

Referee #2

This work reports characterization of a parallel plate DMA (P5). The resolving power and transmission efficiency of the system are measured at different instrument operating conditions and the reasons behind their variations are discussed. Afterwards the DMA is used to characterize sulfuric acid clusters, demonstrating its potential application to atmospheric clusters. This work falls into the scope of AMT and it may be published after major revisions.

Major comments:

1. Line 17: Can the sizing range reach 0? No DMA can size infinitely small particles (e.g., electrons) due to diffusion. Even for ions I believe there is some limit if the size of the ion gets very small.

We thank the reviewer for the comment. We agree with the reviewer that the lower sizing limit of DMA sizing can never reach 0. We have replaced the sizing range to “sub-3.9nm” in the revised manuscript.

2. Line 23: ‘when the V_{DMA} was above 3554.3V’. It is more appropriate to report a flowrate here (and in other similar sentences in the manuscript) since the authors have argued it is a flow field effect that affects the system resolution.

We thank the reviewer for the comment. The sheath gas flowrate/velocity is one of the key parameters for DMA P5 to achieve high sizing resolution. However, the flowrate of DMA P5 is too high for precise measurement. V_{DMA} is constantly monitored and logged, and is tightly connected with sheath gas flowrate and the sizing resolution (R). As can be derived from Eq. (1) $Z = \frac{U \cdot h^2}{L \cdot V_{DMA}}$, the ratio of V_{DMA}/U (sheath flow velocity in the separation region) is a constant value for aerosol with fixed ion mobility. As can be derived from Eq. (4), the R variation is dependent on the change of $\sqrt{V_{DMA}}$. Consequently, we hope to report V_{DMA} in the manuscript. Estimation of sheath flow rate in the symmetry plane going through the center of the inlet slit with different V_{blower} is shown in Fig. S2.

3. Line 166: What is the reason that higher Q_{in} leads to higher signal strength? Are more ions carried to the DMA inlet by the higher flowrate.

We thank the reviewer for the comment. We think the main reason that higher Q_{in} leads to the higher signal strength is the decreased diffusion loss. The mobility diameter of THA^+ (ions used for

characterizing the performance of DMA P5) is 1.47 nm. The diffusive losses are not negligible for these small ions. Higher Q_{in} can decrease the retention time of ions in the nano chamber, and increase the number of ions reaching the DMA inlet.

4. In Figure 2b, I suppose there should be two lines of the counter-flow mode curve corresponding to $Q_{out} = 1L/min$ and $Q_{out} = 2L/min$.

We thank the reviewer for the comment. We have added the curve of counter-flow mode under $Q_{out} = 1L/min$ in Fig. 2b in the revised manuscript.

5. The phrase ‘signal intensity’ is bit ambiguous in the manuscript. In line 162, it seems to refer to the ‘number concentration of the sizing aerosol’. In Line 170, it refers to the total current measured by the electrometer. Please make it clear what signal intensity means exactly throughout the manuscript.

We thank the reviewer for the comment. The output signal of the electrometer is in the unit of Volt (V) and the signal intensity range is 0-2V. The amplification value of the electrometer is 10^{11} V/A for Lynx E11 and 10^{12} V/A for Lynx E12. With the amplification value the output signal can be converted to the actual current intensity. The current intensity can be further converted to number concentration with known flow rate and net charge of the measured ions. We have added the explanation of how the raw output data (in V) convert to ion current (pA) in section 2. The revised part is shown as following “.....*The Faraday cage electrometers (Lynx E11&E12, SEADM, Valladolid, Spain, Fernandez de la Mora et al., 2017) were used as particle counter. The output signal range was 0-2V, with an amplification of 10^{11} V/A and 10^{12} V/A, respectively.*”. Moreover, we have changed the total current to the electrometer output unit (in V) in the revised manuscript (Fig.3), unifying the signal intensity as the direct output value to the electrometer throughout the manuscript.

6. Are the lines in Fig. 4 measured/calculated/taken from literature?

We thank the reviewer for the comment. The dashed lines in Fig 4 are measured with our P5 and HalfMini DMA in our laboratory, while other lines for the commercial DMAs are taken from the literature. In the revised manuscript, we have updated of the commercial DMAs and added explanation of where we cited these values. The added contents are shown as following “.....*The sizing resolution of THA^+ monomer by DMA P5 and Half Mini DMA (Fernandez de la Mora and Kozłowski, 2013), measured in our lab, were compared with the reported results of different types*

of commercial DMAs (Jiang et al., 2011, Stolzenburg et al., 2018). The DMA P5 was operated under counter flow mode at the sheath flow rate of about 1500 L/min (corresponding to the Vblower of 8.5 V). The Half Mini DMA was operated at the aerosol-to-sheath flow ratio of 10/300 L/min. The reported resolution was measured under the aerosol-to-sheath flow ratio of 0.6/6 L/min for the Caltech nanoRDMA, of 6/61.4 L/min for the Vienna DMA, of 2/21.9 L/min for the Grimm nanoDMA, of 2.0/20 L/min for TSI 3085, of 2.5/25 L/min for TSI 3086 and of 1.5/15 L/min for the Caltech RDMA. The aerosol-to-sheath flow ratio for all reported cylindrical DMAs (except HalfMini DMA) is approximately 10, which is the typical flow configuration for particle sizing in both lab and field measurements. ...”.

