until $CaCO_3$ has been completely used up. During dust events, very high concentrations of Ca^{2+} were observed, and high $CaCO_3$ concentrations were therefore expected. For example, the single-particle characterization showed that Asia dust from the Gobi and Inner Mongolian Deserts had rich $CaCO_3$, with a ratio of 4.3-6.7% for reacted $CaCO_3$ and 3.0-4.6% for unreacted $CaCO_3$ (Hwang et al., 2008). Heterogeneous chemical reactions of mineral dust mostly occurred on $CaCO_3$ mineral dust (Hwang and Ro, 2006). However, when Category 1 was considered alone ~~and~~ except for Sample 20100321~~one exterior sample was excluded~~, a good correlation was obtained for $[NH_4^+]_{equivalent\ concentration}$=0.98*$[NO_3^-+SO_4^{2-}]_{equivalent\ concentration}$($R^2$=0.83, P<0.05). The good correlation, together with the slope of 1, strongly indicated that the NO_3^- and SO_4^{2-} were almost completely associated with NH_4^+

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in these dust day samples. ~~It was commonly believed that anthropogenic~~ Anthropogenic ammonium nitrate and ammonium sulfate were thought to be produced by gas, aqueous phase reaction and thermodynamic equilibrium processes and they usually internally mixed ~~,~~ (Seinfeld and Pandis, 1998)~~,~~ ~~and Wang et al., 2017a. And some studies found that this anthropogenic ammonium nitrate and ammonium sulfate was externally mixed with dust particles (Wang et al., 2017a).~~ In reverse, the poor correlation of Ca^{2+} to NO_3^- and SO_4^{2-} showed that the formation of $CaSO_4$ and/or $Ca(NO_3)_2$ was probably negligible. Thus, ammonium salt aerosols very likely~~may be~~ existed separately ~~externally mixed, may externally exist~~ with dust aerosols in these dust day samples. Wang et al. (2017a) also found that coarse mode ammonium was quite low and fine mode dust particles ~~were~~ existed separately~~externally mixed~~ with anthropogenic ammonium nitrate and ammonium sulfate. The observed ~~concentration of~~ NO_3^- and NH_4^+ in Asia dust samples were argued ~~to be~~due to ~~more~~ physically mixing two types of particles~~affected by the dilution of the local pollution by the invaded dust, the dust storm, i.e., the dilution effect,~~ rather than the heterogeneous formation of nitrate and ammonium~~chemical reaction on the dust~~ (Huang et al., 2010)~~.~~ ~~-~~The hypothesis appeared to be valid in Category 1, where NH_4^+ was negatively correlated with Ca^{2+} (Fig. S4). In the ~~exterior~~ Ssample 20100321 collected on 21 March 2010, [NH_4^+] only accounted for ~70% of the observed [NO_3^-+SO_4^{2-}] in an equivalent concentration. This result suggested that ~30% of (NO_3^-+SO_4^{2-}) may be associated with dust aerosols via the formation of metal salts of the two species. This hypothesis was supported by the correlation result, i.e., NO_3^- was positively correlated with NH_4^+ and Cu, and SO_4^{2-} was correlated with K^+, Na^+ and Mg^{2+} (Fig. S4). Scheinhardt et al. (2013) found that Cu^{2+} showed mixed organic and nitrate complexation in aerosol particles, using a thermodynamic model (E-AIM III). ~~And~~ Cu was also detected to be partly in the form of nitrate in aerosol particles by single particle mass spectrometry (Wang et al., 2016a; Zhang et al., 2015). Cu was once used as an effective marker of diesel and biodiesel-blend exhaust (Gangwar et al., 2012), while it can also be derived from copper pyrites ($CuFeS_2$) in Inner Mongolia mines (Huang et al., 2010). The increase of Cu in the mass concentration in dust samples implied dust particles mixed with anthropogenic particles, particularly from industrial emissions, during transport. In addition, many studies showed that SO_4^{2-} ~~mainly~~can exist~~ed~~ in ~~the~~ many forms of metal salts in atmospheric particles, such as Na_2SO_4, K_2SO_4, $K_2Ca(SO_4)_2{\cdot}H_2O$, $Na_2Ca(SO_4)_2$, $Na_2Mg(SO_4)_2{\cdot}4H_2O$, $(NH_4)_2Mg(SO_4)_2{\cdot}6H_2O$, $Na_3(NO_3)(SO_4){\cdot}\ H_2O$ ~~,~~ (Chabas and Lefèvre, 2000; Sobanska et al., 2012; Xie et al., 2005). ~~These reference results further confirm our hypothesis.~~

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For Category 2, no correlation between $[NH_4^+]_{equivalent\ concentration}$ and $[NO_3^-+SO_4^{2-}]_{equivalent\ concentration}$ existed. When Category 2 was considered alone except for one Sample 20110501~~and one exterior sample was excluded~~, the equivalent ratios of NH_4^+ to $NO_3^-+SO_4^{2-}$ were generally much smaller than 1, suggesting that a larger fraction of $NO_3^-+SO_4^{2-}$ may exist as metal salts due to reactions of their precursors with dust aerosols. NO_3^- and SO_4^{2-} showed no correlations with NH_4^+ but did show significant correlations with Pb (Fig. S4). The average concentration of Ca^{2+} in Category 2 (0.43±0.40 μ~~g~~mol/m^3) was evidently higher than that in Category 1 (Ca^{2+}: 0.17±0.04 μ~~g~~mol/m^3), implying the probable formation of $CaSO_4$ and/or $Ca(NO_3)_2$ and the release of NH_3 (gas). Moreover, except for 20080502, the remaining dust samples in Category 2 were transported from the desert relatively enriched with $CaCO_3$ (1-25% in Wt%) (Formenti et al., 2011). A positive correlation between NO_3^- and SO_4^{2-} in Category 2 against a negative correlation in Category 1 also implied that the dust particles enriched with $CaCO_3$ in Category 2 might play an important role to form SO_4^{2-} and NO_3^-. Ca-rich dust particles coated with highly soluble nitrate were observed at Kanazawa in Japan during Asian dust storm periods using SEM/EDX (scanning electron microscopy equipped with an energy dispersive X-ray spectrometer) (Tobo et al.，2010). The single-particle observation conducted by Hwang and Ro (2006) showed that $CaCO_3$ in dust particles was almost completely consumed to produce mainly $Ca(NO_3)_2$ species.

