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Interactive comment on “Impact of Gobi desert dust on aerosol chemistry of Xi’an, inland China during spring 2009: differences in composition and size distribution between the urban ground surface and the mountain atmosphere” by G. H. Wang et al.

Anonymous Referee #1

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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,

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Discussion Paper



b) temporal differences in HNO₃ and H₂SO₄ formation caused large differences between the urban and mountain sites, for example causing:

1. more SO₄ and less NO₃ at the mountain site,
2. greater acidity of fine particles at the mountain site due to continuous SO₄ oxidation and thus greater H₂SO₄ 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 NO₃ to form from heterogeneous reactions in the city and less HNO₃ 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 NO₃ and SO₄ concentrations between the sites was due to differential temporal development of NO₃ and SO₄. Thus some of the conclusions are not strongly supported by the results presented.

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

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, HNO₃ and H₂SO₄ have differential temporal formation rates, and that the difference in these rates is the reason why more NO₃ is observed relative to SO₄ at the urban

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site of Xi'an than at the mountain site of Mt. Hua. However, based on the 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 NO_x 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 NO₃ and SO₄ does not mean that differential formation rates of NO₃ and SO₄ are the explanation for the observations, because it could also mean that there were different NO₃ and SO₄ sources upwind for the mountain dust at some other site.

[Full Screen / Esc](#)[Printer-friendly Version](#)[Interactive Discussion](#)[Discussion Paper](#)

Interactive
Comment

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.

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 PM₁₀ 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.

[Full Screen / Esc](#)[Printer-friendly Version](#)[Interactive Discussion](#)[Discussion Paper](#)

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

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.

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 NH₄ in the coarse mode aerosols. 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. 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 SO₄ is developed most in aged aerosols? Even if this were the case, at some other point in the text they say that Ca²⁺ was also developed over time, which should neutralize some of the SO₄. 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,

Full Screen / Esc

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Discussion Paper



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. d. P. 21373, l. 25: “On the contrary, OC became the highest in the event at both sites, followed by SO₄²⁻, Ca²⁺ and NO₃⁻, 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 SO₄, Ca, and NO₃ concentrations imply the importance of Gobi desert biota? This statement needs to be clarified, at minimum. e. In the abstract and later in the text, the authors claim that heterogeneous NO₃ 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.

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.).

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.

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?

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

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looks more like 0.5?

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.

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.

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?

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)

13) Section 3.3 What about the buffering impacts of HCl phase partitioning and sulfur partitioning shown in Keene 2002? These are not discussed.

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.

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.

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

Full Screen / Esc

Printer-friendly Version

Interactive Discussion

Discussion Paper



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.

17) Please give coordinates of the sampling sites

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.

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.

20) What was the volume of water that the samples were extracted into for the pH determination?

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

22) In the text the authors stated that the units for Table 1 WSON values were $\mu\text{g}/\text{m}^3$, which I assume to mean $\mu\text{g N}/\text{m}^3$, is this correct? To be more inline with literature WSON conventions, the authors might think of changing the units to be in $\text{nmol N}/\text{m}^3$.

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.

[Full Screen / Esc](#)[Printer-friendly Version](#)[Interactive Discussion](#)[Discussion Paper](#)

24) P. 21367, I.5: Re: “The fine fraction of Na⁺ can be explained by a biomass burning contribution, because biomass smoke contains a significant amount of Na₂SO₄ (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).

25) P. 21369, I.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.

26) p. 21370, I.18: “Water-soluble organic nitrogen (WSON) are a class of complex nitrogen-containing compounds such as urea, amines, amino acids, peptides 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.

TECHNICAL CORRECTIONS

- 1) P. 21357, L. 13: Remove the “An” at the beginning of the sentence for proper English
- 2) P. 21357, L. 16: Change to say, “To our knowledge, this is the first simultaneous observation of ...”
- 3) P. 21358, First sentence: Rephrase, e.g. “dust is a major source of particulate matter to the atmosphere.”
- 4) P. 21358, Reference should be McNaughton not Mcnaughton (make sure to make this change later on in the text too).
- 5) P. 21358, L.8 : “These dusts can influence” to “Dust can influence...”

