

Referee #1

This manuscript presents the characterization of a high-resolution planar DMA for cluster classification. I recommend it to be published in Atmospheric Measurement Techniques considering its topic, as transmission and resolution of a DMA are important to the quantification of the measured clusters and nanoparticles. However, I am confused by the presentation in several aspects and recommend a major revision to the manuscript.

Major comments:

1. A major concern is how the audience digest the results and use them in their future studies. The authors need to clarify whether the transmission and resolution are specific to their experimental setup or whether other P5 DMAs are expected to share similar values. Detailed comments are given below:

a. A home-build recirculation system is mentioned several times, making the manuscript very confusing. A home-build system may indicate that the results of this study do not apply to other P5 DMA systems due to the difference in the recirculation system. More discussions are needed when reporting the values from this system: do the authors expect similar transmission and resolution in other P5 systems as a function of different working conditions?

We thank the reviewer for this comment. The original intension of the mention of the home-build recirculation system is that all components can be purchased from the local market. Our reported transmission and resolution are expected to be reproduced under suction mode or counter flow mode of DMA P5 with any recirculation system providing temperature-constant, aerosol-free, stable sheath flow. We agree with the reviewer that emphasizing home-build recirculation system can lead to the misunderstanding that the reported results may not be able to apply to other P5 DMA systems due to the difference in the recirculation system, which is not our original intention. In the revised manuscript, we have removed the descriptions of “*the home-build*”, emphasizing the universal characteristics of the recirculation system that should be applied within DMA P5 system in Section 2. The revised sentences are shown as following “*The recirculation circuit needed for DMA P5 system should be able to provide particle free sheath flow with stable velocity and temperature. The recirculation circuit deployed in this study consists of air blower (Ref 497.3.265-361, Domel), water cooler coupled with constant temperature water bath (DCW-2008, SCIENTZ), particle*

filter adapted for high flow velocity, NW40 and NW 50 corrugated stainless-steel tubes and connectors. The particle filter consists a planar commercial HEPA filter (Ref 34230010, Megalem MD143P3, Camfil Farr) and two stainless assembly. The top side of the assembly is a NW40 connector, while the bottom side fits the geometry of the planar HEPA filter. The HEPA filter is sandwiched between the two bottom surfaces of the assembly, sealed with O-ring and screws. All the components are purchased from the local market. Alternative products with similar performance should not affect the operation of the whole system. ”.

b. More figures and tables on the performance of the P5 DMA under different working conditions will be appreciated. The aim is to help the audience to estimate the resolution and transmission of the P5 DMA in their studies without repeating the same calibration experiment. These figures and tables can go to the SI.

We thank the reviewer for this comment. We have added more figures about the obtained mobility spectrum of standard ions under different working condition and the experimental results of transmission characterization into the SI.

c. Transmission vs penetration. The use of the transmission is rather confusing. I spent quite some time checking section 3.2 and found the authors made no mistakes. However, I am afraid that some readers may be misled as penetration is 1 - particle loss, while transmission is also affected by the peak shape. Their relationship is

transmission = penetration * peakHeight of transfer Function without Loss

For an ideal DMA for sub-micron particles at balanced flow rates, peakHeight of transfer Function without tLoss = 1. However, for clusters and nanoparticles, this value is less than 1.0 as diffusion, turbulence, and other non-idealness will decrease the peak height even though there is no particle loss. Using the transmission may be straightforward when converting the cluster signal to concentration in DMA-MS measurement with a known signal-component cluster sample, yet penetration is used when inverting the signal to cluster/aerosol size distribution. More importantly, different P5 systems may share the same penetration even though the resolution is different due to different levels of turbulence, whereas transmission is expected to vary with the system. I recommend the authors follow the review by Stozenburg (2018) when reporting the DMA parameters, as least reporting penetration together with transmission.

Related to this, Fig. 4 is a bit confusing as none of these DMA peaks reach exactly 1.0 due to diffusional broadening, yet it is probably ok since Fig. 4 is on resolution.