There were only three samples in Category 3. $[NH_4^+]_{equivalent\ concentration}$=0.95*$[NO_3^-+SO_4^{2-}+Cl^-]_{equivalent\ concentration}$ was obtained for Sample 20110418, implying ~~ndicating~~ that ~~relative enough~~the NH_4^+ was not only associated with NO_3^- and SO_4^{2-} but also with Cl^-. In the sample collected on 15 March 2010, $[NH_4^+]$ ~~only~~ accounted for ~~~~78% of the observed $[NO_3^-+SO_4^{2-}]$ in an equivalent concentration. As discussed above, ~20% of ($NO_3^-+SO_4^{2-}$) may be associated with dust aerosols via the formation of metal salts of the two species. The equivalent ratio of NH_4^+ to $NO_3^-+SO_4^{2-}$ was only 0.14 for Sample 20100320, and Ca^{2+} for this sample (0.47 μmol/m^3) was evidently higher than that for Sample 20100315 (Ca^{2+}: 0.12 μmol/m^3) and 20110418 (Ca^{2+}: 0.12 μmol/m^3), suggesting that a larger fraction of $NO_3^-+SO_4^{2-}$ may exist as metal salts. However, the unique changes in NH_4^+ and NO_3^-, different from Category 1 and 2, need further investigation~~As a whole, the limited samples in this Category showed different formation process, which maybe cause the concentration of NH_4^++NO_3^- presented a different variation from Category 1 and 2. However, it's hard to get a formation explanation due to the limited samples.~~

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4.2 Source apportionment of aerosols during dust and non-dust events

The sources of atmospheric aerosols in dust and reference samples were determined by PMF modeling (Paatero and Tapper, 1993; Paatero, 1997). Fig. 4 shows that atmospheric aerosols in the reference samples mainly included six sources, i.e., industry, soil dust, secondary aerosols, sea salt, biomass burning, and coal combustion/other sources~~, with 90% of the scaled residuals falling between -3 and +3; r^2=0.97~~. In these dust samples, including Categories 1-3, oil combustion, industry, soil dust, secondary aerosols, and coal combustion/other sources were identified as five major sources (Table 6). The contribution of soil dust evidently increased from 23% to 36% in the dust samples relative to the reference, consistent with the high concentrations of TSPs and crustal metals observed on dust days. The calculated contribution of nitrate plus ammonium from the soil dust source to the total mass of nitrate plus ammonium in the dust samples greatly increased. The source profile for coal combustion in the dust day samples showed a high percentage of K^+, Cl^-, Ca, Mg, Co, Ni, As, Al and Fe, indicating that coal combustion particles may exist contemporaneously with other anthropogenic pollutants emitted along the transport path. Liu et al. (2014) also found a larger net increase in the contribution of dust aerosols to the mass of PM_{10}, i.e., 31%-40%, on dust days against non-dust days in Beijing which is approximately 600 km upwind of Qingdao. Accordingly, they reported that the contributions of local anthropogenic sources decreased on dust days, especially those from secondary aerosols, consistent with the EF of anthropogenic metals observed on dust days.

4.3 Influence of transport path ways on ~~particulate i~~NH_4^+ and NO_3^-~~norganic nitrogen~~ in dust samples

The calculated air mass trajectories for 13 out of 14 samples showed that the air mass originated from North and Inner Mongolia, China (Fig. 5), generally consistent with the results of Zhang and Gao (2007). The remaining one, with ID of 20110418 originated from Northeast China. The calculated trajectories showed that the entire dust air mass passed over those highly polluted regions with strong modeled emissions of NO_x and NH_3 shown in Fig 6 and experienced different residence times therein. Fig. 5 shows that all air mass trajectories in Category 1 were transported from either the north or northwest over the continent, except for the ~~one exterior s~~Sample 20110502. In Category 2, the air masses always took a 94-255 km trip over the sea prior to arriving at the reception site. NH_3-poor conditions in the marine atmosphere disfavored the formation and existence of ammonium nitrate. On

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the other hand, the humid marine conditions (the calculated average RH ranged in 50-75% over the Bohai and Yellow Seas in 2006-2012~~, calculated~~ using NCEP/NCAR re-analysis data) might have enhanced hetero-coagulation between dust and smaller anthropogenic particles, leading to the release of NH_3 via reactions between preexisting ammonium salts and carbonate salts.

The average mixing layer was less than 900 m along the air mass transport routes for most sampling days in Category 1 (Table 7), favoring the trapping of locally emitted anthropogenic air pollutants in the mixing layer. The air masses in Category 1 took over 11-39 hrs to cross over the highly polluted area with appreciable modeled concentrations of NO_x (5.7±1.4 ppb) and NH_3 (7.6±3.3 ppb). Except for ~~the exterior~~ two samples (ID of 20080529 and 20110319)~~,~~, air masses in Category 2 took less than 10 hrs to cross over the polluted areas with lower concentrations of NO_x (modeled value: 3.6±3.4 ppb) and NH_3 (modeled value: 4.7±4.7 ppb) and the mixing layer height along the route was 916-1194 m (on average) for each dust event. Moreover, the averaged wind speed at sampling site was 2.8 m/s in Category 1, but 6.2 m/s in Category 2. The lower wind speed in Category 1 was unexpected, implying dust particles very likely traveled at aloft with a high speed and then mixed down to the ground through subsidence. This further led to the external mixing of anthropogenic particulate matters and dust. The correlation analysis results in Table S2 indirectly support these conclusions.

The concentrations of PM_{10} and its major components NO_3^- and NH_4^+ over East Asia on dust days and comparison days were modeled using the WRF-CMAQ model (Fig. S5-6). Spatial distributions of simulated PM_{10} during each dust events were consistent with the records in the "Sand-dust Weather Almanac" (CMA, 2009; 2010; 2012; 2013). The dust particles were transported eastward by passing over the sampling site, the China Sea and arriving at the far remote ocean region, except for the local blowing dust sample with ID of 20110415, as mentioned previously. NMB (normalized mean bias) values of simulated NO_3^- were -4% and -12% in dust and non-dust reference samples, respectively, indicating that CMAQ results reasonably reproduce the mass concentrations of NO_3^- (Fig. S6). Simulated NH_4^+ concentrations in dust samples were severely under-predicted with NMB values at -71%. For reference samples, simulated NH_4^+ concentrations sometimes can well reproduce the observational values, but the simulation was sometimes severely deviated from the observation. The deviation could be related to~~totally off because of misprediction of meteorological conditions~~many factors which were out of scope of this study. The ~~external~~ separately mixing mechanism proposed in this study is urgently needed to be included in the model for accurately predicting the concentrations

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during dust events.

