Dear Dr. Ervens,

Thank you very much for your patience and help! Thank you for permission for an extension of the resubmission of the revised manuscript (Ms. Ref. No.:acp-2016-1183). Based on the critical comments of the two reviewers, we have made a substantial revision to our manuscript, including reanalyzing the data, supplementing the model results, enhancing our discussion and changing the references. Most figures and tables have been revised. Additionally, we added 8 supporting files. Due to the substantial changes, the contributions of the authors of this manuscript have changed; hence, we have added two authors and revised the title of the revised manuscript. Detailed item-by-item responses to the comments of Referee #2 are listed below.

Best regards,

Yours sincerely,

Jianhua Qi

Reply to Interactive comment on “The concentration, source apportionment and deposition flux of atmospheric particulate inorganic nitrogen during dust events” by Jianhua Qi et al. Anonymous

Referee #2 Received and published: 27 March 2017

General comments: This paper attempts to study the impact of spring time dust storms in the Chinese deserts on the atmospheric concentration and deposition of inorganic nitrogen (IN) in the coastal Yellow Sea location. The study uses 4 years of particulate matter measurements and their chemical composition to understand how different types of dust storms can affect the abundance of inorganic nitrogen and calculate dry deposition of IN to the coastal Yellow Sea. This type of work is relevant since atmospheric deposition of nutrients and its implications is not a well understood topic and can be important for the regions that receive high atmospheric input like the Yellow Sea. However, the authors have not made best use of the data. There needs to be significant improvement in data interpretation and much more analysis needs to be done to support the main results. I would thus recommend a resubmission and see if there is any substantial improvement in the manuscript. My main comments are given below.

Reply: Thank you for the suggestion. We have reanalyzed the data and supplemented the modeling results using a 3-D air quality model to support the main results in the revised version. We have added "Theoretical analysis of three categories" and "The impact of dust on nitrogen dry deposition flux under anthropogenic activity" to the discussion section. Some figures have been redrawn, and some references have been updated. The data interpretation and the writing have also been improved. In brief, we have made substantial improvements to our manuscript.

Q1. First of all, the manuscript needs an overall improvement in writing. Not sufficient care has been given to the details and there are parts which are difficult to follow. I will provide some examples later, but there are many such cases of improper and awkward sentence constructions.
Second, on what basis are these dusty and non-dusty days decided? More information are needed to show whether the dust storms originating from the deserts are actually passing over the measurement site and it is not the locally produced soil dust in cases of the days with low TSP values. Satellite aerosol products and meteorological data can be used to support this. Again later Al concentration is used to identify dust weather (lines 171-172). Please provide a clear definition of dust days and maintain that throughout the manuscript.

Reply: We have revised all sections of this manuscript, especially the sections "Experimental Method" and "Results and Discussion", to provide detailed methods and a logical analysis of the data. The discussion was revised substantially. Additionally, the writing has also been improved by a native English speaker.

In this study, a dust event was defined according the definition adopted in regulations of surface meteorological observation in China (CMA, 2004;Wang et al., 2008) and was identified based on the meteorological record information from Meteorological Information Comprehensive Analysis and Process System (MICAPS) of China Meteorological Administration. The sampling information is listed in the Table 1. Based on the forecast, we also collected aerosol particle samples immediately before or after the dust event for comparison. The post-dust event samples were collected under clear and sunny weather conditions and at low mass concentrations of PM_{10}.

All air mass trajectories showed that dust storms originated from deserts or Gobi and passed over the measurement site. More information on the dust events has been added to Tables 1 and 4. According to the suggestion, we tried to obtain aerosol particle concentrations using satellite aerosol products; however, the daily satellite data were incomplete. Therefore, we have included modeled dust concentrations over East Asia based on the CFORS model by Uno et al. (2003) (http://www-cfors.nies.go.jp/) and the PM10 modeling results for each dust event using a 3-D air quality model to support our analysis in the revision. The model results for each event are shown in Fig. 2 and Fig.S3. Based on Fig. 2, we found that the Sample 20110418 was mainly affected by local blowing dust. To focus on the impact of long-range transported dust on aerosol samples in downwind areas, this sample 20110415 was not included in the analysis and discussion in later sections.

As the reviewer suggested, the samples can be affected by local soil dust. Therefore, we used the criterion method of total Al concentration in TSP samples proposed by Hsu et al. (2008) to confirm that our samples originated from the sandy sources, as proposed in the previous version of this manuscript. We have added the distribution of hourly dust concentrations modeled over East Asia by the CFORS model for dust events in the revised version. The model results show the dust occurrence and transport patterns, thereby providing much more information than the Al criteria method. Therefore, we revised this section and replaced the Al criteria method with the model results in the revised manuscript.

Table 1. Sampling information for the aerosols 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. 15 th	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
	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
	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
	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
	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 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 figure is Fig. S1.

[b] The sampling duration was reduced to only 2 hrs because of extremely high particle loads.

[c]Note that one exterior dust sample 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, 2011). 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

Table 4. Average concentrations of inorganic nitrogen, TSP, NOx, Relative Humidity (RH) and air temperature for each Category in aerosol samples 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 °C	NOx $\mu g \cdot m^{-3}$	Summary
Category 1	20080301	527	20.5	12.7	57	7.0	36	IN concentrationon dust days higher than that on ND 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	IN concentrationon dust days lower than that on ND days
	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 dust days lower than that on ND days;NH_4^+close to that on ND days
	20100320	3857	5.5	3.4	35	10.6	92	
	20110418	558	3.8	6.6	33	12.6	47	
Non-dust[a]	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	
	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] The corresponding ND day for each dust event see Table 1.

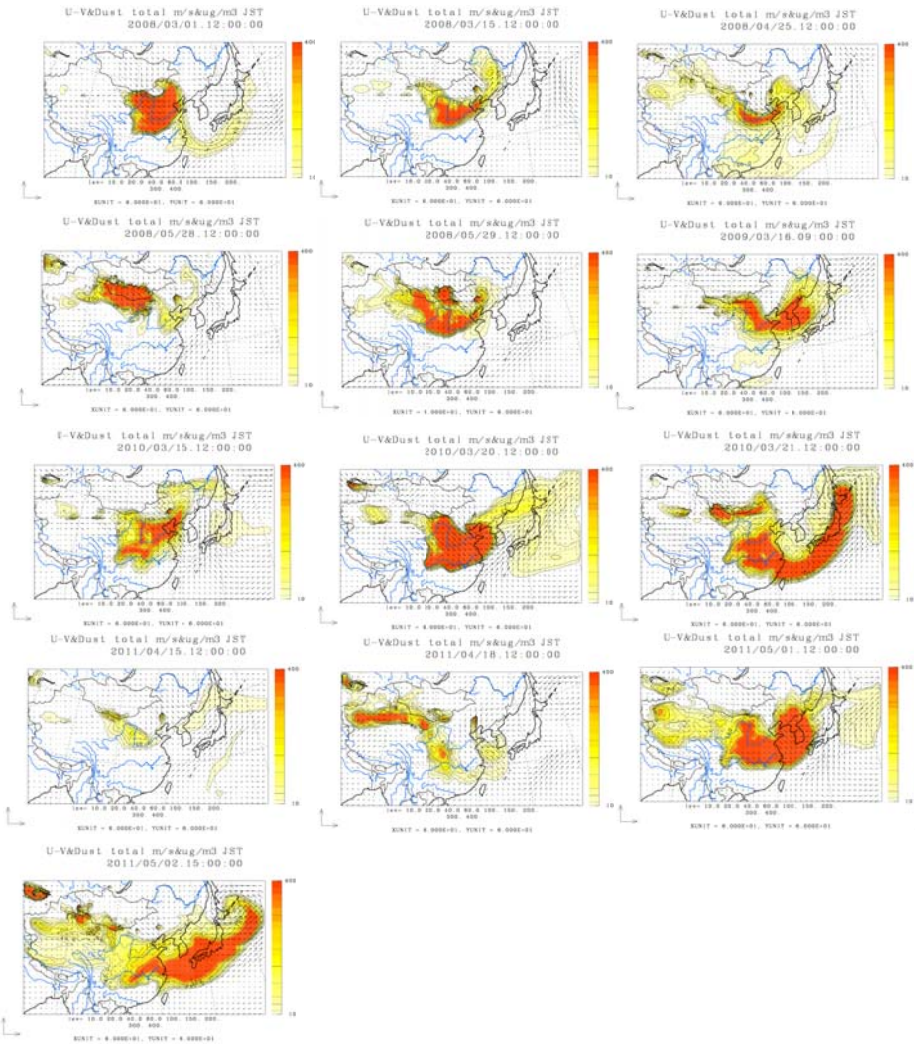
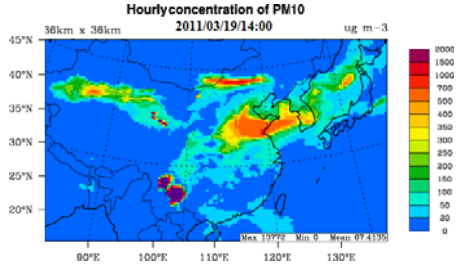


Figure 2. Modeled dust concentrations over East Asia by CFORS model during each dust sampling day from 2008 to 2011 (http://www-cfors.nies.go.jp/). (The figures show the modeled dust concentration in the middle of each sampling duration). No data are available for Mar. 19, 2011, because of the earthquake in Japan. Hourly PM10 concentrations were modeled by the WRF-CMAQ model for each sampling day, and the results are shown in Fig. S3.



FigureS3. Hourly PM10 concentration in China modeled by the WRF-CMA model for Sample 20110418 (The figure shows the modeled PM10 concentration at the middle of the sampling

period).

CMA: Regulations of Surface Meteorological Observation, China Meteorological Press, Beijing, 154–156, 2004.

CMA: Sand-dust weather almanac 2011, China Meteorological Press, Beijing, 36-53, 2013.

Hsu, S. C., Liu, S. C., Huang, Y. T., Lung, S. C. C., Tsai, F., Tu, J. Y., and Kao, S. J.: A criterion for identifying Asian dust events based on Al concentration data collected from northern Taiwan between 2002 and early 2007, J. Geophys. Res-Atmos., 113, 1044-1044, 2008.

Uno, I., Carmichael, G. R., Streets, D. G., Tang, Y., Yienger, J. J., Satake, S., Wang, Z., Woo, J. H., Guttikunda, S., Uematsu, M., Matsumoto, K., Tanimoto, H., Yoshioka, K., and Iida, T.: Regional chemical weather forecasting system CFORS: Model descriptions and analysis of surface observations at Japanese island stations during the ACE-Asia experiment, J. Geophys. Res-Atmos., 108, 1147-1164, 2003.

Wang, Y. Q., Zhang, X. Y., Gong, S. L., Zhou, C. H., Hu, X. Q., Liu, H. L., Niu, T., and Yang, Y. Q.: Surface observation of sand and dust storm in East Asia and its application in CUACE/Dust, Atmos. Chem. Phys., 8, 545-553, 2008.

Q2. The authors have divided dusty days into 3 categories based on inorganic nitrogen (IN) concentrations relative to the non dust (ND) days. They are reporting that in some cases IN concentrations are more than the ND days, in some cases IN concentration are less than the ND days and in some cases nitrate concentration on dust days are less than ND days while ammonium concentrations on dust days are more compared to ND days. Next, the authors are reporting that sand from Duolun (which is a source for dust storms affecting the Yellow Sea region) is poor in nitrate and ammonium content. First of all, trajectory analysis does not seem to point that coastal Yellow Sea region is only affected by dust originating from Duolun region. There are many other dust sources over which the trajectories are passing. And if Duolun is deficient in IN you need to provide a detailed discussion on the possible sources of IN in dust sampled from the coastal region of the Yellow Sea and about the mixing of anthropogenic aerosols with the dusty air mass. The authors have related the 3 cases of IN in dust samples to the wind speed concluding that when wind speed is less IN concentration increased and vice versa . I am not clear how this conclusion is arrived at, especially, with only 5 cases for IN<ND. Forexample, in Table 5 sample number 110501 NO3- and NH4+ concentrations are lowand the wind speed is also low. Again, sample 080315 has higher NO3- and NH4+concentrations at higher wind speed. What is the rationale of using wind speed of 40.5km/h in this study and how is this threshold derived? I would suggest the authors togroup the trajectories according to dust and non dust days and also according to the levels of nitrogen and see which of these trajectories are passing over highly populated regions.

Reply: First of all, East Asia has three major dust aerosol sources: the western China sources, including the Taklimakan Desert and surrounding areas; the Mongolia sources, including the desert and semi-desert areas in southern Mongolia; and the northern China sources, including the BadainJaran Desert, the Tengger Desert, the Ulan Buh Desert and the Hunshandake Desert (Fig. SR1). Duolun and Zhurihe belong to the Hunshandake Desert in northern China. According to studies, 52%-71% of the dust storms that affect the Yellow Sea are from this sand source (Zhang and Gao, 2007; Gao et al., 2010). For our 14 dust samples, 13 events were from northern sources, and the remaining one was from northern sources but was transported to the northeast before being transported to the measurement site (the 72 h trajectory only showed transport from the northeast). Therefore, we collected sand samples at the Zhurihe site and used reported aerosol values from Duolun to characterize the aerosol particles in the source region. As the reviewer suggested, the coastal Yellow Sea region is affected by dust originating not only from the Duolun region but also from other regions. We added the nitrate and ammonium contents in sand samples collected from

other sand sources in China. We also added the reported concentrations of nitrate and ammonium in aerosol particles on dust days at or close to the dust source region in Table 5 in the revised manuscript. All these data verified that the mass concentrations of nitrate and ammonium relative to the total mass of sand particles were very low, i.e., less than 81μg/g. We have added the modeling results of NO_x and NH_3 emissions using a 3-D air quality model and discussed the theoretical analysis of three categories in section 3.3 in the revised manuscript.

To characterize the transport paths of dust day samples more exactly, we reanalyzed the air mass trajectory of each sample at 1500 m altitude and redrew Fig. 5 to distinguish trajectories for samples collected on dust and non-dust days. According to the suggestion, we first we investigated whether these trajectories passed over highly populated regions. All trajectories involved transport from the source over highly populated regions and exhibited different transport times in these regions. We found that the concentrations of nitrate and ammonium depended on the transport distance over the sea, the mixed layer depth and the transport time over highly polluted regions. We have rewritten this section according to the suggestion to discuss the influence of transport paths on nitrate and ammonium contents in the revised manuscript.

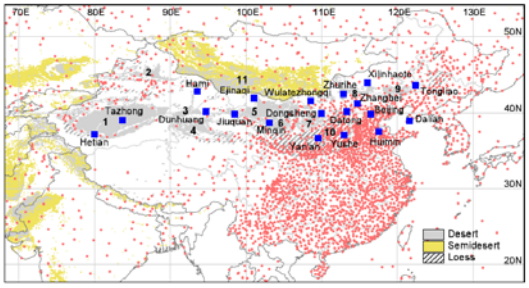


Figure SR1. Map of the main source regions of sand and dust storms in China (from Wang et al., 2008).