We thank the reviewer for this comment. During the transmission experiments, the voltage for Half Mini DMA was fixed and the voltage for the DMA P5 was scanned continuously. The transmission reported is the maximum transmission efficiency calculated based on the ratio of the peak aerosol concentration recorded by the downstream and mean aerosol concentration recorded by the upstream electrometers. We agree with the reviewer that penetration efficiency and transfer function is the necessary to invert the signal to cluster/aerosol size distribution. However, to our best knowledge, there is no reported transfer function for planar DMA. The review by Stozenburg (2018) only provide the recommended transfer function of diffusing particles for cylindrical DMAs and radial DMAs. We are now working on the derivation of the transfer function for planar DMAs. Considering the time consumption, we want to report only the maximum transmission efficiency in this paper, which, to the best of our knowledge has only been reported once. The reported the transmission of planar DMA P4 (former version of P5) with obtained with similar TDMA system to this work. Compared with the precedent, our work provided more detailed information about how to set up TDMA system for characterizing the ion transmission for planar DMAs, and provided the updated transmission value for the latest version of the planar DMA. In the revised manuscript, we have added description of the transmission, which is the maximum transmission efficiency, defined as the peak height of the ratio of downstream electrometer signal to upstream electrometer signal. We have also included more characterization results of ion transmission in the SI. The revised contents are shown as following “*The transmission illustrated is the maximum transmission efficiency, which is defined as the peak height of the ratio of the aerosol number counted by the downstream and upstream electrometer (N_{down}/N_{up}). The experimental results of N_{down}/N_{up} under different working condition is shown in Fig. S7 and S8.*”.

Fig 4 is to compare the sizing resolution of DMA P5 with other cylindrical DMAs, all peak intensity in normalized with the peak height. We have also added description in the figure caption. d. More discussion is needed in Section 3.2 and some parts in 3.1, including more details on different modes of the setup and the applicability of the results to other setups. For instance, when measuring atmospheric clusters, the clusters are introduced to the DMA from the polydispersed aerosol flow inlet, right? Then the discussion related to Fig. 1 can be confusing as clusters are injected into the

DMA using an electrospray. Whether electrostatic losses vary with the working modes also needs to be explained.

We thank the reviewer for this comment. We have added more discussion in section 3.2 about the application of TDMA system for the characterizing the ion transmission efficiency. And more details on setup in both Section 3.1 and Section 3.2. The discussion about Fig.1 is about how we use standard ions generated from electrospray source to characterize the DMA P5. We agree with the reviewer that we have not included the explanation of how the DMA P5 works for measuring the atmospheric clusters. When measuring atmospheric clusters: (1) Under injection mode, the clusters are introduced to the DMA from the polydispersed aerosol flow inlet. The reagent ions are generated via electrospraying of custom selected solutions. The reagent ions charge the oxidation products through secondary electrospray ionization (SESI) (Rioseras et al., 2017). (2) Under counter flow mode, the blocked port (red labeled in Fig. 1b) is used to introduce the atmospheric clusters, while the O_{count} is equal to the sum of the counter flow rate and the sample flow rate. In the revised manuscript, we have added the explanation of how we set up DMA P5 for the measurement of atmospheric clusters in section 3.1. The revised sentences are shown as following ***“It should also be noted the above-mentioned recirculation set ups are applied for the study of aerosols generated from ESI source. For the measurement of atmospheric clusters, secondary electrospray ionization (SESI) (Rioseras et al., 2017) is applied, with the reagent ions generated via electrospraying of custom selected solutions. Under injection mode, the clusters are introduced to the DMA from the polydispersed aerosol flow inlet. Under counter flow mode, the blocked port (red labeled in Fig. 1b) is used to introduce the atmospheric clusters, while the O_{count} is equal to the sum of the counter flow rate and the sample flow rate. Gao et al. (2023) have deployed the SESI-DMA-TOF for the measurement of the products of α -pinene ozonolysis.”***

Reference

Rioseras, A.T., Gaugg, M.T., Martinez-Lozano Sinues, P.: Secondary electrospray ionization proceeds via gas-phase chemical ionization. *Anal. Methods* 9, 5052-5057, 2017.

