Anonymous Referee #1

Summary

The authors presented and compared simultaneous aerosol measurements at two sites: an urban ground level site and a mountain site in the free troposphere. These measurements were taken over a one-month period and include several dust events. Some of the author's main points were:

a) the urban site was more polluted, with 2-22x more EC and OC, and more WSON.

b) temporal differences in HNO_3 and H_2SO_4 formation caused large differences between the urban and mountain sites, for example causing:

1. more SO_4 and less NO_3 at the mountain site,
2. greater acidity of fine particles at the mountain site due to continuous SO_4 oxidation and thus greater H_2SO_4 concentrations, and
3. during dust events, fine and coarse size fractions switched being most acidic between the city and in the mountain, presumably due to greater ability of NO_3 to form from heterogeneous reactions in the city and less HNO_3 formation at the mountain.

General comments

Overall it seems that there are a lot of unique data here. Specifically with regards to their WSON data, the results are quite interesting and unique. In fact, I suggest to the authors that they do a further, more in-depth analysis of the WSON data in order to capitalize on their data as much as possible. However, there are problems with the data interpretation. Specifically, there is strong reason to believe that their mountain samples were not directly comparable with their urban samples, making it difficult to test their hypothesis that the differences in NO_3 and SO_4 concentrations between the sites was due to differential temporal development of NO_3 and SO_4. Thus some of the conclusions are not strongly supported by the results presented.

Overall, I think it should be published with major revisions.

Response: We thank the reviewer's suggestion about WSON. Given that this paper focuses on differences in inorganic ions between Xi'an and Mt. Hua (i.e., composition and size distribution) and length of the current paper is very long, we just gave brief discussion on WSON instead of an in-depth analysis. We agree with the reviewer that the results about WSON are very interesting and the chemistry of WSON in China is very important. We have been analyzing the WSON on a molecular level for investigation the source and formation mechanism. As for the question raised by the reviewer whether the mountain samples were comparable with the urban samples. Air masses reaching Xi'an and Mt. Hua during the DS II period originated from the same Gobi regions and transported via same pathways before simultaneously arriving at the two sites (see Figure 3 in the revised paper), resulting in particles in the DS II event presenting similar levels of loading with a same size distribution pattern in Xi'an and Mt. Hua. Trehalose, a metabolism product of biota in the Gobi desert (Wang et al., 2012), showed comparable concentrations in Xi'an and Mt. Hua in the occurrence of the DS II event. Therefore, we believe that both sites were dominated by a same air mass during the DS II episode and differences in chemical compositions of the dust samples from Xian and Mt. Hua in the dust event were mostly caused by adsorption/reaction of the lowland pollutants with dust. The goal of this paper was to identify the difference in aerosol chemistry between the ground surface and the free troposphere. Such a simultaneous observation has not been conducted in Guanzhong Basin, inland China, where particle loading is among the highest in the world. Therefore, Xi'an and Mt. Hua were selected as our sampling sites, both are located within the same region (i.e., Guanzhong Basin) with a distance about 80 km. As shown in Figure 1, the mountain site is situated on the West Peak of Mt. Hua, which faces directly to the Guanzhong Basin like a tower with a height of ~1500 m above the ground, and thus is an ideal site to identify the vertical difference in aerosol properties by comparison with those on the lowland s of Guanzhong Basin such as Xi'an. Such a comparison of Mt. Hua aerosol with that of Xi'an was also conducted by Rosenfeld et al (Science 2007). Their paper published in Science compared the relation of aerosols with precipitation between Mt. Hua and two cities (i.e., Xi'an and Weinan) in Guanzhong Basin. Observation using aircraft or balloon is another common way to observe the vertical profile of atmospheric aerosol properties, but both are not suitable for a long-term observation. Our simultaneous observation of mountain and ground surface aerosols is similar to the aircraft measurement and has an advantage of long-term measurement. Moreover, compared to those in the Mt. Hua dust samples, increased WSOC, OC Ca^{2+}, K^+ and Mg^{2+} of the urban dust samples due to heterogeneous reaction of lowland pollutants with dust further suggest that a same air mass dominated both sites in the DS II event (see page 11, line 265-280). Therefore, we think the Mt. Hua samples are comparable to those urban samples. The above response has been added into the text, see page 7, line 174-183.

