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Atmospheric Chemistry and Physics Discussions

Interactive comment on "The concentration, source apportionment and deposition flux of atmospheric particulate inorganic nitrogen during dust events" *by* Jianhua Qi et al.

Jianhua Qi et al.

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General Comments: The manuscript titled 'The concentration, source apportionment and deposition flux of atmospheric particulate inorganic nitrogen during dust events' written by Jianhua Qi presented the dust impacts on particulate inorganic nitrogen by analyzing the aerosol samples collected at Qingdao, China. The authors divided dust pattern into three parts, and investigated the dry deposition flux. To estimate the source, PMF receptor model was also used. Based on the above approaches, the authors tried to answer the questions of 'dust event always increase the atmospheric input of nitrogen to the ocean?'. The topic is interested ones because the impact of dust as atmospheric input on ocean ecosystem has been still unclarified. However,

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throughout the manuscript, it is not well organized and hard to follow and understand. Overall, this manuscript will not be acceptable taking into account the high journal quality of Atmospheric Chemistry and Physics.

Reply: We will revise the manuscript according to the comments to improve the manuscript quality.

Q1.Before the discussion, first, the definition of "dust events" cannot be understood well. In L99-101, the authors explained that 'Samples were collected on dust days and selected ND days in spring from March 2008 to May 2011, with sampling duration of 4h for each sample. We refer to the ND days as sunny and cloudy days before and after dust events in the following discussion'. The authors should add the appropriate reference of the Meteorological Information Comprehensive Analysis and Process System (MICAPS) which defined the weather conditions (and also, the subsection 2.4 should be reorganized partly into this explanation). What is the definition of "dust events" here? Visibility? More information of how the dust events are defined in this system should be announced in detail. Total of 14 samples (sample numbers in Table 3) during dust events were analyzed throughout this study. The sampling duration was 4 hrs, so which data are used in the corresponded date in Table 3? All samples in the day? Moreover, what is the sample numbers of ND? The current information in Section 2.1 is severely lacked in the information which the readers can follow the authors methodology. Because this study discussed the dust impact, the explicit and detailed information regarding dust is required. In this sentence, I am worried about the explicit division of dust and non-dust samples. It is well known that some dust events are continued a few days. For example, the samples used in this study during 28-29 May 2008, 20-21 March 2010, 15 and 18 April 2011, and 1-2 May 2011 showed continuous dust events. In such cases, do the authors have confidence to the clear separation of dust and non-dust samples? How about the AI concentration definition (L171-172) of non-dustdays samples? Why were other days samples not collected to clearly separate the dust impacts? The definition of ND is ambiguous. According to the definitions of dust

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and non-dust, the discussion on dust impact might be changed. The reconsideration of dust impact is needed based on the clear definitions of dust.

Reply: In this study, the dust event was defined according the definition adopted in regulations of surface meteorological observation of China (CMA, 2003;Wang et al., 2008) and identified based on the meteorological records information from Meteorological Information Comprehensive Analysis and Process System (MICAPS) of China Meteorological Administration.Each dust sample was collected for 4hrs duration and the sampling started only when the PM10mass concentration available on the website (http://www-cfors.nies.go.jp/ cfors/; http://www.qepb.gov.cn/m2/) was increased greatly. The approach made the dust sample more representative relative to urban background. However, for dust event with duration less than one day, only one sample was collected;; for dust event with longer duration, i.e. multiple days, the sample was collected once a day. The sampling information was listed in the Table S1. Based on the forecast, we also collected aerosol particle samples immediately before or after 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 featured by collecting under a clear and sunny weather condition and lower mass concentration of PM10. 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$\times$2GSD" proposed by Hsu et al. (2008).

CMA: Regulations of Surface Meteorological Observation, China Meteorological Press, Beijing, 154–156, 2004. 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. Wang Y. Q., Zhang X. Y., Gong S. L., Zhou C. H., Hu X. Q., Liu H. L., Niu T., 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.