4.4 Dry deposition fluxes of TSP, ~~particulate~~ NH_4^+, NO_3^- ~~inorganic nitrogen~~ and metals

Dust events are known to increase the deposition fluxes of aerosol particles along the transport path because of high particle loadings. For example, Fu et al. (2014) found that the long-range transported dust particles increased the dry deposition of PM_{10} in the Yangtze River Delta region by a factor of approximately 20. In terms of atmospheric deposition in the oceans, a few studies reported enhancements in oceanic chlorophyll *a* following dust storm events (Banerjee and Kumar, 2014; Tan and Wang, 2014). In addition to those in high-nutrient and low-chlorophyll (HNLC) regions, the input of nitrogen and other nutrients associated with dust deposition is expected to promote the growth of phytoplankton in oceans with varying nutrient limitation conditions. Thus, we calculated the dry deposition fluxes of aerosols particles, $N_{NH4+NO3-}$ and metal elements during dust and reference periods using the measured component concentrations and modeled dry deposition velocities (Table 8). We also compared the calculated dry deposition flux of TSP and $N_{NH4+NO3}$- with previous observations in the literature.

The calculated dry deposition fluxes of atmospheric particulates increased on dust days against the reference to some extent. For example, the particle deposition fluxes varied over a wide range from 5,200 to 65,000 mg/m^2/month in different dust sampling days, with an average of 18,453 mg/m^2/month, in comparison with the dry deposition flux of TSP of 2,800±700 mg/m^2/month from the reference periods in the coastal region of the Yellow Sea. The dry deposition fluxes of $N_{NH4+NO3}$- varied, depending on Category 1, 2 or 3. In Category 1, the dry deposition fluxes of $N_{NH4+NO3}$- increased by 9-75% with increased TSP flux by 86-252% (Table S3). In Categories 2 and 3, the dry deposition fluxes of TSP increased by 126% to 2,226% against the references. ~~Excluding Except for~~ ~~On average, ammonium in Category 3, t~~The dry deposition fluxes of particulate $N_{NH4+NO3}$- decreased by ~~44~~50%, on average, in Categories 2 and 3, ~~though~~although the fluxes of ammonium of two samples in Category 3 increased ~~(on average)~~. A larger decrease against the reference in the ~~concentration~~ flux of nitrate was present in Categories 2 and 3, i.e., decreases of 73% and 46%, respectively. ~~Note that t~~The ~~average~~ ammonium deposition flux also decreased by 47% in Category 2 but increased by 10% in Category 3.

Except for Pb and Zn in Category 2, the calculated dry deposition fluxes of Cu, Pb and Zn increased with those of nitrogen on dust days. Trace metals were found to have a toxic effect on marine

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phytoplankton and inhibit their growth (Bielmyer et al., 2006; Echeveste et al., 2012). Liu et al. (2013) found that inhibition coexisted with the promotion of phytoplankton species in incubation experiments in the southern Yellow Sea in the spring of 2011 by adding Asian dust samples to collected seawater. However, the calculated dry atmospheric deposition fluxes of Fe increased by a factor of 124-2,370% in dust day samples. Wang et al. (2017b) recently reported that Fe can alleviate the toxicity of heavy metals. Moreover, atmospheric inputs of iron to the ocean have been widely proposed to enhance primary production in HNLC areas (Jickells et al., 2005).

Due to anthropogenic activity and economic development, NH_4^+ ~~+~~ and NO_{3x}^- ~~inorganic nitrogen~~ emissions were reported to increase~~d~~ in China from 1980 to 2010 (Fig. S3; Liu et al., 2017). The dry deposition flux of $N_{NH4+NO3}$- should have theoretically increased with the increase in the emission of inorganic nitrogen. Considering the different dry deposition velocities to be used in various studies, we recalculated the dry deposition flux of $N_{NH4+NO3}$- in the literature using the dry deposition velocities of 1 cm/s for nitrate and 0.1 m/s for ammonium, as reported by Duce et al. (1991). We thereby found that dry deposition fluxes of $N_{NH4+NO3}$- over the Yellow Sea during the dust days increased greatly from 1999 to 2007, but the values in Qingdao varied narrowly within a range of 94.75-99.65 mg N/m^2/month during the dust days from 1997 to 2011 (Table 9). The complicated results implied that even more updated works are needed in the future.

5 Conclusion

The concentrations of nitrate and ammonium in TSP samples varied greatly from event to event on dust days. Relative to the reference samples~~non-dust-day samples~~, the concentrations were both higher in some cases and lower in others. The observed ammonium in dust day samples was explained by NH_4^+ was likely either in the form of ammonium salts existing ~~externally~~separately ~~–~~with dust aerosols or as the residual of incomplete reactions between ammonium salts and carbonate salts. NO_3^- in the dust day samples can be due to either mixing or reactions between anthropogenic air pollutants and dust particles or combined both during the transport from the source zone to the reception site. ~~NO_3^- in the dust day samples was attributed to various formation processes during the long-range transport.~~However, this process was generally much less effective and led to a sharp decrease in nitrate in ~~Category 2~~ TSP samples of Category 2. The ~~external~~ existence of ammonium salt aerosols separately

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~~w~~with dust aerosols and the extent of the reactions between ammonium salts and carbonate salts were apparently associated with the transport pathway, metrological conditions and precursor emissions~~moving speeds and metrological conditions~~, ~~among~~nd other factors. Due to a sharp increase in dust loads on dust days, the contribution of ~~soil~~ dust to the total aerosol mass increased against the ~~was higher on dust days than on comparison days~~samples collected on other days. T~~, while t~~he contributions from local anthropogenic sources were accordingly lower on dust days.

Overall, this study strongly suggested that atmospheric deposition of N_{NH4+_NO3-} on dust days varied greatly~~ and that no simple linear increase existed with increasing dust load~~. ~~More observations at various locations are needed to obtain a statistical relationship between dust events and atmospheric deposition of N_{NH4+_NO3-}.~~ A simple assumption of a linear increase in N_{NH4+_NO3-} with increasing dust load, like that in the literature, could lead to a considerable overestimation of the dry deposition flux of nutrients into the oceans and the consequent primary production associated with dust events.

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Table 1. Sampling information for the aerosol samples collected at the Baguanshan site in the coastal region of the Yellow Sea.