Specific comments

Comments most important to address:

1) One of the main points of the paper is that on the aerosols of the Guanzhong Basin, HNO_3 and H_2SO_4 have differential temporal formation rates, and that the difference in these rates is the reason why more NO_3 is observed relative to SO_4 at the urban information presented in this paper, I am not sure the authors have enough proof to support this hypothesis over other possibilities (for example, that the two sites were sampling different air masses). This is because Mt. Hua needs to be downwind of Xi'an in order to support their hypothesis and observations. However, nowhere in the text do the authors discuss whether Mt. Hua actually is downwind of Xi'an. Although it is only 80 km away from Xi'an, Mt. Hua is located in the free troposphere where air mass sources are likely to be very different than from the ground level urban site. In fact, there are multiple lines of evidence throughout the paper to indicate that the two sites are not sampling the same air masses. For example:
a) The differences in OC/EC ratios shown in Fig. 5
b) Later in the text the authors talk about how many aerosol components in Xi'an have primary emission sources in Xi'an. Thus, for Mt. Hua to be comparable to Xi'an, the site must be directly downwind of Xi'an instead of just sampling the larger scale air mass.
c) The later statement: "Our previous study (Wang et al., 2012b) found that during the DS II period secondary organic aerosols in the Mt. Hua air were mostly formed from the local sources rather than being transported from the upwind region..." See comment #2 for more on this topic. The authors refer to Table 2 to "confirm. . . a faster heterogeneous reaction of NOx with dust". However, Table 2 does not really confirm the author's proposed mechanism. If pollution is added in the valley but this pollution is not carried to the mountain, then having different ratios of NO_3 and SO_4 does not mean that differential formation rates of NO_3 and SO_4 are the explanation for the observations, because it could also mean that there were different NO_3 and SO_4 sources upwind for the mountain dust at some other site.
Back trajectories would be useful in showing whether Mt. Hua was likely to be downwind of Xi'an during the sampling period, as would a plot of dust and other constituents in PM10 at both sites (i.e. excluding the fine aerosols at Mt. Hua so that you are looking at the same thing), similar to Figure 3 but where data is shown for both sites. If somehow the authors can prove that Mt. Hua was receiving the same air masses as Xi'an (presumably after only hours to days due to the proximity of the sites), the authors should mention whether this timeframe is even fast enough for the proposed mechanism to occur. If they can't provide more solid proof of this hypothesis, I would recommend that they change this mechanism from a "finding" to just a potential explanation of the data.
Response: We agree that the mountain free troposphere has air mass sources different from that on the urban ground surface. But this is only true in the non-dust event. As we mentioned in the text and above response, during the DS II period both Mt. Hua and Xi'an were dominated by a same mass (see page 7, line 174-183). The objective of this study was to compare the vertical difference of aerosols not the horizontal, so it is not necessary to make upwind or downwind measurement. In fact, Mt. Hua and Xi'an are a pair of parallel sites dominated by a same air mass during the DS II event. (see Figure 3). We agree with the reviewer that back trajectories would be useful. We added a new figure showing the backward trajectories of air masses arriving in Xi'an and Mt. Hua during the non-dust and dust periods (see Figure 3). We also agree to the reviewer's comments "if pollution is added in the valley but this pollution is not carried to the mountain, then having different ratios of NO_3 and SO_4 does not mean that differential formation rates of NO_3 and SO_4 are the explanation for the observations, because it could also mean that there were different NO_3 and SO_4 sources upwind for the mountain dust at some other site". However, this is not the case in our study. Data from documents and our observation showed that concentrations of NO_2 and SO_2 in inland China are similar. For examples, annual means of NO_2 and SO_2 in Xi'an were 43 and 41 $\mu g\ m^{-3}$ during 2006-2007, respectively (Shen et al., 2008). Similar levels of NO_2 and SO_2 were also found in remote sites in inland China, for example, annual mean levels of NO_2 and SO_2 in Waliguan were 0.7±0.4 ppbv and 0.6±0.4 ppbv during 2007-2008 (Meng et al., 2010), and NO_2 and SO_2 in Qinghai Lake in March, 2012 were 2.7±1.6 and 2.2±0.7 ppbv (unpublished data). The above data indicate nitrate precursor level is similar to that of sulfate in inland Chin including Guanzhong Basin. Thus we believe that compared to that in the urban air the lower NO_3^-/SO_4^{2-} ratio in the mountain troposphere is mainly due to different formation rates of NO_3^- and SO_4^{2-}. More discussion was added. See page 9, line 230-236.

2) In a related topic to the first comment, I have some concerns about the Mt. Hua sampling setup; specifically that there is strong reason to believe that their samples were impacted by mixed local and free troposphere sources, which complicates the data interpretation. Unfortunately it is difficult to tell exactly what type of air was sampled at Mt. Hua because the authors do not talk at all (!) about how the samples were collected at the mountain site. This is a major omission and needs to be addressed. As best I can tell, size segregated samples were collected at the mountain top site for 24 hour periods. If this is the case, this complicates the data interpretation greatly because during the day in particular air tends to rise up mountain slopes because of heating. Also there will tend to be an upslope flow because the mountain is an obstacle to wind flow and hence you will get flow around and over the mountain. The study would have been much more solid if some attention had been paid to wind conditions during collection periods and if they

had collected samples only at night for free troposphere data or only during the day for local sources. Were diel PM_{10} measurements taken at Mt. Hua like in Xi'an? If so, why are these data not presented? Perhaps they can provide an idea of what the diel variation caused by mountain wind flow dynamics would have been.
Response: Sampling information on Mt. Hua aerosol collection was added, see page 7, line 172-174, and page 5, section 2.1. The mountain data have been already published, thus we did not tabulate the data again. Instead, we represented them using a figure format in Figure 3 for a comparison with the urban aerosols. As mentioned in the revised paper, the duration of the mountain sampling was 24hr not as that (12hr) in the urban site in order to collect enough materials for organic speciation.