Gao, J., Xu, Z., Cai, R., Skyttä, A., Nie, W., Gong, X., Zhu, L., Cui, S., Pei, X., Kuang, B., Kangashuoma, J., Wang, Z.: Molecular identification of organic acid molecules from α -pinene ozonolysis. *Atmospheric Environment*, 312, 2023.

e. The abstract and conclusion could be sharpened such that the audience can better understand the main contribution of this study to the research community. For instance, "we assessed the performance of a commercial planar DMA integrated with the home-build recirculation system" in the abstract is rather confusing, as it seems to emphasize that the results of this study do not apply to other P5 DMA systems due to the difference in the recirculation system.

We thank the reviewer for this comment. The original intention of mentioning the home-build recirculation system is that all components can be purchased from the local market. Our reported transmission and resolution are expected to be reproduced under suction mode or counter flow mode of DMA P5 with any recirculation system providing temperature-constant, aerosol-free, stable sheath flow. We have re-arranged the abstract and conclusion in the revised manuscript, leaving out the emphasis of the home-made recirculation system.

2. Due to the lack of explanation, the comparison among different DMAs seems to be a bit arbitrary.

a. Taking Fig. 4 as an example, what are the sheath and aerosol flow rates of the DMAs, and why these values are used for comparison? Were the aerosol-to-sheath flow rates the same for all the DMAs or the aerosol flow rate is set to the same value? Are the flow rates chosen to represent typical ambient measurement conditions or they are for different conditions? The underlying question behind these several questions is, if e.g. TSI 3086 DMA works with a resolution of 5 in a setup for ambient cluster/particle measurement, will the P5 DMA provide a resolution higher than 50, or does it simply not usable due to the difference in flow configurations?

We thank the reviewer for this comment. We have added the explanation of Fig. 4, reporting the corresponding sheath and aerosol flow rates of the DMAs. 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. The aim of making this comparison is that although application of DMA P5 on atmospheric particle number size distribution measurements is unpractical due to the high maintaining expenses for keeping the super high sheath flow rate, the exceptional sizing resolution and high ion transmission of DMA P5 is merit of being further exploited by coupling with mass spectrometer for cluster detection with an additional ion mobility dimension. The added contents is shown as following "*.....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. The comparison results show that the planar DMA has the highest sizing resolution, which is 5-16 times higher than conventional cylindrical DMAs (Fig. 4). On one hand, due to the high maintaining expenses for keeping the super high sheath flow rate, application of DMA P5 on atmospheric particle number size distribution measurements is unpractical. On the other hand, high resolution and high ion transmission are almost synonymous for planar DMAs. This advantage is merit of being further exploited by coupling with mass spectrometer for cluster detection with an additional ion mobility dimension.....”.

b. Related to this, it seems the "conventional cylindrical DMAs" in the abstract (line 21) do not include Hermann DMA and the half-mini DMAs. Why?

We thank the reviewer for this comment. Hermann DMA and the half-mini DMAs are conventional cylindrical DMAs. We have corrected the comparison results of the sizing resolution between DMA P5 and conventional cylindrical DMAs. The revised sentences are shown as following “*The sizing resolution of DMA P5 is 5-16 times higher than conventional cylindrical DMAs.*”

c. The results in Fig. 5b need to be improved. TSI 3086 was not developed in 2011 so one cannot only cite Jiang et al. without explanation. It might be better to use the same bar for TSI 3085 and 3086 and cite Stolzenburg et al. (2018). The transmission of the Grimm nanoDMA has been improved and the results can be found in Stolzenburg et al. (2017).

We thank the reviewer for this comment. We have improved the results and citation in Fig. 5b.

d. The resolution in Fig. 2a looks different from Fig. 3 in Amo-González and Pérez (2018). I would like to see a discussion on this difference and how it affects the results.