Sampling year	Sample category	Sampling number	Sampling time	Weather characteristics
2008	Samples on dust days	20080301	From 13:22 a.m. to 17:22 p.m. on Mar. 1st	Floating dust[a]
		20080315	From 13:21 a.m. to 17:21 p.m. on Mar. 15th	Floating dust
		20080425	From 13:14 a.m. to 17:14 p.m. on Apr. 25th	Floating dust
		20080528	From 11:38 a.m. to 15:38 p.m. on May 28th	Floating dust
		20080529	From 10:15 a.m. to 12:15 p. m. on May 29th[b]	Floating dust
	Reference ~~Samples~~ samples ~~on non-dust days~~	20080316	From 13:00 a.m. to 17:00 p.m. on Mar. 16th	Sunny day
		20080424	From 13:00 a.m. to 17:00 p.m. on Apr. 24th	Sunny day
		20080522	From 13:00 a.m. to 17:00 p.m. on May 22nd	Cloudy day with mist
2009	Samples on dust days	20090316	From 8:25 a.m. to 12:25 p.m. on Mar. 16th	Floating dust
	Reference ~~Samples~~ samples ~~on non-dust days~~	20090306	From 13:00 a.m.to 17:00 p.m. on Mar. 6th	Sunny day
2010	Samples on dust days	20100315	From 11:30 a.m.to 15:30 p.m. on Mar. 16th	Mist after floating dust
		20100320	From 10:30 a.m. to 14:30 p.m. on Mar. 20th	Floating dust
		20100321	From 10:30 a.m. to 14:30 p.m. on Mar. 21st	Floating dust
	Reference samples~~Samples on non-dust days~~	20100324	From 11:30 a.m. to 15:30 p.m. on Mar. 24th	Sunny day
2011	Samples on dust days	20110319	From 12:00 a.m. to 16:00 p.m. on Mar. 19th	Floating dust
		20110415	From 12:00 a.m. to 16:00 p.m. on Apr. 15th	Floating dust
		20110418	From 12:25 a.m. to 16:25 p.m. on Apr. 18th	Floating dust[c]
		20110501	From 12:10 a.m. to 16:10 p.m. on May 1st	Floating dust
		20110502	From 16:00 a.m. to 20:00 p.m. on May 2nd	Floating dust

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Reference samples~~Samples on non-dust days~~	20110308	From 12:00 a.m. to 16:00 p.m. on Mar. 8th	Sunny day
	20110416	From 12:00 a.m. to 16:00 p.m. on Apr. 16th	Sunny day
	20110523	From 12:00 a.m. to 16:00 p.m. on May 23rd	Sunny day

[a]Note that one ~~exterior~~ dust sample 20080301 was collected on March 1 when no dust was recorded by the MICAPS. However, the MICAPS information indeed showed dust events in China on March 1. The modeled spatial distribution of the PM_{10} mass concentration for this dust event on March 1 implies that the sample should be classified as a dust sample. The supporting figures are shown in Fig. S1.

[b]The sampling duration was reduced to only 2 hrs because of extremely high particle loads. In addition, the samples with IDs of 20080528 and 20080529 were subjected to two different dust events occurring over two days instead of continuous samples for one dust event (CMA, 2009).

[c]Note that one ~~exterior~~ dust sample 20110418 was collected on April 18 when no dust was recorded by the MICAPS. However, blowing dust occurred and was recorded on April 17 by the Sand-dust Weather Almanac 2011 (CMA, 2013). The modeled spatial distribution of the PM_{10} mass concentration for this dust event on April 18 implies that the sample should be classified as a dust sample. The supporting figure is Fig. S2.

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Table 2. Detection limits, precisions and recoveries of water-soluble ions and metal elements.

Component	Measurement method	Detection limit (μg L^{-1})	Precision (RSD%)	Recovery (%)
NO_3^-	IC	2.72	1.54	97
SO_4^{2-}		1.62	1.55	98
NH_4^+		0.4	1.10	97
Ca^{2+}		0.44	0.79	94
Cu	ICP-MS (Xin et al., 2012)	0.006	4.0	106
Zn		0.009	2.5	102
Cr		0.004	3.0	95
Sc		0.002	2.4	97
Pb		0.008	3.9	104
Al	ICP-AES (Lin et al., 1998)	7.9	0.6	103
Ca		5.0	1.2	99
Fe		2.6	0.7	104
Na		3.0	0.6	99
Mg		0.6	0.6	105
Hg	CVAFS	0.0001	6.6	105
As	CVAFS	0.1	5.0	98

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Table 3. The average concentrations and EFs of metal elements on dust and non-dust days.

Element	Concentration (ng/m^3)		EF*	
	Non-dust days	Dust days	Non-dust days	Dust days
Sc	1.11	13.90	-	-
Al	8.53×10^3	6.86×10^4	3.8	1.4
Fe	4.91×10^3	3.88×10^4	3.	1.2
Ca	1.05×10^4	4.29×10^4	14.0	2.1
Mg	1.62×10^3	1.58×10^4	3.5	1.1
Cu	50.2	124.5	36.3	6.1
Pb	127.9	221.0	389.4	56.1
Zn	340.0	457.7	248.9	20.6
Cr	33.8	244.0	44.0	11.1
Hg	0.26	0.36	176.0	13.8
As	25.5	27.4	707.2	43.9

*EF values less than 10 indicate that the studied element is mainly derived from crustal sources, whereas EF values much higher than 10 indicate an anthropogenic source.

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Table 4. Average measured concentrations of NH_4^+, NO_3^- ~~inorganic nitrogen (DIN)~~, TSP, NOx, relative humidity (RH) and air temperature for each aerosol sample category in Qingdao.