3) I think the authors are missing an opportunity to discuss the implications of their surprising findings for WSON in the Guanzhong Basin. Firstly, this is the only study I know of presenting WSON in aerosols in China (although my knowledge of studies in this area is not comprehensive). Secondly, in Table 1, if I interpreted the units correctly (i.e. the values are ug N/m3 for WSON), then the levels of WSON observed in the samples is REALLY high compared to Saharan dust WSON levels, see Table R1 below. WSON contributing up to 90% of the WSTN in dust samples is near unprecedented and very surprising, because all the studies on WSON contribution to dust so far have total WSON concentrations and relative contributions to total nitrogen being much lower (see Table below). The high total WSON levels, however, are consistent with other measurements of DON in Chinese rain (Zhang et al. (2008), which are much higher other locations observed to date. This indicates that the chemistry and/or emissions of WSON in this region is very different from other regions of the world.
This study is also unique as it is the only study looking at aerosol WSON from Gobi desert dust and not Saharan dust. I agree with the authors that it seems likely that the high concentrations and high relative contributions of WSON are due to anthropogenic WSON absorbed onto dust (also consistent with Zhang et al. (2008)). Again, it seems worth emphasizing that WSON in dust here is 2 orders of magnitude higher than observed in other dust sites. It is also interesting that the WSON levels are higher for the dust than non-dust periods, which indicates an additional chemical mechanism for WSON formation from inorganic N or an additional WSON source upwind that was not present in non-dust periods. This was not discussed but could be.
Response: We thank the reviewer's comments and suggestion. We agree that the results about WSON are very interesting and the chemistry of WSON in China is very important. We have been analyzing the WSON on a molecular level for investigation the source and formation mechanism. In the current paper, we added some discussion and statements into the revised paper. The in-depth analysis of WSON is to be presented in another paper. See page 8, line 201-211.

On p. 21362 the authors say that, "Such an enhancement of WSON was also observed in Miami and Barbados (Zamora et al., 2011) and Eastern Mediterranean (Violaki and Mihalopoulos, 2010) when Africa dust presented." While that may be true, from the table below it seems that the dynamics here are different, or at least are happening on a much larger magnitude than observed before. I think this may be worth mentioning.
As a final note, many known WSON components are highly toxic to humans. Perhaps this is a topic worth suggesting for future research in your conclusions.
Response: Suggestion taken. We added more statements in the revised version. See page 8, line 203-209, and page 20, line 499-500.

Additional comments
4) Throughout the text the authors make various statements that I consider to be too bold, which should either be toned down, changed, or substantiated. Specifically:
a.) P. 21366: It seems a little strong to say "Moreover, ammonia cannot react with coarse particles because of their alkaline nature." While it is true that the majority of ammonium is in the fine mode, this does not mean that under all conditions it cannot react with coarse mode particles. In fact the authors did measure small amounts of NH_4 in the coarse mode aerosols.
Response: Suggestion taken. We deleted this sentence. See page 12-13, line 309-314.

b). P. 21369, l. 20: The authors say, "The small peak of coarse mode of WSOC can be explained by a nature source such as pollen and soil (Fig. 8a), because water-soluble organic compounds like glucose (Graham et al., 2002; Wang et al., 2006a, b, 2009, 2011a, b) and humic acid (Brooks et al., 2004; Dinar et al., 2006; Havers et al., 1998) are enriched in these sources." I don't think from this study that the authors can conclusively say that these natural sources of WSON are the only possible source of coarse mode WSOC. In fact, they observed WSON in the coarse mode, which

they suggest is likely of anthropogenic origin. By definition as organic compounds, there must be some carbon associated with these WSON compounds that is of the same anthropogenic origin.
Response. We agree with the reviewer's comments above. To make the related conclusion more accurate, we revised the statement. See page 16, line 390-395.

c) P. 21372, l. 12: The authors say, "Compared to those in the urban air the alpine aerosols are more aged due to long-range transport, thus in the nonevent fine particles in Mt. Hua are more acidic, rendering the cations/anions ratios smaller in Mt. Hua than in Xi'an (0.6 vs. 0.8) (Fig. 9a, b)." The authors do not supply a reason why aging would mean more acidic aerosols. Is it because they believe SO_4 is developed most in aged aerosols? Even if this were the case, at some other point in the text they say that Ca^{2+} was also developed over time, which should neutralize some of the SO_4. And what about chemistry of organic acids (e.g., formic and acetic acids) and organic bases (e.g., amines)? These compounds are not well understood and data on these compounds is not presented. However, these compounds do impact pH and their concentrations can change with particle aging. Thus I think the connection between aging and acidity in this statement is too bold.
Response: Ca^{2+} and NH_4^+ are the two major cations in fine particles while major anions in fine particles are NO_3^- and SO_4^{2-}. As shown in Figure 11a, NO_3^-/SO_4^{2-} ratio of fine particles in Mt. Hua is much lower than that in Xi'an due to enhanced formation of sulfate in the mountain troposphere. From Figure 7a, we can see that compared to that in Xi'an sulfate content in Mt. Hua is much higher and NH_4^+ and Ca^{2+} contents in the mountain aerosols are slightly lower. Thus the fact that the mountain fine particles during the non-vent are more acidic is most likely caused by an increased formation of sulfate. To make the statement more accurate, we re-wrote the sentence. See page 19, line 464-467.

