**Response to reviewers' comments to ACP-2012-539 by Nie et al.**

Dear Editor,

We have revised the manuscript in response to the comments/suggestions of referee #2. The itemized replies are listed below. The revised texts are highlighted in blue in the manuscript.

**Anonymous Referee #2**

Received and published: 28 August 2012

General comments:

In this study, the authors tried to describe the modifications of dust storm particles as the particles approached the observation site at Mt. Heng with the data they obtained during the passage of the dust event. Although the manuscript includes many solid data and the discussion sounds reasonable, the data and discussion are not sufficient to support the major conclusion that "the mineral dust …. underwent significant modifications during the transport".

My major concern is the lack of the details of the dust plume arrival and passage and the sample collections. Authors used integrated filter samples to study the evolution of metals and ions. There are anthropogenic sources of mineral particles in China, such as coal burning emissions, road dust, and construction dust. They are the major anthropogenic particulate matters and frequently enhance the production of nitrate and sulfate in polluted air in China besides soot particles. I checked the weather charts and found that, from 25 to 27 April, the Mt. Heng areas were covered by a strong high pressure. The air was quite stable in this period and the weather conditions favored the accumulation of anthropogenic pollutants that were emitted from the surrounding areas (Fig.3 supports this expectation). From the evolution of PM2.5 and PM10 in Fig.2, the dust arrival was likely between the midnight 24 April and the morning 25 April. There are many papers reporting that dust plumes' arrival usually causes a short-time decrease of anthropogenic pollutants in the air in eastern China areas. In this regard, it is likely more reasonable, at least to a large extent, to attribute the

results or evolutions shown in Fig.4, Fig.5, Fig.6, Fig.8 and Table 1 to the gradual accumulation of anthropogenic pollutants (including chemical reaction products on anthropogenic mineral and soot particles) than totally to the heterogeneous reactions on the dust storm particles. The effects of the dust storm particles might be small. In many places of the manuscript, the authors actually cited results on anthropogenic pollutants in published literatures to support their discussions, in particular in the subsection 3.3.

Therefore, evidences are necessary to demonstrate that the observed ions were produced, at least most probably, on natural-dust particles (i.e. the dust storm particles) rather than on anthropogenic mineral or soot particles if the conclusion is correct. My concern might be wrong. But authors do need to provide more details and evidences (such as comparisons with non-dust cases; the high-time-resolution evolution of weather conditions, NOx or SO2 or black carbon as in the upper panel in Fig.2; the start and stop time of the filter sample collection; the accurate time and position of air parcels from the backward trajectory calculation; and etc…) to fix the confidence of the conclusion. Otherwise, the results and conclusion may mislead readers and are difficult to be cited correctly and accurately.

Response: We understand the referee's concern and appreciate the helpful comments/suggestions. After re-examining all the information, we are convinced that the coarse particles on April 24-26 at Mt Heng are dominated by long-distance transport of dust.