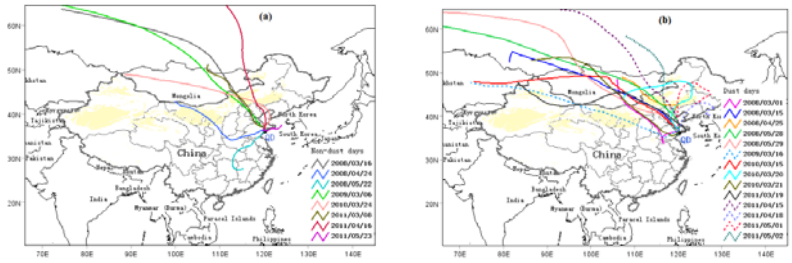


Figure 5. The 72-h backward trajectories for non-dust (a) and dust (b) samples from 2008 to 2011 (the yellow domains in the maps represent the dust source regions in China.

Table 5. Comparison of nitrate and ammonium 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]		Study region and data source	Relative concentration[a]			
	NO_3^-	NH_4^+		NO_3^-	NH_4^+	NO_3^-	NH_4^+
Zhurihe (This study)	25.46± 22.87	4.21± 1.03	Duolun (Cui, 2009)	1200	900	Non-dust: 28,200±24,819	Non-dust: 24,063±21,515
AlxaLeft Banner, Inner Mongolia (NiuandZhang, 2000)	62.1±7.4	79.1±1.1	AlxaRightBanner, Inner Mongolia (NiuandZhang, 2000)	1975[b]	4091[b]	Category 1: 34,892±9570	Category 1: 22,571±7,016
Yanchi, Ningxia (NiuandZhang, 2000)	46.4±2.2	80.9±1.3	Hinterland of theTaklimakan Desert, Xinjiang (Dai et al., 2016)	142-233	2-15	Category 2: 5,542±5,117	Category 2: 4,758±5,698
			Average of SonidYouqi, 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		
			Golmud, Qinghai(Sheng et al., 2016)	892.9	-[c]		
			Hohhot, Inner Mongolia (Yang and Wang, 1995)	588.1	No data		

[a]Relative concentration ofnitrate and ammonium 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.

Cui, W. L.: Chemical transformation of dust components and mixing mechanisms of dust with pollution aerosols during the long range transport from north to south China, M.S. thesis, Department of Environmental Science and Engineering, FudanUniversity, China, 38 pp., 2009.

Dai, Y. J.: Vertical distribution of characteristics of dust aerosols in the near-surface in hinterland of Taklimakan Desert, M.S. thesis,

College of Resources and Environmental Science, Xinjiang University, China, 26 pp., 2016.

Mori, I., Nishikawa, M., Tanimura, T., and Quan, H.: Change in size distribution and chemical composition of kosa (Asian dust) aerosol during long-range transport, Atmos. Environ., 37, 4253-4263, 2003.

Niu, S. J. and Zhang, C. C.: Researches on Sand Aerosol Chemical Composition and Enrichment Factor in the Spring at Helan Mountain Area, Journal of Desert Research, 20, 264-268, 2000.

Sheng, Y., Yang, S., Han, Y., Zheng, Q., and Fang, X.: The concentrations and sources of nitrate in aerosol over Dolmud, Qinghai, China, Journal of Desert Research, 36, 792-797, 2016.

Wang, Q. Z., Zhuang, G. S., Li, J., Huang, K., Zhang, R., Jiang, Y. L., Lin, Y. F., and Fu, J. S.: Mixing of dust with pollution on the transport path of Asian dust — Revealed from the aerosol over Yulin, the north edge of Loess Plateau, Sci. Total. Environ., 409, 573–581, 2011.

Yang, D. Z., Wang, C., Wen, Y. P., Yu, X. L., and Xiu, X. B.: Ananalysis of Two Sand Storms In Spring 1990, Quarterly Journal of Applied Meteorology, 6, 18-26, 1995.

Specific comments:

Q3. L11-13: Reconstruct the sentence.

Reply：These sentences have been rewritten as "Asian dust has been reported to transport anthropogenic reactive nitrogen from source areas to the oceans. In this study, we attempted to characterize the NH_4^+ and NO_3^- in atmospheric particles collected at a coastal site in northern China during spring dust events from 2008 to 2011."

Q4. L20-25: This is not conclusive from the discussions that follow. Statements like “storms were weak or slow moving” and “rapid transport in a strong dust storm” are not supported by proper analysis of the storm characteristics.

Reply: We have written the conclusion.

Q6. L35-36: This is contradicting L32-33.

Reply: We apologize for the confusion in the revision. We will revise the abstract sentence into "The dust deposition was an uncertain source of nitrogen for the ocean".

Q7. L40: These references point to anthropogenic contribution to atmospheric nitrogen deposition.

Reply: We apologize for the confusion in the revision. We have revised the abstract.

Q8. L62-64: The first part of the sentence seems to contradict the last part.

Reply: To avoid confusion, we have revised the sentence to "Although increased concentrations of NO_3^- and NH_4^+ in aerosol particles were observed on dust storm days in northern China relative to those non-dust days prior to the dust storm events, Zhang et al. (2010) also found that the concentrations of the two species were associated with the intensity of the dust storm, i.e., the stronger dust storms corresponded to the smaller increases. In other words, lower NO_3^- and NH_4^+ concentrations occurred during strong dust storm events than during weak dust events (Zhang et al., 2010)."

Q9. L79-81: Meaning is not clear.

Reply: We have deleted this sentence in the revised manuscript because this reference is not closely related to Asian dust.

Q10. L102: Zhurihe in Hunshandake Desert. Later you are using Duolun region which isnot introduced in Section 2. It will be difficult for readers to follow if they are not veryfamiliar with this region.

Reply: Thank you for the suggestion. The data at Duolun was adapted from the reference (Cui et al., 2009b). We have added the citation in the text of section 3.2. According to the former suggestion inQ2, we added new references for the dust components in source region to Table 4 in the revised manuscript (See reply to Q2). Additionally, we introduce Duolun and other study regions using references in Table 4.

Cui W. L.: Chemical transformation of dust components and mixing mechanisms of dust with pollution aerosols during the long range transport from north to south China,M.S. thesis, Department of Environmental Science and Engineering, Fudan University, 32-38 pp., 2009.

Q11. L135: Explain how the PMF model works.

Reply: We have added the PMF work principle in section 2.

Q12. L139: Explain how Williams' model works and cite the original paper.

Reply: We have added the work principle of the Williams' model and cited the original paper.

Q13. L140. Expand U10. In general, all the abbreviations used should be clearly defined.

Reply: We have added definitions for all the abbreviations in our revised manuscript.

Q14. L143: Meteorological data not "climatic" data.

Reply: The term "climatic" has been changed to "meteorological data".

Q15. L146: This heading does not reflect the text content.

Reply: The heading has been revised to "Other data sources and statistical analysis ".

Q16. L152: Please provide details on the MICAPS information used.

Reply: We have supplemented the details on the MICAPS in section 2.1.

Q17. L164: What is the average TSP concentration on dust days?

Reply: We have added the average TSP concentration (1140.3 $\mu g \cdot m^{-3}$) on dust days in the revised manuscript.

Q18. L167: Please provide a brief description on how EF is calculated and what is the significance of using this method.

Reply: We have provided a brief description of EF and the significance of using EF in section 2.3.

Q19. L171-173: The authors were using MICAPS information of dust storm (which has to be explained) and now are relying on Al levels to define AD events. How are these two definitions consistent?

Reply: First, we defined dust days according to MICAPS information in Qingdao. However, the intensity of a dust event decreases gradually during the long-range transport, and high PM10 episodes of anthropogenic origin (dust is regarded as of natural origin) can be erroneously judged as a dust event in downwind regions. Hsu et al. (2008) proposed the "geometric mean ×2 GSD" of Al concentrations in order to identify major Asian dust (AD) events in downwind regions and obtained good results. Our focus was the impact of long-range transported dust on aerosol samples in downwind area; therefore, we verified the long-range AD using reported GSD criterion in former manuscript. Just as discussed above, we have revised this section and replaced the Al criteria method using the model results in the revised manuscript.

Hsu, S. C., Liu, S. C., Huang, Y. T., Lung, S. C. C., Tsai, F., Tu, J. Y., and Kao, S. J.: A criterion for identifying Asian dust events based on Al concentration data collected from northern Taiwan between 2002 and early 2007, Journal of Geophysical Research Atmospheres, 113, 1044-1044, 2008.

Q20. L178-179: This has to be explained with respect to the EF of the anthropogenic elements.

Reply: We have added the calculation and significance of EF in section 2.3 in revised manuscript.

Q21. L214-216: You need to examine the dust sources, transport pathways (if passing over heavy populated regions), the height at which dust is transported together with dust concentration on a case by case basis to conclude these lines. This is very important for the entire paper. How do you decide "stronger a e "stronger a dust storm"?

Reply: Similar to the reply for Q2, we have revised the section according to the suggestion. In addition, the modeling dust concentration, and transport altitude were supplemented.

Q22. L225-230: The average for Case 1 is 700 µg/m3 with values lying well above the average as is evident from Table 5. Again, there are TSP values in Case 2 in Table 5 which are lower than average of Case 1.

Reply: We have rewritten this section. By comparing the relative values of NH_4^+ and NO_3^- in dust day samples to those in comparison samples, the characteristics of dust day samples were discussed, and a theoretical analysis of the three categories was added.

Q23. L230-231: Again, what is your definition of "strong dust storm"? Without sufficient analysis I am not sure how "dust might be transported quickly" is factoring here. This

entire section has to be revisited.

Reply: According to the suggestion inQ2, we have added modeled dust concentrations over East Asia based on the CFORS model for dust event samples (http://www-cfors.nies.go.jp/) and rewritten the entire section considering whether trajectories passed over highly populated regions.

Q24. L242: How is the value 40.5 km/h arrived at? Is it estimated at the dust source region? How many dust storms were studied to derive this value? More explanation is needed.

Reply: According to Asian dust observations in China and Mongolia(He et al., 2008; Li et al., 2006; Natsagdorjaet al., 2003; Zhan et al., 2009), we estimated an average wind speed threshold as 40.5±9.9 km/h during dust storms. However, according to the suggestion inQ2, we have revised the entire section and discussed the influence of transport on the concentrations of nitrate and ammonium in dust day samples.

He, Q., Wei, W., Li, X., Ali, M., and Li, S.: Profile Characteristics of Wind Velocity, Temperature and Humidity in the Surface Layer during a Sandstorm Passing Taklimakan Desert Hinterland, Desert and Oasis Meteorology, 2, 6-11, 2008.

Li, N., Du, Z. X., Liu, Z. Y., Yang, H. J., Wu, J. D., and Lei, Y.: Change of wind speed and soil moisture during occurrence of dust storms, Journal of Natural Disasters, 15, 28-32, 2006.

Natsagdorj, L., Jugder, D., and Chung, Y. S.: Analysis of dust storms observed in Mongolia during1937–1999, Atmos. Environ., 37, 1401–1411, 2003.

Zhan, K. J., Zhao, M., Fang, E., Yang, Z. H., Zhang, Y. C., Guo, S. J., Zhang, J. C., Wang, Q. Q., and Wang, D. Z.: The wind speed characteristics of near-surface vertical gradient of 50m in sandstorm process in 2006, Journal of Arid Land Resources & Environment, 23, 100-105, 2009.

Q25. L250-252 needs explanation. Once the dust storms are properly categorized and the pathways are determined the interpretation of Table 5 might change

Reply: We have reanalyzed the trajectories and rewritten this section.

Q26. L250: Less than 40.5 km/h not 42.4 km/h.
Reply: We have revised this section.

Q27. L281-283: These statements need to be backed by more analysis of the dust events on a case by case basis.

Reply: We cannot discuss the contributions of dust to aerosols for each dust case because of the limited number of samples. The limited number of samples will result in an unrealistic source apportionment. Thus, we have rewritten this sentence.

Q28. L301-311: The text is very confusing. It is very difficult to follow when the authors are referring to TSP and when they are referring to IN or nitrate or ammonium.

Reply: We have rewritten these sentences. Additionally, we added the IN (now $N_{NH4+NO3}$- in revised manuscript) definition in section 3.6 and the flux of $N_{NH4+NO3}$- in Table 8 and revised several mistakes.

Table 8. Dry deposition of TSP (mg/m^2/month), particulate inorganic nitrogen (mg N/m^2/month) and some toxic trace metals (mg/m^2/month) on dust and non-dust 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] The characterization of$N_{NH4+NO3}$- concentration and sample information of the Category see in Table 3.

Technical corrections:

Q29. L260: Colors used in Figure 4 are not clear. Please indicate the dust source regions on the map.

Reply: We have redrawn Fig. 4 (now Fig. 5 in the revised version) to distinguish the trajectory of each sample collected on dust and non-dust days. The dust source regions in China are also now indicated in this figure.

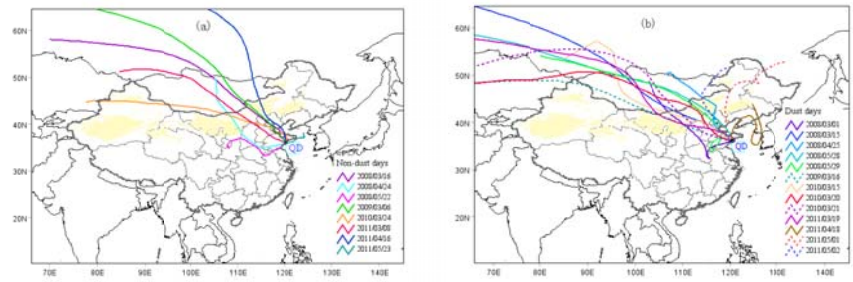


Figure 5.The 72-h backward trajectories for non-dust (a) and dust (b) samples from 2008 to 2011 (the yellow domains in the maps represent the dust source regions in China).

Q30. L343-442: Not proper attention has been given to the References and needs to be corrected.

Reply: We have updated and corrected the reference citations and format.

