Dear Dr. Barbara 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 two reviewers' comments, we have made a substantial revision again to our manuscript, including reanalyzing the model results, enhancing our discussion and changing the figures. Additionally, this resubmitted version has been polished by English editor. We are confident that it is ready for publishing in a high quality journal. Detailed item-by-item responses to the comments are listed below.

Best regards,

Yours sincerely,

Jianhua Qi

Response to Referee report 1

General Comments: First of all, the space was not inserted appropriately in many parts, so it is hard to read and follow. Such a crude revision with low presentation quality should not be sent to reviewers. I have reviewed again this manuscript and found some improvements on the manuscript; however, many replies have not been found in the revised manuscript and/or replied well to my concerns. I feel that the presentation quality is still low as to be published from the high quality journal of Atmospheric Chemistry and Physics. With regret, I have judged to reject this manuscript again.

Response：We are sorry for the space missing problem. We have a double check for our submitted version, but not find this problem. We therefore re-install our software and avoid the problem in the new submission. In the new version, we further improve the quality of the manuscript according to two reviewers' comments. We are confident that it is ready for publishing in a high quality journal.

Q1. I have partly understood my concerning issue regarding the definition of dust event. Further concerning issue is the sampling duration of continuous dust event. Even the dust event continued multiple days, how should we consider the representativeness of the sampling? For instance, sample 20080528 and 20080529 (please note that the sampling time of 20080529 will have typo) had approximately one day interval. Was there large temporal variation of PM10 concentration during continuous dust days? If there was large change on PM10 concentration, why the authors collected on the listed time? The authors should state the reason, and should present the representativeness of 4 hrs sampling. In the revised manuscript, it will be kind for readers to explicitly state that 'http://www-cfors.nies.go.jp/~cfors/' is for forecast model over Asia, and 'http://www.qepb.gov.cn/m2/' is for observed concentration at Qingdao.

Response: Due to no dust events lasting over 12 hrs (Lee et al., 2015; Su et al., 2017; Zhang et al., 2007), we collected one dust sample with a 4-hr duration in a day. The sampling for dust particles started only when the measured PM10 mass concentration in Qingdao (http://www.qepb.gov.cn/m2/) and the forecasted dust mass over Asia (http://www-cfors.nies.go.jp/~cfors/) had greatly increased.

The samples with ID of 20080528 and 20080529 were subject to two different dust events occurring in two days instead of continuous samples for one dust event. On March 20-21, 2010, two dust events subsequently swept Qingdao. The 4 hr dust samples with IDs of 20100320 and 20100321 may not capture the entirety of the two events. However, the on-line data can allow adequate separation of the two dust event samples. The same was true for the dust samples with IDs of 20110501, 20110502.

The link illustration for these two links has been also added properly in the new version.

Lee, Y. G., Ho, C., Kim, J., and Kim, J.: Quiescence of Asian dust events in South Korea and Japan during 2012 spring: Dust outbreaks and transports, Atmos. Environ., 114, 92-101, 2015.
Su X., Wang Q., Li Z., Calvello M., Esposito F., Pavese G., Lin M., Cao J., Zhou C., Li D., Xu H. Regional transport of anthropogenic pollution and dust aerosols in spring to Tianjin — A coastal Su, X., Wang, Q., Li, Z., Calvello, M., Esposito, F., Pavese, G., Lin, M., Cao, J., Zhou, C., Li, D., and Xu, H.: Regional transport of anthropogenic pollution and dust aerosols in spring to Tianjin — A coastal megacity in China, Sci. Total. Environ., 584–585, 381–392, 2017.
Zhang, K., and Gao, H. W.: The characteristics of Asian-dust storms during 2000–2002: From the source to the sea, Atmos. Environ., 41, 9136-9145, 2007.

Q2. I have partly agreed, but I have further question on the application of a 3-D air quality model. First, what is the merit of the application of 3-D air quality model? In the revised manuscript, only the spatial distributions of PM10 were shown (Fig. 2 from CFORS model and Figs. S1-S3 with CFORS and WRF-CMA. Can such application reinforce the authors' discussion points? The behavior of IN were discussed in this manuscript, so what is the purpose to show PM10? The authors stated that '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)' (L199-201). If the consistency between other models is important, why the author calculated on your own model? I cannot follow this reason from the revised manuscript.

Response: Thank you for the suggestion. We have deleted the results by CFORS. The CMAQ model (v5.0.2) was applied to simulate the concentration of PM10, NOx, NH_3, NO_3^- and NH_4^+ over the East Asia area for aerosol samples on dust and comparison days. We have revised the discussion on model results. Distribution of PM10 was used to characterize the dust events. Spatial distributions of PM10 during each dust events were consistent with the records in the "Sand-dust Weather Almanac" (CMA, 2009; 2010; 2012; 2013). The model results indicated that CMAQ results reasonably reproduce the mass concentrations of NO_3^- (Fig. S6). Simulated NH_4^+ concentrations in dust samples were severely under-predicted with NMB values at -71%. For reference samples, simulated NH_4^+ concentrations sometimes can well reproduce the observational values, but sometimes totally off. The external mixing mechanism proposed in this study is urgently needed to be included in the model for accurately predicting the concentrations during dust events.

CMA: Sand-dust weather almanac 2008, China Meteorological Press, Beijing, 10-64, 2009.
CMA: Sand-dust weather almanac 2009, China Meteorological Press, Beijing, 11-59, 2010.
CMA: Sand-dust weather almanac 2010, China Meteorological Press, Beijing, 11-79, 2012.
CMA: Sand-dust weather almanac 2011, China Meteorological Press, Beijing, 10-53, 2013.

Q3. The following specific points also should be revised to clarify the model application.
L189: Centered point is needed because we cannot follow the modeling domain at the current description.

L193: On the INTEX-B emission inventory (Zhang et al., 2009), I suppose that NH3 emissions have not been provided. If so, this description should be changed.
L195, and Figures 6 and 8: So, all calculations were based on the emission level on 2008? Because the temporal resolution of INTEX-B emission inventory is month, I feel that there are no need to display all emissions on all dust samples. These emissions level should be differed only on month. Therefore, I suppose that the averaged (spring time) emissions of NOx and NH3 on each one figure is enough.

Response: We have supplemented the centered point (110°E, 34°N) in the new version.

According to the publications of INTEX-B and TRACE-P Asia emission inventories (Zhang Q et al., 2009; Streets et al., 2003.), INTEX-B inventory was developed based on TRACE-P inventory with NH_3 emission considered (the annual emission amount of NH_3 in China was 13.6 Tg). However, due to the low priority and low variability of NH_3 emission during 2000-2006, NH_3 emission was not updated in INTEX-B inventory, and the NH_3 emission in INTEX-B inventory was consistent with TRACE-P.

Agree and revised.

Zhang Q et al., Asian emissions in 2006 for the NASA INTEX-B mission; Atmos. Chem. Phys., 9, 5131-5153, 2009.

Streets D.G. et al., An inventory of gaseous and primary aerosol emissions in Asia in the year 2000, 108 (D21), DOI: 10.1029/2002JD003093, 2003.

Q4. Figure S1: What is the purpose to show the difference between (b) and (c)? In this caption, what is ‘WRF-CMA’?

Response: We have indicated that one exterior dust sample was collected on 1 March when no dust was recorded in Qingdao by MICAPS. However, the MICPAS information over the whole country indeed showed the dust events in China on 1 March. And the modeled spatial distribution of PM10 and TSP mass concentration for this dust event on 1 March implied that the sample should be classified into dust sample. Therefore we listed all the supporting figures in Fig. S1. Fig. S1 (b) was the weather information from the MICAPS at 8:00 on Mar.2, 2008 and (c) was hourly PM10 concentration modeled by the WRF-CMAQ model at 15:00 on Mar.1, 2008. We guessed that the reviewer maybe refer to the difference between (c) and (d), therefore we deleted (d).

We have revised the caption.

Q5. Figure S3: In the main manuscript, it was stated that ‘each dust sampling day are shown’ in Fig. S3 (L218-219, L895). However, only the hourly concentration of PM10 concentration at 14:00 on 19 Mar 2011 were shown. Please confirm this supplemental figure.

Response: We really modeled the PM10 concentration on each dust sampling day, but

only showed the PM10 concentration at the middle time of the sampling in Fig.S3 (Now Fig.S5 in the new version) due to too many figures. We have revised this section and the sentence has been revised into "The concentrations of PM10 and its major components NO_3^- and NH_4^+ over East Asia on dust days and comparison days were modeled using the WRF-CMAQ model (Fig. S5-6)" in L341-342 in the new version.

Specific comments:
Q6. 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.
Re-comment: cannot find this revision.

Response: The revision in the last round was prepared after the quick response. After a careful consideration, we agreed with the comment and delete the part in the last revised version.

Q7. I have confirmed and understood the meaning. However, is this revision corresponded to L226-230? If so, this revised sentence seems to contain many errors (NOT Table S2 but Table S1?). For example, we can find 410 μg/m3 on dust day sample on 20080315. What is the value of 80-1303%? These increased value were not corresponded to 'Ratio of DD to CS' shown in Table S1.

Response: Yes, this revision corresponded to L226-230 in last version. And the times of dust to non-dust day samples were replaced by the ratio according to the former suggestion. To avoid the confusion, we have revised Table S1 to give the increased ratio.

Q8.L171: Again, I cannot follow the calculation of "1.7-21.9 times (mean: 6.9)".
L175: I cannot follow "10.3 times" for Fe. It can be calculated as 7.90 from the values in Table 2.
L176: "3.6-fold" will not be followed from Fig. 2. It should be listed in Table 2.
Re-comment: So, in this revised manuscript, these statements of the increment ratio on dust-day compared to non-dust day have not been explicitly appeared. In L243, the authors stated 'Table S1', but Table S1 contained not only the information of inorganic nitrogen but also TSP, Al, Fe, and nss-Ca. So, it is appropriate to mention on Table S1 in Section 3.1.

Response: Agree and revised.

Q9.L175: So, please state explicitly regarding this point to the readers. In the current form, nss-Ca was suddenly shown in Fig. 3 without any introduction.

Response: Agree and revised.

Q10. First of all, I cannot find the revision of 'The concentrations of ammonium were increased by 20' anywhere. Is this corresponded to Table S1? I suppose that the

authors discussed regarding this point in L240-L244. Although ratio was shown in Table S1, percentages are discussed here. So it is hard to follow the manuscript. Why the discussion point have not been arranged on the uniformed unit?

Response: Agree and revised.

Q11.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 SO_4^{2-} 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.
Re-comments: I cannot find this revision.

Response: The revision in the last round was prepared after the quick response. After a careful consideration, we completely rewrote the part to avoid confusion in the last revised version.

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Q12. L207: (respectively less than 50 ug/g and 6 ug/g) will be the correct expression for ammonium.
Re-comments: I cannot find this revision.

Response: The revision in the last round was prepared after the quick response. After a careful consideration, we completely rewrote the part into "The ratios of mass concentrations of nitrate and ammonium to the total mass of sand particles were very low, i.e., less than 81 μg/g, which are approximately three orders of magnitude less than the corresponding values in our dust samples." at L230-232.

Q13.I am wondering that the differences of IN concentration between Duolun and Zhurihe. Both are Hunshandadke Desert, however, as is shown in Table 5, IN concentration was much higher in Duolun. Are there some emission source?

Response: Sand samples were collected at a remote site in Zhurihe desert. Little anthropogenic influence is expected. Atmospheric aerosol samples were collected at an urban site in Duolun on dust days for comparison. It is not surprised for a strong signal for anthropogenic sources. This has been clarified in the new version.

Q14. Again, only from the dust spatial distribution, it is hard to state the dust intensity.

Response: We had made a substantial revision on the part in the last round revision and didn't consider dust intensity as an important factor for our unique results.

Q15. 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.
Re-comments: Again, only from the dust spatial distribution, it is hard to state the dust intensity.

Response: We had made a substantial revision on the part in the last round revision and didn't consider dust intensity as an important factor for our unique results.

Q16. I have confirmed the revision, but if the authors discussed on average (L300-303), the averaged values were also needed.

Response: We really had given the average of TSP in form of average±standard deviations at L300-303 in last revision. Now we had made a substantial revision on this part and didn't discuss TSP average concentration.

Q17. 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 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?
Re-comments: Again, I cannot understand the model application results.

Response: We modeled the emission and concentration of NOx and NH_3 over East Asia on the dust and comparison days. The model results showed that the calculated trajectories of the entire dust air mass passed over those highly polluted regions with strong emissions of NO_x and NH_3 shown in Fig 6 and experienced different residence times therein. The average concentration of NO_x and NH_3 during transport were calculated and discussed according to Categories 1 and 2. The air masses in Category 1 took over 11-39 hrs to cross over the highly polluted area with appreciable concentrations of NOx (5.7±1.4 ppb) and NH_3 (7.6±3.3 ppb). Except for the exterior samples, air masses in Category 2 took less than 10 hrs to cross over the polluted areas with lower concentrations of NOx (3.6±3.4 ppb) and NH_3 (4.7±4.7 ppb) and the mixing layer height along the route was 916-1194 m (on average) for each dust event. This further led to the external mixing of anthropogenic particulate matters and dust.

Q18. L230: "strong dust storm" cannot be discussed without any information on dust intensity here.
Re-comments: Again, from the additional information of CFORS, the spatial distribution pattern was found; however, how can we estimate the intensity?