- 6) P. 21358, L.8 : In the first paragraph where it states, “Global annual mean burden of aerosol dust is about 20 Tg (δ 40 %) (Mcnaughton et al., 2009).” I am not sure which measure of variation δ refers to- standard error, deviation, something else? Other readers might have the same problem- could you please specify?
- 7) P. 21358, L.13: Huebert et al., 2003 is not an appropriate reference for the health impacts of aerosols.
- 8) L. 21 p. 21358- sea salt not seal salt
- 9) L. 19, p. 21359- “During the transport” should be changed to “During transport”
- 10) L. 17, p. 21359, from the Gobi desert
- 11) L. 9, p. 21359, change to “where the annual”
- 12) Add units for Table 1
- 13) I found Table 1 difficult to read. I would suggest placing the mean value first with a \pm 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)
- 14) P. 21363, l. 10: “organic matter” not “organic matters”
- 15) P. 21363, l. 19: “Mt. Tai (0.68 ± 0.32 , 0.43–1.0) compared to those in the nonevent” (reference?)
- 16) P. 21365 l. 14: major species in PM10 were
- 17) P. 21365 l. 16: component 1 is associated with
- 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 physico-chemical and optical properties are size-dependent (Hinds, 1999). The size of ambient aerosol varies during transport due to. . .”

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?

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”

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”

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. . .”

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. . .”

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 ”

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).”

26) p. 21368, l.9: “Like that in the nonevent ammonium is still enriched. . .” to “As in the

[Full Screen / Esc](#)[Printer-friendly Version](#)[Interactive Discussion](#)[Discussion Paper](#)

nonevent, ammonium was enriched. . .”

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

28) p. 21370, l.4: reference?

29) p. 21370, l.16: “which was $3.4 \mu\text{g m}^{-3}$ in the DS II event and 10 times higher than that ($0.3 \pm 0.1 \mu\text{g m}^{-3}$) in the nonevent.” Do you mean lower?

30) p. 21373, l. 14: This first sentence is confusing and should be rewritten, “Spring-time of PM10 and size-segregated” to “Springtime PM10 and size-segregated”

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.”

32) Change “Furthermore, dusts are of adverse. . .” to “Furthermore, aerosols can cause adverse. . .”

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[Full Screen / Esc](#)[Printer-friendly Version](#)[Interactive Discussion](#)[Discussion Paper](#)

Interactive
Comment

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Interactive comment on *Atmos. Chem. Phys. Discuss.*, 12, 21355, 2012.

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Table R1: Literature values of aerosol WSON (nmol N m^{-3}) in locations influenced by dust; when available, samples known to be influenced by dust are specifically shown (i.e. for Lesworth et al., 2010 and Zamora et al., 2011).

Reference	Location	Dust source	n	WSON/ WSTN (%)	NH ₄	NO ₃	NH ₄ +NO ₃	WSON
This study: Dust	Xi'an, China	Gobi	4	70%	188	157	345	1163
This study: Non-dust	Xi'an, China	Gobi	60	35%	583	411	995	432
Mace et al., 2003	Erdemli, Turkey	Saharan	39	26±28 %	49	36	85	29
Chen et al., 2006	Gulf of Aqaba, Israel	Saharan	31	11%	23	42	65	10
	Gulf of Aqaba, Israel	Saharan	137	9%	25	39	64	8
Chen et al., 2007	Finokalia, Greece	Saharan	62	13%	79	28	107	17
Violaki and Mihalopoulos, 2010		North Atlantic Ocean	Saharan	14	~23%	~13	~23	~36
Lesworth et al., 2010	Barbados	Saharan	20	6-14%	11	10	22	1
Zamora et al. 2011	Miami, USA	Saharan	25	4-9%	28	26	54	2

Fig. 1. Table R1

[Full Screen / Esc](#)
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[Interactive Discussion](#)
[Discussion Paper](#)
