

## Editor:

In the copy of the response to the review by Referee #1, I replaced all comments that have been addressed satisfactorily by [...]

It seems to me that multiple authors might have worked on the revision of the manuscript and that the response to the referees is based on an early revised version. Several of your responses do not match the submitted revised manuscript, as pointed out at several places by Referee #1.

In addition to addressing all scientific comments below, please make sure that in the next revised version:

- spaces are inserted where needed (e.g. l. 99, 128, 139)
- response to reviews and changes in manuscript are consistent
- proofread carefully the manuscript for typos and grammar errors

## Referee #1 – Re-Review:

### Responses to revision in red

General Comments:

The manuscript titled ‘The concentration, source apportionment and deposition flux of atmospheric particulate inorganic nitrogen during dust events’ written by Jianhua Qi presented the dust impacts on particulate inorganic nitrogen by analyzing the aerosol samples collected at Qingdao, China. The authors divided dust pattern into three parts, and investigated the dry deposition flux. To estimate the source, PMF receptor model was also used. Based on the above approaches, the authors tried to answer the questions of ‘dust event always increase the atmospheric input of nitrogen to the ocean?’. The topic is interested ones because the impact of dust as atmospheric input on ocean ecosystem has been still unclarified. However, throughout the manuscript, it is not well organized and hard to follow and understand. Overall, this manuscript will not be acceptable taking into account the high journal quality of Atmospheric Chemistry and Physics.

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

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.

Q1. Before the discussion, first, the definition of “dust events“ cannot be understood well. In L99-101, the authors explained that ‘Samples were collected on dust days and selected ND days in spring from March 2008 to May 2011, with sampling duration of 4h for each sample. We refer to the ND days as sunny and cloudy days before and after dust events in the following discussion’. The authors should add the appropriate reference of the Meteorological Information Comprehensive Analysis and Process System (MICAPS) which defined the weather conditions (and also, the subsection 2.4 should be reorganized partly into this explanation). What is the

definition of “dust events” here? Visibility? More information of how the dust events are defined in this system should be announced in detail. Total of 14 samples (sample numbers in Table 3) during dust events were analyzed throughout this study. The sampling duration was 4 hrs, so which data are used in the corresponded date in Table 3? All samples in the day? Moreover, what is the sample numbers of ND? The current information in Section 2.1 is severely lacked in the information which the readers can follow the authors methodology. Because this study discussed the dust impact, the explicit and detailed information regarding dust is required. In this sentence, I am worried about the explicit division of dust and non-dust samples. It is well known that some dust events are continued a few days. For example, the samples used in this study during 28-29 May 2008, 20-21 March 2010, 15 and 18 April 2011, and 1-2 May 2011 showed continuous dust events. In such cases, do the authors have confidence to the clear separation of dust and non-dust samples? How about the AI concentration definition (L171-172) of non-dust days samples? Why were other days samples not collected to clearly separate the dust impacts? The definition of ND is ambiguous. According to the definitions of dust and non-dust, the discussion on dust impact might be changed. The reconsideration of dust impact is needed based on the clear definitions of dust.

Reply: In this study, the dust event was defined according the definition adopted in regulations of surface meteorological observation of China (CMA, 2003; Wang et al., 2008) and identified based on the meteorological records information from Meteorological Information Comprehensive Analysis and Process System (MICAPS) of China Meteorological Administration. Each dust sample was collected for 4hrs duration and the sampling started only when the PM10 mass concentration available on the website (<http://www-cfors.nies.go.jp/~cfors/>; <http://www.qepb.gov.cn/m2/>) was increased greatly. The approach made the dust sample more representative relative to urban background. However, for dust event with duration less than one day, only one sample was collected; for dust event with longer duration, i.e. multiple days, the sample was collected once a day. The sampling information was listed in the Table S1. Based on the forecast, we also collected aerosol particle samples immediately before or after the dust event for comparison. These comparison samples were further classified into sunny day samples, cloudy day samples and post-dust samples. The post-dust samples were featured by collecting under a clear and sunny weather condition and lower mass concentration of PM10. Moreover, the concentration of AI referring to the total AI concentration in TSP samples were used to confirm the division of dust or comparison samples according to the criterion "geometric mean $\times$ 2GSD" proposed by Hsu et al. (2008). CMA: Regulations of Surface Meteorological Observation, China Meteorological Press, Beijing, 154–156, 2004. Hsu, S. C., Liu, S. C., Huang, Y. T., Lung, S. C. C., Tsai, F., Tu, J. Y., and Kao, S. J.: A criterion for identifying Asian dust events based on AI concentration data collected from northern Taiwan between 2002 and early 2007, *Journal of Geophysical Research Atmospheres*, 113, 1044-1044, 2008. Wang Y. Q., Zhang X. Y., Gong S. L., Zhou C. H., Hu X. Q., Liu H. L., Niu T., Yang Y. Q.: Surface observation of sand and dust storm in East Asia and its application in CUACE/Dust, *Atmos. Chem. Phys.*, 8, 545–553, 2008.