Response: We had made a substantial revision on the part in the last round revision and didn't consider dust intensity as an important factor for our unique results.

Q19. L233-L234: But NOx concentration was high in Case 3. I cannot follow why the authors concluded on Case 3.
Re-comments: So where did the authors discussed the NOx concentration in the manuscript?

Response: We had made a substantial revision on the part in the last round revision after the quick response. The NO_x concentration was discussed in Section 4.3 in the new revision.

Q20. 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.
Re-comments: Again, model is used only for spatial distribution and not inform the chemical production process.

Response: We had made a substantial revision according to the suggestion. The chemical production process was discussed in Section 4.1 "Theoretical analysis of the three categories". In Category 1, ammonium salt aerosols may externally exist with dust aerosols in these dust day samples and NO_3^- and SO_4^{2-} were almost completely associated with NH_4^+ in these dust day samples; whereas a larger fraction of NO_3^-+SO_4^{2-} may exist as metal salts due to reactions of their precursors with dust aerosols in Category 2. The simulated NO_3^- and NH_4^+ concentrations was compared with the observation in Qingdao, and the results indicated that the external mixing mechanism proposed in this study is urgently needed to be included in the model for accurately predicting the concentrations during dust events.

Q21. I have rechecked the discussion of backward trajectories discussed on Section 3.4. There are many points should be clarified.
Figure 5: Please add the explanation of the trajectory of 20110415 was excluded based on the discussion on Fig. 2. Why the authors displayed "non-dust samples"? What were the differences between non-dust and dust samples trajectories? I feel that these were similar.

Response: Agree and revised.

Q22. L314: What is the 'remaining one'? Please specify the trajectory data. In my opinion, two trajectories of 20110418 and 20110501 originated from northeast China.

Response: Yes, trajectory 20110501 was really from northeast China, however it then passed over the Inner Mongolia, and arrived at Qingdao from north, just like 20110502. Therefore, we grouped the trajectory into the air mass originated from Inner Mongolia, China. However, it was really easy to mislead the readers. Therefore, we accepted the suggestion, and revised the sentences into "The calculated air mass trajectories for 13 out of 14 samples showed that the air mass originated from North and Inner Mongolia, China (Fig. 5), generally consistent with the results of Zhang and Gao (2007). The remaining one, with ID of 20110418 originated from Northeast China.".

Q23. L317: What is the 'one exterior sample'? Please specify.

Response: Agree and revised.

Q24. L319: I cannot see ‘the air masses crossed over the sea for 94-255km’ from Figure 5, because Fig. 5 showed the whole view of trajectories across China. More detailed figure or explanation will be required.

Response: The distance over sea of the air mass for each sample was measured from the trajectory using TrajStat software (Wang et al., 2009).We have added the explanation in Section 2.3.

Wang, Y. Q., Zhang, X. Y., and Draxler, R. R.: TrajStat: GIS-based software that uses various trajectory statistical analysis methods to identify potential sources from long-term air pollution measurement data, Environ. Modell. Softw., 24, 938-939, 2009.

Q25. L328-329: What is the definition of the ‘average mixing layer’. I suppose that the altitude of backward trajectories were so high because most of trajectories were originated outside China on 72 hrs. So, where is the averaged region to calculate ‘900m’ in this sentence?

Response: The average mixing layer was calculated as an average of all points on the air mass back trajectory of each sample. This has been clarified in section 2.4 in the new version.

Q26. I cannot still understand the authors’ conclusion here. As was discussed on L355-361, Fig. 8, and Table 7, although the coal combustion have increased on dust days, the contributions of local anthropogenic sources (especially secondary aerosols) have decreased on dust days. According to the discussion on Section 3.3, the authors concluded that ammonium salts were externally co-exist with dust aerosols in Category 1. So, why the contribution of secondary aerosols were decreased from PMF analysis. I feel that these results have contradicted. More careful discussion is required for this conclusion.

Response: The source of coal combustion on dust days became complicated. “mixture of coal combustion and other pollutants” means these compounds present contemporaneously, because that PMF model can’t show the mixing or existing state. We have revised the sentence into “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 coal combustion presenting contemporaneously with other pollutants emitted along the transport path on dust days.”. Ammonium salts were externally co-exist with dust aerosols in Category 1, but showed lower concentrations in Category 2 likely due to unfavorable conditions for forming ammonium salts. Here the conclusion was a result of source appointment for all dust samples including Category 1 to 3. And we have revised the sentence “In these dust samples, including Categories 1-3, oil combustion, industry, soil dust, secondary aerosols, and coal combustion/other sources were identified as five major sources (Table 6).

Q27.L306: I cannot follow the calculation of “a factor of 1.1-5.8” and “a factor of 1.8-6.3”.
Re-Comments: I cannot find this revision.

Response: We had made a revision on these sentences in the last round revision after the quick response. According to the former suggestion, we changed the factor to ratios. And this sentence was revised to “In Category 1, the dry deposition fluxes of $N_{NH4+ +NO3-}$ increased by 9-75% with increased TSP flux by 86-252% (Table S3)” at L371-372 in the new revision. And we also revised Table S3 to give increased proportion and the calculation method.

Q28. L309: What is the calculation method of “63%” and “46%”?
L310: What is the calculation method of “14%” ?
Re-Comments: I cannot find this revision.

Response: We had made a revision on these sentences in the last round revision after the quick response. We have revised Table S3 to give increased proportion and the calculation method.

Q29. L317: For Fe, it seems that the increased ratio were 2.81-11.08 from Table S3.

Response: This sentence has been revised to “However, the dry atmospheric deposition fluxes of Fe increased by a factor of 124-2370% in dust day samples.” at L383-384 in new revision.

Specific comments:
Q30.Table 6: Missing the note of a and b.

Response: Done.

Q31. Table S2: Please align the right-column, it is hard to follow. What is the meaning of *?

Response: Agree and revised.

Response to Referee report 2

This manuscript focuses on an important issue, specifically the relationship between ammonium and nitrate with mineral dust in China. The variability in previously reported relative concentration trends is worth exploring in detail. There are a number of areas where the paper could use improvement prior to publication, particularly with connecting to the literature and placing the work in context.

Response：We thank the reviewer's constructive comments and revise our manuscript accordingly. We are confident that it is ready for publishing in a high quality journal.

Comments:

Q1 - Throughout the manuscript there are odd spacing issues, where two words are together without a space. As an example in the abstract "For these two groups, NH4+in dust day samples 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."

Response: We are sorry for the space missing problem. We have a double check for our submitted version, but not find this problem. We therefore re-install our software and avoid the problem in the new submission.

Q2 - Line 51 - 54: The authors state the Asian dust has been transported as far as the north pacific, but this understates what has been observed for Asian Dust. Uno et al 2009 Nat Geosci showed that Asian dust can circumnagivate the globe. showed Asian dust impacting California air quality, while Ault et al. 2011 JGR and Creamean 2013 Science showed impacts of Asian dust on orographic precipitation in the Sierra Nevada (in California). Pratt et al. 2009 Nat Geo showed Asian dust influencing clouds over Wyoming.

Response: Thank you very much for the suggestion. We revised the sentence to "Asian dust has been reported to not only frequently cross over the mainland and the China Seas, but also to occasionally reach the remote northern Pacific Ocean or North America (Creamean 2013; Tan and Wang, 2014; Van Curen and Cahill, 2002; Zhang and Gao, 2007). In an extreme case, Asian dust was found to be transported more than one full circuit around the globe in approximately 13 days (Uno et al 2009)." at Line 45-49.

Creamean, J. M., Suski, K. J., Rosenfeld, D., Cazorla, A., DeMott, P. J., Sullivan, R. C., White, A. B., Ralph, F. M., Minnis, P., Comstock, J. M., Tomlinson, J. M., Prather, K. A.: Dust and Biological Aerosols from the Sahara and Asia Influence Precipitation in the Western U.S., Science, 339, 1572-1578, 2013.

Tan, S. C., and Wang, H.: The transport and deposition of dust and its impact on phytoplankton growth in the Yellow Sea, Atmos. Environ., 99, 491-499, 2014.

VanCuren, R., and Cahill, T.: Asian aerosols in North America: Frequency and concentration of fine dust, J. Geophys. Res., 107(D24), 4804, doi:10.1029/2002JD002204, 2002.

Zhang, K., and Gao, H. W.: The characteristics of Asian-dust storms during 2000–2002: From the

source to the sea, Atmos. Environ., 41, 9136-9145, 2007.

Q3 Though the authors note that a native English speaker was utilized for the revision, a considerable improvement in the grammar and proofreading are needed before the writing is at a publishable level.

Response: This resubmitted version has been polished by English editor.

Q4 Line 233 insert comma after “samples”

Response: Done.

Q5 Line 266: Is it really a safe assumption that gas aerosol thermodynamic equilibrium is met for inorganic ions during a dust storm? It would seem that many non‐aqueous (i.e. solid) aerosol would be present that would not have normal equilibrium partitioning. It would be nice to see some evidence of this. This would also help support the conclusion that Ca(NO3)2 and CaSO4 are negligible.

Response: According to this reviewer’s suggestion, the progresses on this issue in literature have been summarized and cited to support our analysis.

Q6 Line 282: The presence of Cu, brings to mind the question of transition metal ions and industrial sources of metal containing particles. How were these accounted for? Particularly since they often have different properties and propensity for generating ROS as Weber and company at Georgia Tech have shown.

Response: Cu was once used as an effective marker of diesel and biodiesel-blend exhaust (Gangwar et al., 2012), while it can also be derived from copper pyrites ($CuFeS_2$) in Inner Mongolia mines (Huang et al., 2010). The increase of Cu in the mass concentration in dust samples implied dust particles mixed with anthropogenic particles, particularly from industrial emissions, during transport.

Gangwar, J. N., Gupta, T., and Agarwal, A.K.: Composition and comparative toxicity of particulate matter emitted from a diesel and biodiesel fuelled CRDI engine, Atmos. Environ., 46, 472-481, 2012.

Huang, K., Zhuang, G., Li, J., Wang, Q., Sun, Y., Lin Y., and Fu J. S.: Mixing of Asian dust with pollution aerosol and the transformation of aerosol components during the dust storm over China in spring 2007, J. Geophys. Res-Atmos, 115, D00k13, Doi:10.1029/2009jd013145, 2010.

Q7. ‐ It should be noted that there is a great deal of uncertainty regarding aerosol pH, particularly in North China, with estimates ranging from 3‐7 pH units. This of course will affect nitrate. The authors could comment on this with respect to their data, though keeping in mind Hennigan et al showing the proxy methods such as NH4+/(NO3‐+SO42‐) are qualitative at best.

Response: We thank the comments. We are not sure whether the estimated aerosols pH from 3‐7 pH units in north China were valid or not, by considering three types of

aerosols, i.e., ammonium salt aerosol, K_2SO_4 or KNO_3 aerosol, and $CaCO_3$. TSP was collected in this study while PM2.5 was used for analysis by Hennigan et al (2015). It is not surprised parts of NO_3^- and SO_4^{2-} to be associated with metals in TSP samples, but we agree that NO_3^- and SO_4^{2-} may overwhelmingly associated with NH_4^+ in PM2.5 as found by Hennigan et al (2015). However, we tried our best to properly interpret the formation of nitrate and sulfate in different Categories in the revised version.

Hennigan, C. J., J. Izumi, A. P. Sullivan, R. J. Weber, and Nenes, A.: A critical evaluation of proxy methods used to estimate the acidity of atmospheric particles, Atmos. Chem. Phys., 15(5), 2775–2790, 2015.

Q8. What is the mineralogy of the Hunshandake Desert? Is it rich in CaCO3? Based on a few assumptions made, documentation of the presence of this mineral from aerosols in the region would be helpful. Perhaps some of Ro and co‐workers analysis of transported dust with SEM‐EDX?

Response: Thank you for the suggestion. The references results show that mineral dust is relatively enriched with Calcite (Matsuki, et al., 2005; Formenti et al., 2011; Nie et al., 2012) and the carbonate content generally de-creases from west to east with exception of the Gurbantunggut desert in China desert (Fig.RS1, Formenti et al., 2011). Except for 20080502, the remaining dust samples in Category 2 were transported from the desert relatively enriched with $CaCO_3$ (1-25% in Wt%) (Formenti et al., 2011). And Huang et al. (2010) found that calcite was one of the main species in the aerosol over Duolun and the high content of $CaCO_3$ (~80% in dust storm days) in the total soluble part of Duolun aerosol. A positive correlation between NO_3^- and SO_4^{2-} in Category 2 against a negative correlation in Category 1 also implied that the dust particles enriched with $CaCO_3$ in Category 2 might play an important role to form SO_4^{2-} and NO_3^-. Ca-rich dust particles coated with highly soluble nitrate were observed at Kanazawa in Japan during Asian dust storm periods using SEM/EDX (scanning electron microscopy equipped with an energy dispersive X-ray spectrometer) (Tobo et al.，2010). The single-particle observation conducted by Hwang and Ro (2006) showed that $CaCO_3$ in dust particles was almost completely consumed to produce mainly $Ca(NO_3)_2$ species.

