

## ***Interactive comment on “Spatial and seasonal variability of PM<sub>2.5</sub> acidity at two Chinese megacities: insights into the formation of secondary inorganic aerosols” by K. He et al.***

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We sincerely thank the reviewer for the valuable comments and suggestions to improve the manuscript.

Comments: Page 10, First paragraph. The authors write ‘Concentrations of SO<sub>4</sub>–2 for all the sites were higher at Chongqing than at Beijing, whereas NO–3 showed the opposite spatial pattern with higher concentrations at Beijing than at Chongqing.’ It is true if only looking at the concentration. But the PM<sub>2.5</sub> concentration is also higher at Chongqing than at Beijing. From the data showed in Table 1, the proportion of SO<sub>4</sub>–2 for MY of Beijing is similar to that of BB in Chongqing rural site, which is not lower than

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Chongqing. Please give some explanation for this and it should be better using the species proportion.

Response: Even from the view of species proportion, the contributions of SO<sub>4</sub><sup>2-</sup> were still generally higher in Chongqing (JB: 0.20; DDK: 0.19; XS: 0.18) in PM<sub>2.5</sub> than in Beijing (TH: 0.13; MY: 0.19), and contributions of NO<sub>3</sub><sup>-</sup> were lower in Chongqing (all: 0.04) than in Beijing (both: 0.09). The high proportion of SO<sub>4</sub> in MY, the background site of Beijing, was probably due to the more homogeneous spatial distribution of sulfate in this region than the other aerosol species, as has been found by other studies (Zhao et al., 2009; Guo et al., 2010). It also indicates that the regional influence of sulfate in Beijing was as important as in Chongqing. The above discussion has been added in the revised manuscript.

Comments: Page 11, Last line. (<10 %; urban: 0.016–0.17, rural: 0.15), the value of 0.016 may be 0.16, please check and correct it.

Response: The number was corrected according to the comments in the revised manuscript.

Comments: Page 12, Last paragraph & Page 13, First Paragraph: The authors list some study results in different cities (Table 2), and conclude that there is no consistent pattern in inter-annual trends. Maybe it is true. But the aerosol sampling time, sites, sampling methods and even laboratory analysis and meteorological conditions may have differences in each study, which would result in big errors among each study. To some extent, the comparison of this is weak or unnecessary to reveal some scientific knowledge in the related studies. Here is one suggestion: conducting more than 3 years sample and analysis in same sites and using the same method to find some trends in inter-annual and more complete understand of.

Response: It is true that there are many factors weakening the relevance of the comparison in Table 2, but their influences have been reduced as we used the ratio of 4 major ions rather than the concentrations. As the reviewer suggested, Table 2 also

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included our previous study in Beijing (He et al., 2001), which used the same method as in this study. Of course, long term observations at more regional sites are still badly needed.

Comments: Page 15, Last paragraph It seems no enough reason choosing the RH to partly explain the inter-annual variation of PM<sub>2.5</sub> acidity. Could the authors give more reasons or explain the function of RH in the formation of PM<sub>2.5</sub> acidity.

Response: It is well known that high RH favors acid formation on particles. In fact, this paragraph intended to use the long term variation of RH as an indicator of behavior of the weather systems rather than explain the influence of RH on aerosol acidity. As discussed in Section 3.4, the inter-annual variation of Asian monsoon systems played an important role in determining the seasonal variation of PM<sub>2.5</sub> acidity in both Beijing and Chongqing.

Comments: Page 16, Third Paragraph The reference of Mu et al., 2008 can not be found in this paper. Fig. 3b and Fig. 3c have no value of coordinate axis, please add them. Response: The reference of Mu et al., 2008 and value of axis missing in Fig. 3 are added in the revised manuscript.

Mu, J. L., Wang, J. J., Li, Z. C.: A study of environment and mesoscale convective systems of continuous heavy rainfall in the South of China in June 2005, *Acta Meteorologica Sinica*, 66(3), 437 – 451, 2008.