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

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Abstract. Asian dust has been reported to carry anthropogenic reactive nitrogen during the transport from its source areas~~zone~~ to 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 ~~at a coastal site in the norther China in every spring~~ from 2008 to 2011.~~To understand the impacts of long-range transport onparticulate inorganic nitrogenassociated with dust in downwind areas, aerosol samples were collected in the Qingdao coastal region on dust and non-dust (ND) days in spring from 2008 to 2011. The concentrations of water-soluble ions were measured by ion chromatography, with metal elements detected using inductively coupled plasma-atomic emission spectroscopy (ICP-AES) and inductively coupled plasma-mass spectrometry (ICP-MS).Compared to atmospheric aerosols collected on ND days, samples from dust days exhibited higher concentrations of particles and crustal elements. Total aerosol particle concentrations increased bya factor of 5.9 on averagedust days. On dust days, the average concentrations of crustal elements (Sc, Al, Fe, Ca and Mg) increased by over a factor of four relative to those on ND days.~~ Based on the mass concentrations of NH_4^+ and NO_3^- in each total suspended particle (TSP), the the samples can be classified into two groups: in Category 1, the concentrations of NH_4^+ and NO_3^- were ~~increased by a factor of~~ 20%-440% ~~on~~ higher in ~~some~~ dust day samples relative to samples collected immediately before or after dust event, while in Categories 2 and 3, these concentrations decreased by 10-75% in ~~other~~ the dust day samples. For these two groups, NH_4^+ in dust day samples

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was present in the form of ammonium salts externally co-existing with dust aerosols or the residual of incomplete reactions between ammonium salt and carbonate salts. The NO_3^- in the dust day samples was attributed to interactions between anthropogenic air pollutants and dust particles during dust transport from the source zone to the receptionsite. Back trajectory analysis showed that the concentrations of NH_4^+ and NO_3^-were apparently affected by the transport distance prior to the reception site, the mixing layer depth and the residence time across highly polluted regions during transport.

~~The inorganic nitrogen content increased 1.2 to 9.2-fold during some dust events in which storms were weak or slow moving and reactionsoccurred during transport. By contrast, nitrate and ammonium exhibited very low concentrations (<20% of ND samples)or decreased concentrations in some cases as a result of the strong dilution effect of low-nutrientdust particlesarising from their rapid transport in a strong dust storm. If air massestraveled fasterthan 40.5±9.9 km/h, the inorganic nitrogen content of most aerosol samples decreased compared to that of ND samples because of the strong dilution effect.~~

~~The concentration of atmospheric particulate inorganic nitrogenwas relatedto not onlythe transport path and speed but also the local emissions and reaction conditions during transport.~~ The positive matrix factorization (PMF) receptor model results showed that the contribution of soil dust ~~dramatically~~ increased from 23% to 36% (90% of the residuals<3.0 andr^2=0.97) on dust days ~~,~~with decreasing ~~while the~~ contributions of local anthropogenic ~~sources decreased~~inputs, especially ~~that of~~ secondary aerosols. The dry deposition flux of atmospheric particulates increased from 2,800±700 mg/m^2/month on non-dust comparison ~~ND~~ days to 16,800±15,900 on dust days. The dry deposition flux of particulate inorganic nitrogen increased by ~~1.1~~9-285% ~~to 5.8-fold under~~ in Category 1~~theweak dilution effects of dust events~~. Relative to the comparison samples, ~~The~~ the average dry deposition flux of nitrate decreased by 46%-73%~~by 46%-63%~~in Category 2, while that of ammonium decreased by ~~1~~47% in Category 3,~~orto ND levels whenstrong dilution occurredduringdust events.~~ The estimated dust deposition flux varied greatly from event to event. Overall, a slight increase in dry deposition flux of particulate inorganic nitrogen associated with dust events in this study relative to values in the literature may reflect the combined effect of anthropogenic nitrogen emissions and the occurrence of natural dust events. ~~atmospheric input of nitrogen tothe oceanwasnot enhanced by dust events, and dust deposition was an uncertain source of nitrogen to the ocean.~~

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

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1 Introduction

~~Nitrogen is a key element for marine phytoplankton growth.~~ Reactive nitrogen carried in dust particles can be transported over ~~a~~ long distances, and the nitrogen deposition ~~deposited to~~ in oceans has been recognized as an important external source of ~~N~~nitrogen to supporting phytoplankton growth ~~across vast distances at high wind speeds~~ (Duce et al., 2008; Zhang et al., 2010b). The~~is~~ hypothesis has been evaluated in incubation experiments, ~~and on-site~~ in situ experiments, and through the use of satellite observational data ~~Additionally, bioavailable nutrients, via dust particle deposition, may enhance phytoplankton growth in some ocean areas~~ (Shi et al., 2012; Guo et al., 2012; Liu et al., 2013; Tan and Wang, 2014). For example, Tan and Wang (2014) found that a phytoplankton bloom with a nearly four-fold increase in chlorophyll concentration occurred 10-13 days after dust deposition ~~with an increase of chlorophyll concentrations increased by four-fold, and a phytoplankton bloom occurred 10-13 days after dust deposition~~. In addition, Banerjee and Kumar (2014) hypothesized that dust-induced episodic phytoplankton blooms are important to the interannual variability of chlorophyll in the Arabian Sea ~~away from active winter convection~~. Dramatic changes have occurred in the reactive nitrogen in anthropogenic emission in the last three decades, e.g., large increases in NH_3 and NO_x in emissions in China and other developing countries in Asia and a substantial decrease in emissions in Europe (Grice et al., 2009; Liu et al., 2017; Ohara et al., 2007; Skjøth and Hertel, 2013). These changes may greatly affect the nitrogencarried by dust particles, but few recent studies have examined this issue.

Asian dust is ~~,~~ one of the main components of dust worldwide~~, affects northern China and the eastern area of the China Sea~~. Asian dust has been reported to cross over the mainland and the marginal seas of China and reach as far as the remote ~~can also affect the~~ northern Pacific Ocean (Zhang and Gao, 2007; Tan and Wang, 2014) ~~via long-range transport by west winds~~. During the long-range transport, ~~the~~ dust ~~storm~~ particles may ~~continually~~ mix with anthropogenic gas and particles, ~~and~~ consequently ~~occur~~resulting in complicated ~~chemcial~~chemical reactions (Cui et al., 2009; Wang et al., 2011; Wang et al., 2015, Wang et al., 2017). However, the extent of these chemical reactions varies widely and depends on the meteorological conditions, such as cloud fraction, wind speed, relative humidityand

atmospheric circulation (Yang et al., 2002; Li et al., 2014; Ma et al., 2012). ~~from local sources along thepathway to carry desert dust and anthropogenic aerosols to downwind areas. The physical and chemical characteristics of atmospheric particulates downwind of dust weather are remarkably different from those normallypresent (Yang et al., 2002; Li et al., 2014; Ma et al., 2012).Additionally, the concentrations and characteristics of atmospheric particulate inorganic nitrogen species in downwind areas are greatly affected by dust weather.~~

For example, a few studies have ~~showedn that Some researchershave found~~ that ~~inorganic nitrogen species in aerosols havehigh concentrationsduring Asian dust events. T~~the concentrations of atmospheric particulate NO_3^- and NH_4^+ on dust storm days were 2-5 times ~~higher~~ larger than ~~those on~~ those ~~non-dust storm (ND)~~ days prior to the events in Beijing (Liu et al., 2014; Liu and Bei, 2016). Xu et al. (2014) also reported ~~found~~ that concentrations of particulate SO_4^{2-} and NO_3^- were simultaneously increased ~~in~~ during dust storm events ~~on in~~along the northern boundary of the Tibetan Plateau ~~because of the enriched dust including more acidic species or anthropogenic aerosols. Compared to those on ND days, higher concentrations of NO_3^- and NH_4^+ in aerosol particles were observed on dust storm days in northern China, and NO_3^- and NH_4^+showed lower concentrations during strong dust storm events than during weak dust events (Zhang et al., 2010).~~ Fitzgerald et al. (2015) found that almost ~~nearly~~ all Asian dust observed in Korea ~~containscontained~~ considerable amounts of nitrate and proposed that ~~because pollution plumes mix with dust~~ the dust from the Gobi and Taklamakan Deserts probably mixed and reacted with anthropogenic air pollutants during the transport ~~and are transported~~ over the Asian continent. Although increased concentrations of NO_3^- and NH_4^+ in aerosol particles were observed on dust storm days in the northern China relative to ~~Compared to~~ those ~~on Nd~~non-dust days prior to the dust storm events~~:~~,

Zhang et al. (2010a) also found that the concentrations of the two species were associated with the intensity of the dust storm, i.e., the stronger dust storms corresponded to the smaller increases. In other words, lower NO_3^- and NH_4^+ concentrations occurred during strong dust storm events than during weak dust events (Zhang et al., 2010a).

~~higher concentrations of NO_3^- and NH_4^+ in aerosol particles were observed on dust storm days in northern China~~, and NO_3^- and NH_4^+showed lower concentrations during strong dust storm events than during weak dust events (Zhang et al., 2010).

On the other hand, ~~However,~~ some studies ~~reporetd~~reported~~observed that~~ ~~the~~ the reverse result~~the~~

~~concentrations of inorganic nitrogen in aerosols decreased on dust storm days~~. For ~~exmaple~~example, a~~A~~t Yulin, a rural site near the Asian dust source region, the concentration of NO_3^- in atmospheric aerosols on dust days was significantly significantly lower in comparison to the concentration measured immediately before or after the event, as a result of the dilution effect~~decreased, in comaprison to the measured immediately before or after the event, in aerosols on dust days~~ (Wang et al., 2016). Even in Shanghai, a mega city being located at a few thousands ~~kilometers~~kilometers from dust source zones in China, t~~T~~he ~~absolute abundances~~ concentrations of NO_3^- and NH_4^+ were notably lower in the ~~oberved~~observed dust plumes than in a polluted air parcel immediately ~~oberved~~ prior to the dust events (Wang et al., 2013). ~~because dust plumesare often separated by the arrival of a cold front in Shanghai, China (Wang et al., 2013).~~ Li et al. (2014) also found that the concentrations of nitrate and ammonium ~~in downwind aerosol particles~~ ~~were~~ decreased on dust storm days ~~tother,~~ with a decreasing ratio of the total soluble inorganic ions to $PM_{2.5}$ in the Yellow River Delta, China. When dust ~~is~~ was rapidly transported from desert regions without passing through ~~a~~ major urban areas and ~~lingers~~ lingering over the Yellow Sea, the concentrations and size distributions of nitrate and ammonium ~~have~~ had no significant variation in heavy Asian dust (AD) plumes (Kang et al., 2013). ~~Nitrate and ammonium also exhibited different concentration variations in other desert regions. Jaafar et al. (2014) found that nitrate was more abundant than ammonium, which showed no concentration variation in non-dust aerosol particles during dust episodesoriginating from both the African and Arabian deserts.~~

The contradictory results highlight the importance of investigating the concentrations of ammonium and nitrate in atmospheric particles during dust events based on a larger database. ~~-~~ In this study, a~~effect of dust events on the inorganic nitrogen concentration in downwind aerosols is complicated because it involves many factors, such as the mixing state and transport pathway. The effect of AD as a nitrogen source on biogeochemical cycles and marine ecology is not adequately understood. Understanding the variations in the concentration and deposition flux of atmospheric particulate nitrogenon dust days is essential to quantifying the impacts of nutrientson the marine environment and primary production. Tounderstand the influence of dust on atmospheric nitrogen,~~ we collected atmospheric aerosol ~~samples~~ particles during and prior to (or ~~post~~after, when no sample was collected prior to) dust events ~~from~~ at a coastal site ~~in the promixty~~adjacent to~~the coastal region of~~ the Yellow Sea ~~in~~ during the~~every~~ spring from 2008 to 2011~~,~~ when ~~a~~ smaller outbreak peak ~~there is a high frequency~~ of dust storms occurred ~~from 2008 to 2011~~. ~~Then, w~~We measured ~~analyzed~~ concentrations of the inorganic nitrogen

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~~concentrations~~in the ~~aerosol~~ samples as well as other components for facilitating analysis. We first characterized ~~The~~ the concentrations of inorganic nitrogen concentrations in various dust events relative to the concentrations in samples collected either prior to or after the events. We then conducted , source apportionment to quantify their sources. Finally, we calculated ~~and~~ deposition flux of atmospheric particulate inorganic nitrogen ~~in~~ during dust events and compared the results with the values in theliterature in order to update the flux values due todynamic changes inanthropogenic emissions and other factors.

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2 Experimental ~~Materials and~~ methods

2.1 Sampling

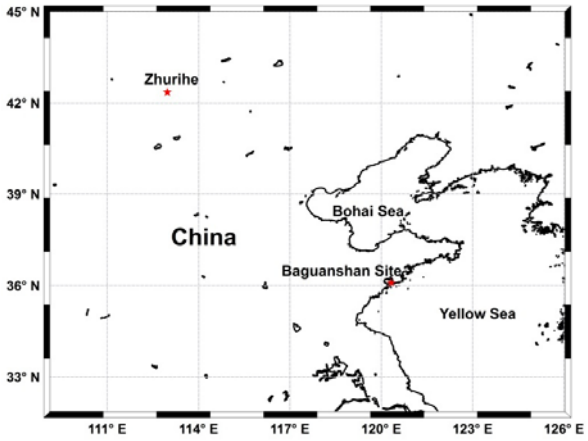
~~As shown in~~ Fig. 1 shows the sampling site, which is situated at the top of a coastal hill (Baguanshan) in Qingdao in northern China (36° 6' N, 120° 19' E, 77 m above sea level)and is approximately 1.0 km from the Yellow Sea tothe east.Ahigh-volume air sampler (Model KC-1000, Qingdao Laoshan Electronic Instrument Complex Co., Ltd.) was set up on the roof of an two-story office buildingto collect total suspended particle (TSP) sampleson quartz microfiber filters~~showed the sampling site, which is situated at the top of a coastal hill(Baguanshan) in the northern China (36° 6' N, 120° 19' E, 77 m above sea level)and approximately 1.0 km from the Yellow Sea in the east. , total suspended particles (TSP) were collected atthe Baguanshan site in the coastal region of the Yellow Sea.The samples were collected with quartz microfiber filters~~ (Whatman QM-A) at a flow rate of 1 m^3/min. Prior to the sampling, the filters were heated at 450°C for 4.5 h 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 Meteorological Information Comprehensive Analysis and Process System (MICAPS) of the China Meteorological Administration.Each dust sample was collected over4 hrs,and the sampling started only when the PM_{10} and dust mass concentration available on the website (http://www-cfors.nies.go.jp/~cfors/;http://www.qepb.gov.cn/m2/) hadincreased greatly. Thisapproach made the dust sample more representative relative to urban background. For dust events with durations of less than one day, only one sample was collected. For dust events with durationsgreater thanone day, a4-hr dust sample was collected once perday. Table 1 lists the sampling information. Based on the forecast, we also collected aerosol particle samples immediately before orafter the dust event for comparison. These comparison samples were further classified into sunny day samples, cloudy day samples and post-dust samples. The post-dust samples were collected under clear and sunny weather conditions and low PM_{10}mass concentrations.