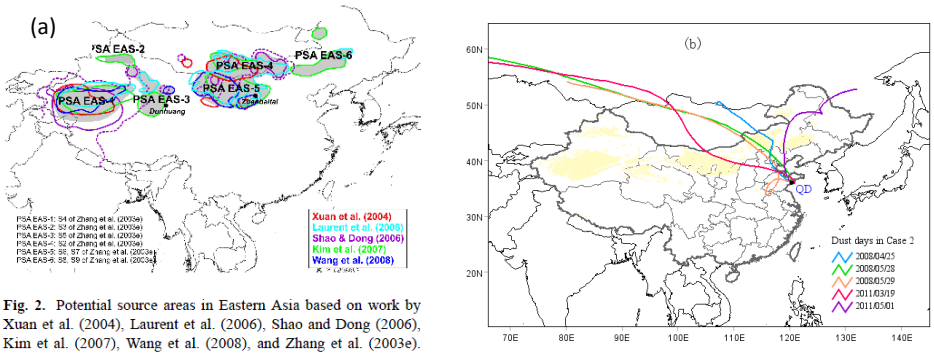

Fig. 2. Potential source areas in Eastern Asia based on work by Xuan et al. (2004), Laurent et al. (2006), Shao and Dong (2006), Kim et al. (2007), Wang et al. (2008), and Zhang et al. (2003e). Outlines of potential source areas (shaded areas) are drawn by hand. PSA EAS-1: Taklamakan; PSA EAS-2: Gurbantunggut; PSA EAS-3: Kumtaq, Qaidam, Hexi corridor; PSA EAS-4: Mongolian (Northern Gobi) deserts; PSA EAS-5: Inner Mongolian (Southern Gobi) deserts: Badain Jaran and Tengger (PSA EAS-5a), Ulan Buh, Hobq, Mu Us (PSA EAS-5b); PSA EAS-6: north-eastern deserts (Otindag Sandy Land, Horquin Sandy Land, Hulun Buir Sandy Land).

Figure RS1. Potential source areas in Eastern Asia adapted from Formenti et al., 2011(a) and The 72-h backward trajectories for samples in Category 2 (b).

Formenti, P., Sch ¨ utz, L., Balkanski, Y., Desboeufs, K., Ebert, M., Kandler, K., Petzold, A., Scheuvens, D., Weinbruch, S., and Zhang, D.: Recent progress in understanding physical and chemical properties of African and Asian mineral dust, Atmos. Chem. Phys., 11, 8231–8256, doi:10.5194/acp-11-8231-2011, 2011.

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Hwang, H. and Ro, C. U.: Direct observation of nitrate and sulfate formations from mineral dust and sea-salts using low-Z particle electron probe X-ray microanalysis, Atmos. Environ., 40, 3869-3880, 2006.

Matsuki, A., Iwasaka, Y., Shi, G. Y., Chen, H. B., Osada, K., Zhang, D., Kido, M., Inomata,Y., Kim, Y. S., Trochkine, D., Nishita, C., Yamada, M., Nagatani, T., Nagatani, M., and Nakata, H.: Heterogeneous sulfate formation on dust surface and its dependence on mineralogy: balloon-borne observations from ballon-borne measurements in the surface of Beijing, China, Water Air Soil Poll., 5, 101–132, 2005.

Nie,W., Wang, T., Xue, L. K., Ding, A. J., Wang, X. F., Gao, X. M., Xu, Z., Yu, Y. C., Yuan, C., Zhou, Z. S., Gao, R., Liu, X. H., Wang, Y., Fan, S. J., Poon, S., Zhang, Q. Z., and Wang, W. X.: Asian dust storm observed at a rural mountain site in southern China: chemical evolution and heterogeneous photochemistry, Atmos. Chem. Phys., 12, 11985-11995.

Tobo, Y., Zhang, D. Z., Matsuki, A., and Iwasaka, Y.: Asian dust particles converted into aqueous droplets under remote marine atmospheric conditions, PNAS Proceedings of the National Academy of Sciences of the United States of America, 107, 17905–17910, 2010.

Q8 - Line 321 some evidence for "humid marine conditions might have enhanced particle - particle coagulation" would be helpful. The number concentrations in the marine boundary layer are unlikely to be > 105 #/cm3 where coagulation is prevalent, more likely in the 102—103 #/cm3. Are the authors referring to fog - processing? That would seem to be the primary way this could happen in a marine environment.

Response: We are sorry for the confusion. The sentence has been revised to "On the other hand, the humid marine conditions (the average RH ranged in 50-75% over the Bohai and Yellow Seas in 2006-2012) might have enhanced hetero-coagulation between dust and smaller anthropogenic particles, leading to the release of NH_3 via reactions between preexisting ammonium salts and carbonate salts.".

Tobo Y., Zhang D., Matsuki A., Iwasaka Y. Asian dust particles converted into aqueous droplets under remote marine atmospheric conditions, PNAS, 2010, 107: 17905–17910.

Q9 - Line 326 The line "ammonium salts mostly co - existed with dust aerosols externally" is confusing as written. Is the population externally mixed with respect to ammonium nitrate and dust? Or are the salts co - existing with dust, but not other particle types? Please rephrase for clarity.

Response: Thank you for the suggestion. We have revised to "ammonium salt aerosols may externally exist with dust aerosols".

Q10 - Overall many of the conclusions on page 12 appear to mostly be speculation with little data to support it. I would recommend sticking to conclusions with more support from the data in the paper.

Response: We have made major revision on this section in the new revision.

Q11 - Line 357: The source profile for coal, could it have dust mixed in? When the author's say that there is a "mixture of coal combustion and other pollutants" are they saying that they are internally mixed or simply present contemporaneously? Clarifying that point would be helpful.

Response: "mixture of coal combustion and other pollutants" means these compounds present contemporaneously, because that PMF model can't show the mixing or existing state. We have revised the sentence into "The source profile for coal combustion in the dust day samples showed a high percentage of K^+, Cl^-, Ca, Mg, Co, Ni, As, Al and Fe, indicating that coal combustion particles may exist contemporaneously with other anthropogenic pollutants emitted along the transport path.".

Q12 - Overall the Figures could use improvement as portions are hard to read and the take home point of each is not always clear. It seems at times as if the authors are simply showing everything they can, as opposed to targeting their figure to the main points of the paper.

Response: Agree and revised.

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

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Abstract. Asian dust has been reported to carry anthropogenic reactive nitrogen during ~~the~~ transport 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. Based on the mass concentrations of NH_4^+ and NO_3^- in each total suspended particle (TSP) sample, the samples can be classified into increasing or decreasing ~~two~~ types~~groups~~. I~~:~~ ~~i~~n Category 1, the concentrations of NH_4^+ and NO_3^- were 20%-440% higher in dust day samples relative to samples collected immediately before or after a dust event~~, while in Categories 2 and 3,~~. T~~t~~hese concentrations decreased by 10-75% in the dust day samples in Categories 2 and 3. Back trajectory analysis suggested ~~showed~~ that multiple factors such as the transport distance prior to the reception site, the mixing layer depth on the transport route and the residence time across highly polluted regions, might ~~afffect~~affect the concentrations of NH_4^+ and NO_3^- ~~were apparently affected by the transport distance over sea prior to the reception site, the mixing layer depth and the residence time across highly polluted regions during transport~~. ~~For these two groups,~~ NH_4^+ in the dust day samples was likely ~~present~~ either in the form of ammonium salts existing externally ~~existing co existing~~ with dust aerosols or as the residual of incomplete reactions between ammonium salts and carbonate salts. ~~The~~ NO_3^- in the dust day samples was attributed to various formation processes ~~interactions between anthropogenic air p~~during the long-range ~~trasnport~~transport~~ollutants and dust particles during dust transport from the source zone to the reception site~~. ~~Back trajectory analysis showed that the concentrations of NH_4^+ and NO_3^- were~~

~~apparently affected by the transport distance over sea prior to the reception site, the mixing layer depth and the residence time across highly polluted regions during transport.~~ The positive matrix factorization (PMF) receptor model results showed that the contribution of soil dust increased from 23% to 36% (90% of the residuals<3.0 and r^2=0.97) on dust days with decreasing contributions ~~of~~ from local anthropogenic inputs~~, especially secondary aerosols~~ and ~~assoacited~~associated secondary aerosols. ~~The dry deposition flux of atmospheric particulates increased from 2,800±700 mg/m^2/month on non-dust comparison days to 16,800±15,900 mg/m^2/month on dust days. The dry deposition flux of particulate inorganic nitrogen increased by 9-285% in Category 1. The average dry deposition flux of nitrate decreased by 46%-73% in Category 2, while that of ammonium decreased by 47% in Category 3.~~ The estimated ~~dust~~ deposition flux of inorganic nitrogen varied greatly from event to event, e.g., the dry deposition flux of particulate inorganic nitrogen increased by 9-285% in Category 1, but decreased by 46%-73% in Category 2. ~~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.~~

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

1 Introduction

Reactive nitrogen carried in dust particles can be transported over a long distance~~s~~, and the atmos~~o~~pheric nitrogen deposition in oceans has been recognized as an important external source of the nitrogen supporting phytoplankton growth (Duce et al., 2008; Zhang et al., 2010b). This hypothesis has been evaluated ~~in~~ through incubation experiments, ~~in situ~~ in situ experiments, and ~~through~~ the use of satellite observational data (Banerjee and Kumar 2014;~~Shi et al., 2012;~~ Guo et al., 2012; Liu et al., 2013; Shi et al., 2012;~~Banerjee and Kumar 2014;~~ Tan and Wang, 2014). ~~For example, Tan and Wang (2014) found that a phytoplankton bloom with a nearly four-fold increase in chlorophyll concentrations 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.~~ However, the process is dynamic ~~because of~~due to the worldwide changing ~~emisisons~~emissions of NO_x and NH_3 ~~worldwidely~~ in the last few decades. For example, China and most of the developing countries in Asia ~~experenced~~experienced a large ~~Dramatic changes have~~

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~~occurred in the reactive nitrogen in anthropogenic emission in the last three decades, e.g., large~~ increase~~s~~ in emissions of NH_3 and NO_x ~~in emissions in China and other developing countries in Asia and~~while a substantial decrease in emissions occurred in Europe over the last three decades (Grice et al., 2009; Liu et al., 2017; Ohara et al., 2007; Skjøth and Hertel, 2013). The~~se~~ change~~s~~ would ~~may greatly~~ affect the nitrogen carried by dust particles to some extent~~., but~~ and updated ~~few recent~~ studies ~~have~~ are thereby essential~~examined this issue~~.

Asian dust is one of three largest dust sources ~~the main components of dust worldwide~~on ~~the~~ earth. Asian dust has been reported to not only ~~to to~~ frequently cross over ~~cross over~~ the mainland and the ~~marginal seas of~~ China Seas ~~frequently and reach~~, but also to occasionally reach ~~as far as~~ the remote northern Pacific Ocean or ~~the continent of the US~~North America ~~occasionally~~ (Creamean 2013; Tan and Wang, 2014; Van Curen and Cahill, 2002; Zhang and Gao, 2007~~; Tan and Wang, 2014~~). In an extreme case, ~~And~~ Asian dust ~~is~~was~~was~~ found ~~that can~~to be transported more than one full circuit around the globe in approximately 13 days (Uno et al 2009). During the long-range transport, dust particles may mix with anthropogenic ~~gas and particles~~air pollutants and~~,~~ consequently undergo ~~resulting in~~ complicated chemical reactions (Cui et al., 2009; Li et al., 2014; Ma et al., 2012; Wang et al., 2011; Wang et al., 2016, Wang et al., 2017a; Xu et al., 2014; ~~Li et al., 2014; Ma et al., 2012;~~ Yang et al., 2002). ~~However, the extent of these chemical reactions varies widely and depends on the meteorological conditions, such as cloud fraction, wind speed, relative humidity and atmospheric circulation (Yang et al., 2002; Li et al., 2014; Ma et al., 2012; Yang et al., 2002).~~ For example, a few studies have shown that the concentrations of atmospheric particulate NO_3^- and NH_4^+ on dust storm days were 2-5 times larger than those prior to the events in Beijing (Liu et al., 2014; Liu and Bei, 2016). ~~Xu et al. (2014) also reported that concentrations of particulate SO_4^{2-} and NO_3^- simultaneously increased during dust storm events along the northern boundary of the Tibetan Plateau.~~ Fitzgerald et al. (2015) found that almost all Asian dust events observed in Korea contained considerable amounts of nitrate ~~and proposed that the dust from the Gobi and Taklamakan Deserts probably mixed and reacted with anthropogenic air pollutants during transport over the Asian continent~~. However, ~~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. (2010a) reported an interesting result, i.e., ~~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 these ions. ~~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).~~This raises the complex for carrying reactive nitrogen by dust particles.