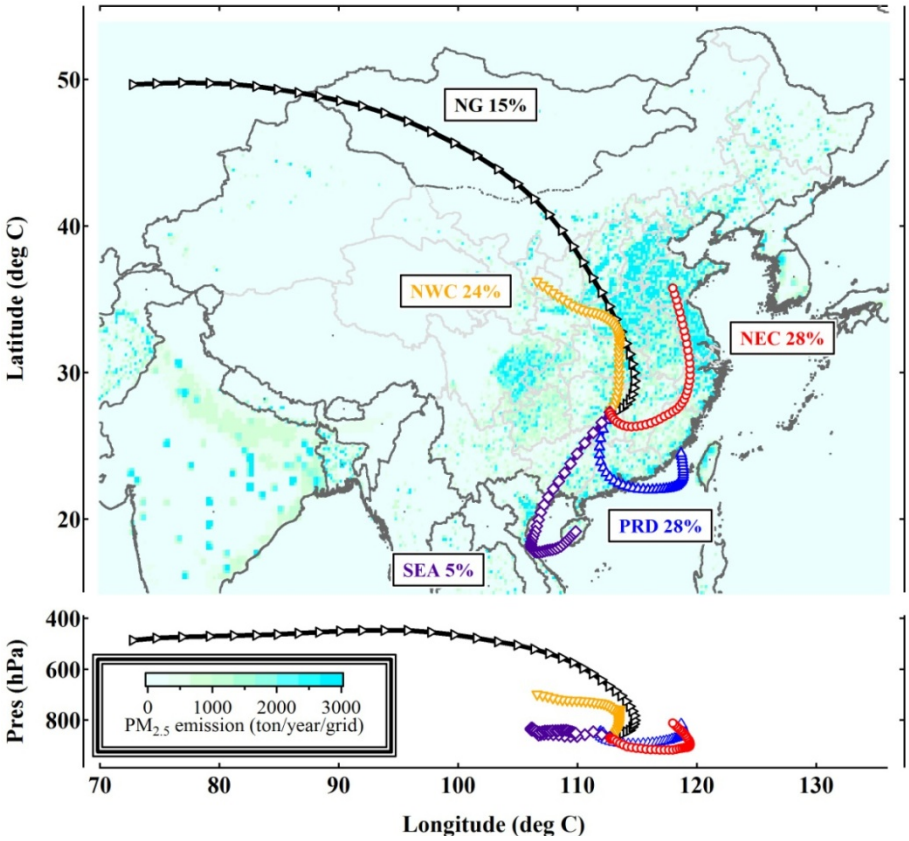
Firstly, the size and chemical composition of aerosol on April 24-26 confirm the arrival of dust storm. On April 24-26, the particle concentrations, especially in the coarse mode, and the chemical tracers of dust particles were much higher than those in the non-dust events, with coarse-mode particles dominating. In the revised manuscript, we have added a paragraph in section 3.1 to compare the aerosol composition and several trace gases for the dust event and non-dust periods.

Secondly, the wind direction and backward trajectories analysis both indicate the long range transported dust from northern China. In the revised manuscript, we have added some meteorological parameters in the upper panel in Fig. 2. Prevailing northerly

winds occurred during the dust storm period; five-day back trajectories during the dust storm show that the air mass belongs to a group named as “North Gobi, NG”, which originated from the Gobi areas (i.e., Siberia and Mongolia) passing over north China at higher altitudes with a high speed, and descended before arriving Mt.Heng (Gao et al., 2012; Zhou et al., 2012, see the figure below).

Thirdly, the April 24-26 dust event was also observed at three other sites at Mt. Hua in the Shaanxi province of central China (Fig. 1a), Mt. Tai in the Shandong province of central-eastern China (Wang et al., 2010) and the Island of Taiwan. MODIS images also confirm this dust storm covering a large geographical region including Mt Heng.

Our analysis are focused on the supermicron range particles ($PM_{>1}$), whereas the diameter of anthropogenic mineral or soot particles are usually smaller than 1 μm. Therefore, we believe the observed dust storm is a natural dust storm, and the observed ions are produced on the natural dust particles.




Transport pattern of Mt. Heng during March to May, 2009 based on five-day back trajectories.

*"Gao, X., Xue, L., Wang, X., Wang, T., Yuan, C., Gao, R., Zhou, Y., Nie, W., Zhang, Q., and Wang, W.: Aerosol ionic components at Mt. Heng in central southern China: Abundances, size distribution, and impacts of long-range transport, Science of The Total Environment, 433, 498-506, 10.1016/j.scitotenv.2012.06.095, 2012."*

*"Zhou, S., Wang, Z., Gao, R., Xue, L., Yuan, C., Wang, T., Gao, X., Wang, X., Nie, W., Xu, Z., Zhang, Q., and Wang, W.: Formation of secondary organic carbon and long-range transport of carbonaceous aerosols at Mount Heng in South China, Atmospheric Environment, 10.1016/j.atmosenv.2012.09.021, "*

*"Wang, G., Li, J., Cheng, C., Hu, S., Xie, M., Gao, S., Zhou, B., Dai, W., Cao, J., and An, Z.: Observation of atmospheric aerosols at Mt. Hua and Mt. Tai in central and east China during spring 2009 – Part 1: EC, OC and inorganic ions, Atmos. Chem. Phys., 11, 4221-4235, 2011."*