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Q2.The second concern is the “dilution effect” which the authors claimed as the key factor for the discussion of inorganic nitrogen. Again, without the explicit definition of dust and non-dust, the dilution effect cannot be understood well. In this discussion, although the authors introduced the air mass speed, there were no implications on the intensity of dust events itself. Why the upwind (i.e., near desert) information was not used here to describe the dust intensity? The dilution is not so simple, hence more information are required to reinforce the authors finding. The authors discussed the inorganic nitrogen behavior. In these cases, what is the counter ion of NH4+ and NO3-? Are the main counter ions metal elements? If NH4NO3 are formed, due to its chemical unstablity according to the temperature and relative humidity, it is not simple to discuss only the viewpoint of “dilution effect”. In addition, the authors used NO2 data to investigate the inorganic nitrogen, but how about NH3? Only from NO2 data, it is insufficient to estimate the inorganic nitrogen variation. On the above reasons, the reconsideration is required to publish this manuscript from Atmospheric Chemistry and Physics.

Reply: In revision, the part reads as “Inorganic nitrogen (IN) concentrations highly varied in different dust samples (Table 3). According to the concentrations relative to those in comparison samples, they can be classified into three categories, i.e., Category 1 in which higher IN concentrations were observed in dust samples, Category 2 in whichlowerIN concentrations were observedin dust samples, and Category 3 in which lower nitrate concentrations with slightly higherconcentrationsof ammoniumin dust samples. Category 1 was usually associated with a lower moving speed of dust air mass or a longer distance over the ocean (Table 5) while the reverse was true for Category 2. The moving speed and distance over the ocean of dust air mass in Category 3 was generally between them. Theoretically, lower moving speed of dust air mass favors reactions between dust particles and anthropogenic gaseous precursors of IN due to a longer reaction time. Largemoving speed of dust air mass was frequently associated with a large wind speed in the lower layer atmosphere (Gao et al., 2010; Gillette and Passi, 1988; Peng et al., 2007; Yue et al., 2008), leading to anthropogenic

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gaseous precursors therein to be better diluted. Shorter reaction time and reduced concentrations of anthropogenic gaseous precursors likely lowered IN in Category 2. Moreover, the relative concentration of IN per aerosol particle mass in μg/g was analyzed and compared with those values in literature. .." It is questionable for using NOx observed in Qingdao to argue the generation of IN in dust samples. We agree this because most of IN observed in dust samples should be derived from secondary reactions upwind of Qingdao by considering a low conversion rate of NOx to IN. The former study (Liu et al., 2010) showed that NOx and NH3 generally capture the spatial distribution patterns with high values over eastern China and relatively lower values over central and western China, where dust source regions are located(Fig. S1-S3). Thus, we will add modeling results using a 3-D air quality model to support our analysis in revision.

Gao, Q X., Ren Z H. et al. : Dust events and its impacts on atmospheric environment, Science press, Beijing, 2010. Gillett e D A, Passi R.: Modeling dust emission caused bywind erosion, J G R., 1988, 93: 14234- 14242. Liu X. H., Zhang Y., Cheng S. H., Xing J., Zhang Q., Streets D. G., Jang C., Wang W. X., Hao J. M.:Understanding of regional air pollution over China using CMAQ, part I performance evaluation and seasonal variation, Atmospheric Environment , 44,2415-2426, 2010. Peng, Z., Liu X. M., Hong Z. X., Wang B. L.: Characteristics of Atmospheric BoundaryLayer Structure and Turbulent FluxTransfer during a Strong Dust StormWeather Process over Beijing Area, Climatic and Environmental Research, 2007, 12(3): 268-276. Qi J.H., Gao H.W., Yu L.M. , Qiao J.J.: Distribution of inorganic nitrogen-containing species in atmospheric particles from an island in the Yellow Sea, Atmospheric Research, 101,938-955, 2011. Wang Y. Q., Zhang X. Y., Gong S. L., Zhou C. H., Hu X. Q., Liu H. L., Niu T., 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. Yue P., Niu S. J., Liu X. Y.: Dust Emission and Transmission during Spring Sand-dustStorm in Hunshandake Sand-land, Journal of Desert Research, 2008, 28(2): 227-230.

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Specific comments: Q3.L35-36: This conclusion does not match to the manuscript contents. The authors stated that input of nitrogen to the ocean depends on the dust events.