~~usinga Ahigh-volume air sampler (Model KC-1000, Qingdao Laoshan Electronic Instrument Complex Co., Ltd.)with a flow rate of 1 m^3/minwas set up on the roof of an two-story office buildingforcollecting total suspended particles (TSP)on quartz microfiber filters (Whatman QM-A)with~~

~~a flow rate of 1 m^3/min(36° 6' N, 120° 19' E, 77 m above sea level) approximately 1.0 km from the shore.Prior to the sampling, The the filters were heated at 450°C for 4.5 h to remove organic compounds. Moreover, the concentration of Alreferring to the total Al concentration in TSPsamples were used to confirm the division of dust or comparison samples according to the criterion "geometric mean×2GSD" proposed by Hsu et al. (2008).~~

Since Asian dust events at the sampling site mostlyoccurin the spring, we collected samples duringevery spring, i.e., from March to May,from2008-2011. A smaller peak inAsian dust was observed in eastern China in 2008-2011, which followeda larger peakin 2000-2003 duringthis century (Fig. 6).Overall, atotal of 14 sets of dust samples and 8 sets of comparison samples were collected for this study.

To facilitate the coastal sampling data analysis, t~~T~~he sand samples were collected at the Zhurihe site (42°22'N, 112°58'E) in the Hunshandake Desert, one of main Chinese sand deserts, in April 2012. ~~After s~~Sand samples were packed in clean plastic sample bags~~, the samples~~ and were stored below -20°C before the transfer. An ice-box was used to store the samples ~~which were~~during transported to the lab for chemical analysis.



~~**Figure 1.**Location of the aerosol and dust sampling site.~~

2.2 Analysis

The aerosol samples were allowed to achieve equilibrium in a air-conditioned chamber at a constant relative humidity and temperature for 24hrs ~~balanced in a air-condition relative humidity and temperature-controlled chamberat a constant relative humidity and temperature for 24 hrs until the particle~~before weight~~s remained constant~~ing. ~~The mass concentrations of TSPwere calculated~~

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~~according to the particlemassesand the sampling volume.~~The sample membranes were then cut into several portions for analysis.

One portion of each aerosol sample ~~and 0.1 g of parallel sandsample were~~ was ultrasonically extracted with ultra-pure water in an ice water bath, and the concentration of inorganic water-soluble ions was determined ~~via~~ using ICS-3000 ion chromatography (Qi et al., 2011). The ~~parallel~~ sand samples collected at the Zhurihe site~~from the Hunshandake Desert~~ were analyzed using the same procedure. We refer to dissolved inorganic nitrogen (DIN),thesum of nitrate and ammonium, in the later discussion due to the very low concentration of nitrite in the samples.

One portion of each aerosol filter was cut into 60 cm^2 pieces and digested with HNO_3+$HClO_4$+HF (5:2:2 in volume) at 160°C using an electric heating plate. A blank membrane was also analyzed using the same procedure to ensure analytical precision. Cu, Zn, Cr, Sc and Pb were measured ~~by~~ using an inductively coupled plasma-mass spectrometry (Thermo X Series 2), while Al, Ca, Fe, Na and Mg were detected ~~by~~ using an inductively coupled plasma-atomic emission spectroscopy (IRIS Intrepid II XSP). The membrane blanks have been corrected for in the calculation of the.~~The membrane blanks have beeb deducted to calculate t~~The metal concentrations ~~were determined by calibrating the measured concentrations of samples using m~~for analysis below.~~embrane blanks~~.

~~**Table 1.**Detection limits, precisions and recoveries of water-soluble ions and metal elements.~~

~~Component~~	~~Measurement method~~	~~Detection limit ($\mu g \cdot L^{-1}$)~~	~~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~~

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~~Al~~	~~ICP-AES (Lin et~~	~~7.9~~	~~0.6~~	~~103~~
~~Ca~~	~~al., 1998)~~	~~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~~

One portion of each aerosol sample was digested by adding ~~with~~ HNO_3 solution (10% HNO_3, 1.6 M) at 160°C for 20 min in a microwave digestion system (CEM, U.S.). 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 ~~1~~2.

2.3 Computational modeling

The enrichment factor of metal elementswas given by

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

where subscripts i and Re refer to the studied metal and the reference metal; (Xi/XRe)aerosols is the concentration ratio of metal i to metal Re in the aerosol samples; and (Xi/XRe)crust is the ratio of metal i to metal Re in the Earth 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'scrust given by Taylor (1964) was adopted.

~~To determinethe origins of sampled air masses, t~~The 72 h air mass back trajectories were calculated for each TSP sample using TrajStat software (Wang et al., 2009) and the NOAA GDAS archive data (http:// www.arl.noaa.gov/ready/hysplit4.html). The air mass back trajectories were calculated at an altitude of 1~~0~~500 m to identify the dust origin.

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

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by a matrix X of i by j dimensions, in which i number of samples and j chemical species were measured, with uncertaintyu. 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. ~~receptor modeling method (Paatero and Tapper, 1993; Paatero, 1997)~~

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

where Eij is the residual for species j of the i-th sample.

The aim of the model is to minimize a 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.

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

EPA PMF 3.0 model was used to obtain the source apportionment of atmospheric particulates on dust and comparison~~ND~~ days. The correlation coefficient between the predicted and observed concentrations was 0.97.

Dry deposition velocities were obtained using Williams' model (Williams, 1982)~~and~~ by accounting for particle growth (Qi et al., 2005). Williams' model is a two-layer model to calculated the dry velocity of size-segregated particle over the water. At upper layer below a reference height (10 m), the deposition of aerosols particles is governed by turbulent transfer and gravitational settling. In the deposition layer, gravitational settling of particles is affected by particle growth due to high relative humidity. To obtain the deposition velocity of different particle size, Williams' model need many input parameters, such as the wind speed at 10m height (U10), air/water temperature, relative humidity. Relative humidity, air temperature and U10 obtained from the National Centers for Environmental Prediction (NCEP)were used in ~~the model as the meteorological inputs~~this study. Surface seawater temperature was collected from the European Centre for Medium-Range Weather Forecasts (ECMWF).The meteorological~~climatic~~ 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 ~~ND~~ comparison days.

The CMAQ model (v5.0.2) was applied over the East Asia area to simulate the concentrations of PM10, NOx and NH3for 14 samples collected during11 dust events.The simulated domain contains 164×97 grid cells with a 36 km spatial resolution. The vertical resolution includes 14 layers from surface to tropopause, with the first model layer height of 36m above the ground level. The

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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. The emissions of NOX and NH3over East Asia for each dust event were also modeled using the CMAQ model according to the emission inventory in 2008, which was generated by extrapolating the 2006 activity data 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. Validation of the application of the CMAQ model in China has been reported by Liu et al. (2010a, b). The spatial distribution of PM10 concentrations for each dust event was consistent with the model results of dust by the Chemical Weather Forecast System (CFORS) by Uno et al. (2003)~~he Chemical Weather Forecast SystemCFORS~~.

2.4 ~~Statistical analysis~~Other data sources and Statistical analysis

Meteorological data were obtained from the Qingdao Meteorological Administration (http://qdqx.qingdao.gov.cn/zdz/ystj.aspx) and the ~~Meteorological Information Comprehensive Analysis and Process System (~~MICAPS~~)~~ of the Meteorological Administration of China. ~~NO_2 and air quality index (AQI) data were downloadedfrom the Qingdao Environmental Protection Bureau (http://www.qepb.gov.cn/m2/).~~ Different weather characteristics, such as sunny days, cloudy days and dust days, were defined according to information of MICAPS ~~information~~and Qingdao Meteorological Administration. The Meteorological Information Comprehensive Analysis and Process System (MICAPS)~~he Chemical Weather Forecast SystemCFORS~~.

According to the altitude, longitude and latitude of the72 h air mass back trajectoryof each dust sample, thepressure level, temperature and relative humidity data along the path of theair 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 sampleswasobtained from the HYSPLIT Trajectory Model (http://ready.arl.noaa.gov/hypub-bin/trajasrc.pl) using the same method. 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.

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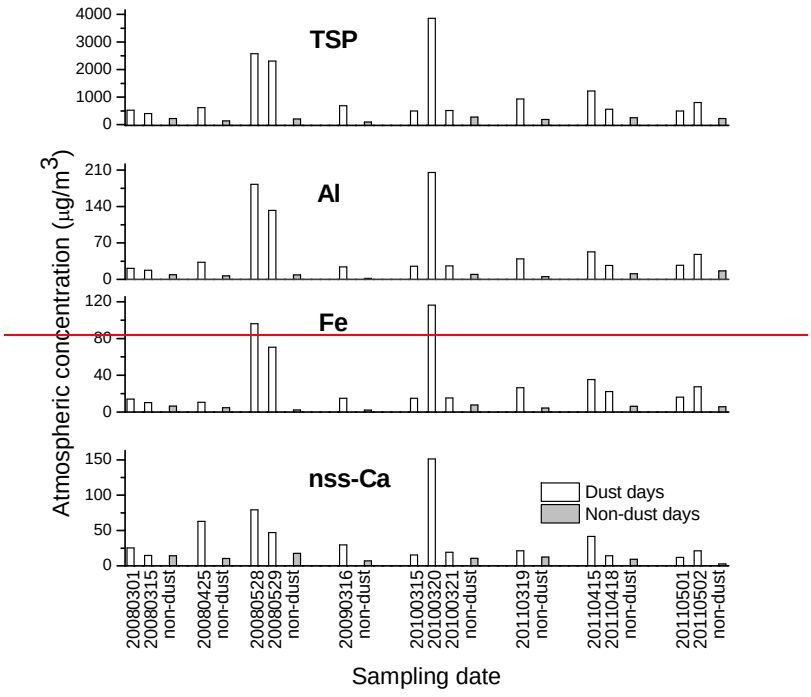
3 Results and discussion

3.1 Characterization of aerosol samples collected during dust events

To support our analysis, the dust intensity and influence range of thedust events modeled byUno et al. (2003) were analyzed, and the spatial distributions of dust concentration over East Asiaduringeach dust sampling day are shown inFig. 2and Fig.S3.Almost alldust events originated innorthern or northwestern Chinaand passed over thesampling site. However, Sample 20110415was judged to belocal blowing dust because no correspondinghigh dust concentrations were observed in the dust source areas; this sample was thereforeexcluded from furtheranalysis.



~~**Figure 2**~~~~3.Concentrations of TSP, Al, Fe and nss-Ca in aerosol samples collected in the coastal region of the Yellow Sea on non-dust and dustdays from 2008 to 2011.~~

Before characterizing the inorganic~~To understand the impact of dust events on atmospheric particulate~~ nitrogen in atmospheric particles at Baguanshan site,~~downwind areas, we collected aerosol samples in the coastal region of the Yellow Sea in the spring from 2008 to 2011.W~~we first examined

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the mass concentrations of ~~particles~~ TSP~~and~~ as well as concentrations of crustal and anthropogenic metals to compare samples collected on dust days and immediately before or after days. ~~in aerosol samples. Although metal concentrations in aerosols collected on different dust days varied, some characterizations~~ The comparative results are highlighted below. ~~The concentration of atmospheric particulate increased on dust days. On non-dust (ND) days~~For these comparison samples, ~~aerosol particles~~ the TSP concentrations ~~varied in the~~ range~~d of~~ from 94 to~~-~~ 275μg·m^{-3}, with an average of 201 μg·m^{-3} (Fig. 2). The TSP mass concentration ~~Particle concentrations~~were increased substantially ~~increased~~ to 501-3857μg·m^{-3}~~on~~ in dust day samples, with an average of 1140.2 μg·m^{-3}. In each dust day-comparison day sample pair, the mass concentration of ~~The~~ TSPs increased by ~~concentrationon dust days was 1-~~80-~~14.0~~1303% with the median value of 537%~~times(mean: 5.9) higher thanthat on ND days~~. ~~The~~ A similar increase was present in the crustal elements ~~increased considerably with the increasing particle concentrations when dust events occurred~~in each pair of samples. For example, the mean concentrations of Sc, Al, Fe, Ca and Mg were increased by over a factor of four in dust day samples relative to the comparison samples. In addition, ~~As shown in Table 2,~~ the enrichment factors (EF) of Al, Fe, Ca, and Mg were less than three in dust day samples but ~~lower~~ less than ~~ten~~ 14~~on ND days~~in comparison samples ~~and decreased to less than three on dust days~~in each pair (Table 3). ~~These~~ lower ~~data~~ values are indicative of these elements from the the primar~~il~~y crustal origin~~s of these elements~~. ~~We found that the mean concentrations of Sc, Al, Fe, Ca and Mgincreased by over a factor of four as compared to those on ND days. Al concentrations in dust weather increased 1.7 to 21.9-fold (mean: 6.9) on ND days. The Al concentration of the "geometric mean×2GSD" (where GSD is the geometric standard deviation) was used as a criterion to define major AD events in areas of East Asia (Hsu et al., 2008). Al concentrationswere higher than the criterion level in all dust samples, which indicated that the samples we collected on dust days were truly affected by dust events. Fe was 10.3 times higher on dust days than on ND days. Additionally, nss-Ca, a typical dust index, increased 3.6-fold on dust days (Fig. 2). The EF of the anthropogenic metal elements decreased on dust days. Cu, Pb, Zn, Cr, Hg and As had high EFs,much greater than 10,on ND days, which indicated that these metal elements were mainly from anthropogenic sources.~~ Although ~~The~~ the average mass concentrations of ~~these~~ anthropogenic elements such as Cu, Pb, Zn, Cr, Hg and As~~on~~ in dust day sample ~~–~~increased 1.~~1~~07% to 7.~~2-fold~~22% ~~on average~~relative ~~compared~~ to those in comparison samples, the EF of the anthropogenic metal elements decreased in the dust day samples~~on ND days~~.~~Additionally, the EFs of~~

these anthropogenic elements decreased on dust days. These data are consistent with the very low EFs of these elements in dust particles. ThusThis pattern, indicated a decreasing relative contribution of the influence of anthropogenic sources on atmospheric particulates decreasedto the total TSP mass on in dust day samples.

Table 2. Theaverage 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

*The EF factor was calculated using Scandium as the reference element (Han et al., 2010).