	Sample number	TSP ($\mu g \cdot m^{-3}$)	NO_3^- ($\mu g \cdot m^{-3}$)	NH_4^+ ($\mu g \cdot m^{-3}$)	RH (%)	T (℃)	NOx ($\mu g \cdot m^{-3}$)	Summary
Category 1	20080301	527	20.5	12.7	57	7.0	36	NH_4^+ and NO_3^- ~~DIN~~ concentration ~~on~~ in dust day samples higher than ~~that~~ reference samples~~on non-dust days~~
	20080315	410	19. 5	29.9	62	11.0	59	
	20090316	688	15.9	17.2	27	16.0	75	
	20100321	519	16.5	9.4	51	8.8	76	
	20110502	810	21.0	11.0	49	17.7	62	
Category 2	20080425	622	6.8	2.0	30	18.0	40	NH_4^+ and NO_3^- ~~DIN~~ concentration ~~on~~ in dust day samples lower than ~~that on non-dust days~~reference samples
	20080528	2579	9.2	2.7	17	27.0	34	
	20080529	2314	17.5	4.8	60	20.0	29	
	20110319	939	12.3	9.4	16	12.6	93	
	20110501	502	4.5	5.3	23	21.6	66	
Category 3	20100315	501	5.4	4.3	30	7.2	73	NO_3^- concentration ~~on~~ in dust day samples lower than ~~that on non-dust days~~reference samples; NH_4^+ close to that on reference samples~~non-dust days~~
	20100320	3857	5.5	3.4	35	10.6	92	
	20110418	558	3.8	6.6	33	12.6	47	
~~Non-dust~~ Reference	20080316	225	12.6	8.4	28	11.0	60	
	20080424	137	21.7	7.2	49	18.0	53	
	20080522	206	27.4	16.6	78	20.0	60	

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samples[a]	20090306	94	2.9	3.0	29	7.00	51
	20100324	275	7.2	2.4	23	9.0	82
	20110308	194	13.0	13.1	20	11.5	111
	20110416	252	5.6	5.4	26	14.1	55
	20110523	224	15.2	10.2	42	20.6	49

[a]For the corresponding ~~non-dust day~~reference samples for each dust event, see Table 1.

Table 5. Comparison of the NH_4^+ and NO_3^- ~~inorganic nitrogen~~ ~~(DIN)~~ content in sand and aerosol particles on dust days or close to the dust source region (unit: μg/g).

Sands sampled in dust source regions			Aerosols in or close to dust source region on dust days			Aerosols in the coastal region of the Yellow Sea	
Study region and data source	Relative concentration[a] NO_3^-	Relative concentration[a] NH_4^+	Study region and data source	Relative concentration[a] NO_3^-	Relative concentration[a] NH_4^+	NO_3^-	NH_4^+
Zhurihe (This study)	25.46 ± 22.87	4.21 ± 1.03	Duolun (Cui, 2009)	1200	900	~~Non-dust~~Reference samples: 28,200 ±24,819	~~Non-dust~~Reference samples: 24,063 ±21,515
Alxa Left Banner, Inner Mongolia (Niu and Zhang, 2000)	62.1 ±7.4	79.1 ±1.1	Alxa Right Banner, Inner Mongolia (Niu and Zhang, 2000)	1975[b]	4091[b]	Category 1: 34,892 ±9570	Category 1: 22,571 ±7,016
Yanchi, Ningxia (Niu and Zhang, 2000)	46.4 ±2.2	80.9 ±1.3	Hinterland of the Taklimakan Desert, Xinjiang (Dai et al., 2016)	142-233	2-15	Category 2: 5,542 ±5,117	Category 2: 4,758 ±5,698
			Average of Sonid Youqi, Huade (Inner Mongolia), Zhangbei (Hebei) (Mori et al., 2003)	253	710	Category 3: 6,359 ±4,697	Category 3: 7,059 ±5,591
			Yulin, the north edge of Loess Plateau (Wang et al., 2011)	216.4	80.6		

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	Golmud, Qinghai(Sheng et al., 2016)	892.9	-[c]
	Hohhot, Inner Mongolia (Yang et al., 1995)	588.1	No data

[a]Relative concentration of NH_4^+ ~~+~~ and NO_3^- ~~DIN~~ per aerosol particle mass

[b]Samples collected on a floating dust day (horizontal visibility less than 10000 m and very low wind speed)

[c]The ammonium concentration was lower than the detection limit of the analytical instrument.

Table 6. Sources and source contributions (expressed in %) calculated for aerosol samples collected during dust and non-dust events

Dust event		Comparison days	
Source	% of TSP	Source	% of TSP
Soil dust	36	Soil dust	23
Industrial	21	Industrial	24
Secondary aerosol	6	Secondary aerosol	23
Oil combustion	6	Biomass burning	16
Coal combustion and other uncertain sources	31	Coal combustion	5
		Sea salt	9

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Table 7. Concentrations of TSP, NO_3^-, and NH_4^+; transport speed; transport distance over the sea; transport distance; air temperature; RH; average mixed layer during transport and transport time in polluted region for atmospheric aerosol samples on dust days.

Group	Sample number	TSP (μg/m^3)	NO_3^- (μg/g)	NH_4^+ (μg/g)	Speed (km/h)	Distance over the sea (km)	Transport altitude (m)	Mixed layer depth (m)	R-time[a] (h)	T[b] (℃)	RH[c] (%)
Category 1 $NH_4^+ > RS^d$ $NO_3^- > RS^d$ ~~IN>ND~~	080301	527	38,984	24,107	40.1	0	1,160±702	864±745	39	-2.9±11.7	29±10
	080315	410	47,611	34,130	79.1	0	4,921±1,870	950±525	13	-32.5± 16.4	34±16
	090316	688	23,050	25,012	86.2	0	3,739±1083	702±665	11	-19.1±11.7	42±17
	100321	519	31,741	18,155	87.2	0	3,407±1,249	1,113±760	19	-23.0±13.6	42±22
	110502	810	25,995	13,632	30.2	177	3,666±1,371	747±957	26	-13.2±15.8	31±13
Category 2 $NH_4^+ < RS^d$ $NO_3^- < RS^d$ ~~IN<ND~~	080425	256	4,089	372	29.6	0	887±656	1,161±1,040	10	-2.7±6.1	66±13
	080528	2579	232	72	88.2	244	4,336±1461	1,064±830	8	-15.5±13.6	31±16
	080529	2314	26	166	63.7	94	2,148±1,725	1,194±816	43	3.6±18.4	25±17
	110319	939	13,088	10,067	70.6	132	4,271±1867	790±719	27	-26.3±20.0	48±32
	110501	502	8,924	10,631	35.1	252	3,212±810	916±1,114	5	-13.4±8.5	39±13
Category 3	100315	501	10,767	8,515	57.3	0	5,009±1410	1,110±365	7	-40.4±13.3	45±29
$NO_3^- <$ ~~ND~~ RS^d	100320	3857	1,418	884	76.9	0	1,284±401	525±371	10	-12.2±6.3	61±16
$NH_4^+ \geqq$ ~~ND~~ RS^d	110418	558	6,891	11,778	35.6	931	1,344±780	695±672	2	-0.1±8.2	52±28

[a]Residence time of the air mass passing over parts of highly polluted regions according to the

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trajectories of samples.

[b]Average air temperature with the definition in Section 2.4.