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.

Q2. The second concern is the “dilution effect” which the authors claimed as the key factor for the discussion of inorganic nitrogen. Again, without the explicit definition of dust and non-dust, the dilution effect cannot be understood well. In this discussion, although the authors introduced the air mass speed, there were no implications on the intensity of dust events itself. Why the upwind (i.e., near desert) information was not used here to describe the dust intensity? The dilution is not so simple, hence more information are required to reinforce the authors finding. The authors discussed the inorganic nitrogen behavior. In these cases, what is the counter ion of  $\text{NH}_4^+$  and  $\text{NO}_3^-$ ? Are the main counter ions metal elements? If  $\text{NH}_4\text{NO}_3$  are formed, due to its chemical unstability according to the temperature and relative humidity, it is not simple to discuss only the viewpoint of “dilution effect”. In addition, the authors used  $\text{NO}_2$  data to investigate the inorganic nitrogen, but how about  $\text{NH}_3$ ? Only from  $\text{NO}_2$  data, it is insufficient to estimate the inorganic nitrogen variation. On the above reasons, the reconsideration is required to publish this manuscript from Atmospheric Chemistry and Physics.

Reply: In revision, the part reads as “Inorganic nitrogen (IN) concentrations highly varied in different dust samples (Table 3). According to the concentrations relative to those in comparison samples, they can be classified into three categories, i.e., Category 1 in which higher IN concentrations were observed in dust samples, Category 2 in which lower IN concentrations were observed in dust samples, and Category 3 in which lower nitrate concentrations with slightly higher concentrations of ammonium in dust samples. Category 1 was usually associated with a lower moving speed of dust air mass or a longer distance over the ocean (Table 5) while the reverse was true for Category 2. The moving speed and distance over the ocean of dust air mass in Category 3 was generally between them. Theoretically, lower moving speed of dust air mass favors reactions between dust particles and anthropogenic gaseous precursors of IN due to a longer reaction time. Large moving speed of dust air mass was frequently associated with a large wind speed in the lower layer atmosphere (Gao et al., 2010; Gillette and Passi, 1988; Peng et al., 2007; Yue et al., 2008), leading to anthropogenic gaseous precursors therein to be better diluted. Shorter reaction time and reduced concentrations of anthropogenic gaseous precursors likely lowered IN in Category 2. Moreover, the relative concentration of IN per aerosol particle mass in  $\mu\text{g/g}$  was analyzed and compared with those values in literature. ...” It is questionable for using  $\text{NO}_x$  observed in Qingdao to argue the generation of IN in dust samples. We agree this because most of IN observed in dust samples should be derived from secondary reactions upwind of Qingdao by considering a low conversion rate of  $\text{NO}_x$  to IN. The former study (Liu et al., 2010) showed that  $\text{NO}_x$  and  $\text{NH}_3$  generally capture the spatial distribution patterns with high values over eastern China and relatively lower values over central and western China, where dust source regions are

located (Fig. S1-S3). Thus, we will add modeling results using a 3-D air quality model to support our analysis in revision.

Gao, Q X., Ren Z H. et al. : Dust events and its impacts on atmospheric environment, Science press, Beijing, 2010.

Gillett e D A, Passi R.: Modeling dust emission caused by wind erosion, J G R., 1988, 93: 14234-14242.

Liu X. H., Zhang Y., Cheng S. H., Xing J., Zhang Q., Streets D. G., Jang C., Wang W. X., Hao J. M.: Understanding of regional air pollution over China using CMAQ, part I performance evaluation and seasonal variation, Atmospheric Environment , 44,2415-2426, 2010.

Peng, Z., Liu X. M., Hong Z. X., Wang B. L.: Characteristics of Atmospheric Boundary Layer Structure and Turbulent Flux Transfer during a Strong Dust Storm Weather Process over Beijing Area, Climatic and Environmental Research, 2007, 12(3): 268-276.

Qi J.H., Gao H.W., Yu L.M. , Qiao J.J.: Distribution of inorganic nitrogen-containing species in atmospheric particles from an island in the Yellow Sea, Atmospheric Research, 101,938-955, 2011.

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

Yue P., Niu S. J., Liu X. Y.: Dust Emission and Transmission during Spring Sand-dust Storm in Hunshandake Sand-land, Journal of Desert Research, 2008, 28(2): 227-230.

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. 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. Figure S1:

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

Specific comments:

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

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

I cannot find this revision.

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

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

I have confirmed that the authors use the wording of ‘dust storm’ in the introduction.

[...]

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

Reply: Each sample on dust day had its corresponding non-dust sample (Table S2). The 1.8-14.0 times was calculated as a ratio of the TSP concentration on a given dust day to the values in the comparison samples. The concentration and the ratio of samples on dust days were listed in Table S2.