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3.2 Concentrationsdistribution of inorganic nitrogen in dust eventsday samples

As discussed above, the concentrations of TSP and metal elements increased on dust days compared to those on ND days. However, When the mass concentrations of inorganic nitrogen species NH_4^+ and NO_3^-in each pair of TSP samples were compared, exhibited different variations in particles and crustal metal elements.Tthe concentrations of ammonium NH_4^+ increased by a factor of 1.220%-5.747oin some dust day samples (20080301, 20080315, 20090316, 20100315, 20100320, 20100315. 20110415 and 20110418), but slightly or greatly and decreased by 40-85%or had a very low

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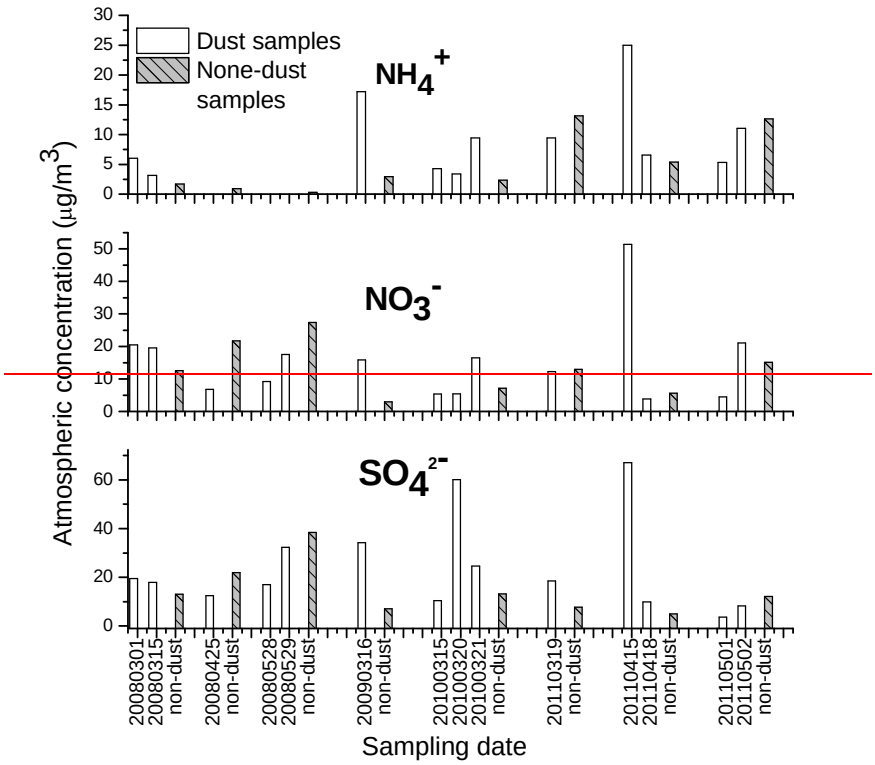
~~concentration (less than 20% of that on ND days) o~~in other dust day ~~s~~amples (Fig. 3, Table S2). The same was general true for the measured concentration of NO_3^-. ~~Similar to ammonium, nitrate displayed two different concentration variationson dust days. Nitrate concentrationsincreased by a factor of 1.4-9.2on some dust daysand decreased on other dust days.The secondary inorganic ionSO_4^{2-} exhibited concentration variations that were similar to those ofnitrate. Therefore, the influence of dust on these secondary ions was different from that on crustal metal elements, andthe effect of dust on inorganic nitrogen differed during different types ofdust events.~~



~~**Figure 34. Mass concentration of** NH_4^+, $^+$ and$NO_3^-$and SO_4^{2-}in aerosol samples collected at the Baguanshan site on dust and comparison days during March – May in the coastal region of the Yellow Sea on non-dust and dust days from 2008 to 2011.~~

Considering the relative values of NH_4^+ and NO_3^- in dust day samples relative to comparison samples, the dust day samples can be classified into three categories (Table 4). In Category 1, the mass concentrations of NH_4^+ and NO_3^- were larger in dust day sample than in the comparison samples. In

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Category 2, the reverse was true. In Category 3, the mass concentrations of NO_3^- were lower in the dust samples than in the comparison samples, whereas the reverse was true for NH_4^+. Considering that the Yellow Sea was mainly affected by dust storms from the Hunshandake Desert (Zhang and Gao, 2007), we compared our observations with the sand particles collected from this desert (Table 5). The relative mass concentrations of nitrate and ammonium to the total mass of sand particles were very low, i.e., less than 81μg/g, approximately three orders of magnitude less than the corresponding values in our dust samples. Moreover, the values obtained from atmospheric aerosols at Duolun (Cui, 2009) and Alxa Right Banner (NiuandZhang, 2000) were also more than one order of magnitude lower than the corresponding values in this study (Table 5). This suggested that NO_3^- and NH_4^+ observed in the dust day samples were very likely due to interactions and mixing between anthropogenic air pollutants and dust particles during transport from the source zone to the reception site (Cui et al., 2009,Wang et al., 2011; Wu et al., 2016). However, along the different transport paths of Asian dust, air pollutant emissions, meteorological conditions, chemical reactions, and other factors can affect the abundance of NH_4^+ and NO_3^- in atmospheric particles. These factors canvary greatly among different dust events, hence leading to the three different categories.

~~Considered relative values of NH_4^+ and NO3- in dust day sample against in comparison sample, these dust day samples can be classified into three categories When we focused on inorganic nitrogen (IN), we found that IN concentrations could be grouped into three cases (Table 3). InN Category 1, the mass concentrations of NH_4^+ and NO_3^- were larger in dust day sample than in comparison sample.In Category 2, the reverse was true. In Category 3, the massconcentrationof NO_3^-swere less in dust samplehigher on dust days than on in comparison sampleND days for Case 1, while IN was loweron dust days for Case 2. For Case 3, nitrate concentrationson dust days were less than on ND days, while ammonium concentrationson dust days were slightly higher than those on ND days the reverse was true for NH_4^+.~~

~~Considering that To understand the influence of dust on the nitrogen concentration, we compared the IN content in aerosols from the coastal regionof the Yellow Sea with sand particles and atmospheric aerosols from Duolun, a site very close to the Zhurihe Sand Desert. The Yellow Sea is was mainly affected by dust storms from the Zhurihe Sand desertthis sand source (Zhang and Gao, 2007).), we compared our observation with those sand particles collected at the desert (Table 4). The relative mass concentrations of nitrate and ammonium to the total mass of sand particles were much low, i.e., less~~

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~~than 81μg/g, which were three orders of magnitude less than the corresponding values inherently with our dust samples. Moreover, the values obtained from atmospheric aerosols at Duolun, a site very close to the Zhurihe Sand Desert, in literature (Cui et al., 2009), were also more than one order of magnitude less than the corresponding values in this study (Table 4). This suggested that NO_3^- and NH_4^+ observed in the dust day samples were related to interactions of anthropogenic air pollutants and dust particles during the transport from the source zone to the reception site (Zhang et al., 2016; Cui et al., 2014,). However, along the different transport paths of Asian dust, air pollutants' emissions, meteorological conditions, chemical reactions, etc., can affect the abundance of NH_4^+ and NO_3^- in atmospheric particles. These factors could vary a lot in different dust events, leading to three different categories.~~

3.3 Theoretical analysis of three categories

Ammonium salts are common in atmospheric particles with diameters of less than 2 μm (Yao et al., 2003; Yao and Zhang, 2012). Gas-aerosol thermodynamic equilibrium is widely assumed to be fully attained for inorganic ions, including ammonium salts in $PM_{2.5}$, in all regional air quality modeling studies. Reasonably good agreements between ammonium salt modeling results and observations reported in literature support the validity of this assumption (Chen et al., 2016; Penrodet al., 2014; Walker et al., 2012). Assuming that 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, much high concentrations of Ca^{2+} wereobserved, and high $CaCO_3$concentrations were therefore expected. When Category 1 was considered alone and one exterior sample was excluded, a good correlation however, was obtained for $[NH_4^+]_{\text{equivalent concentration}} = 0.98 * [NO_3^- + SO_4^{2-}]_{\text{equivalent concentration}}$ ($R^2 = 0.83$, $P < 0.05$). 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^+ in these dust day samples. The formation of $CaSO_4$ and/or $Ca(NO_3)_2$ was probably negligible. Thus, ammonium salt aerosols may externally co-exist with dust aerosols in these dust day samples. In the exterior sample collected on 21 March 2010, $[NH_4^+]$ only accounted for ~70% of the observed $[NO_3^- + SO_4^{2-}]$ in equivalent concentration. This result suggested that ~30% of $(NO_3^- + SO_4^{2-})$ may be associated with dust aerosols viathe formation of metal salts of the two species. The 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). Note that only samples in Category 1 showed NH_4^+ to be negatively correlated with Ca^{2+} (Fig.S4).

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 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), implying that $NO_3^-+SO_4^{2-}$ existed as metal salts. The average concentration of Ca^{2+} in Category 2 ($0.43\pm0.40\ \mu g/m^3$) was clearly higher than that in Category 1 (Ca^{2+}: $0.17\pm0.04\ \mu g/m^3$), implying the probable formation of $CaSO_4$ and/or $Ca(NO_3)_2$ and the release of NH_3 (gas), resulting in a decrease in NH_4^+. However, the concentration of total Ca was $1.11\pm0.70\ \mu g/m^3$ in Category 1 and $0.74\pm0.49\ \mu g/m^3$ in Category 2. In Category 1, NO_3^- was negatively correlated with SO_4^{2-} (Fig.S4), suggesting competition for NH_3 under NH_3-poor dust days during long-range transport. However, NO_3^- was positively correlated with SO_4^{2-} in Category 2. The latter relationship can be explained by the fact that the amount of $CaCO_3$ was sufficient to absorb the precursors of both SO_4^{2-} and NO_3^-. Due to the absence of TSP concentration data along the transport pathway, we compared TSP concentrations at the sampling site and found that the average value of Category 2 ($1391\pm981\ \mu g/m^3$) was substantially higher than that of Category 1 ($591\pm158\ \mu g/m^3$). This implied that dust events in Category 2 were even stronger. Note that the NO_2 concentrations in Category 2 ($1.35\pm2.45\ \mu g/m^3$) were lower or comparable to those in Category 1 ($1.51\pm2.16\ \mu g/m^3$). The potential formation of nitrate metal salts was expected to be similar between the two categories, while favorable formation conditions for ammonium nitrate greatly increased the mass concentrations of nitrate and the contributions to the TSPs in Category 1.

Overall, the higher ammonium concentrations observed in the dust day samples in Category 1 were likely associated with external co-existence of ammonium salt aerosols. However, the lower concentrations in Category 2 were likely due to unfavorable conditions for forming ammonium salts. The observed ammonium was just the residual of incomplete reactions between preexisting ammonium salt and carbonate salts. More discussion on this issue will be presented in Section 3.4.

~~From Table 4, we found that nitrate and ammonium concentrations in the source sand particles were~~

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very low (less than 50 μg/g). Therefore, the dust particlesin this th source area that affectthe Yellow Seaarenutrient poor.Although the IN content in aerosols particles at Duolunin dust days was higher than that th in sand particles, the nitrate and ammonium concentrations were much lower than osein the coastal region of the Yellow Sea.Other researchers also found the nitrate concentration in dust particles in downwind area higher than that at source region (Zhang et al., 2016; Cui et al., 2014). Therefore, we believe that the dust particles from the source have a dilution effect on atmospheric particulate nitrogen because of the low IN concentration in sand particles, if the dust particles just mixing with anthropogenic particles and no effective heterogeneous reaction on dust particles during the transport. When dust events occurred and no effective formation of nitrate and ammonium reaction happen, the content of nitrogen per particle mass decreasedbecause of the dilution of particulate nitrogen resulting from theincreasednumber of nutrient-poor dust particles rapidlyleaving the source area, . And we really found some samples in Case 2, such as 20080425, 20080528, 20080529, 20110319 and 20110501, had a lower IN content in aerosols particle on dust days thanthatonND.The nitrate concentration decreased 30-95% and ammonium decreased 16-72% for these samples. We think these dilution effect resulted by mixing is related with dust intensity of dust events and the distance from the dust source. In the region close to the dust sources, the IN content was very low in aerosols in dust days due to IN poor dust particlesandno effective emission or absorption and reaction occur during such short transport time. However, during long range transport, the concentration of IN will increase by reacting with gas emitted into the air under appropriate reaction conditions or by mixing withanthropogenic aerosol particles from a local source. Therefore, IN concentrations will increase in aerosols in downwind areas because of reactions on the dust surfaces or mixing with anthropogenic particles along the transport path. Cui et al. (2009) observed one dust event at four sites on transport path (Duolun, Beijing, Taishan, Shanghai), and found the enhancement and complex of mixing of dust with anthropogenic particles during transport from source to downwind region. In addition, the dilution effect was affected by dust intensity. For our samples in Case 2, Sample 200805028corresponded to a severe dust storm; 080529 and 110501 was related a dust storm; 110319 was a blowing dust (Table 3). IN concentration decreased by 66-84%, 36-71% and 5-28% for samples collected on severe dust storm, dust storm andblowing dust, respectively. The results showed The the stronger a dust storm is and the closer to the source, the stronger the dilution effect is.

Table 34. Average concentrations of inorganic nitrogen, TSP, NOx, Relative Humidity (RH) and T for

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eachcaseinaerosolsamplesin the coastal region of the Yellow Sea

	Sample numbers	TSP $\mu g \cdot m^{-3}$	NO_3^- $\mu g \cdot m^{-3}$	NH_4^+ $\mu g \cdot m^{-3}$	RH %	T °C	NOx $\mu g \cdot m^{-3}$	Summary
Case 1	080301, 080315, 090316, 100321, 110415, 110502	696	24.1	14.9	46.6	13.8	62.7	IN>ND
Case 2	080425, 080528, 080529, 110319, 110501	1199	3.1	2.6	29.2	19.8	52.3	IN<ND
Case 3	100315, 100320, 110418	1639	4.9	4.7	10.1	10.1	70.7	NO_3^-<ND NH_4^+≅ND
Non-dust		212	5.5	4.6	42.2	13.7	59.7	

Therefore, IN concentrations will increase in aerosols in downwind areasbecause of reactions on the dust surfacesor mixing with anthropogenic particles along the transport path. If no effective emissionor

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absorption and reaction occur during transport, theIN content per particle mass (μg/g) will decrease in atmospheric aerosols in the downwind area. In Case 1, the particle concentration in the dust events wasless than 700 μg/m^3, and the IN concentration in atmospheric aerosols increased by a factor of more than three in dust events in the coastal region of the Yellow Sea, which might be a result of slow transport or a weak dilution effect. High relative humidity (RH), low temperature and high NO_xtransport over a long distance and at a low speed arebeneficial to the formation of nitrate and ammonium. In Cases 2 and 3, the particle concentrationswere very high, with an average higher than 1100μg m^{-3}, which indicatedthat the samples were affected by a strong dust storm or that the dust might be transported quickly. Concentrations of IN in aerosols in dust events decreased at the downwind site in Case 2 because the low RH, high temperature and low NOxduring rapid transport were not advantageous to the formation of IN. In Case 3, the low IN content was a result of a strong dilution effect and low RH.

In addition, the transport path affected the IN content of aerosol particles in the downwind area, and this influence will be discussed in Section 3.3.