[c]Average relative humidity with the definition in Section 2.4.

[d]Reference samples collected on days immediately before or after dust event

Table 8. Dry deposition of TSP (mg/m^2/month), $N_{NH4+NO3}$ ~~particulate inorganic nitrogen~~ (mg N/m^2/month) and some toxic trace metals (mg/m^2/month) on dust and ~~non-dust~~reference days.

	Dry deposition flux							
	TSP	NO_3^--N	NH_4^+-N	$N_{NH4+NO3}$	Fe	Cu	Pb	Zn
Category 1[a]	8,000±1800	65±9	24±14	90±17	533±179	2±0.3	0.3±0.3	6±2
Category 2[a]	18000±11,000	13±18	8±4	21±22	1300±1000	3±2	0.08±0.04	4±1
Category 3[a]	29,000±31,000	26±6	17±8	42±12	2100±2200	6±1	0.20±0.02	5±3
Non-dust	2,800±700	48±33	15±8	63±39	190±110	1±1	0.09±0.1	5±4

[a]For the characterization of $N_{NH4+NO3}$ concentration and sample information of the category, see Table 3.

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Table 9. Comparison of dry deposition flux and normalized flux of TSP (mg/m^2/month) and $N_{NH4++NO3}$- (mg N/m^2/month) with observations from other studies (mg N/m^2/month)

Source	Year	Area		TSP	$N_{NH4++NO3}$-	Normalized average flux of $N_{NH4++NO3}$-[a]
This work	2008-2011	Qingdao, coastal region of the Yellow Sea	~~Non-dust~~Reference day	2,800±700	63±39	93.90
			Dust day	10,138±15,940	58±36	101.39
			Average of dust and ~~non-dust~~reference			97.64
Qi et al., 2013	2005-2006	Qingdao, coastal region of the Yellow Sea	Average of nine months samples	159.2 - 3,172.9	1.8-24.5	94.75
Zhang et al., 2011	1997-2005	Qingdao	Average of annual samples		132	99.65
Zhang et al., 2007	1999-2003	The Yellow Sea			11.43	9.91
Shi et al., 2013	2007	The Yellow Sea	Reference~~Non-dust~~ day		19.2	132.17
			Dust day		104.4	227.07

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	Average of dust and Reference~~non-dust~~	179.62

[a]The calculation method of the normalized flux of $N_{NH4+NO3}$ was discussed in Section 3.7.

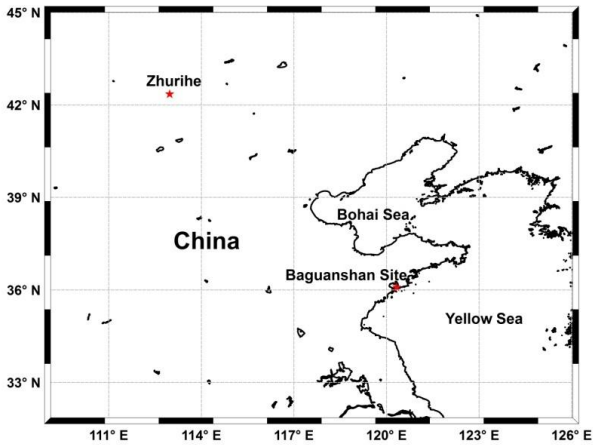

Figure 1. Location of the aerosol and dust sampling sites.

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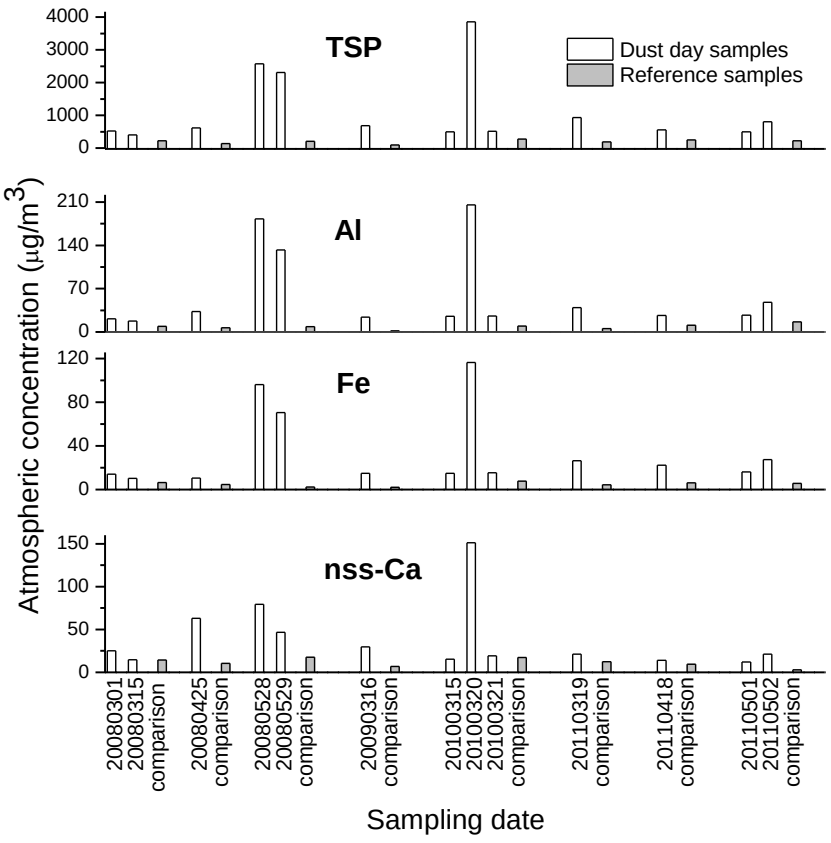
TSP
Al
Fe
nss-Ca
Dust day samples
Reference samples
Atmospheric concentration (μg/m3)
Sampling date
20080301
20080315
comparison
20080425
comparison
20080528
20080529
comparison
20090316
comparison
20100315
20100320
20100321
comparison
20110319
comparison
20110418
comparison
20110501
20110502
comparison