d) P. 21373, l. 25: "On the contrary, OC became the highest in the event at both sites, followed by SO_4^{2-}, Ca^{2+} and NO_3^-, indicating the importance of input of biota in Gobi desert." As far as I could tell, the authors did not discuss in this paper the impact of secondary aerosols at Mt. Hua as a source of OC. Also, how do SO_4, Ca, and NO_3 concentrations imply the importance of Gobi desert biota? This statement needs to be clarified, at minimum. In the abstract and later in the text, the authors claim that heterogeneous NO_3 formation is more favorable in the city, but it was not clear to me why they think that is. Unless they can prove this, this statement should be changed or removed.
Response: In our previous paper we measured the biomarkers like n-alkanes, fatty acids, fatty alcohols, and sugars in the Mt. Hua samples. Based on the variation and composition of these biomarkers, we found an increased OC during the dust storm periods was mostly caused by input of biota in Gobi desert (Wang et al., 2012). These statements have been complemented in the revised version. See page 4, line 104-105, and page 16, line 394-400. NO_3^- precursors are much higher on the urban ground surface than in the mountain free troposphere, thus we think heterogeneous NO_3 formation is more favorable in the city. However, we did not measured the NO_3^- precursors, thus removed the related statement.

5) The terms "event" and "nonevent" are used a great deal throughout the text. I think these terms are a little confusing, and suggest changing these terms to "dust event" and "non-dust event" since there could be other kinds of events (e.g., biomass burning events, etc.).
Response: Suggestion taken. See the changes throughout the paper.

6) In the introduction, more can be done addressing what previous studies have found and what questions were left that this study helps answer. In other words, it would help to write more clearly about why this study is unique and interesting.
Response: Suggestion taken. We added a brief introduction on our previous related work in the introduction section. See page 4, line 102-106. We also briefly introduced the importance of the current study in the introduction section. See page 4, line 112-116.

7) The OC/EC ratios were given at the beginning of the results section, but the implications of these data were not discussed. It would help to talk a little about the implications of this ratio for aging of the aerosols and secondary organic aerosol formation. I also suggest placing these observed ratios in context of other sites in the region and in the world. The OC/EC results indicate that the dust samples are more aged than the non-dust samples- surely this is relevant for the data interpretation?
Response: As mentioned above, input of biota in Gobi desert contributed significantly to the OC in the samples during the dust events. And plant emission in the mountain region is an important source of OC for the mountain samples during the non-dust periods (Li et al., 2011; Li et al., 2012; Wang et al., 2012). Thus, we think the higher OC/EC ratio does not mean that the dust samples are more aged than the non-dust samples. Furthermore, due to

different sources of organic aerosols, it is difficult to discuss aerosol ageing and secondary organic aerosol formation only based on the OC/EC ratio. Therefore we think in the current paper it is not necessary to give more discussion on OC/EC ratio. In fact, we analyzed organic aerosols in the Xi'an samples using a GC/MS technique, and one more paper has been preparing, which will discuss aerosol ageing and SOA formation, as as well as OC/EC ratio. We will take the suggestion above in this forthcoming paper.

8) Maybe I am misunderstanding something, but the WSON data presented in Table 1 seem inconsistent with the data discussed in the text. Where does the value of 0.7 for the WSON/WSTN ratio come from (p. 21462, l. 19)? From the values in Table 1 it looks more like 0.5?
Response: As shown in Table 1, WSON/WSTN should be 0.7 and 0.8±0.3, not 0.7±0.3. We corrected the number. See Table 1 in page 28.

9) P. 21365, l. 6: it would be really interesting if you could do the same calculation for WSON and WSOC, since anthropogenic WSON is supposed to be important.
Response: Suggestion taken. In our previous work (Wang et al., 2012), we did not measure WSOC in the mountain samples. In this revision stage, we first determined WSOC in the mountain samples and then calculated WSOC formed by local pollutants with dust using the equation. Because of the limited materials in the mountain filters, WSON was not determined. Thus WSON formed by the local urban pollutants was not calculated for the revised paper. Based on the WSOC calculation, we found that compared to the mountain dust samples increased OC in the urban dust samples is equal to that of WSOC, which are 7.4 $\mu g\ m^{-3}$ for WSOC and 7.6 $\mu g\ m^{-3}$ for OC. In other words, compared to OC in the mountain dust the increased OC in the urban dust was caused by heterogeneous reaction and/or adsorption of gaseous organics onto dust (e.g., organic acids and amines), which are water-soluble. The related discussion has been added into the text. See page 11, lines 267-269, and 274-277.