Table 45. Comparison of the IN content in dust particles according to the dust source region (unit: μg/g)

Sands sampled in Zhurihe		Aerosols in Duolun*		Aerosols in the coastal region of the Yellow Sea	
NO_3^-	NH_4^+	NO_3^-	NH_4^+	NO_3^-	NH_4^+
25.46±22.87	4.21±1.03	1200	900	Non-dust:	Non-dust:
				28,200±24,819	24,063±21,515
				Case 1:	Case 1:
				34,892±9570	22,571±7016
				Case 2:	Case 2:
				5542±5117	4758±5698

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~~Case 3:~~	~~Case 3:~~	
~~6359±4697~~	~~7059±5591~~	

~~* Adapted from Cui (2009)~~

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3.~~3~~ 4Influence of transport pathways on particulate inorganic nitrogenin dust samples

The calculated air mass trajectoriesof 13 out of 14 samplesshowed that the air mass originatedfromInner Mongolia, China (Fig. 5), generally consistent with the results by Zhang and Gao (2007). Theremaining one originated from Northeast China. Figs. 6 and 7 show a few areas with high emissions of NO_x and NH_3, e.g., Liaoning, Beijing-Tianjin-Hebei, Shandong, Henan and Jiangsu in China. The calculated trajectories showed that all the air mass passed over parts of these highly polluted regions and experienced different residence time in these regions. In Fig. 5, except for the one exterior sample, all trajectories in Category 1 showed that the air masses were transported from either the north or northwest over the continent. In Category 2,the air massescrossed over the sea for 94-255 km prior to arriving at the reception site. NH_3-poor conditions in the marine atmosphere disfavored the formation and existence of ammonium nitrate. On the other hand, the humid marine conditions might have enhanced particle-particle coagulation and might have led to the release of NH_3 via reactions between preexisting ammonium salts and carbonate salts. Moreover, we also examined the links among the measured concentrations of particulate ammonium and nitrate, the mixing layer along the back trajectories, and the residence time of air masses crossing over the highly polluted zones. The results supported our hypothesis, i.e., ammonium salts mostly co-existed with dust aerosolsexternally. For example, except for 20080425, all dust day samples mostly traveled at an altitude above the mixing layer before mixing down to ground level. For most sampling days in Category 1,the average mixing layer was less than 900 m, favoring the trapping of locally emitted anthropogenic air pollutants in the mixing layer. In addition, the air massesat this elevation apparently moved slowly and took over 10 hr to cross over the highly polluted area. Even lower speeds were expected for air masses atthe bottom of the mixing layer, as wind speed decreases with height. Except for exterior samples, the sampling days in Category 2 featured a mixing layer that was, on average, higher than 900 m. The air masses at this

elevation took less than 10 hr to cross over the highly polluted areas and generally had higher speeds. Theoretically, a lower mixing layer and a lower wind speed favored the accumulation of air pollutants and the formation of ammonium nitrate to some extent. The transport of dust air masses above the mixing layer reduced the possibility for internal mixing of ammonium salts and reactionwith dust aerosols along the long transport path. The shorter time for dust air masses mixing down to ground level before arriving at the reception site also increased the possibility for external co-existence between ammonium salt aerosols and dust aerosols in Category 1. The reverse could be argued to explain the observations for Category 2. The correlation analysis results in Table S2 indirectly supportthese conclusions. In fact, previous studies proposed that nitrate is rarely formed on the surface of dust particles (Zhang and Iwasaka, 1999). Therefore, much lower nitrate concentrations were observed in Category 2. Noted that the exteriors with ID of 20110415 and 20110502 have not yet been explained.

~~The reported threshold of windspeed for dust mobilization in the Gobi Desert ranges from 10-12 m/s(Choi and Zhang, 2008).We estimated 40.5±9.9 km/hhas the average wind speed during a dust storm according to Asia dust observations (). If air masses were transported faster than40.5 km/h,we found that the INcontent in most atmospheric aerosol samples was lower on dust days than on ND daysbecause of the strong dilution effect. This effect was observed in samples 080528, 080529, 110319 and 100315 (Table 5). If an air mass was transported over the ocean for some distance (ratio of oversea to totaldistance of at least 10%), no matter how fast the transport velocity, the IN content decreased because of theinput of clean marine air, such as in samples080425, 100320, 110418and110501. If the air mass was transported slowly (less than 42.4 km/h) or transported onlya short distance over the sea, with an oversea to total distance ratio of less than 10%, the IN content increased in samples collected in the downwind area, such as in samples 080301, 090316, 100321 and 110502. Therefore, the IN content is related to not only the transport path and speed but also local emissions and reaction conditions during transport.~~

~~**Table 5**~~**6.** ~~IN content, RH, NOx, transport speed and transport distance over the sea for atmospheric aerosol samples on dust days~~

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Group	Sample number	TSP ($\mu g/m^3$)	NO_3^- ($\mu g/g$)	NH_4^+ ($\mu g/g$)	Speed (km/h)	Distance over the sea (km)	Ratio of the distance over the sea to the total distance (%)
	080301	527	38984	24107	35.1	0	0
Case 1 IN>ND	080315	410	47611	34130	53.4	262	6.8
	090316	688	23050	25012	59.5	0	0
	100321	519	31741	18155	49.2	0	0
	110415	1225	41970	20390	57.0	258	6.3
	110502	810	25995	13632	31.2	121	5.4
Case 2 IN<ND	080425	256	4089	372	29.3	253	12.0
	080528	2579	232	72	79.8	259	4.5
	080529	2314	26	166	78.0	229	4.1
	110319	939	13088	10067	55.4	404	10.1
	110501	502	8924	10631	30.6	298	13.5
Case 3 NO_3^-<ND NH_4^+≧ND	100315	501	10767	8515	58.7	183	4.3
	100320	3857	1418	884	38.0	254	9.3
	110418	558	6891	11778	23.0	380	22.9

Figure 6. The 72-h backward trajectories for non-dust (a) and dust (b) samples from 2008 to 2011.

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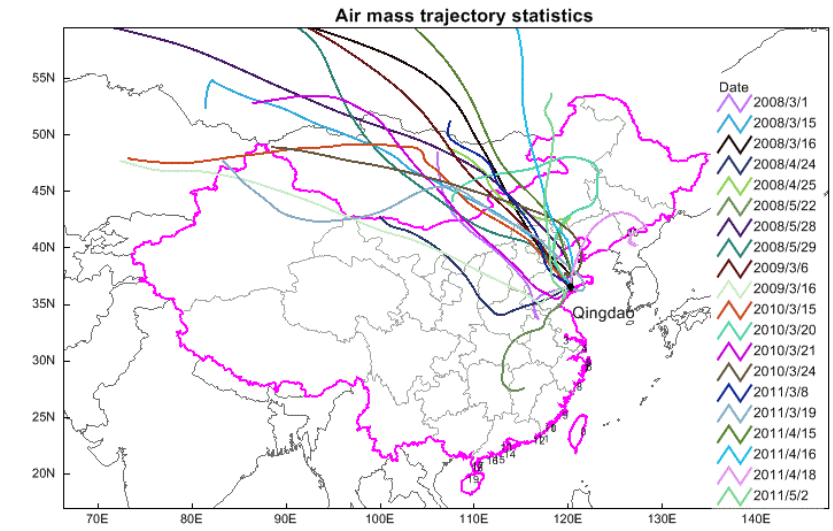
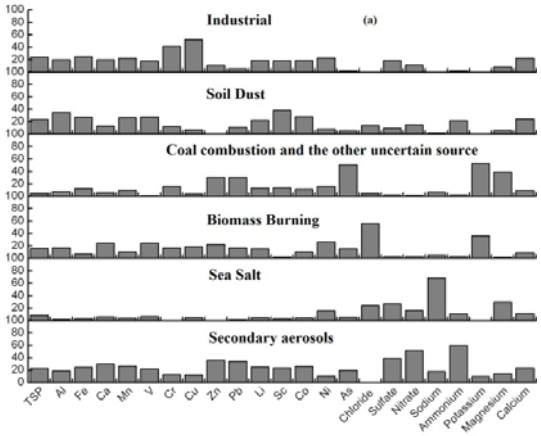


Figure 4. The 72-h backward trajectories for non-dust and dust samples from 2008 to 2011

Fig. Daily mean emission of NOxdust

3.4-5 Source apportionment of aerosols from dust and non-dust events



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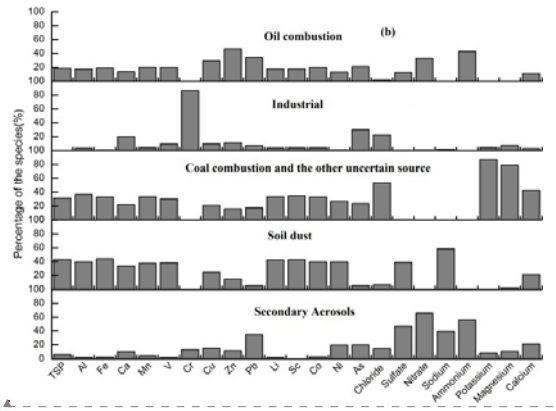

Figure 5. Source profiles of atmospheric aerosol samples collected on non-dust (a) and dust (b) days using the PMF model

The sources of atmospheric aerosols on dust and ND days were determined by running the PMF model (Paatero and Tapper, 1993; Paatero, 1997). As shown in Fig. 58 showed -that atmospheric aerosols on ND comparison days were mainly from consisted of six sources: industry, soil dust, secondary aerosols, sea salt, biomass burning, and coal combustion and/ the other uncertain sources (, with 90% of the scaled residuals falling between -3 and +3; r^2=0.97). On dust days, the sources of aerosols differed from those on ND days, mainly including oil combustion, industry, soil dust, secondary aerosols, and coal combustion and /other uncertain sources. These value are compared in Table 7. We compared the contributions of aerosol sourcesin dust and ND periods.As shown inTable6, the The contribution of soil dust increased from 23% to 36% on dust days relative to comparison days, which is consistent with the high concentrations of TSPs and crustal metals observed on dust days. Liu et al. (2014) also found an even larger increase inthe contribution of dust aerosols to PM_{10}, i.e., 31%-40%, on dust days relative to non-dust days. that the contributions of dust aerosols to PM_{10} increased by 31%-40% on dust days, which is greater than the 10%-20% contribution of local soil dust on ND days. Accordingly, The the contributions of local anthropogenic sources decreased on dust day samples, especially those of secondary aerosols, which verifiedthat the influence of anthropogenic sources on atmospheric particulates decreased on dust days. The source profile for coal combustion in dust day samples showed a high percentage of K^+, Cl^-, Ca, Mg, Co, Ni, As, Al and Fe, indicating a mixture of coal combustion and other pollutants emitted along the transmission path on dust days. This source increased due to the coal combustion emission mixing with other uncertain source emitted

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to the air under strong wind. ~~Coal combustion emissions were mainly a mixture of coal combustion and other pollutants emitted along the transmission path on dust days.~~

The calculation results also showed that the contribution of dust aerosol mass(the sum of nitrate and ammonium associated with the dust source) to the total aerosol mass (the total nitrate and ammonium) greatly increased on dust days.

~~Therefore, the sources of aerosol particles changed on dust days. Dust events had a great impact on aerosol sources in the downwind area. The influence of soil dust on aerosols and IN-loaded particles was greater than that on local soureson dust days. In fact, the contribution of soil dust to aerosols was related to the intensity of the dust storm and the transport path. However, we could notdetermine the contributions of dust to aerosols for the different dust casesbecause of the limited number of samples.~~

~~**Table 6.** Sources and source contributions (expressed as %) calculated for aerosolsamples collected during dust and non-dust events~~

~~Dust event~~		~~Non-dust event~~	
~~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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3.~~5~~ 6 Dry deposition fluxes of ~~aerosol particles~~TSP, particulate inorganic nitrogen and metals

Dust events are known to increased the concentration and deposition of aerosol particles during long-range transport along the transport path. For example, Fu et al. (2014) found that the long-range transport of dust particles increased the dry deposition of PM_{10} in the Yangtze River Delta region by a factor of approximately ~~239~~820%. Some studies ~~observed~~ reported enhancements in oceanic chlorophyll α following ~~a~~ dust storm events (Tan and Wang, 2014; Banerjee and Kumar, 2014).

However, ~~The~~ the deposition ~~magnitude~~ fluxes of dust varied greatly among different dust storms, and only a few ~~some~~ dust episodes were followed by increases in chlorophyll *a* (Banerjee and Kumar, 2014). In addition to those in high-nutrient and low-chlorophyll (HNLC)regions, the input of nitrogen input and other nutrients associated with dust deposition is expected to promote the growth of phytoplankton. However, the extent can vary greatly depending on the nutrient limitation conditions in the oceans. A similar principle holds for the occurrence or absence of algal blooms following dust events. Thus, we calculated the dry deposition fluxes of aerosols particles, $N_{NH4+NO3}$ and metal elements during dust and comparison periods using the measured component concentrations and modeled dry deposition velocities (Table 8). We then compared the dry deposition flux of TSP and $N_{NH4+NO3}$with the previous observations in literature.

~~The role of dust deposition as a nutrient source leading toan increase in algalblooms has not been adequately addressed. To understand the influence of dust weather on the nitrogen deposition flux, we calculated the dry deposition fluxes of aerosols particles,IN and metal elements during dust and ND periods using the measured component concentrations and modeled dry deposition velocities obtainedfrom Williams' model (Qi et al., 2005)(Table 7).~~

~~–~~The dry deposition fluxes of atmospheric particulates increased on dust days relative to comparison days. All increases or decreases in this section reflected the value on dust days relative to comparison days, if not specified. For example,

~~Compared to that on ND days, the dry deposition flux of atmospheric particulateincreasedon dust days.On ND days,~~ the dry deposition flux of ~~particles~~ TSP was 2,800±700 mg/m^2/month in the coastal region of the Yellow Sea~~, . and t~~The particle flux varied over a wide range from 5,200-65,000 mg/m^2/month under different dust ~~conditions~~sampling days, with an average of ~~16~~18,~~800~~ 453 mg/m^2/month. ~~The results verified that dust eventsenhanced the dry deposition flux of atmospheric particulates.~~ However, the dry deposition flux of $N_{NH4+NO3}$ did not follow this pattern~~IN showed variations with particles~~. In ~~Case~~ Category 1, the dry deposition fluxes of $N_{NH4+NO3}$ ~~IN~~ increased by ~~a factor of 1.19-5.8~~285%, corresponding to the increase in theTSP flux of 86-252% (Table S3)~~and the flux of atmospheric particles increasedby a factor of 1.8-6.3~~. In Categories 2 and 3, the dry deposition fluxes of TSP increased by 126% to 2226% compared to that on comparison days. Except for ammonium in Category 3, the dry deposition fluxes of particulate $N_{NH4+NO3}$, however, decreased by 41% (on average). A larger relative decrease was found for the concentration of nitrate, i.e., decreases of 73%

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and 46% in Category 2 and 3, respectively. Note that the average ammonium deposition flux decreased by 47% in Category 2 but increase in Category 3.