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

Q4.L57-L67: In this paragraph, the authors used “ND days” simply. However, this wording should be used carefully; because the definition of non-dust days will be different in each study. Please consider to carefully define this wording.

Reply: Thank you for this suggestion. To avoid confusion, we will use "non-dust storm days" according to the original reference in L57-L67.

Q5.L146: Some information should be replaced on Section 2.1 appropriately.

Reply: We will move this information to Section 2.1 in the revised version.

Q6.L162: “atmospheric particulate” is “TSP”?

Reply: We apologize for the confusion. The term “atmospheric particulate” will be revised to "total suspended particulates". Atmospheric particulate concentrations were obtained by weighting TSP samples. We will revise the sentence and the corresponding figures.

Q7.L165: I cannot follow the calculation of “1.8-14.0 times (mean: 5.9)“. The mean concentration have not been stated for dust days.

Reply: Each sample on dust day had its corresponding non-dust sample (Table S2).The 1.8-14.0 times was calculated as a ratio of the TSP concentration on a given dust day to the values in the comparison samples. The concentration and the ratio of samples on dust days were listed in Table S2. Q8.L167: The EF of Ca is 14.0 in Table 2. L168: The statement of “decreased to less than three” cannot be followed from the valued

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listed in Table 2.

Reply: We apologize for this error and will revise the incorrect description to read "the enrichment factors (EFs) of Al, Fe, and Mg were lower than ten on ND days and decreased to less than three on dust days. These data are indicative of the primarily crustal origins of these elements. Furthermore, the EF of Ca was 14.0 on ND days, which indicated that Ca had a partially anthropogenic source on dust days".

Q9.L171: Again, I cannot follow the calculation of “1.7-21.9 times (mean: 6.9)“.

Reply: We apologize for the confusion. The calculation method is the same as that for TSP (see the reply to Q7). The correct concentrations and the ratios of samples on dust days are listed in Table S2.

Q10.L173-L174: To clarify the separation of dust and non-dust days, the information of criterion for samples on non-dust days will be needed. Reply: As discussed above for Q1, the information will be supplemented in Section 2.1.

Q11.L175: I cannot follow “10.3 times” for Fe. It can be calculated as 7.90 from the values in Table 2.

Reply: The calculation method is the same as that for TSP (see the reply to Q7). The concentrations and corrected mean ratios of samples on dust days are listed in Table S2.

Q12.L175: In Figure 2, nss-Ca was shown, but nss-Ca was not listed in Table 2. What is the authors intention to introduce nss-Ca here?

Reply: Follow others’ study, we calculated the EF of Ca in Table 2. The EFs of Ca on ND days indicated that Ca was affected by anthropogenic sources. nss-Ca usually was used as a typical dust index. Therefore we showed the nss-Cain Fig.2 and discussed the influence of dust on crustal elements using nss-Ca.

Q13.L176: “3.6-fold” will not be followed from Fig. 2. It should be listed in Table 2.

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Reply: The calculation method is the same as that for TSP (see the reply to Q7). The concentrations and the corrected mean ratios of samples on dust days are listed in Table S2.

Q14.L177: The EF of Ca on dust days is also greater that 10.

Reply: The EF of Ca was 14, not much greater than 10, indicating that the Ca was mainly from a natural source mixed with an anthropogenic source.

Q15.L183: The increasing ratio of concentration between dust days and non-dust days will be helpful to understand the discussion on Section 3.1.

Reply: We will replace the times with ratios in our revised manuscript.

Q16.L189: What is the comparison method on some dust days? The sample date are shown in Figure 3, so why the authors explicitly mention the date? I cannot follow the calculation of “a factor of 1.2-5.7 “.

Reply: It will be revised as “The concentrations of ammonium were increased by 20

Q17.L190: What means “less than 20

Reply: We apologize for the confusion. The sentence has been revised to read " The concentrations of ammonium were increased by 20

Q18.L191: Again, what is the comparison method on some dust days? I cannot follow the calculation of “a factor of 1.4-9.2 “.