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  $\mu\text{g}/\text{m}^3$  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.

[...]

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

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

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

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

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

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

[...]

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

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

[...]

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

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

Q17.L190: What means “less than 20% of that on ND days”? Averaged data over ND days? Reply: We apologize for the confusion. The sentence has been revised to read "The concentrations of ammonium were increased by 20.

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

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

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?

Q19.L194-L195: In this sentence, the authors stated “the effect of dust on inorganic nitrogen differed during different types of dust events“. Why the authors suddenly focused on inorganic nitrogen here? In L192-193, it was mentioned “inorganic ion SO<sub>4</sub><sup>2-</sup> exhibited concentration variations that were similar to those of nitrate”. L197: The figures for inorganic nitrate will be helpful information here, if the authors focused on inorganic nitrogen.

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

I cannot find this revision.

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

Reply: We have incorporated this suggestion.

I cannot find this revision.

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

Reply: Duolun and Zhurihe belong to the Hunshandake Desert in Inner Mongolia, one of the main Chinese sand deserts. According to studies, the Yellow Sea is mainly affected by dust storms from this sand source with a probability of 52 Zhang, Z K., and Gao, H.: The characteristics of Asian-dust storms during 2000–2002: From the source to the sea, Atmospheric Environment, 41, 9136-9145, 2007. Gao, Q X., Ren Z H.: Dust events and its impacts on atmospheric environment, Science press, Beijing, 2010.

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

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

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

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

[...]

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



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

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

[...]

Q27.L227-L228: The favorable condition to form ammonium cannot be discussed without the information of NH<sub>3</sub>. 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?

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

Again, I cannot understand the model application results.

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

Reply: We will add modeling results of dust distribution to support our analysis in revision. Again, from the additional information of CFORS, the spatial distribution pattern was found; however, how can we estimate the intensity?

Q29. L233-L234: But NO<sub>x</sub> concentration was high in Case 3. I cannot follow why the authors concluded “the strong dilution effect” on Case 3.

Reply: Among three cases, the NO<sub>x</sub> concentration was the highest with an average value of 70.7 for Case 3 and increased by 17.8

So where did the authors discussed the NO<sub>x</sub> concentration in the manuscript?

[...]

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

Reply: As discussed above (see the Reply to Q2), We will add modeling results using a 3-D air quality model to support our analysis in revision.

Again, model is used only for spatial distribution and not inform the chemical production process.

Q32. L256: RH and NO<sub>x</sub> information are not shown in Table 5.

Reply: We apologize for the mistake. We have revised the title of Table 5.



I have confirmed the information of RH and NO<sub>x</sub> on the revised Table 6.

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

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

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. L314: What is the ‘remaining one’? Please specify the trajectory data. In my opinion, two trajectories of 20110418 and 20110501 originated from northeast China. L317: What is the ‘one exterior sample’? Please specify. 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. 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?

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

Reply: The source of coal combustion on dust days became complex. The source profile showed high percentages of K<sup>+</sup>, Cl<sup>-</sup>, Ca, Mg, Co, Ni, As, Al and Fe, indicating a mixture of coal combustion and other pollutants emitted along the transmission path on dust days, such as industry and building dust. This source increased due to the coal combustion emissions mixing with other uncertain sources emitted into the air in strong winds.

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.

[...]

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

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

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

I cannot find this revision.

[...]

Q39. L309: What is the calculation method of "63%" and "46%"?

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

I cannot find this revision.

Q40. L310: What is the calculation method of "14%" ?

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

I cannot find this revision.

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

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

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

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

Reply: We apologize for the confusion. "aerosol particles" was revised to read "TSP". The former digits were revised according to the editor's suggestion. We will consider revising again to confirm the significant digits.

The revision for TSP was found in Table 9.

Technical Corrections:

[...]

Specific comments: Table 6: Missing the note of a and b. Table S2: Please align the right-column, it is hard to follow. What is the meaning of \*?

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### Referee #3:

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.

#### Comments:

- 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, NH<sub>4</sub><sup>+</sup> 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.”*

- 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 circumnavigate the globe. VanCuren and Cahill 2002 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.

- 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.

- Line 233 insert comma after “samples”

- 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(NO<sub>3</sub>)<sub>2</sub> and CaSO<sub>4</sub> are negligible.

- 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.

- 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 NH<sub>4</sub><sup>+</sup>/(NO<sub>3</sub><sup>-</sup>+SO<sub>4</sub><sup>2-</sup>) are qualitative at best.

- What is the mineralogy of the Hunshandake Desert? Is it rich in CaCO<sub>3</sub>? 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?

- 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  $> 10^5$  #/cm<sup>3</sup> where coagulation is prevalent, more likely in the  $10^2$ — $10^3$  #/cm<sup>3</sup>. Are the authors referring to fog-processing? That would seem to be the primary way this could happen in a marine environment.

- 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.

- 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.

- 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.

- 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.