A few contradictory results ~~was~~were also reported in the literature, which made the scientific issue even more complicated.~~On the other hand, some studies reported the reverse result.~~ For example, ~~at Yulin, a rural site near the Asian dust source region,~~ the concentration of NO_3^- in atmospheric aerosols on dust days was significantly lower in comparison to the concentration measured immediately before or after the event~~, as a result of the dilution effect~~ at a rural site ~~at~~ in Yulin near the Asian dust source region (Wang et al., 2016). The phenomenon was also observed in Shanghai, a mega city at a few thousands of kilometers from dust source zones in China, and more downwind sites ~~Even in Shanghai, a mega city located at a few thousands kilometers from dust source zones in China, the concentrations of NO_3^- and NH_4^+ were notably lower in the observed dust plumes than in a polluted air parcel observed immediately prior to the dust events (~~-Kang et al., 2013; Li et al., 2014; Wang et al., 2013). ~~Li et al. (2014) also found that the concentrations of nitrate and ammonium decreased on dust storm days with a decreasing ratio of the total soluble inorganic ions to $PM_{2.5}$ in the Yellow River Delta, China. When dust was rapidly transported from desert regions without passing through major urban areas and lingering over the Yellow Sea, the concentrations and size distributions of nitrate and ammonium had no significant variation in heavy Asian dust (AD) plumes (Kang et al., 2013).~~

To ~~upadte~~update and improve our knowledge on reactive nitrogen carried by dust particles, ~~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,~~ we collected atmospheric aerosol particles during and prior to (~~(~~or post, but ~~after,~~ only when no sample was collected prior to~~)~~ dust events) at a coastal site adjacent to the Yellow Sea ~~during the~~ in each spring ~~from~~ of 2008 ~~to~~ - 2011~~ when smaller outbreak peaks of dust storms occurred~~. ~~We measured t~~The concentrations of inorganic nitrogen ~~in the samples as well as~~ and other components were ~~determiend~~determined for ~~to facilitate our~~ analysis. In this study, w~~W~~e first characterized the concentrations of inorganic nitrogen ~~concentrations~~ in ~~various~~ dust samples by comparing them with ~~events relative to the concentrations~~the values in atmospheric particles ~~in samples collected~~measured either prior to or post~~after the events~~ the event. We then conducted source apportionment to quantify

their sources. Finally, we calculated and discussed the deposition flux of atmospheric particulate inorganic nitrogen during dust events ~~and compared the results with the values in the literature in order to update the flux values due to dynamic changes in anthropogenic emissions and other factors~~.

2 Experimental methods

2.1 Sampling

Fig. 1 shows the sampling site, which is situated at the top of a coastal hill (Baguanshan) in Qingdao in northern China (36 °6' N, 120 °19' E, 77 m above sea level) and is approximately 1.0 km from the Yellow Sea to the east. A high-volume air sampler (Model KC-1000, Qingdao Laoshan Electronic Instrument Complex Co., Ltd., China) was set up on the roof of a~~n~~ two-story office building to collect total suspended particle (TSP) samples on quartz microfiber filters (Whatman QM-A) at a flow rate of 1 m^3/min. Prior to ~~the~~ sampling, the filters were heated at 450 ℃ for 4.5 hrs to remove organic compounds. Our sample collection strategy involved collecting dust samples representing long-range transported particles. We followed the definition of dust events adopted in the regulations of surface meteorological observations of China (CMA, 2004; Wang et al., 2008) and identified dust events based on the meteorological records (Weather Phenomenon) of Qingdao from the Meteorological Information Comprehensive Analysis and Process System (MICAPS) of the China Meteorological Administration. Due to no dust events without enhancive-source lasting over 12 hrs ~~(~~Lee et al., 2015; Su et al., 2017; Zhang et al., 2007), we collected ~~only~~ one dust sample with a 4-hr duration in a day ~~was collected for dust events with durations of less than one day~~. ~~And t~~The sampling for dust particles started only when the measured PM_{10} mass concentration ~~observed~~ in Qingdao ~~on the website~~ (http://www.qepb.gov.cn/m2/) and the forecasted dust mass over Asia (http://www-cfors.nies.go.jp/~cfors/) had greatly increased.

~~I~~On ~~20-21~~ March 20-21, 2010, two dust events subsequently swept Qingdao. The 4~~-~~ hr dust samples with IDs of 20100320 and 20100321 may not capture the entirety of the two ~~whole~~ events. However, the on-line data can allow adequate ~~well seperating~~ separation of the~~separating~~ two dust event samples. The same was ~~ture~~true for the dust samples with IDs of ~~For dust events with durations greater than one day due to supplementing from enhancive-source, such as 20100320, 20100321 and~~ 20110501, 20110502. ~~, a 4-hr dust sample was collected once per day according to temporal variation of PM10~~

~~concentration. This can be clearly identified from the on-line data of PM10 concentration.~~ ~~Each dust sample was collected over 4 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/) had increased greatly.~~ ~~4 hrs samplings may not full represent these dust event samples since some events lasted over 4 hrs, and in factbecause off-line samplings are practically hard to exactly cover the whole dust event period. However~~ ~~Tthis approach made the dust sample more representative relative to urban background~~ ~~under off-line samplings.~~ ~~This approach 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 durations greater than one day, a 4-hr dust sample was collected once per day.~~ Table 1 lists the sampling information. Based on the forecast, we also collected aerosol particle samples immediately before, which were regarded as the reference samples ~~or after the dust event for comparison~~. These reference ~~comparison~~ samples were further classified into sunny day samples and~~,~~ cloudy day samples. For those events missing sampling prior to dust events, we collected ~~and~~ post-dust samples ~~. The post-dust samples were collected un~~under clear and sunny weather conditions ~~and low PM_{10} mass concentrations~~as early as possible.

~~Since~~ Asian dust events ~~weer~~were ~~at the sampling site~~ mostly observed ~~occur~~ in the spring at the sampling site.~~,~~ Our intensive samplings were concentrated in the period of ~~we collected samples during every spring, i.e., from~~ March to May in ~~, from~~ 2008-2011, ~~where~~when a~~. A~~ smaller outbreak ~~peak for~~~~in~~ Asian dust events was observed in northern ~~eastern~~ China ~~in 2008-2011, which followed a larger peak in 2000-2003 during this century~~ (Fig. ~~6~~S5). Overall, a total of 14 sets of dust samples and 8 sets of comparison samples were ~~avaiable~~available for analysis~~collected for~~ in this study.

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

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2.2 Analysis

The aerosol samples were weighted according to the ~~standrad~~standard ~~protocoal~~protocol~~allowed to~~

achieve equilibrium in an air conditioned chamber at a constant relative humidity and temperature for 24hrs before weighing. The sample membranes were then cut into several portions for analysis. One portion of each aerosol sample was ultrasonically extracted with ultra-pure water in an ice water bath, and the concentration of for determining inorganic water-soluble ions was determined using ICS-3000 ion chromatography (Qi et al., 2011). The sand samples collected at the Zhurihe site were analyzed using the same procedure. We later refer to dissolved inorganic nitrogen (DIN) as the sum of nitrate and ammonium later, in the later discussion by excluding nitrite because of its due to the very low concentration. of nitrite in the samples.

One portion of the each aerosol filter was cut into 60 cm^2 pieces and digested with HNO_3+$HClO_4$+HF (5:2:2 by volume) at 160 ℃ using an electric heating plate. TA blank membrane was also analyzed using the same procedure to ensure analytical precision. The concentrations of Cu, Zn, Cr, Sc and Pb were measured using inductively coupled plasma mass spectrometry (Thermo X Series 2), while the concentrations of Al, Ca, Fe, Na and Mg were measured using inductively coupled plasma atomic emission spectroscopy (IRIS Intrepid II XSP). Field bThe membrane blanks have been corrected for in the calculation of the metal concentrations.A blank membranes wasere also analyzed for correctionusing the same procedure to ensure analytical precision. T

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

2.3 Computational modeling

The enrichment factor of metal elements was given by

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

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

elements in the Earth's crust given by Taylor (1964) was adopted.

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

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

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

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

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

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

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

Dry deposition velocities were obtained using Williams' model (Williams, 1982) by accounting for particle growth (Qi et al., 2005). Williams' model is a two-layer model used to calculate the dry velocity of size-segregated particles over the water. In an upper layer below a reference height (10 m), the deposition of aerosol~~s~~ particles is governed by turbulent transfer and gravitational settling. In the deposition layer, the gravitational settling of particles is affected by particle growth due to high relative humidity. To obtain the deposition velocity of different particle size~~s~~, Williams' model needs many input parameters, such as the wind speed at ~~10~~ 10-m height (U_{10}), air/water temperature, and relative humidity. Relative humidity, air temperature and U_{10} ~~obtained~~ from the National Centers for

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Environmental Prediction (NCEP) were used in this study. Surface seawater temperature data was collected from the European Centre for Medium-Range Weather Forecasts (ECMWF). The meteorological and seawater temperature data had a six-hour resolution. According to a previously reported method (Qi et al., 2013), the dry deposition fluxes of the particles and the nitrogen species were calculated for dust and comparison days.

The CMAQ model (v5.0.2) was applied over the East Asia area to simulate the concentrations of PM10, NO_x and NH_3 for 14 samples collected during 11 dust events. The simulated domain contains 164×97 grid cells with a ~~36~~ 36-km spatial resolution, and the centered point was 110 °E, 34 °N. The vertical resolution includes 14 layers from the surface to the tropopause, with the first model layer at a height of 36 m above the ground level. The meteorological fields were generated by the Weather Research and Forecasting (WRF) Model (v3.7). Considering that the simulated area is connected to the Yellow Sea, the CB05Cl chemical mechanism was chosen to simulate the gas-phase chemistry. The emissions of NO_X and NH_3 over East Asia for each dust event were also modeled using the CMAQ model according to the emission inventory in 2008, which 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. The ~~Validation~~ 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).~~

2.4 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 MICAPS of the Meteorological Administration of China. Different weather characteristics, such as sunny days, cloudy days and dust days, were defined according to information from the MICAPS and Qingdao Meteorological Administration. According to the altitude, longitude and latitude of the ~~72~~ 72-hr air mass back trajectory of each dust sample, the pressure level, temperature and relative humidity (RH) data along the path of the air mass were derived from the NCEP/NCAR re-analysis system

(http://www.esrl.noaa.gov/psd/data/gridded/data.ncep.reanalysis.html) for each sample. The mixed layer depth during the air mass transport of dust samples was obtained from the HYSPLIT Trajectory Model (http://ready.arl.noaa.gov/hypub-bin/trajasrc.pl) using the same method. Then the average mixing layer, transport altitude, air temperature and RH were calculated as an average of all points on~~during~~ the air mass back trajectory of each sample. Spearman correlation analysis was applied to examine the relationships of nitrate and ammonium with transport parameters, and P values of <0.05 were considered to be statistically significant.

3 Results ~~and discussion~~

3.1 Characterization of aerosol samples collected during dust events

~~To support our analysis, the spatial distributions of PM_{10} concentration over East Asia for each dust event were modeled by WRF-CMAQ model (Fig. S3)~~are shown in Fig. 2 and Fig. S3. ~~From Fig.S 3, we can found that almost all dust events originated in northern or northwestern China and passed over the sampling site and then transported eastward to the China Sea and furtherother ocean region. However, Sample 20110415 was judged to be local blowing dust because no correspondinghigh dust concentrations were observed in the dust source areas; this sample was therefore excluded from further analysis.~~ All these dust events, affectedobserved in Qingdao, had a characteristicsa characteristic of large influence range and relatively strong intensity. Though the dust intensity was judged by visibility in China (CMA, 2004), the PM10 concentration during a dust event can also show the dust intensity indirectly. Generally, strong dust storm had a high PM_{10} value, such as 2010320, consistent with the dust records (CMA, 2009). The dust intensity and influence range of these dust events varied greatly, depending on the dust event. The different dust events were expected to had different impact on the composition of aerosols in downwind areaintensity and influence range of the dust events modeled by Uno et al. (2003) were analyzed, and the spatial distributions of dust concentration over East Asia during each dust sampling day ~~are shown in Fig. 2 and Fig. S3.~~Almost all dust events originated in northern or northwestern China and passed over the sampling site. ~~However, Sample 20110415 was judged to be local blowing dust because no correspondinghigh dust concentrations were observed in the dust source areas; this sample was therefore excluded from further analysis.~~

~~Before characterizing the inorganic nitrogen in atmospheric particles from the Baguanshan site, W~~we

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first examined the mass concentrations of TSP samples and the concentrations of crustal and anthropogenic metals therein through a comparison with the ~~reference to compare samples~~reference samples collected on dust days and immediately before or after days, providing the background information for our target specie~~i~~s analyzed later. The comparative results are highlighted below. For these reference~~comparison~~ samples, the TSP mass concentrations ranged from 94 to 275 μg·m^{-3}, with an average of 201 μg·m^{-3} (Fig. 2, Table S1). The TSP mass concentration increased substantially to ~~501~~410-3857 μg·m^{-3} in dust day samples, with an average of 1140.~~2~~ 3 μg·m^{-3}. In each ~~dust day comparison day sample~~ pair of dust day sample ~~against~~against reference sample, the net increase in the mass concentration of TSPs ~~increased by~~was ~~80~~82-1303%, with a median value of ~~537~~403% (Table S1). A similar increase was present in the crustal elements in each pair of samples. For example, the mean concentrations of Sc, Al, Fe, Mg ~~Ca~~ and nss-Ca (usually used as a typical dust index) ~~Mg~~ increased by ~~over~~ more than a factor of ~~four~~ two ~~in dust day samples relative to comparison samples~~. On the other hand~~In addition~~, the enrichment factors (EF) of Al, Fe, Ca, and Mg were less than three in dust day samples with ~~, but were~~the values ~~–~~less than 14 in the reference ~~comparison s~~samples (Table 3). Lower values are indicative of elements from ~~the~~ a primarily crustal origin. ~~Although t~~The average mass concentrations of anthropogenic elements, such as Cu, Pb, Zn, Cr, Hg and As, in dust day samples increased by 107% to 722% ~~relative to~~against t~~t~~hose in ~~comparison~~ the reference samples~~, ~~; however, the EF of the anthropogenic metal elements ~~however~~ decreased in the former~~dust day samples~~. ~~This pattern indicates a decreasing relative contribution of anthropogenic sources to the total TSP mass in dust day samples.~~This indicates that dust particles likely carried more anthropogenic elements, although their relative contribution to the total mass was lower than that in the reference sample. Note~~d~~ that ~~s~~Sample 20110415 was excluded for further analysis ~~aforementioned~~. It was judged as a local blowing dust event because ~~of~~ no corresponding dust event existed upwind.