Reply: The calculation method is the same as that for ammonium (see the reply to Q16). The concentrations and the increasing factors of samples on dust days are listed in Table S2.

Q19.L194-L195: In this sentence, the authors stated “the effect of dust on inorganic nitrogen differed during different types of dust events“. Why the authors suddenly focused on inorganic nitrogen here?In L192-193, it was mentioned “inorganic ion SO42-

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exhibited concentration variations that were similar to those of nitrate". L197: The figures for inorganic nitrate will be helpful information here, if the authors focused on inorganic nitrogen.

Reply: The part will be revised as "Similar to ammonium, nitrate concentrations were sometimes increased by a factor of 1.4-9.2 relative to the comparison sample while they were decreased in others. Unlike substantially increased concentrations of crustal metal elements in dust samples, the concentrations of IN were likely determined by meteorological conditions as well as surface areas provided by dust particles."

Q20.L207: (respectively less than 50 ug/g and 6 ug/g) will be the correct expression for ammonium.

Reply: We have incorporated this suggestion.

Q21.L211: So what is the source of atmospheric particulate nitrogen? The location of Duolun and Zhurihe Sand Desert is very close.

Reply: Duolun and Zhurihe belong to the Hunshandake Desert in Inner Mongolia, one of the main Chinese sand deserts. According to studies, the Yellow Sea is mainly affected by dust storms from this sand source with a probability of 52

Zhang, Z K., and Gao, H.: The characteristics of Asian-dust storms during 2000–2002: From the source to the sea, Atmospheric Environment, 41, 9136-9145, 2007. Gao, Q X., Ren Z H. : Dust events and its impacts on atmospheric environment, Science press, Beijing, 2010.

Q22.L214-L216: Without more information of the intensity of dust, the discussion on 'dilution effect' seems to be lacked in scientific understanding. This part should be fully revised based on not only dilution effect but also dust intensity.

Reply: As discussed above, we will add modeling results of dust distribution to support our analysis in revision.

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Q23.L217: Averaged information were listed here, however, will the each sample information be valuable? The equation shown in summary column cannot be understood form (e.g., IN and ND were not comparable index).

Reply: Thank you for the suggestion. According to the suggestion, we revised Table 3 and listed the sample information.

Q24.L219: It seems that the discussion on this paragraph (e.g., "700 ug/m3 in Case 1" and "higher than 1100 ug/m3 in Cases 2 and 3") are based on Table 3. Please reorganize the paragraph, or please refer appropriate information here. It is hard to follow these values.

Reply: We will revise this paragraph and refer to the appropriate information in the revised manuscript according to revised Table 3.

Q25.L219-L222: So what is the local source? What is the definition of the wording of "local" here? There was no information of the emissions here. It is hard to understand the "reaction" without the information of emissions intensity around dust source and downwind regions.

Reply: Local source refers to the gas or particle emissions from a local pollutant source, such as industry emission, coal burning, vehicle exhaust and agricultural activity, in the downwind region during the dust transport, which is not from the dust event itself. As we discussed above, the NOx and NH3 emissions increase greatly from the dust source region to the downwind region (see the reply to Q2). We have supplemented the modeled emissions intensity of NOx and NH3 in the revised manuscript.

Q26.L224: "particle" is "TSP"?

Reply: We apologize for the confusion. We will revise "particle" to read "total suspended particles".

Q27.L227-L228: The favorable condition to form ammonium cannot be discussed without the information of NH3. In addition, Table 3 indicated the aerosol samples in the

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coastal region of the Yellow Sea. How about the status over air mass path? Is it sufficient to conclude only from the downwind information to the formation of inorganic nitrogen?

Reply: We will add modeling results using a 3-D air quality model to support our analysis in revision.

Q28.L230: “strong dust storm” cannot be discussed without any information on dust intensity here.

Reply: We will add modeling results of dust distribution to support our analysis in revision.

Q29.L233-L234: But NOx concentration was high in Case 3. I cannot follow why the authors concluded “the strong dilution effect” on Case 3.

Reply: Among three cases, the NOx concentration was the highest with an average value of 70.7μgâĂćm-3 for Case 3 and increased by 17.8

Q30.L244-L246: Because the Table 5 was lack in the information of ND days, we cannot follow the authors conclusion. The information of ND days on Table 5 will be required.