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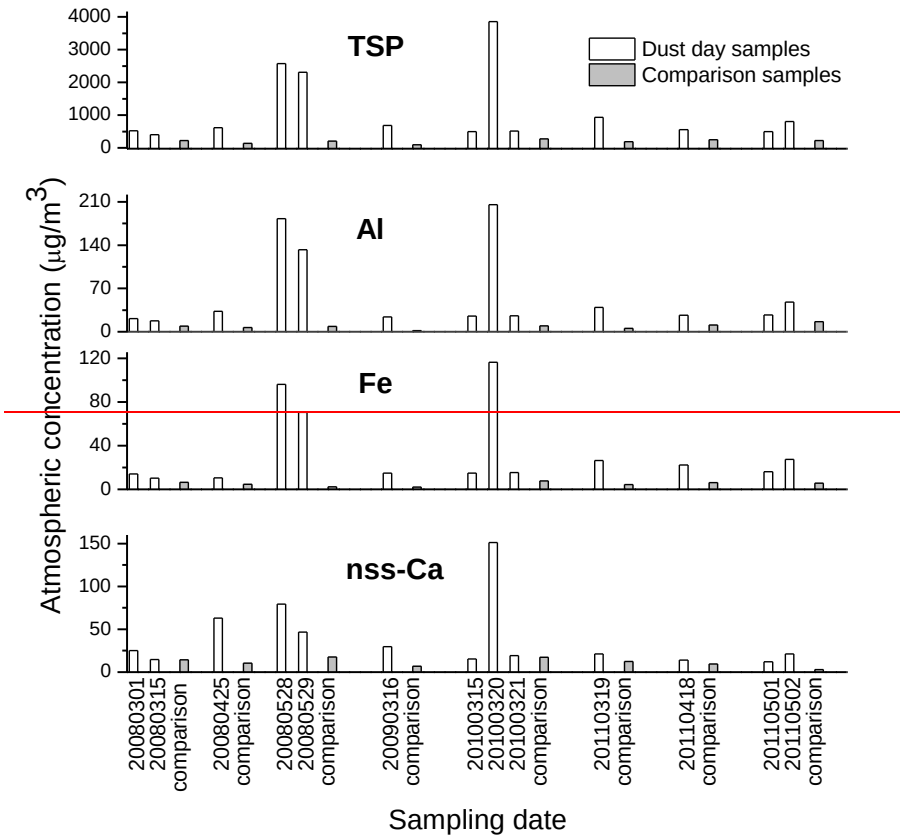

Figure 2. Mass concentrations of TSP, Al, Fe and nss-Ca in aerosol samples collected at the Baguanshan site on dust and ~~comparison~~ reference days from 2008~~-~~ to 2011.

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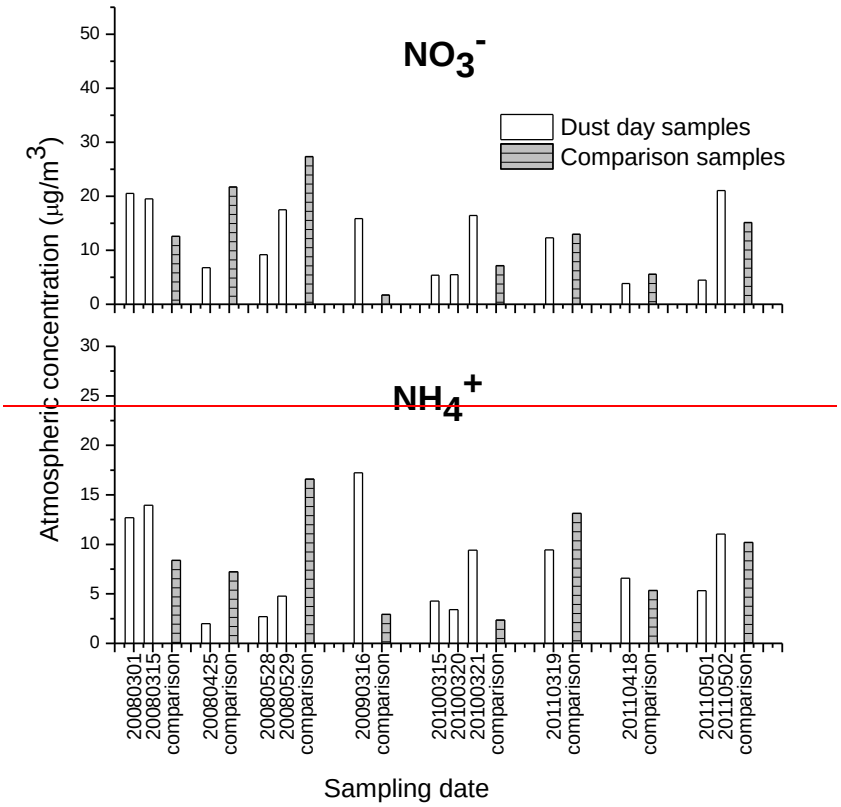
NO_3^-
Dust day samples
Comparison samples
Atmospheric concentration (μg/m3)
NH_4^+
20080301
20080315
comparison
20080425
comparison
20080528
20080529
comparison
20090316
comparison
20100315
20100320
20100321
comparison
20110319
comparison
20110418
comparison
20110501
20110502
comparison
Sampling date

1247

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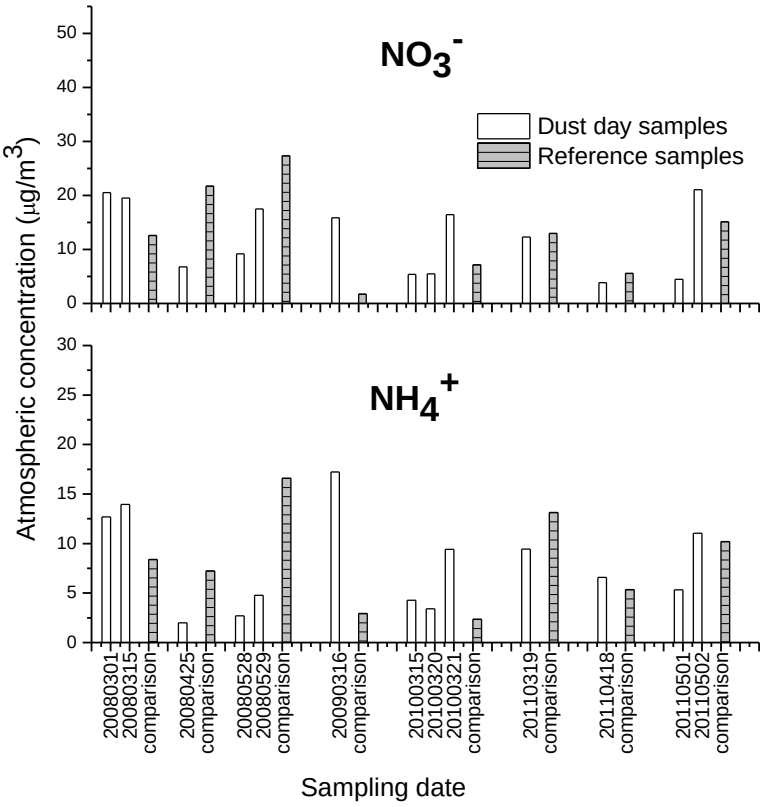

Figure 3. Mass concentrations of NH_4^+ and NO_3^- in aerosol samples collected at the Baguanshan site on dust and ~~comparison~~ reference days during March-May in 2008 to 2011.