Comments: Page 18: Asian desert dust. It is not enough to discuss the detailed dust event's influence on PM<sub>2.5</sub> acidity using the seasonal average results. It need to select several processes of dust event days and non-dust event days to further investigate the aerosol species' proportion or differences among dust and non-dust days. Then hope give the believable and concrete explanation of dust influence on aerosol acidity.

Response: Our study was focused on the inter-seasonal influence of Asian dust with variable strength, rather than the inter-seasonal influence of dust events. Therefore, the

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average results could represent the seasonal trends of aerosol components. Besides, our weekly samples can not separate the dust and non-dust days, as dust events generally only last for a short period.

Comments: Page 18, Third Paragraph. It seems that the Caption of Fig. 6a is inconsistent with the analysis in this paragraph. For example, the paper text shows that 'the Ca<sup>2+</sup> concentration at Beijing was 23.8% to 30.6% higher in the spring of 2006 than of 2005', but when using  $\frac{2005-2006}{2006}$  as list in the figure caption, it will be the opposite description, namely, the Ca<sup>2+</sup> concentration at Beijing was higher in 2005. Please check and give the reasonable explanation. Why do the author use the different calculation expression for Beijing and Chongqing? It causes much difficulty to understand.

Response: The sentence in Fig. 6 should be "which were calculated based on (2006–2005)/2005 and (2005–2006)/2006, respectively" and it has been corrected in the revised manuscript. The different calculation expressions for Beijing and Chongqing are used to highlight the influence on the season of more dust strength relative to the season of less dust strength.

Comments: Page 19, First paragraph: Why the difference of NO<sub>3</sub><sup>-</sup>/SO<sub>4</sub><sup>2-</sup> is so much at TH and MY in Beijing? The explanation of this paper is not enough. It need add further study and explanation, as well as to explain the different aerosol acidity between TH and MY.

Response: The higher ratio of NO<sub>3</sub><sup>-</sup>/SO<sub>4</sub><sup>2-</sup> at TH than MY suggested higher contribution from vehicles in urban area in Beijing, as has been discussed in Section 3.2.1. In the previous response, we also addressed that the comparable concentrations of SO<sub>4</sub><sup>2-</sup> for TH and MY were probably due to the more homogeneous spatial distribution of sulfate in this region than the other aerosol species. The more significant difference between the two springs in TH than in MY was also explained in Section 3.4.2.

Comments: Page16-17: In Fig. 4, authors select air mass trajectory at 500m, but the

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aerosol sample conducted near surface. It should be better when using lower layer trajectory.

Response: The altitude of ~500m has been widely used in trajectory analysis to represent the movements of regional air masses in previous studies and its results generally are similar as those calculated at lower level of altitude. Besides, the mountainous landscape in Chongqing would also limit the precision of modeling results if lower altitude was used.

Comments: Page 20, Wet deposition Why only discuss the wet deposition of  $\text{NH}_4^+$ ? How about the deposition of  $\text{NO}_3^-$  and  $\text{SO}_4^{2-}$ ? This also influences the aerosol acidity. Although the precipitation is much less in northern cities than that in southern cities, it also plays important role in aerosol and its species deposition. How does the wet deposition play in the formation of aerosol acidity in summer when the precipitation occurs frequently.

Response: In fact, we have already discussed the influence of  $\text{SO}_4^{2-}$  deposition in the precipitation at Chongqing in this section, as the ratios of  $\text{NH}_4^+/\text{SO}_4^{2-}$  were compared between  $\text{PM}_{2.5}$  and precipitation.  $\text{NO}_3^-$  was not of our concern due to its low content in both aerosol and rain water. The discussion on wet deposition was focused on Chongqing, as its aerosol was much more frequently influenced by precipitation than Beijing and the dataset of precipitation in Beijing was also too limited to be compared with  $\text{PM}_{2.5}$ . The influences of wet deposition on aerosol acidity in summer at northern cities are clearly important subjects for future studies.

Comments: Please check the caption and denotation in Fig. 7.

Response: We did not find anything needed for improvement in Fig. 7(b).

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