

Response to Editor's comment:  
Comment

I've read your response to the referee report and revised manuscript, and found satisfied to most of the answers. Only one remaining issue is about the explanation to the coarse mode sulfate (raised by both ref #2 and ref #3). I am not really convinced or do not full understand why you reject the suggestion of other possible sources, such as heterogeneous reactions etc. It would be nice if you could provide some re-evaluation or further explanation on that.

Response: Thanks for the opportunity to clarify the point on contribution of heterogeneous reactions to coarse mode sulfate. The wording in our manuscript was not clear. What we intend to mean is that heterogeneous reactions are unlikely the major cause for coarse sulfate in factor 2, as this factor has abundant presence of  $\text{NH}_4^+$  and sulfate in this factor mainly exists as  $\text{NH}_4\text{HSO}_4$  while nitrate exists as  $\text{NH}_4\text{NO}_3$ . The alkaline compounds (i.e.  $\text{Ca}^{2+}$ ,  $\text{Na}^+$ ,  $\text{K}^+$ ,  $\text{Mg}^{2+}$ ) in dust and the cation  $\text{Na}^+$  tied to sea salt make it difficult for ammonia to partition into dust and sea salt particles (Xu and Penner 2012).

Heterogeneous reactions do contribute to coarse-mode sulfate and these processes are responsible for coarse sulfate in factor 3 (mixture of aged sea salt and dust particles). This latter point was missed and not clearly spelled-out in our manuscript.

We have made the following revisions to clarify the point on heterogeneous reactions. The new text is also marked in blue in the revised manuscript.

- (1) The sentence below has been deleted from the paragraph describing coarse sulfate factor 2 (starting at Line 492)

~~“The possibility of heterogeneous reaction of ammonia, sulfuric acid, and nitric acid on sea salt or dust particles could be ruled out since when exposed to abundant acids, the alkaline sea salt or dust particles tend to react with them to form more stable salts (e.g.  $\text{CaSO}_4$  and  $\text{NaNO}_3$ )”.~~

The text below in blue is added (lines 494-496):

“The alkaline species (i.e.  $\text{Ca}^{2+}$ ,  $\text{Mg}^{2+}$ , and  $\text{Na}^+$ ) in dust and sea salt make it difficult for ammonia to partition into dust and sea salt particles. Considering the abundant presence of  $\text{NH}_4^+$  in this factor, we therefore attribute coarse-mode sulfate apportioned to this factor (termed as  $[\text{SO}_4^{2-}]_{\text{C}_F2}$  hereafter) to coagulation of fine  $\text{NH}_4\text{HSO}_4$  and  $\text{NH}_4\text{NO}_3$  particles with coarse sea salt/ dust particles or re-suspension of dust particles that contain  $\text{NH}_4\text{HSO}_4$  and  $\text{NH}_4\text{NO}_3$ ...”

- (2) The following text is inserted into the paragraph describing factor 3 (starting at Line 515)  
Lines 516-518

“Heterogeneous reactions of sulfuric acid and nitric acid on sea salt or dust particles (e.g., reactions R2 and R4) are mainly responsible for this source of coarse-mode sulfate.”

Reference:

Xu, L. and Penner, J. E.: Global simulations of nitrate and ammonium aerosols and their radiative effects, Atmos. Chem. Phys., 12, 9479-9504, doi:10.5194/acp-12-9479-2012, 2012.