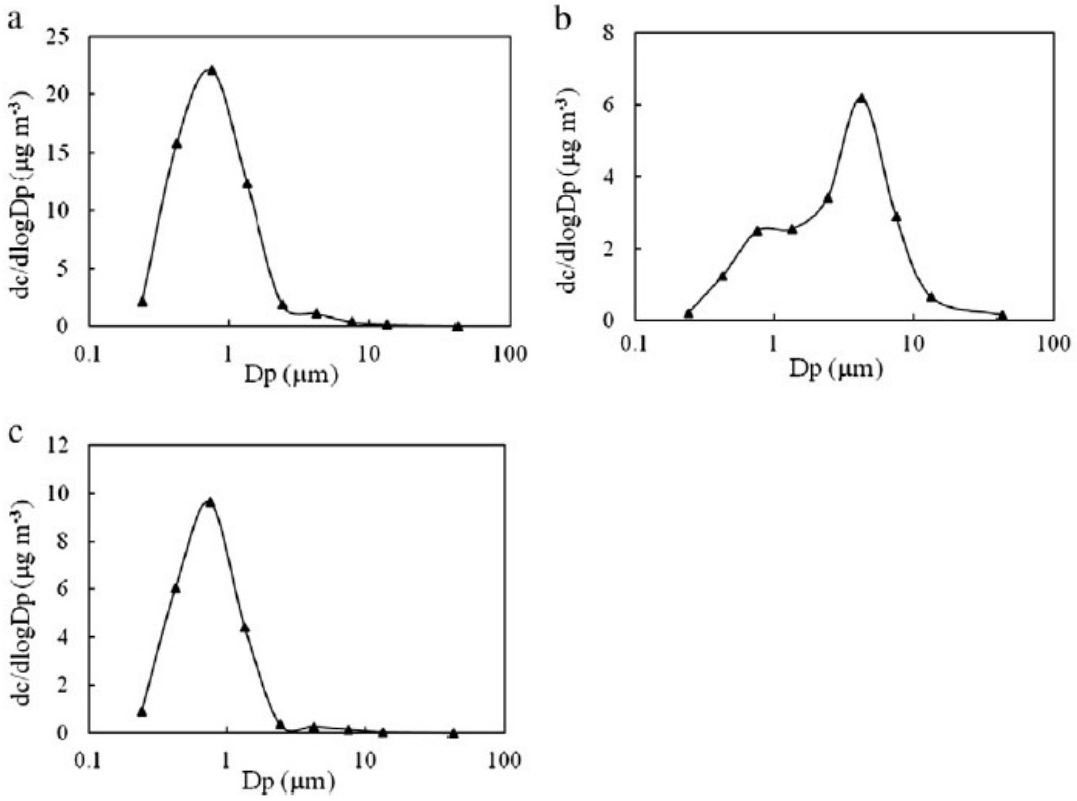
Other comments:

(1) The comparison between Mt. Heng and Mt. Hua needs to be conducted carefully by showing clearly if the comparison was on the same air parcel. This is because pollutants are usually not homogeneously distributed within the territory of a high pressure.

Response: We agree that the dust events observed at Mt. Hua and Mt. Heng may be not exactly in the same air parcel. From the back trajectories analysis, we can clearly see that this dust storm was originated in the Gobi deserts, moved southward, and arrived Mt. Heng. Mt Hua can be viewed a site close to the dust source region, and the aerosol composition there can represent relatively fresh dust, whereas Mt. Heng is a downwind region receiving this dust storm. Therefore, the comparison between the two mountainous sites can provide valuable insights into the dust particles at source and the chemical evolution during long-range transport to a southern region. We have modified some wording to clarify their relationship.

(2) Bimodal and monomodal distributions as shown in Fig. 4 can be frequently detected in polluted air with filter-based samples. They are not exceptionally meaningful in case of dust storm particles to the extent of the current manuscript contents.

Response: we have added some information on the particle size distributions on non-dust days in the revised manuscript, which are significantly different from those during the dust event. For the non-dust events, sulfate and ammonium showed single peaks in the fine particle mode, while nitrate had bimodal peaks in the fine and coarse modes (see the following figures from Gao et al., 2012). During the dust event, the mass of these three major ions moved to the coarse mode (see Fig. 4 in the revised manuscript).




Average size distributions of (a) $SO_4^{2-}$, (b) $NO_3^-$ and (c) $NH_4^+$ at Mt. Heng during the non-dust events (Gao et al., 2012).

(3) Fig.6: I cannot find why PM>1 was used in this figure and when these samples were collected. There is no explanation for it. Are the time periods of the sample collection similar to or different from those shown in Fig.8?