We thank the reviewer for this comment. The theoretical resolution of DMA P5 is different from the calculation in Amo-González and Pérez (2018). Dr. Amo-González, as the co-author of this paper, have pointed out that there was a mistake on the formulas in his published paper. We have used the corrected formula in this paper, which showed higher theoretical value of sizing resolution. The detailed derivation of Eq. (4) has been added in the SI (Section 3).

3. The novelty of the study can be better emphasized by shortening Section 3.3 (moving some parts to the SI) and leaving more space for 3.1 and 3.2. Fig. 8 can be moved to Section 3.1. The authors are encouraged to emphasize more on the characterization results instead of emphasizing the high resolution of P5 without restricting the working conditions, as it is known that P5 can reach a high resolution of > 100 at certain conditions.

We thank the reviewer for this comment. In the revised, we have added more discussions in section 3.1 and 3.2, emphasizing the characterization results of P5. Giving more details about how to set up DMA P5 system for characterizing its performance with standard ions generated from electrospray sources and for conducting experiments for characterizing transmission efficiency through TDMA system. We think the revised version is better organized for the reader to follow. On the other hand, the advantage of DMA P5, compared to cylindrical DMAs, is its exceptional ion transmission and the capability of coupling with MS. With the current setup, this technique can be very useful in laboratory studies of atmospherically relevant clusters, we have also added more details about the interpretation of the 2D spectrum of sulfuric acid clusters.

Minor comments:

4. Please revise the title to "Characterization of a planar differential mobility analyzer (DMA P5): resolving power, transmission efficiency and its application towards atmospheric cluster measurements". It seems none of the measured clusters in this study are sampled from the atmosphere.

We thank the reviewer for this comment. Our measured clusters were not sampled from the really 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.

5. The P5 DMA is described as newly developed, which is a bit confusing. Is it a new model or the same as the one reported by Amo-González and Pérez (2018)? Also, the DEG-SMPS in 2011 cannot be described as "newly developed".

We thank the reviewer for this comment. We agree that newly developed is not an appropriate description for both DMA P5 and DEG-SMPS. In the revised manuscript, we have removed “newly developed” for both DMA P5 and DEG-SMPS.

6. a. line 17, page 1, abstract, "0-3.9 nm". Better to use sub-3.9 or start with a very small diameter. 0 nm does not practically make sense.
- b. line 23, page 1, abstract, "stopped linearly increase". increasing?
- c. line 23, page 1, abstract, "enter a plateau". Entered
- d. line 24, page 1, abstract, "one factor of magnitude". one order of magnitude or a factor of 10
- e. "thorough" in multiple places. through.
- f. line 149, page 6, "much closer". Significantly closer. It is still far from the ideal resolution.
- g. Table 1, diameter. Please specify which diameter it is in the caption or the table header.
- h. line 278, page 14. "is 7-16 times higher". Can be. "is" is too strong and hence incorrect as it depends on the flow configurations.
- i. Please check the colon in the title

We thank the reviewer for the comment. All the above-mentioned inappropriate words or grammar errors have been corrected.

References:

Stolzenburg, D., Steiner, G., and Winkler, P. M.: A DMA-train for precision measurement of sub-10 nm aerosol dynamics, *Atmospheric Measurement Techniques*, 10, 1639-1651, 10.5194/amt-10-1639-2017, 2017.

Stolzenburg, M. R.: A review of transfer theory and characterization of measured performance for differential mobility analyzers, *Aerosol Science and Technology*, online available, 10.1080/02786826.2018.1514101, 2018.

Stolzenburg, M. R., Scheckman, J. H. T., Attoui, M., Han, H.-S., and McMurry, P. H.: Characterization of the TSI Model 3086 Differential Mobility Analyzer for Classifying Aerosols down to 1 nm, *Aerosol Science and Technology*, 52, 748-756, 10.1080/02786826.2018.1456649, 2018.

Amo-González, M. and Pérez S.: Planar Differential Mobility Analyzer with a Resolving Power of 110, *Analytical Chemistry*, 90, 6735–6741, 10.1021/acs.analchem.8b00579, 2018