~~Cases 2 and 3, the dry deposition flux increased 2.3 to 23.2-fold compared to that on ND days. Except for ammonium in Case 3, the dry deposition flux of particulate IN decreased by an average of 41% in the case of high particle concentrations. The concentration of nitrate decreased 63% and 46% in Cases 2 and 3, respectively. Additionally, the ammonium flux decreased by 14% in Case 2, while in Case 3, ammonium was higher thanthat on ND days.We found that dust events sometimes led to an increase in the nitrogen input to the ocean relative to that during ND events, but it did not always occur depending on the chemical composition of the dust particles. As discussed, dust particles may carry abundant reactive nitrogen when they travel through polluted continental atmosphere. However, the relatively pure dust particles may be transported when no air pollution occurs along the dust transport route to oceans.~~

The dry atmospheric deposition fluxes of Fe ~~in atmospheric particulates~~ increased by a factor of ~~2~~124-2370%~~5~~ on dust days ~~compared to that on ND days~~. Atmospheric inputs of iron to the ocean ~~can~~ have been proposed to enhance primary production in ~~high-nutrient, low-chlorophyll regions (~~HNLC~~)~~ areas (Jickells et al., 2005). ~~However~~Moreover, except for Pb and Zn in ~~Case~~ Category 2, the dry deposition fluxes of Cu, Pb and Zn increased with those of nitrogen and iron on dust days. T~~hese t~~race metals were found to have a toxic effect on marine phytoplankton and inhibit their growth (Bielmyer et al., 2006; Echeveste et al., 2012). Liu et al. (2013) found that this inhibition coexisted with the promotion of some phytoplankton species in incubation experiments involving the addition of Asian dust samples in the southern Yellow Sea in the spring of 2011.~~In Case 3, dust wasdeposited in the ocean, the atmospheric supply of nitrogen decreased, and the atmospheric inputs of Fe and some toxic metals increased. Moreover, phytoplankton growth was affected by the addition of nutrient elements andtoxic elements. The overall effect of dust deposition on primary productivity was a combinationof these two effects. This is likely the reason why inhibition coexisted with the promotion of some phytoplankton species in incubation experimentsusing additions of AD in the southern Yellow Sea in the spring of 2011 (Liu et al., 2013).~~

~~The contribution of dust eventsto marine nitrogen input and primary production will be overestimated if the nutrient flux simply considers dust concentrations and a constant ratio of nutrients to particles. The atmospheric input of nitrogen to the ocean on dust days depends on the 'dilution effect'~~

of a dust event.Dust subjected to long-range transportdoes not always increase the atmospheric input of nitrogen. Long-term observations of dust eventsmust be performed to evaluate the contributions of dust to the biogeochemistry of nitrogen and primary production in oceans.

Table 7. Dry deposition of aerosol particles (mg/m^2/month), particulate inorganic nitrogen (mg N/m^2/month) and some toxic trace metals (mg/m^2/month) on dust and non-dust days

	Dry deposition flux						
	Particles	NO_3^--N	NH_4^+-N	Fe	Cu	Pb	Zn
Case 1 IN>ND	9600±4300	87±53	25±13	650±340	2±1	0.3±0.2	6±3
Case 2 IN<ND	18000±11,000	13±18	5±7	1300±1000	3±2	0.08±0.04	4±1
Case 3 NO_3^-<ND NH_4^+≈ND	29,000±31,000	26±6	17±8	2100±2200	6±1	0.20±0.02	5±3
Non-dust	2800±700	48±33	8±8	190±110	1±1	0.09±0.1	5±4

This work	2008-2011	Qingdao, coastal region of the Yellow Sea	Non-dust day	2800±700	63±39	
			Dust day	10138±15940	66±61	
Shi et al., 2013	2007	The Yellow Sea	Non-dust day		19.2	

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~~Dust day~~	~~104.4~~
~~Average of dust and non-dust~~	

3.7 Potential impacts of nitrogen dry deposition flux associated with dust influenced by anthropogenic activity

Due to anthropogenic activity and economic development, inorganic nitrogen emissionsincreasedin China from 1980 to 2010 (Fig.S5). Accordingly, the $N_{NH4+NO3}$ dry deposition flux should have theoretically increased with the increase in inorganic nitrogen emissions. However, from the limited data shown in Table 9, we did not find the expected increase in dry deposition flux of inorganic nitrogen during the dust days. Considering the uncertainty in dry deposition velocity, we normalized the dry deposition flux of $N_{NH4+NO3}$ using the concentration of nitrate and ammonium reported in the literature and the recommended dry deposition velocity of 1 cm/s for nitrate and 0.1 m/s for ammonium in coastal areas reported by Duce et al. (1991). We then found that dry deposition fluxes of $N_{NH4+NO3}$ over the Yellow Sea during the dust days increased greatly from 1999 to 2007. The fluxes of $N_{NH4+NO3}$ in Qingdao, including during the dust days, varied narrowly in a range of 94.75-99.65 mg N/m^2/month from 1997 to 2011(Table 8). The complicated results may reflect the combined effects of NOx and NH_3 emissions in northern China, the occurrence frequency and intensity of dust events and metrological conditions affecting the transport pathways and moving speeds of dust air masses and chemical reactions occurring therein. For example, dust events commonly exhibited a periodic variation from 2000 to 2011 (Fig.S5).

4 Conclusion

The concentrations of nitrate and ammonium in TSP samples varied greatly from event to event on dust days. Relative to 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 ammonium salt aerosols co-existing externally with dust aerosols or the residual of incomplete reactions between ammonium salts and carbonate salts. NO_3^- in the dust day samples was partially related to mixing and reactions between anthropogenic air pollutants and dust particles during the transport from the source zone to the reception site. However, this process was generally much less effective and led to a sharp decrease in nitrate in Category 2 TSP samples. The external co-existence of ammonium salt aerosols

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with dust aerosols and the extent of the reactions between ammonium salts and carbonate salts were apparently associated with the transport pathway, moving speeds and metrological conditions, among other factors.

The concentration of ~~particulate IN~~ nitrate and ammonium in TSP samples varied greatly~~exhibited a large variation~~ from event to event on dust days~~, and a dust event did not simplyincrease nutrient concentrations~~. Relative to 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 ammonium salt aerosols co-existing externally with dust aerosols or the residual of incomplete reactions between ammonium salts and carbonate salts. NO_3^- in the dust day samples was partially related to mixing and reactions between anthropogenic air pollutants and dust particles during the transport from the source zone to the reception site. However, this process was generally much less effective and led to a sharp decrease in nitrate in Category 2 TSP samples. The external co-existence of ammonium salt aerosols with dust aerosols and the extent of the reactions between ammonium salts and carbonate salts were apparently associated with the transport pathway, moving speeds and metrological conditions, among other factors.

~~The effect of dust events on particulate nitrogen in the downwind region was determined by the dilution effect of a dust event, which depends on many factors, such as the dust storm intensity, transport speed and path, local sourceemissions during transport, meteorological state and atmospheric reactions.~~ Due to a sharp increase in dust loads on dust days, the contribution of soil dust to the total aerosol mass was higher on dust days than on comparison days, while the contributions from local anthropogenic sources were accordingly lower.

~~Dust events affect the source apportionment of aerosols. The contribution of soil dust to aerosols increased,while local anthropogenic sources decreasedduring a dust event.The contribution of dust to aerosols must be studied further under different IN conditions.~~

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 statisticalrelationship between dust events andatmospheric deposition of $N_{NH4+-NO3-}$. A simple assumption ofa linear increase in$N_{NH4+-NO3-}$ with increasing dust load, like that in the literature, could lead to considerable overestimation ofthe dry deposition flux of

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nutrientsinto the oceans and the consequent primary productionassociated with dust events.

~~Dust events enhance the input of atmospheric particulates via dry deposition. However, the influence of dust events on the input of nitrogen to the ocean is still uncertain. The dry deposition flux of IN on dust days decreased when a strong dilution effect was present. The contribution of dust events to marine nitrogen inputs and primary production could be overestimated if the dry deposition flux of nutrients is estimated using onlyparticulateconcentrationson dust days.~~

Acknowledgments. This work was supported by the Department of Science and Technology of the P. R. China through the State Key Basic Research & Development Program under Grant No. 2014CB953701 and the National Natural Science Foundation of China (No. 41375143). We thank Prof. Yaqiang Wang and Jinhui Shi for the valuable discussion regarding this research. We also express our appreciation to Tianran Zhang for help with sand sampling and Qiang Zhang for data collection.~~We thank Prof. Xiaohong Yao for the valuable discussion regarding this research. We alsoexpress our appreciation to Tianran Zhang for help with sand sampling.~~

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Table 1. Sampling information for the aerosol samples collected at the Baguanshan site in thecoastal 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
	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
	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
	Samples on	20100324	From 11:30 a.m. to 15:30	Sunny day

	non-dust days		p.m. on Mar. 24th	
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
	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 was collected on March 1 when no dust was recorded by the MICAPS. However, the MICAPSinformation indeed showed dust events in China on March 1. The modeled spatial distribution of thePM_{10} mass concentration for this dust event on March 1 implies that the sample should be classified as adust sample. The supporting figureisFig. S1.

[b]The sampling duration was reduced to only 2 hrs because of extremely high particle loads.

[c]Note that one exterior dust sample was collected on April 18when 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 eventon April 18implies that the sample should be classified as adust sample. The supporting figure is Fig. S2.

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

Component	Measurement method	Detection limit ($\mu g \cdot L^{-1}$)	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

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 valuesless than 10 indicate that the studied element is derived mainly from crustal sources,whereasEF valuesmuch higher than 10 indicatea anthropogenic source.

Table 4. Average concentrations of inorganic nitrogen (DIN), TSP, NOx, relative humidity (RH) and air temperature for each aerosol samplecategoryin Qingdao.

	Sample number	TSP μg·m^{-3}	NO_3^- μg·m^{-3}	NH_4^+ μg·m^{-3}	RH %	T °C	NOx μg·m^{-3}	Summary
Category 1	20080301	527	20.5	12.7	57	7.0	36	DIN concentrationon dust days higher than that on non-dustdays
	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	DIN concentrationon dust days lower than that on non-dust days
	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 dust days lower than that on non-dust days;NH_4^+close to that on 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[a]	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	

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-dustday for each dust event, see Table 1.

Table 5. Comparison of the inorganic nitrogen (DIN) content in sandand 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]		Study region and data source	Relative concentration[a]			
	NO_3^-	NH_4^+		NO_3^-	NH_4^+	NO_3^-	NH_4^+
Zhurihe (This study)	25.46±22.87	4.21±1.03	Duolun (Cui, 2009)	1200	900	Non-dust: 28,200±24,819	Non-dust: 24,063±21,515
AlxaLeft Banner, Inner Mongolia (NiuandZhang, 2000)	62.1±7.4	79.1±1.1	AlxaRightBanner, Inner Mongolia (NiuandZhang, 2000)	1975[b]	4091[b]	Category 1: 34,892±9570	Category 1: 22,571±7,016
Yanchi, Ningxia (NiuandZhang, 2000)	46.4±2.2	80.9±1.3	Hinterland of theTaklimakan Desert, Xinjiang (Dai et al., 2016)	142-233	2-15	Category 2: 5,542±5,117	Category 2: 4,758±5,698
			Average of SonidYouqi, 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
Golmud, Qinghai(Sheng et al., 2016)	892.9	-[c]
Hohhot, Inner Mongolia (Yang et al., 1995)	588.1	No data

[a]Relative concentration of DIN per aerosol particle mass

[b] Samples collected on a floating dust day (Horizontalvisibility 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. 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)	Residence time[a] (h)	T[b] (℃)	RH (%)
Category 1 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 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	100320	3857	1,418	884	76.9	0	1,284±401	525±371	10	-12.2±6.3	61±16
NH_4^+≅ND	110418	558	6,891	11,778	35.6	931	1,344±780	695±672	2	-0.1±8.2	52±28

Table 7. Sources and source contributions (expressed in%) calculated for aerosolsamples 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

Table 8. Dry deposition of TSP (mg/m^2/month), particulate inorganic nitrogen (mg N/m^2/month) and some toxic trace metals (mg/m^2/month) on dust and non-dust 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 in Table 3.

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 day	2,800±700	63±39	93.90
			Dust day	10,138±15,940	58±36	101.39
			Average of dust and non-dust			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	Non-dust day		19.2	132.17
			Dust day		104.4	227.07

	Average of dust and non-dust	179.62

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

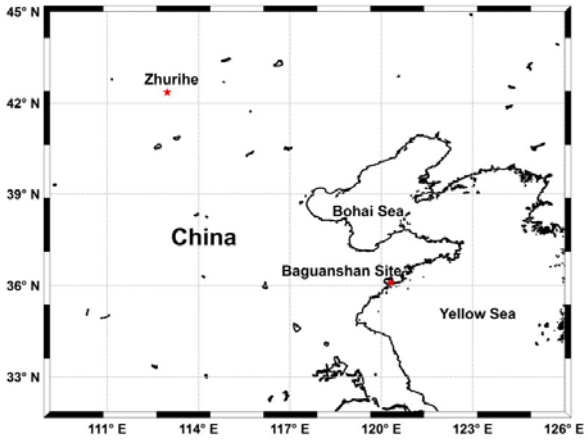

Figure 1. Location of the aerosol and dust sampling sites.

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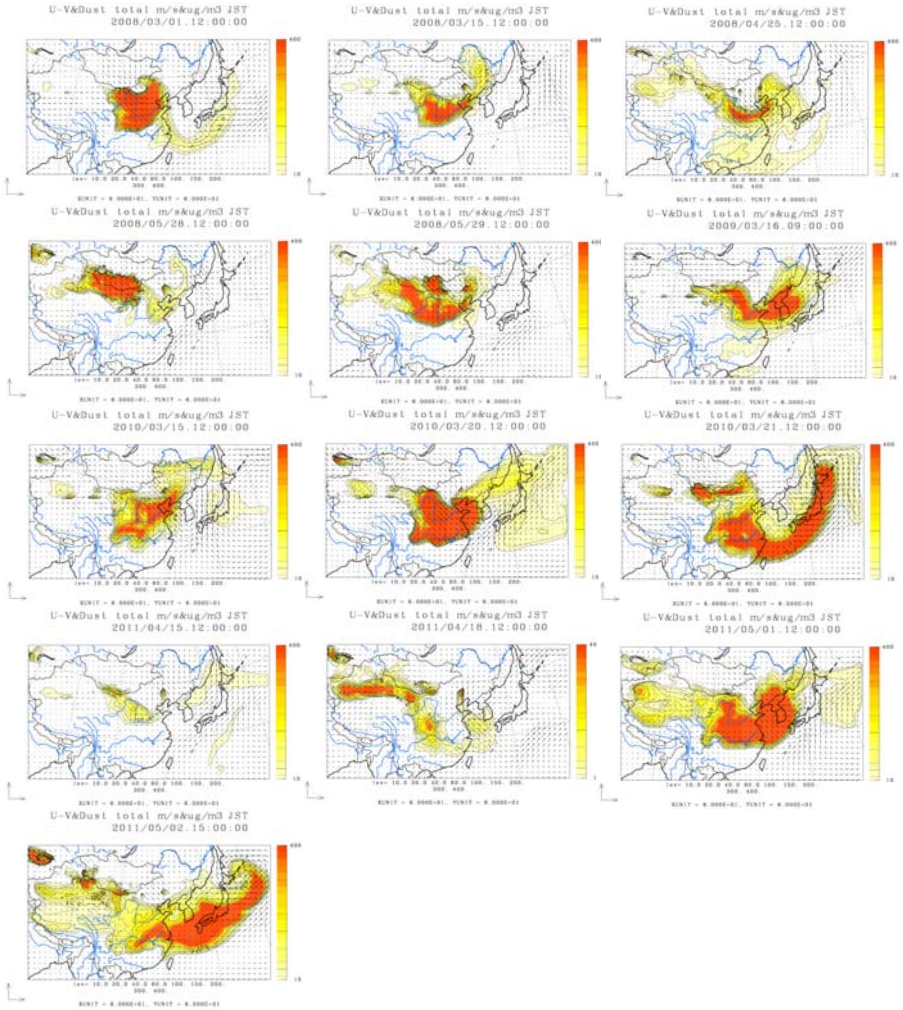

Figure 2. Modeleddust concentrations over East Asia by CFORSmodel during each dust sampling day from 2008 to 2011(http://www-cfors.nies.go.jp/). (The figures showthe modeled dust concentration inthe middle of each sampling duration). Nodata are available forMar.19, 2011,because of the

earthquake in Japan.Hourly PM10 concentrationsweremodeled by the WRF-CMAQ model for eachsampling day,and the resultsareshownin Fig. S3.