10) Section 3.2.2. This section reads a little bit like a data dump and not like a compelling scientific story. It would improve the paper if the authors could make this section more focused.
Response. Suggestion taken. We modified this section.

11) Section 3.3. I am confused. In the methods it was stated that pH was measured directly. But in this section only equivalent ions are presented, which to my understanding, are not as indicative of actual aerosol acidity because the influences of buffering and acid displacement are not accounted for (Keene et al., 2004). Why not just present measured pH values?
Response: In this paper the pH values presented are related to the particle loadings on the filter. The volume of pure water used for extraction is fixed, but particle concentration of each filter is quite variable. For a less amount of acidic sample, the measured pH value is probably larger than a sample with high amount of particles. Thus, we think equivalent ratio is more reasonable than the direct measured pH for discussing our sample acidity.

12) Section 3.3 How long were samples stored until analysis and under what conditions? This might impact post-collection production of acidity via oxidation of sulfur or via HCl volatilization, see (Keene et al., 2002)
Response. Our samples were dried in a desiccator and stored in a freezer at –20°C prior to analysis, which is a typical method for aerosol sample storage. See page 5, line 128-129.

13) Section 3.3 What about the buffering impacts of HCl phase partitioning and sulfur partitioning shown in Keene 2002? These are not discussed.
Response. We think the buffering impacts of HCl phase partitioning and sulfur partitioning are negligible in this study, because the particles Keene et al (2002, 2004) studied is marine aerosol, of which major component is NaCl. NaCl is hygroscopic and may uptake water under a marine condition, thus buffering impacts of HCl phase partitioning and sulfur partitioning is important. However, it is not the case for our study, because what we studied is continental aerosol, which typically contains more than 30% mineral dust.

Additional, more minor comments
14) The abstract is quite long, I would suggest shortening it. ACP does not require that the abstract be only one paragraph, but it is my personal feeling that the article might be more citation-friendly if the abstract were limited to one paragraph.
Response. Suggestion taken. We re-wrote the abstract. See page 2, the abstract section.

15) EC, OC are not defined in the abstract. Since these abbreviations are not considered standard outside of the atmospheric chemistry community, to be on the safe side I might define them in the abstract.
Response. Suggestion taken. See page 2, line 45.

16) To avoid confusion in the introduction, specify “mineral dust” and/or “aerosols,” and avoid using the term “dust.” For example, this is currently a problem when the authors reference the health impacts of what they call “dusts,” because it is not clear whether they are talking about mineral dust or aerosols in general. To my knowledge, the health impacts of mineral dust are still unclear; however, aerosols in general of certain size class are thought to have health impacts.
Response. Suggestion taken. We changed as “dust event” and “non-dust event” throughout the paper.

17) Please give coordinates of the sampling sites.
Response. Suggestion taken. See page 4, line 96-97.

18) The authors might mention in the methods that there are problems associated with sonication, which include mechanical rupture of particles that might lead to greater ion release and particle solubility than would naturally occur.
Response. Suggestion taken. See page 6, line 154-157.

19) P. 21361, l. 4: “One fourth of the filter was cut into pieces and extracted 3 times. . .”. I got confused about the actual procedure from this sentence. Did the authors mean 3 different quarters were extracted separately one time each, or one quarter was subsequently extracted 3 different times and then the extractants were combined? Please specify.
Response. Suggestion taken. We re-wrote the sentence as “one fourth of the filter was cut into pieces and extracted with Milli-Q pure water under sonication for three times in each”. See page 6, line 145-146.

20) What was the volume of water that the samples were extracted into for the pH determination?
Response. The volume of water for pH determination is 15 mL. We added this information into the text. See page 6, line 145.

21) Since Mt. Tai is periodically referred to, it should also be put it in the map in Figure 1.
Response. Suggestion taken. See Figure 1.

22) In the text the authors stated that the units for Table 1 WSON values were ug/m^3, which I assume to mean ug N/m3, is this correct? To be more inline with literature WSON conventions, the authors might think of changing the units to be in nmol N/m^3.
Response. In this table we also presented WSOC and other species concentrations such as particle mass and inorganic ions. In order to let readers easily compare these data, we think a consistent unit is better, thus we did not change the WSON as nmol N/m^3.

23) P. 21365, l. 16: Regarding the following statement: “component 1 is associate with high loadings with F−, Cl−, K$^+$, WSOC, EC and OC, respectively, and represents fossil fuel and biomass combustion source, because F−, K+ and EC are largely derived from fossil fuel and biomass burning emission in China (Dan et al., 2004; Kline et al., 2004; Zhang et al., 2011).” Note that a major source of WSOC is also from fossil fuel and biomass burning.
Response. We agree with the reviewer’s comment that a major source of WSOC is also from fossil fuel and biomass burning. We revised the statement. See page 11, line 284-287.