Reply: We have supplemented the information for ND days in Table S1 and S2.

Q31.L254-L255: The authors simply mentioned “local emissions” here. Because the samples were collected on downwind regions in the coastal region of the Yellow Sea, I guess that the discussion on emission characteristics of each (or, at least, some categorized) air mass should be discussed in detail. The inorganic nitrogen concentrations are highly related to the local conditions both on emissions strength and meteorological parameters, so the discussion only on air mass speed and air mass path over ocean are insufficient.

Reply: As discussed above (see the Reply to Q2) , We will add modeling results using

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a 3-D air quality model to support our analysis in revision.

Q32.L256: RH and NOx information are not shown in Table 5. Reply: We apologize for the mistake. We have revised the title of Table 5.

Q33.L260: The colors are overlapped, hence we cannot distinguish each trajectory. Some paths (e.g., thick green color: 2008/5/22 or 2011/4/15) are apparently indicated the west or south part of China. Are these events really related to dust events?

Reply: We apologize for the confusion. We have provided all trajectories of samples collected on dust and non-dust days. Fig.4 has been redrawn to distinguish each trajectory for samples collected on dust and non-dust days.

Q34.L278-L280: The source of coal combustion have increased compared to non-dust days. Short explanation will be needed here.

Reply: The source of coal combustion on dust days became complex. The source profile showed high percentages 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, such as industry and building dust. This source increased due to the coal combustion emissions mixing with other uncertain sources emitted into the air in strong winds.

Q35.L305: If the authors discuss the dry deposition flux of “IN”, the information should be inserted in Table 7. Table 7 only contained NO3- and NH4+ independently.

Reply: We inserted the flux of IN in Table 7 and corrected several mistakes.

Q37.L306: I cannot follow the calculation of “a factor of 1.1-5.8” and “a factor of 1.8-6.3”.

Reply: These factors were the flux ratio of each dust sample in Case 1 to the ND average. The flux and ratio of each sample are listed in Table S3. We recalculated the increasing factors according to the revised values. The sentence was revised to read

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"Compared with the average flux on ND days, the dry deposition flux of IN increased by a factor of 1.1-3.9, and the flux of atmospheric particles (TSP) increased by a factor of 1.8-6.3 in Case 1"

Q38.L307: “the dry deposition flux” of what?

Reply: We apologize for the mistake in the revision. The passage has been revised to read "the dry deposition flux of atmospheric particles (TSP)".

Q39.L309: What is the calculation method of “63

Reply: We apologize for the mistake. The sentence has been revised to read "Compared with the average dry deposition flux on ND days, the average nitrate flux of samples in Cases 2 and 3 decreased by 73

Q40.L310: What is the calculation method of “14

Reply: We corrected the calculation error and revised this sentence to read "Additionally, the average ammonium flux decreased by 47

Q41.L317: I cannot follow the calculation of “a factor of 2-25”.

Reply: The factor was calculated by comparing the flux of the sample on dust days with the average Fe flux on ND days (see Table S3).

Q42.L339: “aerosol particles” is “TSP”? In Table 7, please confirm the significant digits for each specie.

ReplyïijŽWe apologize for the confusion. “aerosol particles” was revised to read “TSP”. The former digits were revised according to the editor’s suggestion. We will consider revising again to confirm the significant digits.

Q43. Technical Corrections: L31: Comma is needed on ‘2800’.

Reply: We have added a comma according to the suggestion.

Q44.L199: ‘IN’ should be defined in L194.

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Reply: Due to the very low concentration of nitrite, in this manuscript, IN represents inorganic nitrogen, mainly including nitrate and ammonium. We have provided this definition in L194.

Q45.L236: Need appropriate comma for all numbers. L301: Comma is needed on '2800±700'.

Reply: We have added a comma according to the suggestion.

Please also note the supplement to this comment:
http://www.atmos-chem-phys-discuss.net/acp-2016-1183/acp-2016-1183-AC1-supplement.pdf

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Interactive comment on Atmos. Chem. Phys. Discuss., doi:10.5194/acp-2016-1183, 2017.