3.2 Concentrations of inorganic nitrogen in dust day samples

When the mass concentrations of NH_4^+ and NO_3^- in each pair of TSP samples were compared, the concentrations of NH_4^+ increased by ~~45~~8%-~~487~~473% in some dust day samples (20080301, 20080315, 20090316, 20100315, 20100320, ~~20100315~~20100321~~, ~~20110415 ~~~~20110418~~ and 20110502), but decreased by 28-84% in other dust day samples (Fig. 3, Column NH_4^+ and NO_3^- in Table S1~~Table S1~~). The same was generally true for the measured concentrations of NO_3^-.

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Considering the relative values of NH_4^+ and NO_3^- in dust day samples relative to the reference ~~comparison~~ samples, we classified 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 samples ~~against~~against ~~than in~~ the reference ~~comparison~~ samples. In Category 2, the reverse was true. In Category 3, the mass concentrations of NO_3^- ~~–~~were lower in the dust samples than in the reference ~~comparison~~ samples, whereas the concentrations of~~reverse was true for~~ NH_4^+ ~~was~~were close to the reference~~comparison ones~~.

As reported, ~~Considering that~~ the Yellow Sea ~~encounterred~~encountered ~~was mainly affected by~~ dust storms mainly derived from the Hunshandake Desert (Zhang and Gao, 2007). W~~, w~~e thereby compared our observations with the sand particles collected from this desert (Table 5). The ratios ~~relative mass concentr~~of mass concentrations ~~ations~~ of nitrate and ammonium to the total mass of sand particles were very low, i.e., less than 81 μg/g~~due to little anthropogenic influence expected~~, which are approximately three orders of magnitude less than the corresponding values in our dust samples. T~~Moreover, t~~he values obtained from atmospheric aerosols at ~~an~~ the urban sites of Duolun (Cui, 2009) and Alxa Right Banner (Niu and Zhang, 2000), which are closer to the desert, ~~–~~increased on dust days~~increased~~, but were still ~~also more than~~ over 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~~ The mixing and chemical interaction~~s and mixing~~ between anthropogenic air pollutants and dust particles during ~~the~~ transport from the source zone to the reception site likely played an important role ~~to~~in increas~~e~~ing the ratios, leading to ~~an~~ extremely larger ratio values at this site relative to those in source dust and in upwind atmospheric particles (Cui et al., 2009, Wang et al., 2011; Wu et al., 2016). However, the increase or decrease in the mass concentration absolute of nitrate and ammonium in different dust samples ~~against~~against the reference implied the complex for the interactions~~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 can vary greatly among different dust events, hence leading to the three different categories.~~.

4. Discussion and conclusion

~~3~~4.1~~3~~ Theoretical analysis of the three categories

Ammonium salts are common in atmospheric particles with diameters of less than 2 μm (Yao et al., 2003; Yao and Zhang, 2012). Many modeling studies have shown that the ~~Gas~~gas-aerosol

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thermodynamic equilibrium is ~~widely~~ assumed to be fully attained for inorganic ions, including ammonium salts in $PM_{2.5}$ (Dentener et al., 1996; Underwood et al., 2001; Wang et al., 2017a; Zhang et al., 1994; Zhang and Carmichael, 1999)~~, in all regional air quality modeling studies~~. Reasonably good agreements between ammonium salt modeling results and observations reported in the literature support the validity of this assumption (Chen et al., 2016; Penrodet al., 2014; Walker et al., 2012). Suppos~~ed~~ing ~~Assuming~~ that a thermodynamic equilibrium had been attained by the ammonium salts in Category 1, the reactions between carbonate salts and ammonium salts, such as 1) $(NH_4)_2SO_4$+ $CaCO_3 \Rightarrow CaSO_4 + NH_3$ (gas) $+CO_2$ (gas) $+H_2O$ and 2) $2NH_4NO_3 + CaCO_3 \Rightarrow Ca(NO_3)_2 + 2NH_3$ (gas) $+CO_2$ (gas) $+H_2O$, will release NH_3 (gas) until $CaCO_3$ has been completely used up. During dust events, ~~much~~ very high concentrations of Ca^{2+} were observed, and high $CaCO_3$ concentrations were therefore expected. For example, t~~T~~he single-particle characterization showed that Asia dust from the Gobi and ~~the~~ Inner Mongolian Deserts had rich $CaCO_3$, with a ratio of 4.3-6.7% for reacted $CaCO_3$ and 3.0-4.6% for unreacted $CaCO_3$ (Hwang et al., 2008). Heterogeneous chemical reactions of mineral dust mostly occurred on $CaCO_3$ mineral dust (Hwang and Ro, 2006). However, ~~When~~ when Category 1 was considered alone and one exterior sample was excluded, a good correlation ~~however,~~ was obtained for $[NH_4^+]_{equivalent\ concentration}=0.98*[NO_3^-+SO_4^{2-}]_{equivalent\ concentration}$ (R^2=0.83, P<0.05). The good correlation, together with the slope of 1, strongly indicated that the NO_3^- and SO_4^{2-} were almost completely associated with NH_4^+ in these dust day samples. It was commonly believed ~~generally accepted~~ that anthropogenic ammonium nitrate and ammonium sulfate were produced by gas, aqueous phase reaction and thermodynamic equilibrium processes, and ~~these~~this anthropogenic ammonium nitrate and ammonium ~~sulfate were~~sulfate was externally mixed with dust particles (Wang et al., 2017a). In ~~reverese~~reverse, ~~And T~~the poor correlation of Ca^{2+} to NO_3^- and SO_4^{2-} showed that ~~T~~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. Wang et al. (2017a) also found that coarse mode ammonium was quite low and fine mode dust particles were externally mixed with anthropogenic ammonium nitrate and ammonium sulfate. ~~The results on Asia dust samples verified that~~ NO_3^- and NH_4^+ in Asia dust samples were ~~argured~~argued to be~~verified that~~ more physically affected by the dust storm ~~for Asia dust samples,~~, i.e., the dilution effect, rather than the chemical reaction on the dust (Huang et al., 2010). The hypothesis appeared to be valid ~~Note that only samples~~ in Category 1, ~~showed~~ NH_4^+ ~~to be~~ was negatively correlated with Ca^{2+} (Fig.S4). In the exterior sample collected on

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21 March 2010, [NH_4^+] only accounted for ~70% of the observed [NO_3^-+SO_4^{2-}] in an equivalent concentration. This result suggested that ~30% of (NO_3^-+SO_4^{2-}) may be associated with dust aerosols via the formation of metal salts of the two species. ~~The~~ This hypothesis was supported by the correlation result, i.e., NO_3^- was positively correlated with NH_4^+ and Cu, and SO_4^{2-} was correlated with K^+, Na^+ and Mg^{2+} (Fig.S4). Cu was once ~~was generally~~ used as an effective marker~~s~~ of ~~vehicular emission sources from~~ diesel and biodiesel-blend exhaust (Gangwar et al., 2012), while it ~~also~~ can also be ~~and was also~~derived from copper pyrites ($CuFeS_2$) in Inner Mongolia mines (Huang et al., 2010). The increase of Cu in the mass concentration in dust samples implied ~~, indicating~~ dust particles mixed with ~~the~~ anthropogenic particles, particularly from ~~industrail~~industrial emissions, during transport. ~~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)~~, an effective marker of vehicular emission sources, implying that NO_3^-+SO_4^{2-} existed as metal salts~~. The average concentration of Ca^{2+} in Category 2 (0.43±0.40μg/m^3) was ~~evidnetly~~evidently ~~clearly~~ higher than that in Category 1 (Ca^{2+}: 0.17±0.04μg/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^+~~. Moreover, e~~However, the concentration of total Ca was 1.11±0.70 μg/m^3 in Category 1 and 0.74±0.49 μg/m^3 in Category 2.~~ ~~E~~Except for 20080502, the remaining dust ~~rest~~ samples in Category 2 ~~was~~were transported from the ~~middle~~ desert ~~in China carrying dust~~ relatively enriched with $CaCO_3$ (1-25% in Wt%) (Formenti et al., 2011). A positive correlation between ~~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~~and SO_4^{2-} in Category 2 against a negative correlation in Category 1 ~~alos~~also implied that the dust particles enriched with $CaCO_3$ in Category 2 might play an important role to form~~. 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^- ~~in these dust days~~. ~~Tobo et al. (2010) provide field evidence for the existence of~~ Ca-rich dust particles coated with highly soluble nitrate were observed at

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Kanazawa in Japan during Asian dust storm periods using SEM/EDX (scanning electron microscopy equipped with an energy dispersive X-ray spectrometer) ~~Tobo et al. (~~Tobo et al.，2010). The single-particle observation conducted by Hwang and Ro (2006) ~~characterization~~ showed that $CaCO_3$ in dust ~~species~~ particles was almost completely consumed ~~reacted~~ to produce ~~mostly~~mainly $Ca(NO_3)_2$ species~~ (Hwang and Ro, 2006)~~. ~~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±981 μg/m^3) was substantially higher than that of Category 1 (591±158 μg/m^3). This implied that dust events in Category 2 were even stronger. Note that the NO_3^- concentrations in Category 2 (1.35±2.45 μg/m^3) were lower or comparable to those in Category 1 (1.51±2.16 μg/m^3). The potential formation of nitrate metal salts was expected to be similar between the two categories, while unfavorable formation conditions for ammonium nitrate greatly increased decreased the mass concentrations of nitrate and the contributions to the TSPs in Category 12. Note that the NO_3^- concentrations in Category 2 (1.35±2.45 μg/m^3) were lower or comparable to those in Category 1 (1.51±2.16 μg/m^3).~~

~~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 and dust particles in some dust days. However, the lower concentrations in Category 2 were likely due to unfavorable conditions for forming ammonium salts in some dust days. The observed ammonium was just the residual of incomplete reactions between preexisting ammonium salt and carbonate salts depending on dust event, atmospheric chemical state, etc. More discussion on this issue will be presented in Section 3.4.~~

~~3.4 Influence of transport pathways on particulate inorganic nitrogenin dust samples~~

~~The calculated air mass trajectoriesof 13 out of 14 samples showed that the air mass originated from north and Inner Mongolia, China (Fig. 54), generally consistent with the results by Zhang and Gao (2007). The remaining one 20110418, originated from Northeast China. Figs. 6 and 75 (taking emission in 2008 as an example) 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. 54, except for the one exterior sample 20110502, all trajectories in Category 1 showed that the air masses were transported from either the north or northwest over the~~

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continent. In Category 2, the air masses crossed 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 (the average RH beingranged in 50-75% over the Bohai and Yellow Sea in Qingdaoin 2006-2012) might have enhanced hetero-coagulation between dust and smaller anthropogenic particlesparticle-particle coagulation and might have led to the release of NH_3 via reactions between preexisting ammonium salts and carbonate salts. Tobo et al. (2010) suggested that the conversion of insoluble $CaCO_3$ to $Ca(NO_3)_2$ tends to be dominated over urban and industrialized areas of the Asian continent, while relatively moist conditions in the marine boundary layer (usually, RH >60%), it is highly likely that the production of $CaCl_2$ exceeds that of $Ca(NO_3)_2$ by modifying Ca-rich particles in dust storms. Therefore we think the input of marine air during the transport was one reason for the low concentration of NH_4^++NO_2^- in Category 2.