24) P. 21367, l.5: Re: “The fine fraction of Na$^+$ can be explained by a biomass burning contribution, because biomass smoke contains a significant amount of Na_2SO_4 (Andreae et al., 1998).” For your interest, note that due to anthropogenic Na$^+$ sources, the Asian continent is actually a larger source of Na$^+$ to the coastal regions than the ocean is (e.g., Ooki et al., 2002, Sources of sodium in atmospheric fine particles, Atmospheric Environment, Volume 36, Issue 27, 2002, Pages 4367–4374).
Response. We thank the reviewer for the comments above. Biomass burning, i.e., biofuel burning, is an important anthropogenic source of Na$^+$ in inland China.

25) P. 21369, l.15: Re: “Several studies have reported that particulate WSOC in an urban environment is largely formed from photochemical oxidation of organic gases (Agarwal et al., 2010; Ram and Sarin, 2010; Salma et al., 2007;

Wang et al., 2012a, b; Yu et al., 2004, 2005).” It is important to note that this is only the case for sites where biomass burning is low.
Response. We agree with the comment above and revised the statements. See page 15, line 386-389.

26) p. 21370, l.18: “Water-soluble organic nitrogen (WSON) are a class of complex nitrogen-containing compounds such as urea, amines, amino acids, eptides and proteins, which are derived from urban pollution, biomass burning and dust (Altieri et al., 2012; Cape et al., 2011; Chen and Chen, 2010; Mace et al., 2003a, b; Violaki and Mihalopoulos, 2011; Zhang and Anastasio, 2003).” Note that there are other sources of WSON also, such as sea spray.
Response. We agree that see spray is a source of WSON and revised the sentence. See page 17, line 418.

TECHNICAL CORRECTIONS
1) P. 21357, L. 13: Remove the “An” at the beginning of the sentence for proper English
Response. Based on the previous comments, we have shortened the abstract. This sentence has been delete.

2) P. 21357, L. 16: Change to say, “To our knowledge, this is the first simultaneous observation of . . .”
Response. We deleted this sentence when re-writing the abstract.

3) P. 21358, First sentence: Rephrase, e.g. “dust is a major source of particulate matter to the atmosphere.”
Response. Suggestion taken. See page 3, line 68.

4) P. 21358, Reference should be McNaughton not Mcnaughton (make sure to make this change later on in the text too).
Response. Suggestion taken. We corrected the mistake.

5) P. 21358, L.8 : “These dusts can influence” to “Dust can influence. . .”
Response. Suggestion taken. See page 3, line 69.

6) P. 21358, L.8 : In the first paragraph where it states, “Global annual mean burden of aerosol dust is about 20 Tg (_ 40 %) (Mcnaughton et al., 2009).” I am not sure which measure of variation _ refers to- standard error, deviation, something else? Other readers might have the same problem- could you please specify?
Response. Suggestion taken. We changed the number format. See page 3, line 69.

7) P. 21358, L.13: Huebert et al., 2003 is not an appropriate reference for the health impacts of aerosols.
Response. Suggestion taken. We changed the reference. See page 3, line 74.

8) L. 21 p. 21358- sea salt not seal salt
Response. Suggestion taken. We corrected it. See page 3, line 80.

9) L. 19, p. 21358- “During the transport” should be changed to “During transport”
Response. Suggestion taken. See page 3, line 79.

10) L. 17, p. 21359, from the Gobi desert
Response. Suggestion taken. See page 4, line 101.

11) L. 9, p. 21359, change to “where the annual”
Response. Suggestion taken. See page 4, line 93.

12) Add units for Table 1
Response. Suggestion taken. See Table 1 in page 28.

13) I found Table 1 difficult to read. I would suggest placing the mean value first with a ± and then the standard deviation followed by the max and min in brackets next to it so that it is easier to read, for example for the first value: 250±79 (58-420)
Response. Suggestion taken. We changed the format. See Table 1 in page 28.

14) P. 21363, l. 10: “organic matter” not “organic matters”
Response. We changed the sentence, see page 9, line 220-222.

15) P. 21363, l. 19: “Mt. Tai (0.68 ± 0.32, 0.43–1.0) compared to those in the nonevent” (reference?)
Response. Suggestion taken. See page 9, line 226.

16) P. 21365 l. 14: major species in PM10 were
Response. Suggestion taken. See page 11, line 283.

17) P. 21365 l. 16: component 1 is associated with
Response. Suggestion taken. See page 11, line 285.

18) P. 21366, l. 2: change “Almost all physicochemical and optical properties of aerosol are dependent on its size (Hinds, 1999). The size of ambient aerosol is not constant, which is always variable during the transport via. . .” to “Almost all aerosol physicochemical and optical properties are size-dependent (Hinds, 1999). The size of ambient aerosol varies during transport due to. . .”
Response. Suggestion taken. See page 12, line 296-298.