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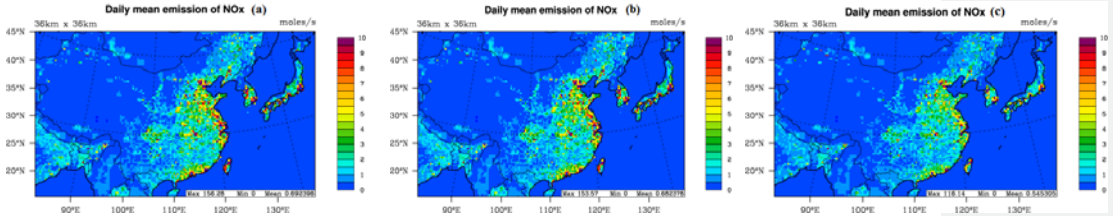

Fig. S1 Daily mean NOx emission in China on Apr.24th (a), 25th (b) and 26th (c), 2008 (From Liu et al., 2010).

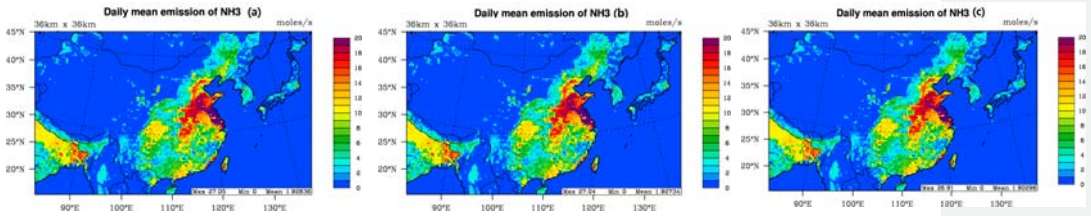

Fig. S2 Daily mean NH_3emission in China on Apr.24th (a), 25th (b) and 26th (c), 2008 (from Liu et al., 2010).

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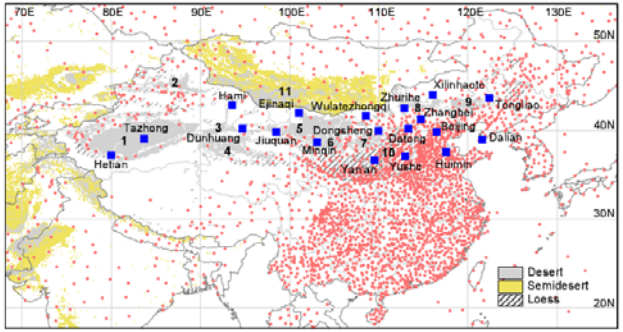

Fig. S3 Map of the main source regions of sand and dust storms in China (from Wang et al., 2008).

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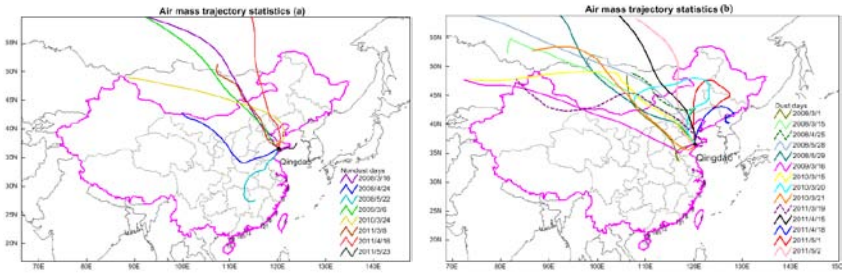

Figure 4. The 72-h backward trajectories for non-dust (a) and dust (b) samples from 2008 to 2011

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Table S1. 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 22th	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	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. 21th	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
		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 23th	Sunny day

[a]Note that one exterior dust sample was collected on 1 March when no dust was recorded by MICAPS. However, the MICPAS information indeed showed the dust events in Qingdao on 29 February and 2 March. Both the PM_{10} mass concentration and our measured Al in TSP on 1 March implied that the sample should be classified into dust sample.

[b] In addition, the sampling duration was reduced down to only 2 hrs because of extremely high particle loadings.