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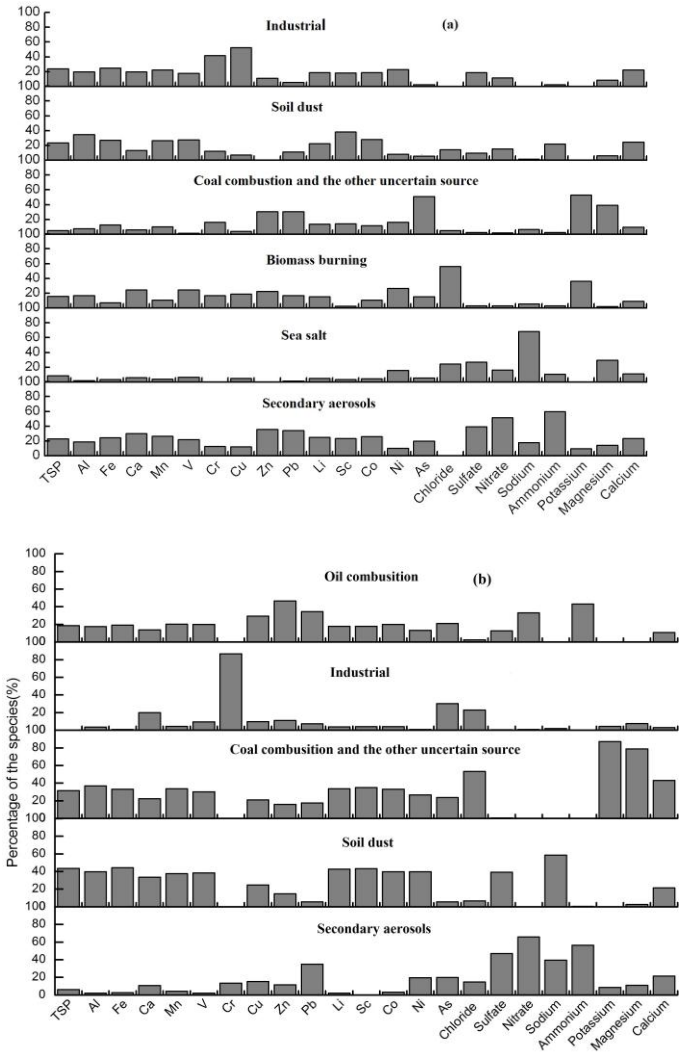

Figure 4. Source profiles of atmospheric aerosol samples collected on ~~non-dust~~reference (a) and dust (b) days using the PMF model.

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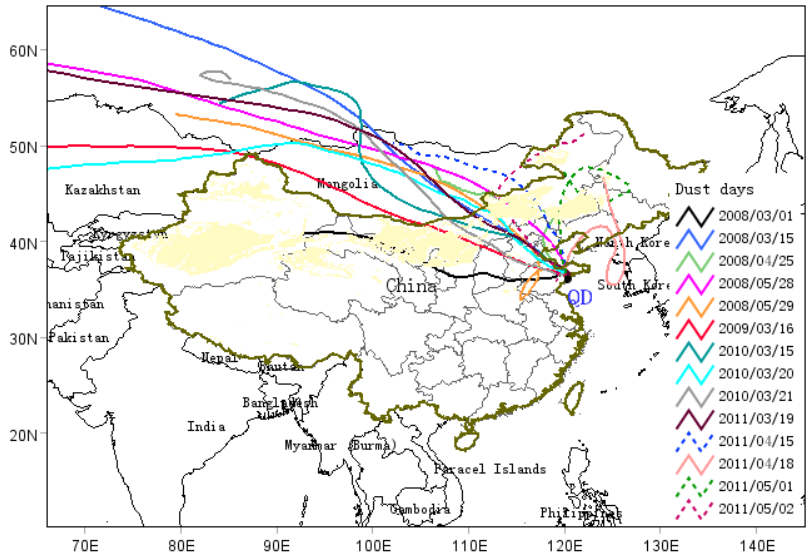

Figure 5. The 72-h backward trajectories for dust samples from 2008 to 2011(the yellow domains in the map represent the dust source regions in China).

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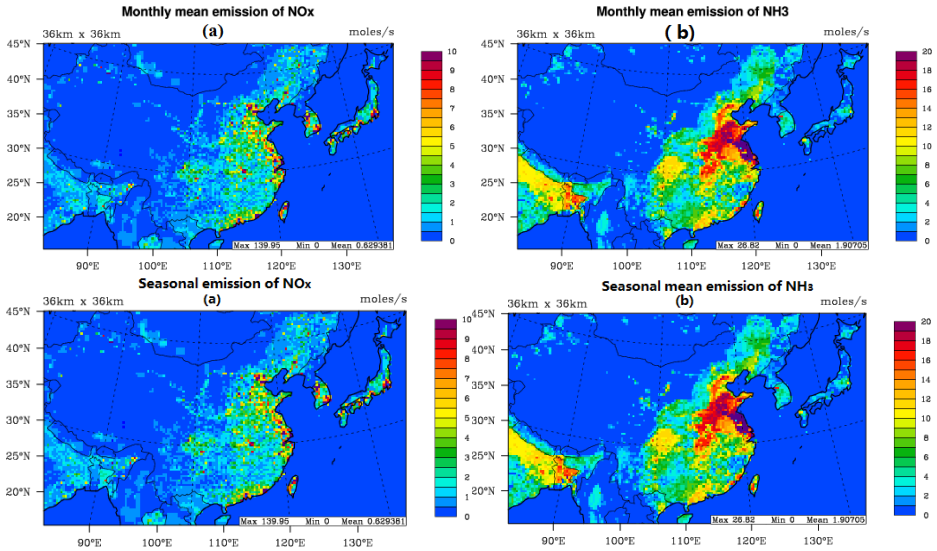

Figure 6. Seasonal mean emissions of NOx (a) and NH_3 (b) over East Asia from March-May 2008.

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