19) P. 21366, l. 26: “The small fraction of ammonium in the coarse fraction is mostly derived from suspended soil containing fertilizer. Generally, fine mode of K+ is mostly derived from biomass burning while the coarse mode K+ can be attributed to suspended soil (Fig. 7e).”- References?
Response. Suggestion taken. See page 13, line 318.

20) P. 21367, l.2: “Mg^{2+} and Ca^{2+} are crust species,” change to “Mg^{2+} and Ca^{2+} have dominant crustal origins, and”
Response. Suggestion taken. See page 13, line 320.

21) p. 21367, l.4: “Na+ is an important component in soil, thus it occurred abundantly in coarse fraction” to “Na+ is an important component in soil, thus it occurred abundantly in the coarse fraction”
Response. Suggestion taken. See page 13, line 321-323.

22) p. 21367, l.15: “The disappeared peak of Cl^- in the fine mode at the mountain site indicates that the biomass burning derived aerosols was more aged when they arrived in the mountaintop. . .” to “That the fine mode peak of Cl^- was not evident at the mountain site indicates that the biomass burning derived aerosols were aged when they arrived at the mountaintop. . .”
Response. We deleted this sentence and related discussion to make this paragraph more concisely according to the reviewer’s comments above.

23) p. 21367, l.20: “In addition, the coarse mode of sulfate is in part directly originated from soil since loess and desert dust also contain certain amount of sulfate . . .” to “In addition, sulfate in the coarse mode fraction also has a soil primary emission source, since loess and desert dust also contain a certain amount of sulfate. . .”
Response. Suggestion taken. See page 13-14, line 336-338.

24) p. 21367, l.23: “labile to decompose into gaseous NH_3 and HNO_3” to “easily converts to gaseous NH_3 and HNO_3”
Response. Suggestion taken. See page 14, line 339.

25) p. 21368, l.7: “Compared to those in the nonevent particle mass at both sites dominated in the coarse mode with a disappeared peak in the fine mode (Fig. 7s, t).” to something like, “Compared to particle size distributions sampled in the nonevent, a greater portion of particles during the dust event were present in the coarse mode (Fig. 7s, t).”
Response. Suggestion taken. See 14, line 352.

26) p. 21368, l.9: “Like that in the nonevent ammonium is still enriched. . .” to “As in the nonevent, ammonium was enriched. . .”
Response. Suggestion taken. See page 14, line 353.

27) P. 21369, l.10: “responsible for such a coarse mode of increase” to “responsible for the shift towards the coarse mode”

Response. Suggestion taken. See page 15, line 379.

28) p. 21370, l.4: reference?
Response. Suggestion taken. See page 16, line 403.

29) p. 21370, l.16: "which was 3.4 $\mu g\ m^{-3}$ in the DS II event and 10 times higher than that (0.3±0.1 $\mu g\ m^{-3}$) in the nonevent." Do you mean lower?
Response. No, we mean WSIC became higher in the DS II event. We re-wrote the sentence to make it clear. See page 17, line 414-415.

30) p. 21373, l. 14: This first sentence is confusing and should be rewritten, "Springtime of PM_{10} and size-segregated" to "Springtime PM_{10} and size-segregated"
Response. Suggestion taken. See page 20, line 493.

31) p. 21373, l. 20: "High level of WSON was found in the DS II time due to the deposition of anthropogenic WSON onto dust and the input of biological organism in Gobi desert dust." to "High levels of WSON were found in the DS II samples, due to the deposition of anthropogenic WSON onto dust and the input of biological organisms in Gobi desert dust."
Response. Suggestion taken. See page 20, line 497.

32) Change "Furthermore, dusts are of adverse. . ." to "Furthermore, aerosols can cause adverse. . ."
Response. Suggestion taken. See page 3, line 72.

Anonymous Referee #2

General comments
The manuscript by Wang et al. conducted the synchronous measurements of aerosol composition and size distributions at a ground site and a mountain site in the free troposphere in central China during the spring of 2009. The results were compared with their previous observations at Mt. Hua to investigate the aerosol chemistry differences in the two different environments. In particular, the impact of dust storm from Gobi deserts on the aerosol chemistry at the two sites was discussed in detail. The data is very interesting and unique. Some important findings are reported. For example, the authors found that the heterogeneous formation of nitrate could result in urban fine particles more acidic during dust storm, which is surprising. Such finding might have important implications since the NOx level in China continues to increase in recent years, and the nitrate chemistry would become more important than before. It's also interesting that the WSON and organic matters were enhanced during dust storm. The long-range transport of dust with high WSON, thus, might have a potential impact on the productivity of marine phytoplankton. Overall, this study increased our understanding of aerosol chemistry between the ground surface and the free troposphere over central China. It should be accepted for publication. The referee #1 has given detailed comments on this manuscript, I, here, have a few comments listed below.
Response. We thank the reviewer's comments above.