Fig. 4. Table

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Table S2. Sampling information for the aerosol samples collected at the Baguanshan site in thecoastal region of the Yellow Sea.

Sampling Month	Sample category	Sampling number	TSP		Al		Fe		nss-Ca		NH_4^+		NO_3^-	
			Concentration ($\mu g/m^3$)	Ratio of DD to NDS	Concentration ($\mu g/m^3$)	Ratio of DD to NDS	Concentration ($\mu g/m^3$)	Ratio of DD to NDS	Concentration ($\mu g/m^3$)	Ratio of DD to NDS	Concentration ($\mu g/m^3$)	Ratio of DD to NDS	Concentration ($\mu g/m^3$)	Ratio of DD to NDS
Mar., 2008	Dust days (DD)	20080301	526.7	2.3	21.3	2.4	14.1	2.2	25.2	1.8	12.7	1.5	20.5	1.6
		20080315	409.5	1.8	17.5	1.9	10.3	1.6	14.6	1.0	29.9	3.6	19. 5	1.5
	Non-dust days (NDS)	20080316	225.1		9.0		6.5		14.3		8.4		12.6	
Apr., 2008	Dust days (DD)	20080425	622.2	4.5	33.2	5.0	10.6	2.3	63.1	6.1	2.0	0.3	6.8	0.3
	Non-dust days (NDS)	20080424	137.5		6.6		4.7		10.4		7.2		21.7	
May, 2008	Dust days (DD)	20080528	2578.7	12.5	182.8	20.9	96.1	41.4	79.2	4.5	2.7	0.2	9.2	0.3
		20080529	2313.8	11.2	132.7	15. 2	70.5	30.4	46.8	2.7	4.8	0.3	17.5	0.6
	Non-dust days (NDS)	20080522	206.1		8.8		2.3		17.6		16.6		27.4	
Mar., 2009	Dust days (DD)	20090316	688.4	7.3	24.2	13.7	14.8	7.7	29.6	4.2	17.2	5.7	15.9	5.4
	Non-dust days (NDS)	20090306	94.3		1.8		1.9		7.0		3.0		2.9	
Mar., 2010	Dust days (DD)	20100315	501.1	1.8	25.6	2.7	14.8	2.0	15.2	0.9	4.3	1.8	5. 4	0.8
		20100320	3856.7	14.0	205.4	21. 9	116.3	15.3	151.1	8. 7	3.4	1.4	5.5	0.8
		20100321	518.6	1.9	25.8	2. 8	15.3	2.0	19.2	1.1	9.4	4.0	16.5	2.3
	Non-dust days(NDS)	20100324	274.8		9.38		7.6		17.4		2.4		7.2	
Mar., 2011	Dust days (DD)	20110319	938.6	4.8	39.3	7.0	26.3	6.0	21.2	1.7	9.4	0.7	12.3	0.9
	Non-dust days (NDS)	20110308	194.1		5.6		4.4		12.4		13.1		13.0	
Apr., 2011	Dust days (DD)	20110415	1224.6	4.9	52.8	4.8	35.3	5.7	41.6	4.4	25.0	4.7	51. 4	9.2
		20110418	557.9	2.2	26.6	2.4	22.3	3.6	14.2	1.5	6.6	1.2	3.8	0.7

Fig. 5. Table

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Table 3. Concentrations of inorganic nitrogen, TSP, NOx, Relative Humidity (RH) and T for aerosol samples of different casein the coastal region of the Yellow Sea.

	Sample number	TSP μg·m^{-3}	NO_3^- μg·m^{-3}	NH_4^+ μg·m^{-3}	RH %	T ℃	NOx μg·m^{-3}	Summary
Case 1	20080301	527	20.5	12.7	57	7.0	36	IN concentration on 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	
	20110415	1224	51. 4	25.0	34	22.3	68	
	20110502	810	21.0	11.0	49	17.7	62	
Case 2	20080425	622	6.8	2.0	30	18.0	40	IN concentration on 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	
Case 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 S2.

