

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

Anonymous Referee #1

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General comments:

This is a very fine study, presenting crucial insights into the occurrence of severe haze episodes in the North China Plain during winter. Convincing evidence is provided that hydroxymethanesulfonate (HMS) is a key player in this process. Use is made of aerosol time-of-flight mass spectrometry for the real-time detection of HMS in ambient aerosols and a novel and accurate analytical method has been developed for the measurement of HMS in fine particulate matter (PM_{2.5}). This method has the advantage that it does not suffer from overestimation of sulfate. A comprehensive dataset has been obtained and interpreted in great detail. The results obtained in this study should be very useful for pollution abatement measures in Beijing. The scientific content of this manuscript is in a very good shape but the English usage should be improved. A list of suggested

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technical corrections is provided below.

Technical corrections:

Line 20 (and many places elsewhere, e.g. line 23, 31, 32,): winter haze occurs and threatens ...[Note: “haze” should be in singular].

Line 24: single particle mass spectrometry and an optimized ion chromatography method,

Line 25: in Beijing during winter.

Line 29: using traditional ion chromatography.

Line 41: in the NCP during winter.

Line 46: in the NCP during winter, which increase

Line 53: overestimation of the observed

Line 55: evidence for the presence of HMS

Line 59: haze, unambiguous identification and accurate quantification

Line 65: mass spectrometry and an optimized ion

Line 66: in Beijing during winter and

Lines 67-73: I suggest to use the present tense here: “We demonstrate water is an important pathway that contributes to, not only accounting for a winter haze favor heterogeneous HMS formation. to 2016 indicate that Finally, we discuss the”

Line 76: located at Tsinghua

Line 85: the ATOFMS instrument

Line 90: A bipolar reflectron mass analyzer. [Note: the abbreviation “MS” stands

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for “mass spectrometry” and should not be used to denote a mass analyzer or the instrument. See the IUPAC guidelines for terms relating to mass spectrometry: K. K. Murray, R. K. Boyd, M. N. Eberlin, G. J. Langley, L. Li, Y. Naito. Definitions of terms relating to mass spectrometry. IUPAC Recommendations 2013. Pure Appl. Chem., 85, 1515-1609, 2013].

Line 93: of the ATOFMS instrument

Line 95 (and many places elsewhere, e.g., lines 163, 165,): It is more correct to write “..... the peak at m/z 111 in the negative ion mode as”. [Note: the m/z value is not negative].

Line 101: Here, we

Line 102: in the day- and nighttime.

Line 103: baked in a Muffle, put in the cassettes, and packed

Line 106:, ultrasonic agitation for

Line 108: overestimation, whereas extraction with

Line 113: under conventional conditions

Line 114: Here, we used

Line 117: HMS dissociates into

Line 118:, and the HMS concentration

Line 119: Thus, the ion chromatography

Line 151: in previous work

Line 162: In aerosols from Beijing during winter. We found that

Line 165: According to the natural isotopic distribution, the contribution of KCl_2^- was found to be insignificant.

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Line 170:, the peak at m/z 111 unlikely corresponds to methylsulfate

Line 172: Therefore, the peak at m/z 111 in ambient particles can safely be assigned to

Line 173: in Beijing during winter contain

Line 179: I suggest to use the past tense consistently in this section. Thus: “We found that the HMS”

Line 180: The HMS concentration , and was consistent with the variation of the relative humidity (RH).

Line 178:, we determined the HMS concentration

Line 180: also showed a similar HMS evolution as that of

Line 183: , the HMS concentration

Line 184:, and the HCHO concentration increase, while the O₃ concentration

Line 199: during severe winter haze could be overestimated

Line 199: S(IV) oxidation reactions compete with

Line 213:, whereas it decreases the

Line 215: of the reaction rate constant

Line 220: . . . The HMS concentration

Line 222: With the increase of RH, the atmospheric sulfur distribution shifts toward the particle phase and more particulate sulfur exists in

Line 229: The calculations based

Line 233: conditions, HMS formation is favored, whereas decomposition of

Line 233: under high HCHO concentrations, low oxidant levels,

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Line 239: , the atmospheric oxidation with the weak photochemical activity.

Line 248 (Niu et al., 2016); in this study, we further

Line 253: from primary emissions and

Line 256: formation, should be considered in future pollutant control

Line 261: In Beijing during winter.

Figure 1, legend: The peak at m/z 111 in the negative ion mode is The peaks at m/z 80 [Note: see comment made above with regard to the omission of the minus sign].

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