Characteristics and mixing state of amine-containing particles at a rural site in the Pearl River Delta, China

By Cheng et al.

General:

This paper has been significantly improved from the first version. I have a few comments that should be addressed before publication.

Major Comments:

My main comments are:

1. Figure 8 is a bit problematic. Correlations between sulfate and nitrate on amine-containing particles is used to infer how amines are formed. Sulfate and nitrate should be separated out in this figure in order to determine the formation of different aminium salts. Also, the correlations are quite misleading since amine-containing particles were compared against each other instead of sulfate, nitrate, and amine markers on all particles.
2. The conclusions could be significantly strengthened by moving the commentary on lines 382-398 that links sulfate formation in Chinese haze to mixing state and amines to the conclusions.

Specific Comments:

Introduction

1. I suggest adding one few more reference on amines on lines 89-90. Please add [*Facchini et al.*, 2008].
2. Lines 111-113: Please also add [*Gaston et al.*, 2013; *Qin et al.*, 2012; *Zauscher et al.*, 2013].

Methods

1. Was a silica gel drier used during sampling to reduce particle phase water?
2. Lines 191-195: Please also cite [*Gaston et al.*, 2013; *Qin et al.*, 2012].
3. In table 1, include references for each ion peak.
4. Line 220: Please also add to the end of the sentence “and are likely sea salts”. This will help avoid confusion.

Results

1. Line 231: From Figure 1, it looks like the open ocean trajectory has the fewest amines. Perhaps your amines aren’t “marine sources” per say but are derived from coastal emissions.
2. Line 232: Cluster 4 for the winter is very stagnant. I would guess that those stagnant conditions also facilitate the partitioning of amines.
3. Line 263-266: What about the role of temperature?
4. Figure 4 needs to have actual m/z values on the ion peaks. For example, instead of CH_3NH, show $^{30}CH_3NH^+$.
5. Lines 287-291: was the size distribution for amines any different than the size distribution for all particles? If not, then this figure and discussion is not very important.
6. Line 298: I suggest removing “formation processes” in the title of section 3.3. I am not completely convinced that you can completely deduce this information.

7. Lines 299-308: I suggest comparing your seasonal trends of amines and ammonium to Qin et al., 2012, which also contains observations of ammonium nitrate and amines.
8. I find Figure 7 to be very interesting. What is the R^2 between amines and ammonium-rich particles? It looks high in the winter.
9. Lines 333-335: RH has been shown to exert an influence on compounds such as ammonium nitrate. I suggest revising this sentence to reflect that your findings indicate that source seems more important than RH for explaining seasonal trends.
10. Figure 8 just shows a peak area comparison on amine comparing particles. The trends reported in lines 339-341 are misleading because only amine-containing particles were selected so if these particles also had sulfate, the correlation would be high. How do the correlations hold if you compare the ion peak areas for all particles instead?
11. Lines 347-349: The authors should explicitly state that this method was developed by Pratt et al., 2009 for single particle work since the authors use the exact same methodology.
12. Lines 386-398 should be moved to the conclusions section.

Conclusions:

1. I recommend removing the sentence on lines 417-419. Correlation does not equal causation.
2. This section has conclusions but no implications. I recommend removing lines 429-436 and replacing it with lines 386-398.

Technical Comments:

1. Line 49: change “count” to “counts”
2. Line 214: change “ion of m/z” to “an ion peak at m/z”
3. Line 250: change “similar variation pattern” to “a similar pattern”
4. Line 255 and 412: remove “special”
5. Line 258: change “count” to “counts”
6. Line 299: change “aging” to “mixing”
7. Line 316: change “During entire” to “During the entire”
8. Line 319: change “aging” to “mixing”
9. Line 379: remove “Besides”
10. Line 386: change “hence the mixing state…” to “As pointed out in Pratt et al., 2009 and in this work, the mixing state…”

References:

Facchini, M. C., S. Decesari, M. Rinaldi, C. Carbone, E. Finessi, M. Mircea, S. Fuzzi, F. Moretti, E. Tagliavini, D. Ceburnis, and C. D. O'Dowd (2008), Important source of marine secondary organic aerosol from biogenic amines, *Environmental Science & Technology*, *42*(24), 9116-9121.

Gaston, C. J., P. K. Quinn, T. S. Bates, J. B. Gilman, D. M. Bon, W. C. Kuster, and K. A. Prather (2013), The impact of shipping, agricultural, and urban emissions on single particle chemistry observed aboard the R/V Atlantis during CalNex, *Journal of Geophysical Research-Atmospheres*, *118*, doi:10.1002/jgrd.50427.

Qin, X., K. A. Pratt, L. G. Shields, S. M. Toner, and K. A. Prather (2012), Seasonal comparisons of single-particle chemical mixing state in Riverside, CA, *Atmospheric Environment*, *59*, 587-596.

Zauscher, M. D., Y. Wang, M. J. K. Moore, C. J. Gaston, and K. A. Prather (2013), Air quality impact and physicochemical aging of biomass burning aerosols during the 2007 San Diego wildfires, *Environmental Science & Technology*, DOI: 10.1021/es4004137.