1. Page 21357, line 12-15. It appears that the increased nitrogen-containing species during the dust event is primarily from Gobi desert, are any other sources for WSON?
Response: Deposition of anthropogenic pollutants like amines onto dust is an important origin of WSON in the dust samples. See page 11, line 277, and page 17, line427-432.

2. Section 2.1, How about filter blanks in this study? A brief description is needed.
Response: Suggestion taken. See page 5, line 131-135.

3. Page 21371, line 2-6, Could authors give more explanation how the water-soluble organic nitrogen compounds were adsorbed onto dust?

Response: In the current stage we cannot give a clear explanation about the mechanism of adsorption of water-soluble organic nitrogen compounds onto dust. Instead, we only give a brief explanation. For example, see page 17, line 427-432.

4. Section 3.3. The results that fine particles in Xi'an during the DS II episode became more acidic are very interesting. The authors claimed that such phenomenon was observed for the first time in China. I am wondering if there are any lab studies on heterogeneous reactions, e.g., HNO_3^+ dust, to support your conclusion.
Response: Yes, there are several lab studies supporting our explanation. For example, Sullivan et al (2009) reported that their lab work showed fine particulate calcite can rapidly convert into $Ca(NO_3)_2$, which will subsequently uptake water, forming a aqueous phase on the dust surface and further promoting HNO_3 partitioning into the aqueous phase (Goodman et al., 2000), resulting in fine particle being acidic (He et al., 2012). The related discussion has been added into the text. See page 18, line 448-458.

5. Page 21363, paragraph 3. Cloud processing at the summit of Mt. Hua should be very important for the sulfate formation, which might also explain the lower ratio of NO_3/SO_4 observed at Mt. Hua than the ground site.
Response: We agree with the reviewer that cloud processing at the summit of Mt. Hua is very important for the sulfate formation. We added this statement into the text. See page 9, line 230-236.

6. Page 21357, line 8, Suggest to replace "event" by "dust event" in the text, which is clearer to readers.
Response: Suggestion taken. We replaced "event" by "dust event" in the text.

Anonymous Referee #3

This paper provides an insight on the interaction between dust and pollution. I would like to make minor comments on that. Dusts enhance the heterogeneous reaction but it depends on the mineralogy of dust particles. For example Al_2SiO_5 has an affinity to SO_2 and it helps to convert SO_2 to sulfate. More caution is required for the explanation about the interaction between dust particles and air pollution.
Response. We thank the reviewer's the comments above.

1. The authors said that the enhancement of the heterogeneous formation of nitrate in the dust event makes the urban fine particles more acidic. If more nitrate may be the reason, in the sense of thermodynamic equilibrium, a certain amount of cation is needed to convert nitric acid to nitrate. This seemed not to be the case in Fig. 5. It seemed that less ammonium cannot be ignored for the less acidity in addition to that.
Response: We think there are two reasons causing the urban fine particles become acidic, one is an enhanced formation of nitrate, another is a less amount of ammonium. Thus, we modified related discussion. Figure 6, which is named as Figure 5 in the ACPD version, shows the composition of PM_{10} not fine particles. The detailed discussion was revised. See page 19, line 471-485.

2 The authors explained less acidity in the mountain by decreasing formation of nitrate in the event. Both sulfate and nitrate were formed less in the event than in the nonevent and it seemed that more cation such as ammonium may be contributed to the less acidity. Further explanation needs to be explored for acidity issues.
Response: We agree that more cation such as ammonium contributed to the less acidity during the non-dust event in comparison to the dust event. Indeed, as mentioned by the reviewer that both sulfate and nitrate were formed less in the event than in the nonevent, but this is referred to PM10 (see Table 1). Since we talked about the acidity of fine particles, Figure 11 can give more reasonable explanation. As shown in Figures 11a and 11b, for the urban samples we can find that relative abundance of NH_4^+ plus Ca^{2+} to NO_3^- plus SO_4^{2-} was lower in the event than in the non-event, and NO_3^-/SO_4^{2-} ratio was higher in the event than in the non-event. Therefore we think urban fine particles during the event became more acidic is due to an enhanced formation of nitrate.

3. The same air mass should be a necessary condition to explore the interaction between dust and air pollution. In a nonevent, two sites may be under a mountain-valley circulation which turns over its direction during a day. Of course, when the synoptic circulation is dominant, the local circulation is ignored. Authors might need to check it.
In addition to the above issues, I hope authors might elaborate on exploring the issues of OC dominating over the dust event.

Response: Suggestion taken. As for the issues of same air mass, we checked it by using backward trajectories. The air mass trajectory analysis, and other data such as particle loading, size distribution patterns, and chemical composition concentrations clearly demonstrated that both Xi'an and Mt. Hua during the DS II event were dominated by a same air mass. A detailed response can be found in our answer to the referee #1. For example, see the backward trajectory in Figure 3, and related discussion in page 11, line 267-282, page 4, line 105-109, and page 7, line 174-183.

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