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 aerosols externally. For example, except for 20080425, most of the time all dust day samples mostly traveled at an altitude above the mixing layer before mixing down to ground level. The transport of dust air masses above the mixing layer reduced the possibility for internal mixing of ammonium salts and reaction with dust aerosols along the long transport path. 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 masses in Category 1 at this elevation apparently moved slowly and took over 10 11-39 hr to cross over the highly polluted area. Even lower speeds were expected for air masses at the 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, higher than 900 m on average (916-1194m), higher than 900 m. The air masses in this Category 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 reaction with dust aerosols along the long transport path. The shorter time for dust air masses mixing down to ground

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~~level before arriving at the reception site and lower wind speed (mean of 2.8m/s at sampling 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 (average wind speed being 6.2 m/s at sampling site). The single particle characterization also showed that the Asian dust particles collected in Korea were mixed with sea-salts entrained over the Yellow Sea, as well as air pollutants from the eastern China coastal areas for a slow-moving, low-altitude air-mass (Hwang et al., 2008). The correlation analysis results in Table S2 indirectly support these conclusions. In fact, previous studies proposed that nitrate is rarely formed on the surface of dust particles for some dust events(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.~~

~~3~~4.~~5~~ 2 Source apportionment of aerosols during dust and non-dust events

The sources of atmospheric aerosols in ~~on d~~dust and reference samples ~~comparison days~~ were determined by ~~the~~ PMF modeling (Paatero and Tapper, 1993; Paatero, 1997). Fig. ~~8~~ 6 shows that atmospheric aerosols in the reference samples ~~on comparison days~~ mainly included ~~mainly consisted of~~ six sources, i.e., ~~:~~ industry, soil dust, secondary aerosols, sea salt, biomass burning, and coal combustion/other sources, with 90% of the scaled residuals falling between -3 and +3; r^2=0.97. In these dust samples, ~~On all dust days inc~~including Categor~~y~~ies 1-3., ~~the sources of aerosols mainly included o~~oil combustion, industry, soil dust, secondary aerosols, and coal combustion/other sources were identified as five major sources (Table 7). ~~These values are compared in Table 7.~~ The contribution of soil dust evidently increased from 23% to 36% in the dust samples ~~on dust days~~ relative to the reference~~comparison days~~, consistent with the high concentrations of TSPs and crustal metals observed on dust days. ~~Liu et al. (2014) also found an even larger increase in the contribution of dust aerosols to PM_{10}, i.e., 31%-40%, on dust days relative to non-dust days. Accordingly, the contributions of local anthropogenic sources decreased on dust days, especially those of secondary aerosols, consistent with EF of anthropogenic metals observed 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 coal combustion presenting contemporaneously with a mixture of coal combustion and other pollutants emitted along the transport path on dust days.~~ The calculation results also showed that the ed contribution of nitrate plus ammonium from the soil dust source ~~aerosol mass (the sum of nitrate and~~

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~~ammonium associated with the dust source)~~ to the total ~~aerosol~~ mass ~~(the total~~of nitrate plus~~and~~ ammonium~~)~~ in the dust samples greatly increased ~~on dust days. However, we could not determine the contributions of above sources to aerosols for the different dust Categories because of the limited number of samples.~~The source profile for coal combustion in the dust day samples showed a high percentage of K^+, Cl^-, Ca, Mg, Co, Ni, As, Al and Fe, indicating that coal combustion particles may exist contemporaneously with other ~~anthoropogenic~~anthropogenic pollutants emitted along the transport path. Liu et al. (2014) also found a~~n even~~ larger net increase in the ~~the~~ contribution of dust aerosols to the mass of PM_{10}, i.e., 31%-40%, on dust days agaist ~~relative to~~ non-dust days in Beijing which is approxiately 600 km upwind of Qingdao. Accordingly, they reported that the contributions of local anthropogenic sources decreased on dust days, especially those ~~of~~ from secondary aerosols, consistent with the EF of anthropogenic metals observed on dust days.

4.3 Influence of transport path ways on particulate inorganic nitrogen in dust samples

The calculated air mass trajectories for 13 out of 14 samples showed that the air mass originated from ~~a~~North and Inner Mongolia, China (Fig. 4), generally consistent with the results ~~by~~of Zhang and Gao (2007). The remaining one, with ID of 20110418 originated from Northeast China. The calculated trajectories showed that ~~all~~ the entire dust air mass passed over those highly polluted regions with strong emissions of NO_x and NH_3 shown in Fig 5 and experienced different residence times therein. Fig. 4 show~~ed~~s that all air mass trajectories in Category 1 were transported from either the north or northwest over the continent, except for the one exterior sample 20110502. In Category 2, the air masses always took a 94-255 km trip over the sea prior to arriving at the reception site. NH_3-poor conditions in the marine atmosphere disfavored the formation and existence of ammonium nitrate. On the other hand, the humid marine conditions (the average RH ranged in 50-75% over the Bohai and Yellow Seas in 2006-2012) might have enhanced hetero-coagulation between dust and smaller anthropogenic particles, leading to the release of NH_3 via reactions between preexisting ammonium salts and carbonate salts.

The average mixing layer was less than 900 m along the air mass transport routes for most sampling days in Category 1 (Table 7), favoring the trapping of locally emitted anthropogenic air pollutants in the mixing layer. The air masses in Category 1 took over 11-39 hrs to cross over the highly polluted area with appreciable concentrations of NO_x (5.7±1.4 ppb) and NH_3 (7.6±3.3 ppb). Except for the

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exterior samples, air masses in Category 2 took less than 10 hrs to cross over the polluted areas with lower concentrations of NO_x (3.6±3.4 ppb) and NH_3 (4.7±4.7 ppb) and the mixing layer height along the route was 916-1194 m (on average) for each dust event. Moreover, the averaged wind speed at sampling site was 2.8 m/s in Category 1, but 6.2 m/s in Category 2. The lower wind speed in Category 1 was unexpected, implying dust particles very likely traveled at aloft with a high speed and then mixed down to the ground through subsidence. This further led to the external mixing of anthropogenic particulate matters and dust. The correlation analysis results in Table S2 indirectly support these conclusions.

The concentrations of PM10 and its major components NO_3^- and NH_4^+ over East Asia on dust days and comparison days were modeled using the WRF-CMAQ model (Fig. S3). Spatial distributions of PM10 during each dust events were consistent with the records in the "Sand-dust Weather Almanac" (CMA, 2009; 2010; 2012; 2013). The dust particles were transported eastward by passing over the sampling site, the China Sea and arriving at the far remote ocean region, except for the local blowing dust sample with ID of 20110415, as mentioned previously. NMB (normalized mean bias) values of NO_3^- were -4% and -12% in dust and non-dust reference samples, respectively, indicating that CMAQ results reasonably reproduce the mass concentrations of NO_3^- (Fig. S4). Simulated NH_4^+ concentrations in dust samples were severely under-predicted with NMB values at -71%. For reference samples, simulated NH_4^+ concentrations sometimes can well reproduce the observational values, but sometimes totally off. The external mixing mechanism proposed in this study is urgently needed to be included in the model for accurately predicting the concentrations during dust events.

~~All these dust events, observed in Qingdao, had a characteristic of large influence range and relatively strong intensity. Though the dust intensity was judged by visibility in China (CMA, 2004), the PM10 concentration during a dust event can also show the dust intensity indirectly. Generally, strong dust storm had a high PM_{10} value, such as 2010320, consistent with the dust records (CMA, 2009). The dust intensity and influence range of these dust events varied greatly, depending on the dust event. The different dust events were expected to had different impact on the composition of aerosols in downwind area.~~

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~~Moreover, we also examined the links among the measured concentrations of particulate ammonium and nitrate, the mixing layer along the back trajectories, concentration of NO_x and NH_4 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 existed with dust aerosols externally. For example, except for 20080425, most of the time all dust day samples traveled at an altitude above the mixing layer before mixing down to ground level. The transport of dust air masses above the mixing layer reduced the possibility for internal mixing of ammonium salts and reaction with dust aerosols along the long transport path. For most sampling days in Category 1, the average mixing layer was less than 900 m with high concentration of NO_x and NH_4 in range of 13,000-48,000 (Table 5), favoring the trapping of locally emitted anthropogenic air pollutants in the mixing layer. In addition, the air masses in Category 1 took over 11-39 hr to cross over the highly polluted area. Except for exterior samples, the sampling days in Category 2 featured a mixing layer that was higher than 900 m on average (916-1194m) and much low concentration of NO_x and NH_4 in range of 26-13,100 (Table 5). The air masses in this Category took less than 10 hr to cross over the highly polluted areas. Theoretically, a lower mixing layer, high concentration of NO_x and NH_4 and a lower wind speed favored the accumulation of air pollutants and the formation of ammonium nitrate to some extent. The shortlonger residence time over the highly polluted zones for dust air masses mixing down to ground level before arriving at the reception site and lower wind speed (mean of 2.8m/s at sampling site) also increased the possibility for external existence between ammonium salt aerosols and dust aerosols in Category 1. The reverse could be argued to explain the observations for Category 2 (average wind speed being 6.2 m/s at sampling site). The single particle characterization also showed that the Asian dust particles collected in Korea were mixed with sea salts entrained over the Yellow Sea, as well as air pollutants from the eastern China coastal areas for a slow moving, low altitude air mass (Hwang et al., 2008). The correlation analysis results in Table S2 indirectly support these conclusions. In fact, previous studies proposed that nitrate is rarely formed on the surface of dust particles for some dust events (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.~~

~~3~~4.~~6~~ 4 Dry deposition fluxes of TSP, particulate inorganic nitrogen and metals

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

The dry deposition fluxes of atmospheric particulates increased on dust day~~s~~ ~~agaist~~against the reference~~relative to comparison days~~ to some extent. ~~All increases or decreases in this section reflected the value on dust days relative to comparison days, if not specified.~~ For example, ~~the dry deposition flux of TSP was only 2,800±700 mg/m^2/month on comparison days in the coastal region of the Yellow Sea. T~~the particle deposition fluxes varied over a wide range from 5,200 to 65,000 mg/m^2/month ~~under~~ in different dust sampling days, with an average of 18,453 mg/m^2/month, in comparison with ~~,~~the dry deposition flux of TSP ~~to be~~of 2,800±700 mg/m^2/month ~~on~~from the reference periods in the coastal region of the Yellow Sea. ~~However, t~~The dry deposition fluxes of $N_{NH4+NO3}$ varied, depending on Category 1, 2 or 3~~did not follow this pattern~~. In Category 1, the dry deposition fluxes of $N_{NH4+NO3}$ increased by 9-~~285~~75% with~~, corresponding to the~~ increased ~~in the~~ TSP flux ~~of~~ by 86-252% (Table S3). In Categories 2 and 3, the dry deposition fluxes of TSP increased by 126% to 2,226% against the reference~~compared to that on comparison days~~s. Exclud~~ed~~ing~~cept for~~ ammonium in Category 3, the dry deposition fluxes of particulate $N_{NH4+NO3}$~~, however~~, decreased by ~~41~~44% (on average). A larger ~~relative~~ decrease in the concentration ~~for~~of nitrate was present ~~found for~~ in Categor~~y~~ies 2 and 3~~the~~

~~concentration of nitrate~~, i.e., decreases of~~–~~ 73% and 46%~~ in Category 2 and 3~~, respectively. Note that the average ammonium deposition flux decreased by 47% in Category 2 but increased by 10% in Category 3.

~~The dry atmospheric deposition fluxes of Fe increased by a factor of 124-2370% on dust days. Atmospheric inputs of iron to the ocean have been proposed to enhance primary production in HNLC areas (Jickells et al., 2005). Moreover, e~~Except for Pb and Zn in Category 2, the dry deposition fluxes of Cu, Pb and Zn increased with those of nitrogen ~~and iron~~ on dust days. Trace 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 the~~ inhibition coexisted with the promotion of ~~some~~ phytoplankton species in incubation experiments in the southern Yellow Sea in the spring of 2011 by adding ~~involving the addition of~~ Asian dust samples to collected seawater~~s in the southern Yellow Sea in the spring of 2011~~. However, the dry atmospheric deposition fluxes of Fe increased by a factor of 124-2370% in dust day samples. Wang et al. (2017b) recently reported that Fe can alleviate the toxicity of heavy metals. Moreover, ~~A~~atmospheric inputs of iron to the ocean have been widely proposed to enhance primary production in HNLC areas (Jickells et al., 2005).

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~~**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 emissions increased in China from 1980 to 2010 (Fig. S5). ~~Accordingly, t~~The ~~$N_{NH4++NO3}$~~ dry deposition flux of $N_{NH4++NO3}$ should have theoretically increased with the increase in ~~inorganic nitrogen~~ the emission~~s~~ of inorganic nitrogen. ~~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 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. Consider~~~~ed~~~~ing~~ the different ~~the uncertainty in~~ dry deposition ~~velocity~~ velocities to be used in various studies, we recalculated ~~normalized~~ the dry deposition flux of $N_{NH4++NO3}$ in the literature using ~~the concentration of nitrate and ammonium reported in the literature and~~ the ~~recommended~~ dry deposition ~~velocity~~ velocities of 1 cm/s for nitrate and 0.1 m/s for ammonium, as ~~in coastal areas~~ reported by Duce et al. (1991). We ~~then~~ thereby found that dry deposition fluxes of $N_{NH4++NO3}$ over the Yellow Sea during the dust days increased greatly from 1999 to 2007, but the

values . The fluxes of $N_{NH4+NO3}$ in Qingdao, including during the dust days, varied narrowly within a range of 94.75-99.65 mg N/m^2/month during the dust days from 1997 to 2011 (Table 8). The complicated results implied that even more updated works are needed in the future. 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). 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.

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 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.

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.

Overall, this study strongly suggested that atmospheric deposition of $N_{NH4+NO3}$ on dust days varied greatly and that no simple linear increase existed with increasing dust load. More observations at various locations are needed to obtain a statistical relationship between dust events and atmospheric deposition of $N_{NH4+NO3}$. A simple assumption of a linear increase in $N_{NH4+NO3}$ with increasing dust load, like that in the literature, could lead to considerable overestimation of the dry deposition flux of

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~~nutrients into the oceans and the consequent primary production associated with dust events.~~

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

Sampling ~~Year~~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 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	Floating dust

		p.m. on May 1st	
	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 ~~figureis~~figures ~~was~~are shown in Fig. S1.

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

[c]Note that one exterior dust sample 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~~ Weather ~~almanac~~ Almanac 2011 (CMA, 2013). The modeled spatial distribution of the PM_{10} mass concentration for this dust event on April 18 implies that the sample should be classified as a dust sample. The supporting figure is Fig. S2.

