### Responses to Anonymous Referee #1

Thank you for your positive review of our manuscript. We sincerely appreciate the efforts you have put in the review process, and we improve this work based on your comments and suggestions. Below we will respond to your comments one by one. Your comments are in bold italics, and my responses are in plain text. All the changes have been included in the newest version of our manuscript.

## Major comments:

1. One major comment arises from the lacking descriptions about the measurement techniques, H-TDMA. I could not get any information about the experimental protocols and technical details about the study methods, such as the configuration of HTDMA used in this study, how aerosols were generated and dried, how aerosols get charged (soft X-ray ionization or corona discharge), the number concentration of aerosols, etc. Elaborating these points would put this study in better context.

Response: Thank you for your comment. Based on your suggestion and concern, more information on this part has been added in our new manuscript in section 4.

2. Page 1, line 21-26: This paragraph needs improvements. The authors tried to describe the significance of aerosol's hygroscopicity to state their motivations to study the effects of multi-charge on aerosols. However, the current version was too short and brief to state the environmental, climate as well as the health effects of atmospheric aerosols. Furthermore, citing more classical and recent references.

Response: The introduction has been updated to include more information about the climatic and environmental effects of atmospheric aerosols, especially specifying the role of hygroscopic properties. The following is cited from the new introduction:

'Atmospheric particles can scatter solar radiation and absorb longwave radiation, imposing direct effects on the Earth's radiation balance (Haywood and Boucher, 2000;Bond et al., 2013). They can also indirectly affect the climate through acting as cloud nuclei and modify the cloud optical properties and life cycle (ALBRECHT, 1989;Twomey, 1974;CHARLSON et al., 1992). Both these two effects are closely related to aerosol particle's hygroscopicity, which describes the particle's ability to absorb water at sub or supersaturated conditions (e.g. McFiggans et al., 2006). Aerosol hygroscopicity also plays a vital role in environmental aspects. It has been reported to be an important factor regulating environmental visibility because it can greatly enhance the particle's light scattering efficiency and degrade visibility under relatively high relative humidity (Chen et al., 2012;Xu et al., 2020). It can increase aerosol particle's

liquid water content, affect the multiphase chemistry and local photochemistry, and facilitates particle formation and aging processes (Wu et al., 2018; Herrmann et al., 2015; Ervens et al., 2011). For human health, aerosol hygroscopicity directly determines the particle's size, thus modifying the deposition pattern of inhaled particles in the human respiratory tract (Heyder et al., 1986; Löndahl et al., 2007). In general, aerosol particle's hygroscopicity is one of the most important properties when quantifying the particle's climatic and environmental effects. It's also useful to characterize the particle's detailed chemical information. Therefore, it's necessary to offer a correct and detailed measurement of aerosol hygroscopicity.'

3. Section 3.2: For ambient aerosols, there are different mixing states, e.g., internal, external and core-shell structures, whether this factor has an impact on the multi-charge correction results and has been considered in the algorithm?

Response: The mixing state doesn't impact our multi-charge correction results because the charge distribution and DMA sizing process is only related to the particle size (Wiedensohler et al., 1986), not affected by the mixing state. In the algorithm, only the measured mean hygroscopicity is used in the correction.

#### Minor comments:

- Authors should fix the typo and format mistakes of references through the whole manuscript, especially in the References section.
  Response: Thank you for your comment. We double checked our manuscript and corrected the typos and format mistakes.
- 2. Page 1, line 28: I notice that aerosol hygroscopicity measurement techniques have been reviewed in a recent study (Tang et al., 2019, ACP), please cite it. Response: This citation has been added to our new manuscript.
- 3. Page 1, line 29, 30: Swietlicki et al., 2017 should be Swietlicki et al., 2008. Response: We updated this citation in the new manuscript.
- 4. Page 4, line 100: The results are summarized in Fig. 3b and Table 1. Response: Thank you for your comment. It has been added in the text.
- 5. Page 6, line 171: Please give some information about the sampling site. Response: We added some site descriptions in the section 4.

# 6. Page 7, line 195: How large? I suggest that authors provide more discussions based on their field measurement results.

Response: We have another academic paper focusing on the measurement results (Shen et al., 2020). We added this citation in the manuscript for reference.

#### Reference

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