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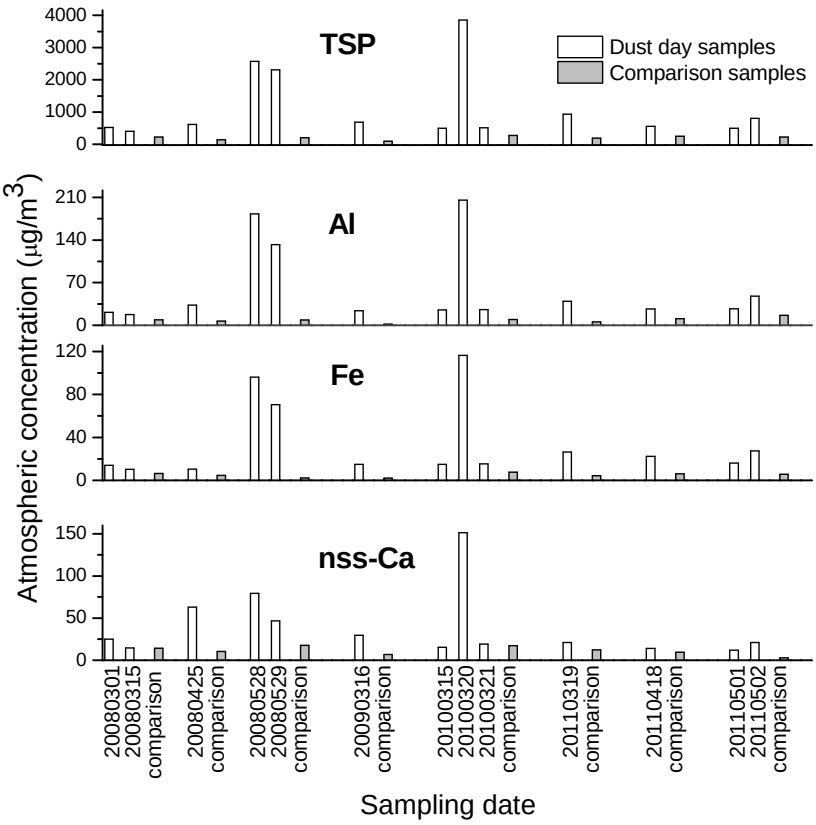

Figure 3. Mass concentrations of TSP, Al, Fe and nss-Ca in aerosol samples collected at the Baguanshan site on dust and comparison days from2008-2011.

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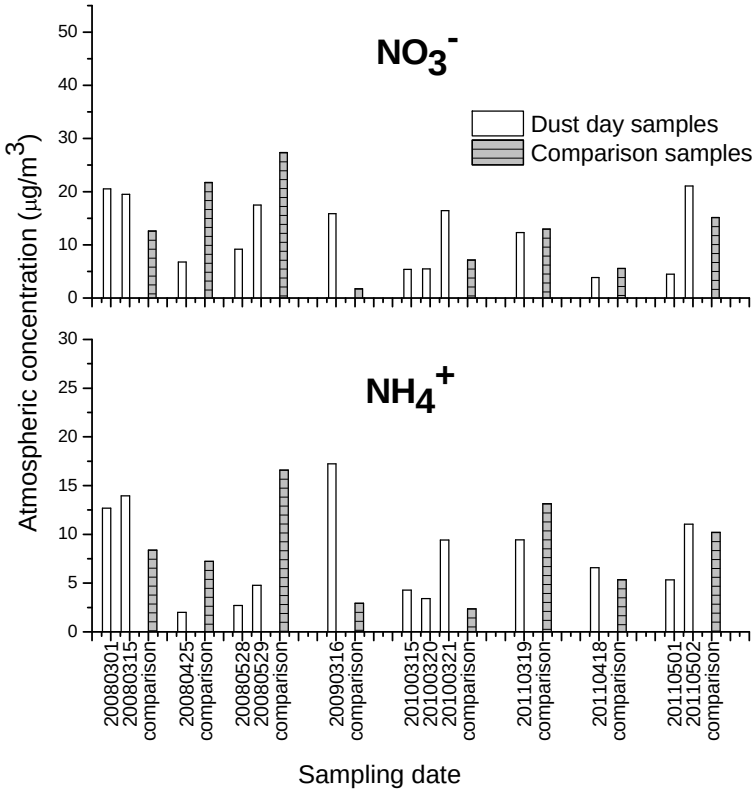

Figure 4. Mass concentrations of NH_4^+ andNO_3^-in aerosol samples collected at the Baguanshan site on dust and comparison days during March-May in 2008 to 2011.

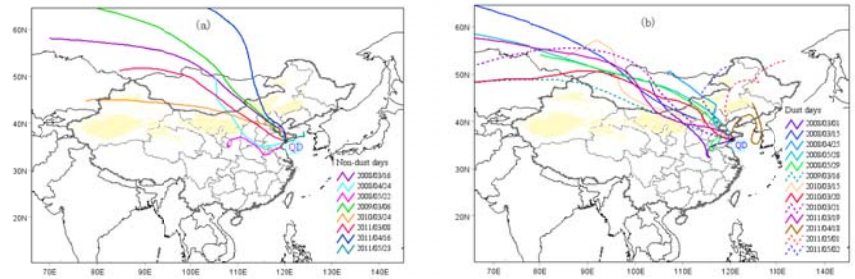

Figure 5. The 72-h backward trajectories for non-dust (a) and dust (b) samples from 2008 to 2011(the yellow domains in the maps represent the dust source regions in China).

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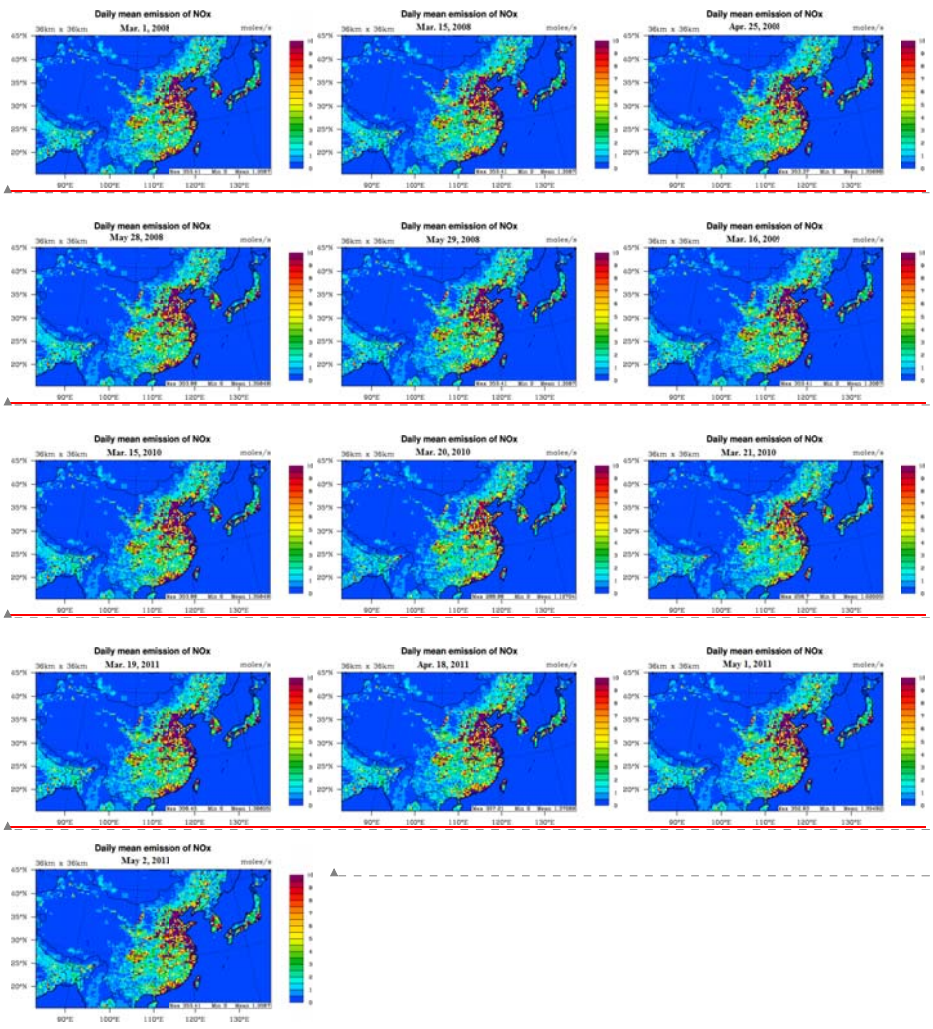

Figure 6. Daily mean emission of NOxover East Asia on the dust daysfrom 2008 to 2011.

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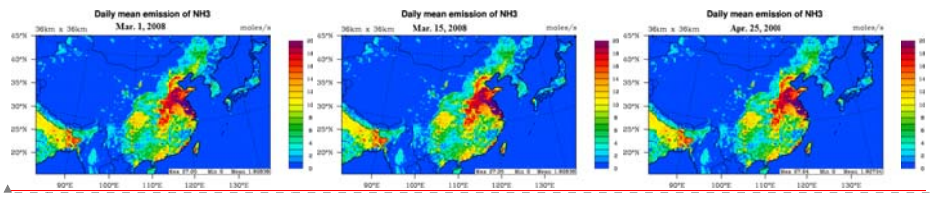

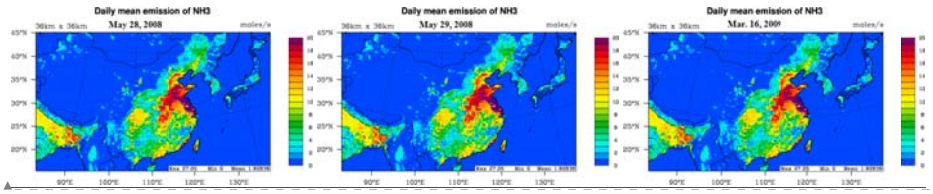

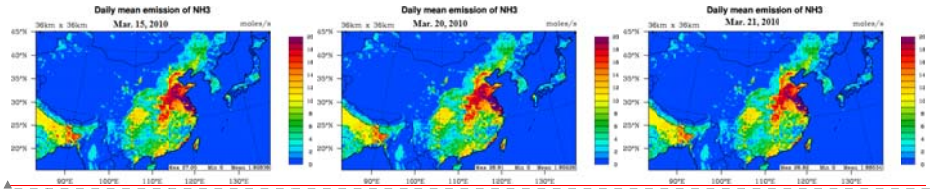

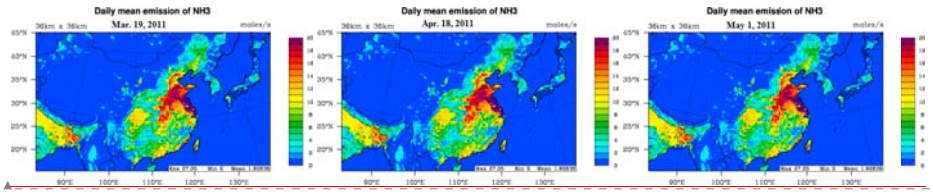

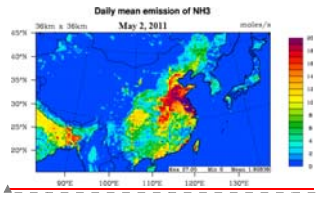

Figure 7. Daily mean emission of NH_3 over East Asia on the dust days from 2008 to 2011.

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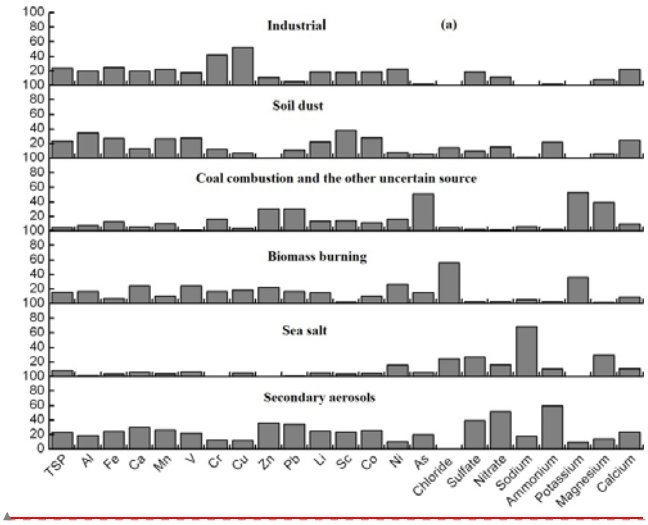

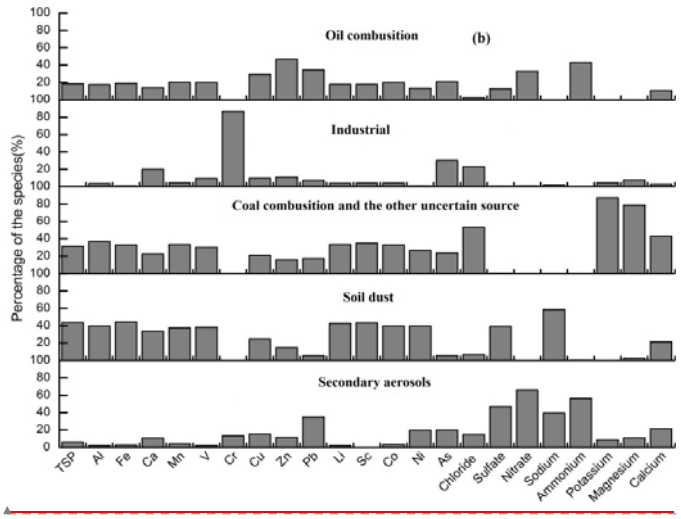

Figure 8. Source profiles of atmospheric aerosol samples collected on non-dust (a) and dust (b) days using the PMF model

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