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

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

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

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

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

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

	Sample number	TSP ($\mu g \cdot m^{-3}$)	NO_3^- ($\mu g \cdot m^{-3}$)	NH_4^+ ($\mu g \cdot m^{-3}$)	RH (%)	T (℃)	NOx ($\mu g \cdot m^{-3}$)	Summary
Category 1	20080301	527	20.5	12.7	57	7.0	36	DIN concentration on dust days higher than that on non-dust days
	20080315	410	19. 5	29.9	62	11.0	59	
	20090316	688	15.9	17.2	27	16.0	75	
	20100321	519	16.5	9.4	51	8.8	76	
	20110502	810	21.0	11.0	49	17.7	62	
Category 2	20080425	622	6.8	2.0	30	18.0	40	DIN concentration on 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	

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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.

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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
Alxa Left Banner, Inner Mongolia (Niu and Zhang, 2000)	62.1±7.4	79.1±1.1	Alxa Right Banner, Inner Mongolia (Niu and Zhang, 2000)	1975[b]	4091[b]	Category 1: 34,892±9570	Category 1: 22,571±7,016
Yanchi, Ningxia (Niu and Zhang, 2000)	46.4±2.2	80.9±1.3	Hinterland of the Taklimakan Desert, Xinjiang (Dai et al., 2016)	142-233	2-15	Category 2: 5,542±5,117	Category 2: 4,758±5,698
			Average of Sonid Youqi, Huade (Inner Mongolia), Zhangbei (Hebei) (Mori et al., 2003)	253	710	Category 3: 6,359±4,697	Category 3: 7,059±5,591
			Yulin, the north edge of Loess Plateau (Wang et al., 2011)	216.4	80.6		

	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 (~~Horizontal~~ 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.

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

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

Table ~~6~~7. Concentrations of TSP, NO_3^-, and NH_4^+; transport speed; transport distance over the sea; transport distance; air temperature; RH; average mixed layer during transport and transport time in polluted region for atmospheric aerosol samples on dust days.

Group	Sample number	TSP (μg/m^3)	NO_3^- (μg/g)	NH_4^+ (μg/g)	Speed (km/h)	Distance over the sea (km)	Transport altitude (m)	Mixed layer depth (m)	Residence time[a] (h)	T[b] (℃~~°C~~)	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 NO_3^-<ND NH_4^+≅ND	100315	501	10,767	8,515	57.3	0	5,009±1410	1,110±365	7	-40.4±13.3	45±29
	100320	3857	1,418	884	76.9	0	1,284±401	525±371	10	-12.2±6.3	61±16
	110418	558	6,891	11,778	35.6	931	1,344±780	695±672	2	-0.1±8.2	52±28

[a]Residence time of the air mass passeding over parts of highly polluted regions according to the trajectories of samples.

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

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

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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

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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.

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

Source	Year	Area		TSP	$N_{NH4+NO3}$-	Normalized average flux of $N_{NH4+NO3}$-[a]
This work	2008-2011	Qingdao, coastal region of the Yellow Sea	Non-dust 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 the normalized flux of $N_{NH4+NO3}$- was discussed in ~~section~~ Section 3.7.

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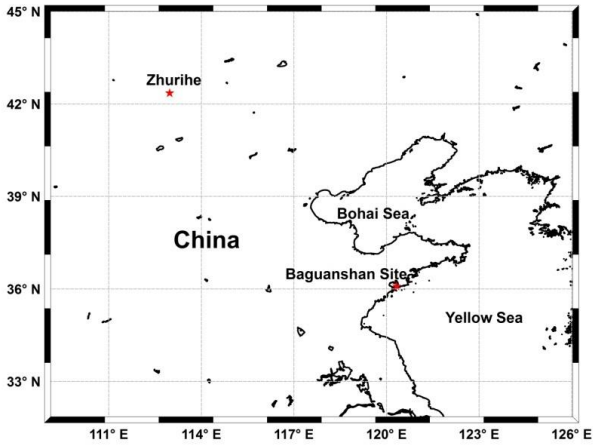
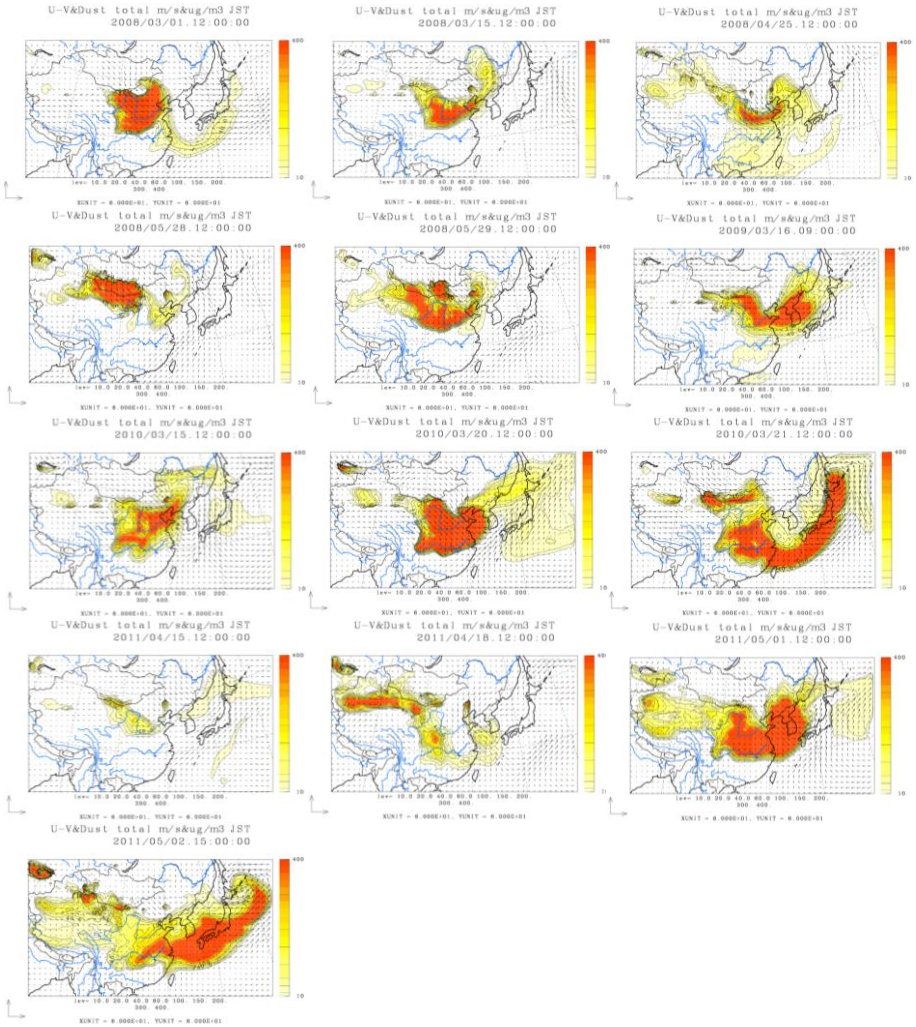


Figure 1. Location of the aerosol and dust sampling sites.



~~**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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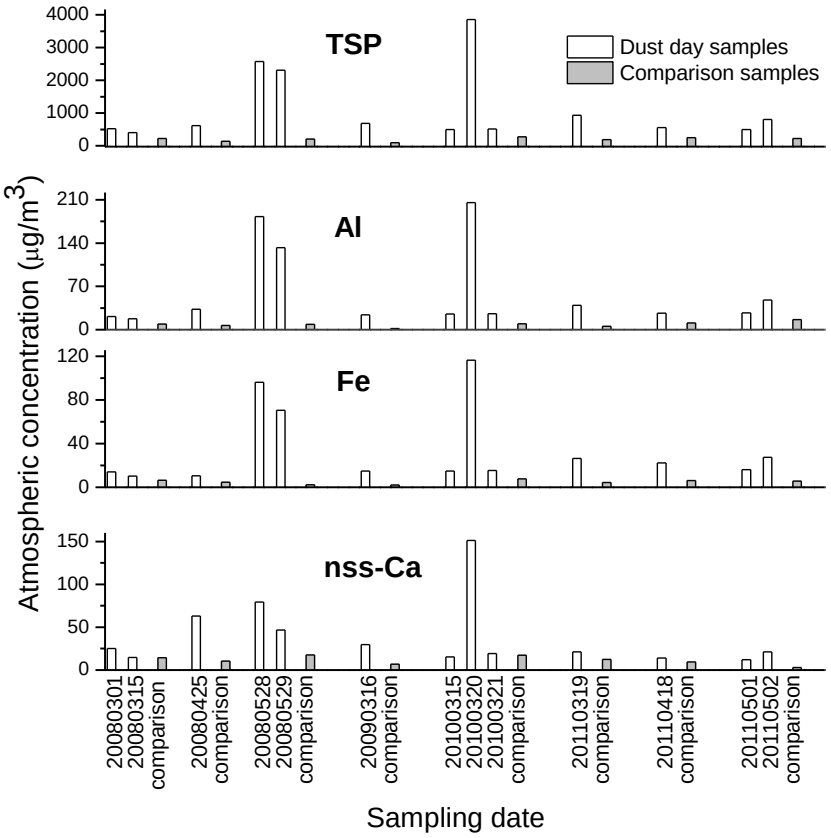

Figure ~~32~~. Mass concentrations of TSP, Al, Fe and nss-Ca in aerosol samples collected at the Baguanshan site on dust and comparison days from 2008-2011.

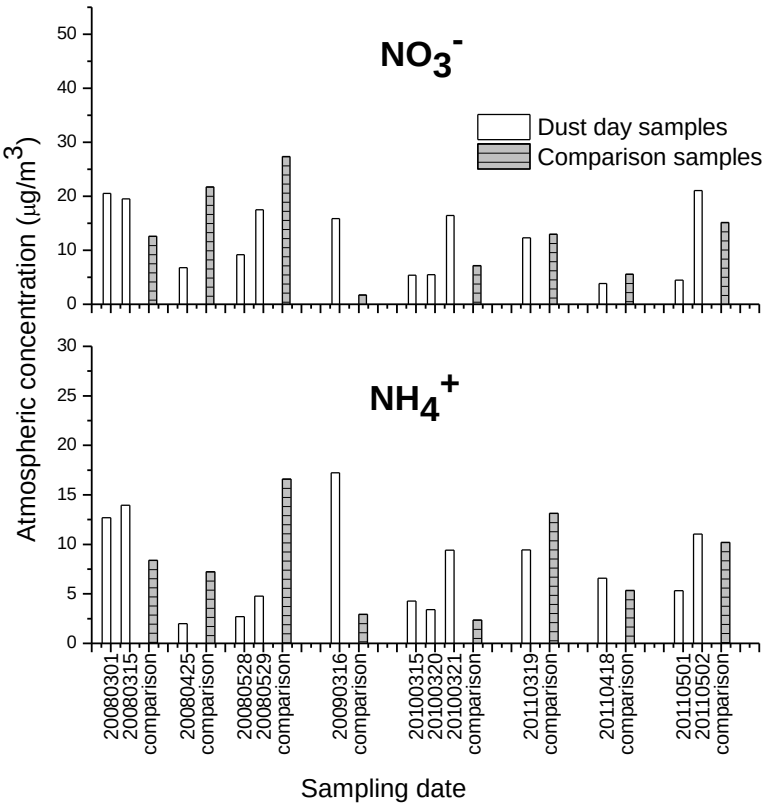

Figure 43. Mass concentrations of NH_4^+ and NO_3^- in aerosol samples collected at the Baguanshan site on dust and comparison days during March-May in 2008 to 2011.

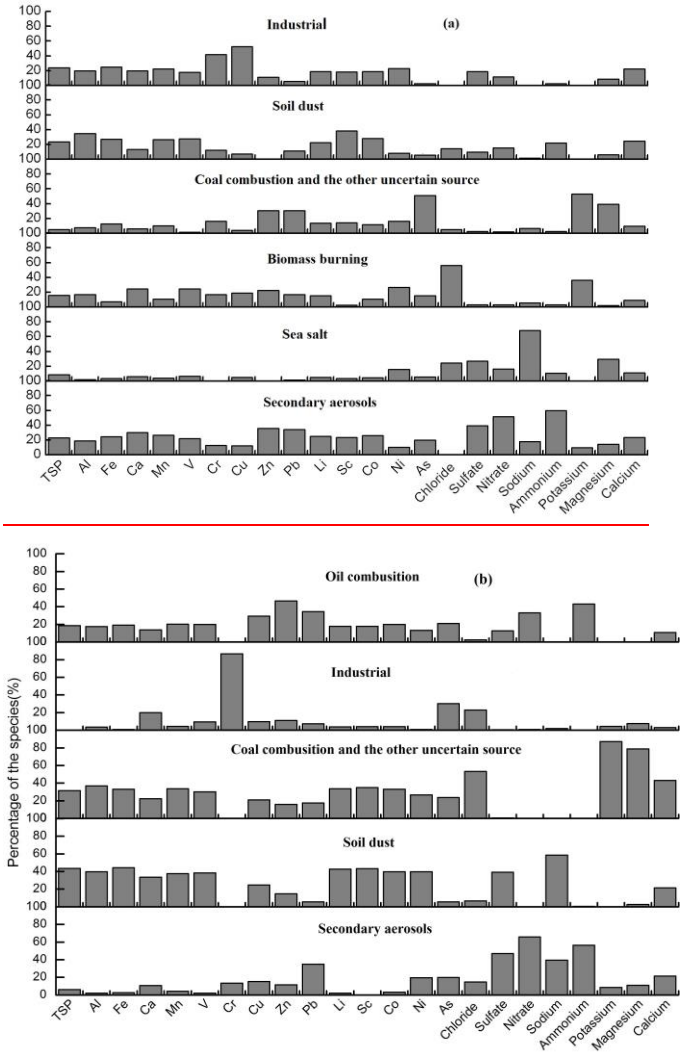

Figure 4. Source profiles of atmospheric aerosol samples collected on non-dust (a) and dust (b) days using the PMF model.