Fig. 6. Table

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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							
	TSP	NO_3^--N	NH_4^+-N	IN	Fe	Cu	Pb	Zn
Case 1[a]	9600± 4300	87±53	28±16	114±64	650±340	2±1	0.3±0.2	6±3
Case 2[a]	18000± 11,000	13±18	8±5	21±22	1300±1000	3±2	0.08±0.04	4±1
Case 3[a]	29,000± 31,000	26±6	17±8	42±12	2100±2200	6±1	0.20±0.02	5±3
Non-dust	2800± 700	48±33	15±8	63±39	190±110	1±1	0.09±0.1	5±4

[a] The characterizations of IN concentrations and sample information of the Cases are provided in Table 3.

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Table S3. Dry deposition flux of aerosol particles (mg/m^2/month), particulate inorganic nitrogen (mg N/m^2/month) and some toxic trace m
non-dust days

Case	Sample number	Particles (TSP)		NO_3^--N		NH_4^+-N		IN		Fe		Cu		P
		Flux	Ratio[a]	Flux	Ratio[a]	Flux	Ratio[a]	Flux	Ratio[a]	Flux	Ratio[a]	Flux	Ratio[a]	F
Case 1	20080301	7680	2.7	78.4	1.6	23.8	1.6	102.2	1.6	509	2.7	2.1	1.7	0.
	20080315	5207	1.9	62.1	1.3	47.7	3.2	109.8	1.8	295	1.6	1.5	1.2	0.
	20090316	9254	3.3	54.6	1.1	13.3	0.9	67.9	1.1	483	2.6	1.2	1.0	0.
	20100321	8049	2.9	67.7	1.4	19.2	1.3	86.9	1.4	588	3.1	1.2	1.0	0.
	20110415	17782	6.3	193.6	4.1	47.6	3.2	241.1	3.9	1260	6.7	4.0	3.2	0.
	20110502	9887	3.5	63.8	1.3	16.9	1.1	80.7	1.3	789	4.2	1.6	1.3	0.
	Average	9643	3.4	86.7	1.8	28.1	1.8	114.8	1.8	654	3.5	1.9	1.6	0.
Case 2	20080425	11356	4.0	5.2	0.1	5.1	0.34	10.3	0.2	424	2.2	1.7	1.4	0.
	20080528	31391	11.2	1.8	0.04	4.1	0.27	5.9	0.1	2631	13.9	6.0	4.9	0.
	20080529	28053	10.0	0.2	0.004	7.3	0.48	7.4	0.1	2020	10.7	2.0	1.6	0.
	20110319	12682	4.5	42.5	0.9	14.8	0.98	57.3	0.9	847	4.5	1.1	0.9	0.
	20110501	6340	2.3	14.3	0.3	8.5	0.56	22.8	0.4	454	2.4	2.0	1.6	0.
	Average	17964	6.4	12.8	0.27	8.0	0.53	20.7	0.33	1275	6.7	2.6	2.1	0.
Case 3	20100315	12174	4.3	32.2	0.7	23.6	1.6	55.8	0.9	666	3.5	6.8	5.4	0.
	20100320	65267	23.3	24.5	0.5	7.9	0.5	32.4	0.5	4675	24.7	5.0	4.0	0.
	20110418	10695	3.8	19.9	0.4	17.9	1.2	37.9	0.6	951	5.0	6.1	4.9	0.
	Average	29379	10	25.6	0.54	16.5	1.1	42.0	0.67	2097	11	5.9	4.8	0.
ND	HDT080316	2840		39.7		13.3		52.9		193		0.6		0.
	HDT080424	2851		102.6		17.9		120.5		199		0.8		0.
	HDT080522	2705		91.7		27.6		119.3		73		0.9		0.
	HDT090306	1596		13.2		6.8		20.0		110		0.9		0.
	HDT100324	3992		27.3		4.6		31.9		449		4.2		0.
	HDT110308	2573		43.6		22.8		66.3		135		1.0		0.
	HDT110416	3236		18.1		8.6		26.7		198		0.9		0.
	HDT110523	2658		44.4		18.7		63.1		156		1.2		0.
	Average	**2806**		**47.6**		**15.0**		**62.6**		**189**		**1.2**		**0.**

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