

Interactive comment on “Spatial and seasonal variability of PM_{2.5} acidity at two Chinese megacities: insights into the formation of secondary inorganic aerosols” by K. He et al.

K. He et al.

zhaog@mails.tsinghua.edu.cn

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We sincerely thank the reviewer for the valuable comments and suggestions to improve the manuscript.

Comments: The authors attempted to explain the enhancement of aerosol acidity during Asian dust in Beijing because of possible heterogeneous reactions. The enhancement of aerosol acidity during Asian dust is definitely a new result to research community and is contradictory to the common knowledge. However, it is well known that the response of ionic species in high concentration could be non-linear when IC is used to detect these ions. The non-linear response could be even stronger for NH₄⁺ than

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other ions. This reviewer suggested that the authors should detail how to solve the non-linear response in chemical analysis. Response: The non-linear response of ions in the solution of high concentration could be strong in IC analysis, but this is not for our case. Firstly, aerosols collected on the filter were of small amount as the sampling flow rate was very low (0.4L/min), filtering only 4m³ air during a whole week. Secondly, the ionic species were extracted and diluted in 10mL of ultra purity water, and the concentrations of major ions were low. For instance, the average concentrations of NH₄⁺, SO₄²⁻ and NO₃⁻ in the solutions for all samples in Chongqing were (3.07±1.63)μg/mL, (1.64±1.39)μg/mL and (9.02±4.28)μg/mL, which are of the normal range for IC analysis.

Comments: In Discussion Section, this reviewer has problem to follow the logic and strongly encourages the authors rearranging most of discussion. Response: We have improved the discussion according to the comments of reviewers in the revised manuscript.

Comments: 25563-Line 13, References are needed here. Response: We have introduced the method of equivalent ratio of cations/anions to indicate the neutralizing level of aerosol with citations in the introduction section of the original manuscript. For the selection of ions in the ratio function, however, there is no universal standard and we chose the four components with the reason discussed in Section 3.1.

Comments: 25571-Line 2, the sentence does not sound scientific. Rewrite by including the average or the mean value. Response: We have added the average values for [H⁺]_{ins} and [H₂O] according to the comments in the revised manuscript.

Comments: 25571-Line 11, the sentence is problematic, correct it. Response: The sentence is corrected: resulting in a significant increase of in situ pH and thus much lower acidity of PM_{2.5}.

Comments: 25572-Line 5, it will be easy for the reader to follow the discussion if the authors can summarize all factors before detailed discussion. Response: A general

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introduction was already given for each factor in Section 3.4 and the major findings were summarized and extended in Section 4 of the original manuscript.

Comments: 25579-Line 4, why was Rc/a at 0.9 used a threshold to judge the extent of neutralization? Response: RC/A is an approximated indicator of aerosol acidity in this study. With a good fit of linear regression, a ratio of 0.9 is the best estimate that could divide the samples into a group of more acidic aerosols and a group of less acidic aerosols. The above discussion has been given in the original manuscript.

Comments: 25582-Line 1-3 The contribution from coal combustion in southwestern China is not a new finding here, so it is better for the author to give a value here to describe how significant it contributed. Response: It is well known that the coal combustion significantly contributes to the high aerosol concentration in this region, but few studies have quantitatively estimated its contribution, especially for PM_{2.5}. This is clearly the subject of future studies related to this work.

Comments: 25582-Line 22, why “Chongqing’s lower levels of NO₃- suggest that vehicle sources play a more important role in Beijing.”? Response: The sentence should be “the higher levels of NO₃- in Beijing suggest that its vehicle sources play a more important role”. It has been corrected in the revised manuscript.

Comments: Table.2 What does “0.95/0.80” mean? The uncertainties are missing and should be included. Response: “0.95/0.80” means the RC/A for the spring of 2005 (0.95) and 2006 (0.80), and it has been made clearer in Table 2 in the revised manuscript. The uncertainties were not included here, as the space is too limited to cover with so many sites and seasons.

Comments: Figure.3b & 3c are not readable, and Chongqing and Beijing should be labeled out. Response: Labels for Beijing and Chongqing are added in the two figures in the revised manuscript.

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