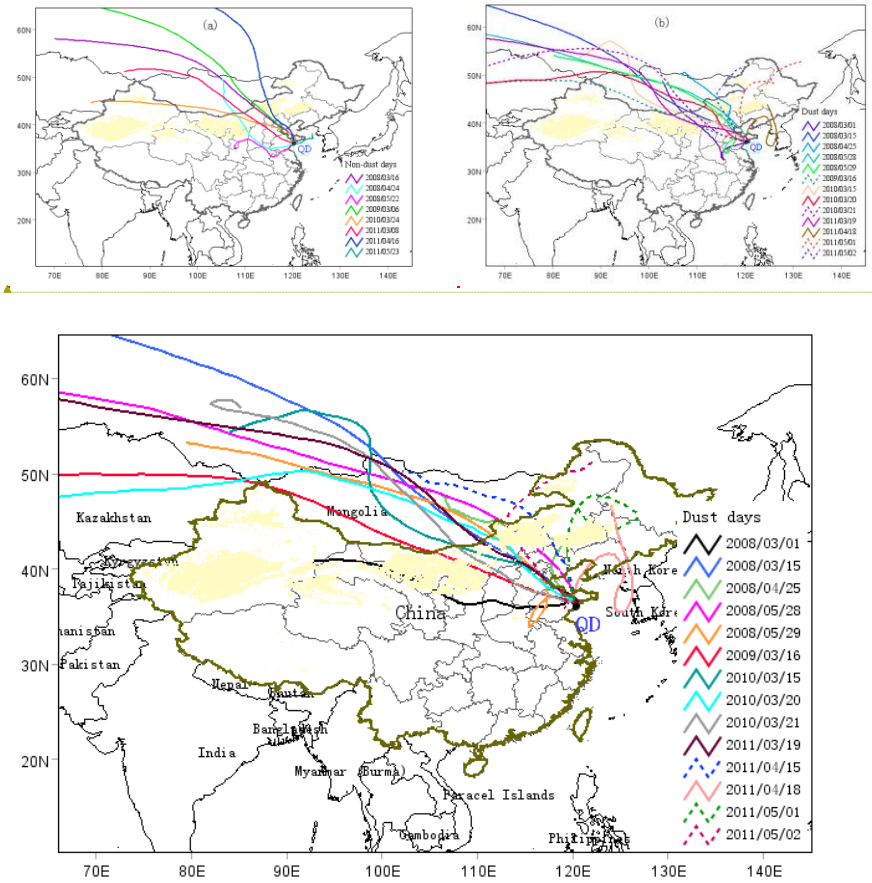

Figure ~~55~~**.** The 72-h backward trajectories for ~~non-dust (a) and~~ dust ~~(b)~~ samples from 2008 to 2011(the yellow domains in the map~~s~~ represent the dust source regions in China).

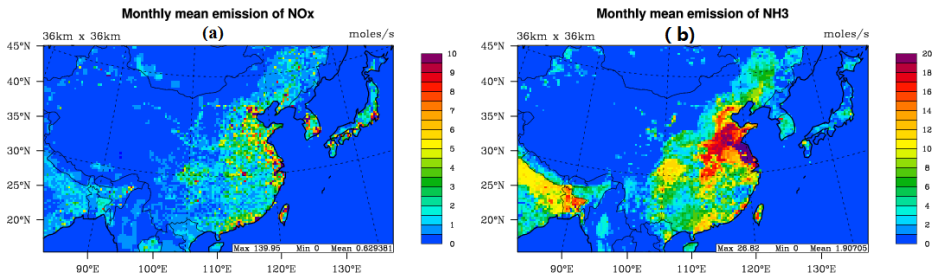

Figure 6. Seasonal mean emissions of NOx (a) and NH_3 (b) over East Asia from March-May 2008.

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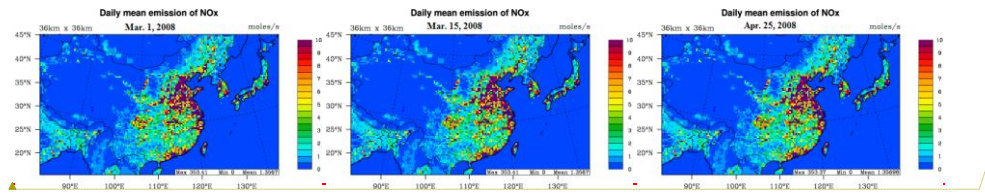

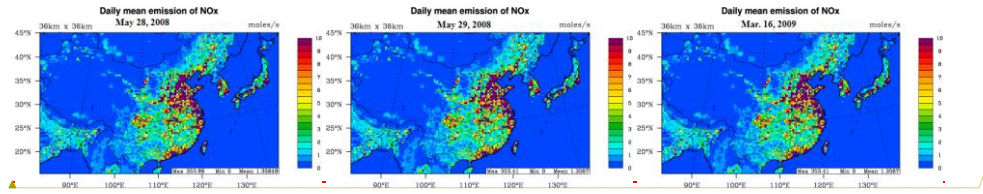

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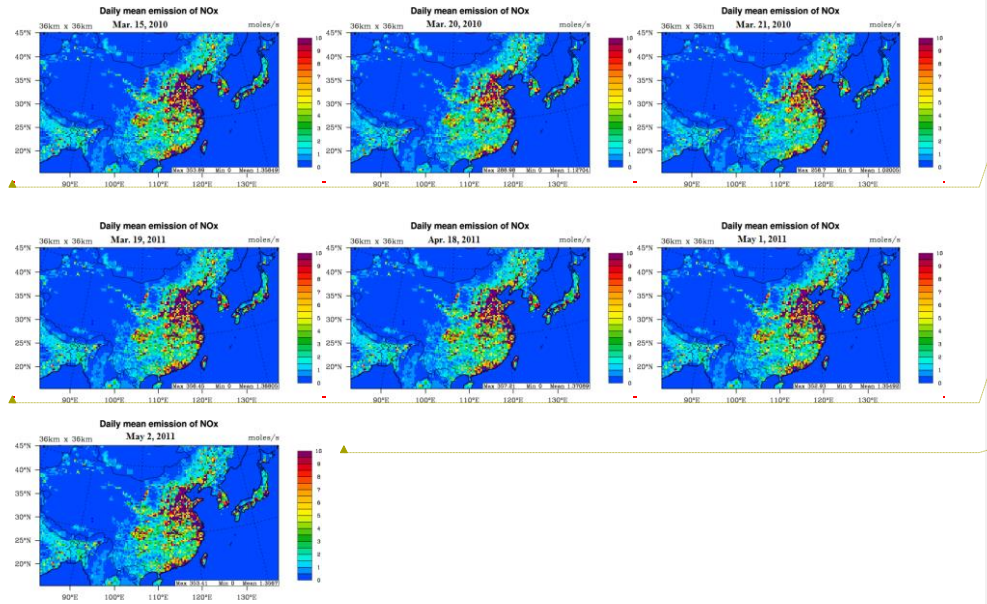
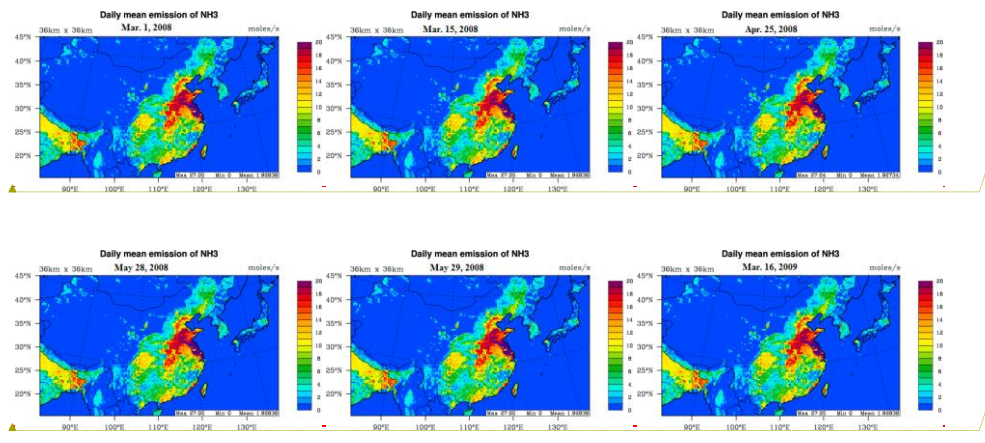


Figure 6. Daily mean emission of NOx over East Asia on the dust days from 2008 to 2011.



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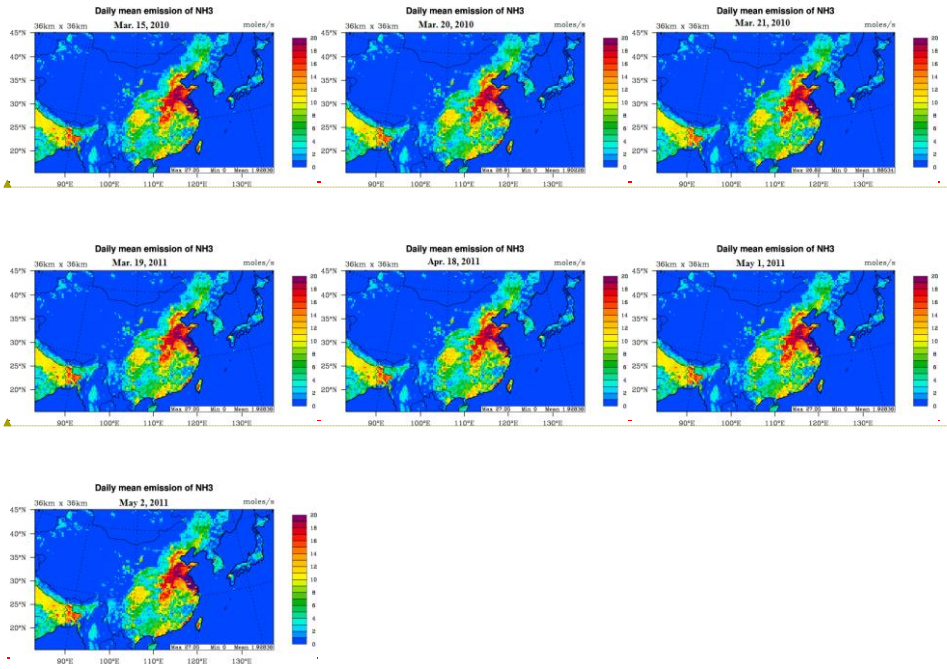

Figure 7. Daily mean emission of NH_3 over East Asia on the dust days from 2008 to 2011.

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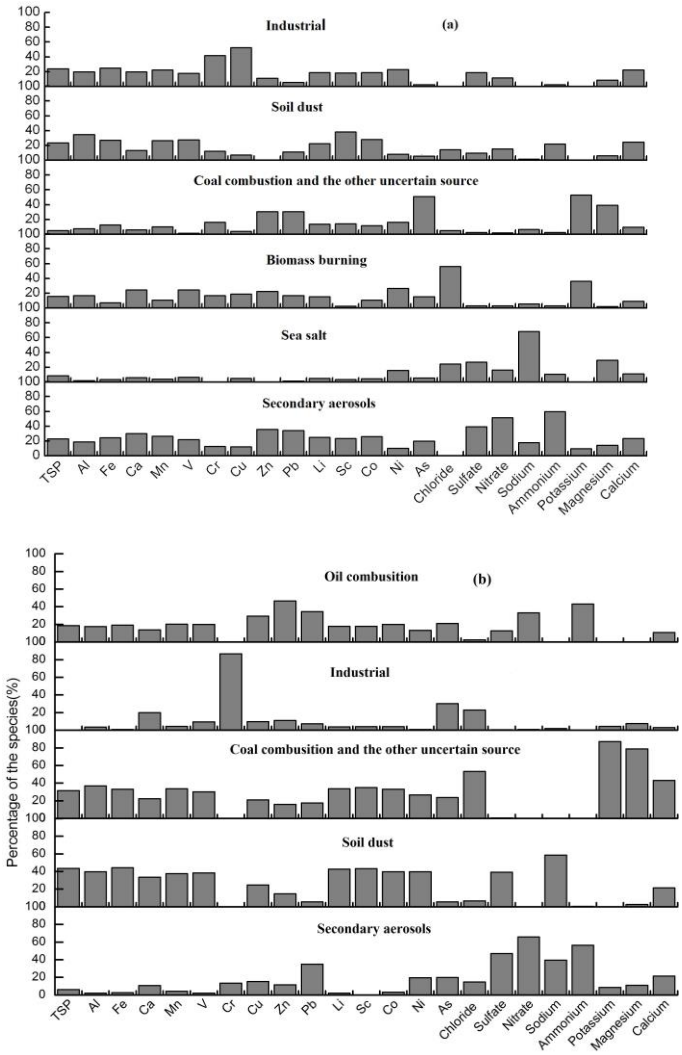

Figure 8. Source profiles of atmospheric aerosol samples collected on non-dust (a) and dust (b) days using the PMF model