

Interactive comment on “Contribution of hydroxymethanesulfonate (HMS) to severe winter haze in the North China Plain” by Tao Ma et al.

This paper discusses field measurements of aerosol composition of HMS, one of important S(IV) and organic species, which obtains more attention recently. They did very rigorous work to identify and quantify HMS from the atmospheric particles. The main finding is that HMS, which was usually observed in cloud/fog events, is ubiquitous in Beijing winter. The HMS concentration, its mole ratio to sulfate and contribution to organic matter increased with the deterioration of winter haze. And discussed the reasons for such a high abundance of HMS in winter Beijing. While the results are interesting and novel, and important to improve the prediction of OM and sulfate.

The study relies on a combination of single particle mass spectrometry and IC. It mainly focus on bulk measurements. The analysis and interpretation of the main results is valid and almost satisfied most of my curious of HMS in winter Beijing. However I still have more questions about this study and look forward to some further investigations. A minor revision is required for finalizing the manuscript.

Major comments:

1. The author did very rigorous work to isolate and quantify HMS from other easily mixed species. That is very good job. But I am curious, the filter extraction found just using water as extraction may overestimate the sulfate concentration due to the transformation of HMS to sulfate over time. I am wondering how about the MARGA data? Looks like the author use MARGA sulfate to estimate the pH of particles. Will the data be influenced by the overestimation if MARGA has the same problem?
2. The author can generally compare the data observed in this study with previous studies. For the first impression, I was surprised by such a high abundance of HMS measured by ATOF. Previous studies usually found HMS only during cloud/fog event and only account for a small portion to the particle, usually with only several to hundreds' particles of HMS by number, or no more than 4% to the total particle number during the events. However the characteristics of HMS is kind of very typical HMS particles. But I do think it is necessary to compare the spectrum of HMS observed in Beijing with other studies. As the HMS particles observed in this study is very mixed with other species (e.g. nitrate, elemental carbons...) no matter in the average spectrum or the digital spectrum. Is there any other data can be compared?
3. Moderate pH is one of the critical factors to promote HMS formation. Have you compared the difference of pH between clean, high pollution and sever haze period? Is there any finding? Clean days with high solar radiation and less RH usually have very low particle water which decreases the aerosol pH. Compared with 2015, does the pH also influence the high contribution and concentration of HMS in 2016?
4. The author noted for some periods day and night samples were sampled separately. Is there any difference between these D and N samples?
5. Since HMS is so easy to transform from S(IV) to sulfate, what's the fate of HMS in the urban atmosphere?

Minor comments:

1. Section 2, total numbers of filter samples should be noted in the experiment section. In the discussion section, clean polluted and serve haze day should be also noted with the sample number. The reader may be confused when find some figures show the winter data, some figures show the filter information for four seasons.
2. The figure caption of Figure 2, 95th, 75th, 50th ...
3. Line 219, atmospheric sulfur distribution shifts toward particle/liquid phase?
4. Figure 3. Evolution of sulfur distribution with the increase of RH in the winter of a) 2015 and b) 2016. (Solid circle stands for the molar ratio of HMS to sulfate, with colored by the HMS concentrations and sized by the PM2.5 concentrations; The gray crosses represent the particulate sulfur molar percentage. Particulate sulfur molar percentage....)

Or

Figure 3. Relationship between the molar ratio of HMS to sulfate, particulate sulfur molar percentage, and RH in the winter of a) 2015 and b) 2016.(.....)