7. Section 3.2: The P5 was operated at fixed voltages corresponding to the THA⁺ monomer peak?

We thank the reviewer for the comment. When applying the TDMA system for ion transmission measurement, the upstream Half Mini DMA was operated at fixed voltage corresponding to the THA⁺ monomer peak. Downstream to the Half Mini DMA, the monodispersed THA⁺ monomer passed through a flow splitter, reaching DMA P5 and the first electrometer. The DMA P5 was operated under scan mode and was connected to the second electrometer, to obtain the full mobility spectrum of THA⁺ monomer. We have added the description of DMA P5 operation when characterizing the transmission efficiency in the revised manuscript. As well as detailed discussion in Section 3.2. The description of the operation status of DMA P5 is shown as following “... ***During the experiments, the voltage for Half Mini DMA was fixed and the voltage for the DMA P5 was scanned continuously. ...***”.

8. Fig 5a: It is interesting to know if there is an upper limit for the positive relation between ion transmission and Q_{out}.

We thank the reviewer for the comment. We believe that there is an upper limit for the positive relation between ion transmission and Q_{out}. Higher Q_{out} can not only compensate the electrical velocity generated from the electric field between outlet electrode with a high negative voltage and the grounded electrometer, but also decrease the diffusive losses from the aerosol beam splitter to the inlet slit at the inlet electrode of DMA P5. The reason we do not try higher Q_{out} is that 3 L/min is a quite high value with respect to the geometry of exit slit (1.0mm in diameter) at the exit electrode and the inlet slit (0.6mm width, 7mm length). Since the original outlet slit of the parallel plate DMA was designed to be coupled to the vacuum system of a mass spectrometer. Based on our current

experimental condition, it is hard to obtain Q_{out} large enough to find the turning point, after which the relation of ion transmission and Q_{out} reaches the plateau. It should be noted that under conventional DMA P5 configuration (both outlet electrode and detectors are grounded), the ion loss due to the electrical dragging force is negligible. Our results represented the lower limit of the DMA P5 ion transmission efficiency operated under conventional configuration. This lower limit value (54.3%) is 4.5-17.5 times higher than other commercial cylindrical DMAs.

9. Fig 5a: Another interesting comparison would be comparing the transmission of ions with different sizes at the same flowrate (using ions presented in Fig 6). It would be interesting to know if a single transmission can be applied to different ions at a given flow configuration.

We thank the reviewer for the comment. During the experiments, the voltage for Half Mini DMA was fixed and the voltage for the DMA P5 was scanned continuously. The transmission efficiency reported in this study is the maximum ratio of the aerosol concentrations recorded by the downstream and upstream electrometers. To the best of our knowledge, there was only one paper, reporting the transmission of planar DMA P4 (former version of P5). The reported value was about 50%. Our results indicated that the lower limit transmission of DMA P5 was ~5% higher than its former version. It should also be noted that the characterization of transmission of DMA P4 and other cylindrical DMAs used THA^+ as standard ions, due to the intensively studied ion mobility and the capability of generating monodispersed THA^+ monomer. The reason we reported only THA^+ is to compare the performance of DMA P5 with other DMAs. The exceptional transmission indicates that DMA P5 deserved to be further exploited for atmospheric cluster studies by coupling with MS. The combination of DMA P5 with API-TOF-MS shows that it is already a useful tool in the laboratory studies of atmospherically relevant clusters.

We agree with the reviewer that it is interesting to conduct transmission characterization for different tetra alkyl ammonium halides. We think the transmission of different ions at a given flow configuration is different, because of the different effect of diffusion broadening for ions with different mobility. Clarifying the transmission of different ions in DMA P5 needs not only further experimental studies, but also theoretical studies of the transfer function of planar DMA, which, to our best knowledge, have not been reported before. Consequently, the transmission of different tetra alkyl ammonium halides was not studied in this paper.

10. Atmospheric clusters -> atmospherically relevant clusters. For the DMA-electrometer or DMA-

MS system, one challenge to detect the atmospheric clusters is their low concentration. It has not been shown that atmospheric clusters can actually be measured by the parallel plate DMAs in this manuscript.

We thank the reviewer for the comment. Our measured clusters were not sampled from the real atmosphere, but generated by electrospray. These clusters have the same (or similar) element composition and physicochemical properties with the atmospheric clusters. We agree with the reviewer that the current title cannot precisely reflect the content of our experiment. We have changed our title to "*Characterization of the planar differential mobility analyzer (DMA P5) : resolving power, transmission efficiency and its application to atmospheric relevant cluster measurements*" in the revised manuscript. Though the detect limit of our system need to be further evaluate for ambient measurement, it, with its current form, can be a good tool for studying the physicochemical properties of atmospherically relevant clusters in the lab.

Technical corrections:

Line 58: parallel plate

We thank the reviewer for the comment. We have changed the "parallel electrodes" to "parallel plates" in the revised manuscript.

Line 94: springer?

We thank the reviewer for pointing out this spelling mistake. We have corrected the "springer" to "syringe" in the revised manuscript.

Eq. (4): what is $\Delta L_{0.5}$?

We thank the reviewer for the comment. The DMA sizing resolution is defined as the mean ion mobility divided by the full mobility width at half-maximum (fwhm). In $\frac{\Delta L_{0.5}}{L}$, $\Delta L_{0.5}$ represents fwhm, L represents the mean mobility. To avoiding misunderstanding, we have modified Eq. (4) in the revised manuscript, following the expression of Eq. (3).