Response: The PM>1 was calculated from the size resolved samples (MOUDI). We have added the information on MOUDI's cutoff sizes in section 2.2 in the revised manuscript. The exact sampling periods have been marked in Fig. 5 in the revised manuscript.

(4) “carbonate-rich Asian dust” may cause confusion. Carbonate is always one of the major components of Asian dust. It is true that some dust particles are carbonate-rich and some are not. But the methods of this study could not separately identify them and there is no such information in the manuscript. The discussion in the subsection 3.2.3 was completely a speculation without any evidences from this study. Fig.7 is not supported by the data in this paper. It is meaningful if the authors, with this model, can quantitatively (not qualitatively) explain the ammonium they speculated on dust storm particles.

Response: We have changed the description of “carbonate-rich” to “carbonate-containing” in the revised manuscript.

For the conceptual model, we do have some observation evidence to support it. In the revised manuscript, we have added a figure (the new Fig. 6 on the ratio of ammunition to PM) and re-arranged other figures in order to better facilitate the discussion on photochemical age and the proposed concept model.. Our evidence to support the 4-stage evolution is shown in the following figures (Fig. 4c and Fig. 6). For the dust sample of Mt. Hua near the dust source region, no accumulated ammonium was detected in the supermicron range (Fig. 4c). For the first three samples of Mt. Heng in southern China, ammonium appeared in the coarse mode, but at moderate level (Fig. 5c and Fig. 6). For the forth sample at Mt. Heng (see the second figure), the abundance of ammonium increased significantly. We have added this information in the revised manuscript.

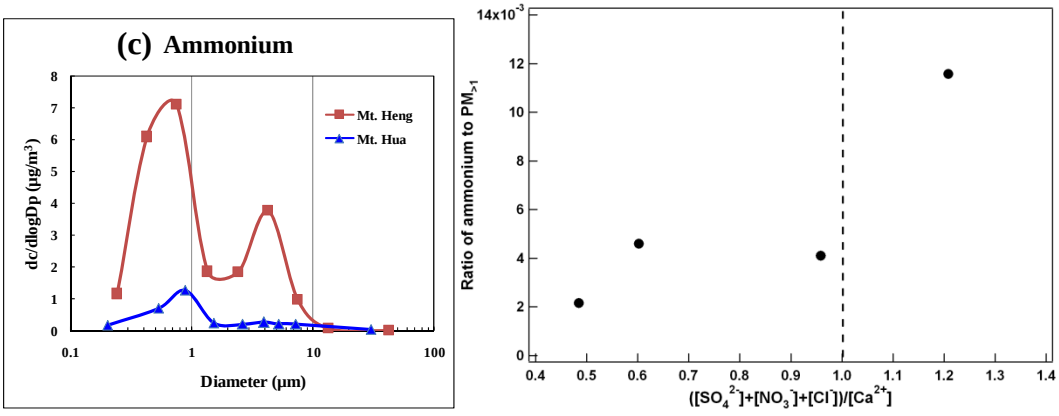



Fig. 4c and Fig. 6.

In the second paragraph of Section 3.2.3, we try to explain why the ammonium abundance increased sharply in the forth sample comparing to the first three samples at Mt. Heng. As shown in Fig. 6, the difference between the first three samples and the forth sample is the aerosol acidity (or the neutralization of carbonate). In the first three samples, the calcium carbonate was not fully neutralized, accumulated ammonium on dust particles was observed at moderate level; in the fourth sample, the calcium carbonate was fully neutralized, and the ammonium abundance increased sharply. These results indicate that the dust particles can have some accumulated ammonium after it has been coated with acidic species (sulfate or nitrate), but the abundance should not be much until all the alkaline mineral substance is fully neutralized. These observational results provide some supports to the subsequent conceptual model.

We have also added some information on the limitation of the 4-stage evolution process: (1) the conceptual model can only apply to the carbonate-containing dust particles; (2) ambient ammonia concentrations will influence the development of the 4 stages.

(5) I am not sure when Sample-I, -II, -III, and –IV were collected. Descriptions for Fig.4 and Fig.8 in the text are not consistent with the figure legends or what the figures show. I consider them as shown in Fig.8 in this review.

Response: we have added more information to describe the sampling of the four MOUDI samples in Section 2.2 in the revised manuscript. The exact sampling periods are provided in Fig. 5 in the revised manuscript.