Response to comments by editor:

Thank you for making changes in response to the referee comments, and for an interesting and well-written manuscript. I have a conceptual question and then several technical corrections:

Response: We are grateful to editor for her/his comments and suggestions to improve our manuscript. We have implemented changes based on these comments in the revised manuscript. We repeat the specific points raised by the editor in italic font, followed by our response. The pages numbers and lines mentioned are with respect to the third version that uploaded on 16.08.2020.

Technical comments:

1) Line 367: I don't understand the heat source from the center electrode. Are you assuming it is at different temperature than the air, and if so, why? Please explain all the variables in the equation.

Response: Yes, we assume that the inner electrode is at different temperature than the air. We observed that temperature of excess flow is ~0.2 °C higher than that of sheath flow at the inlet of nano-DMA2, while temperature of sheath flow is equal to that of aerosol flow at the inlet of nano-DMA2 during the measurements. Thence, a small temperature difference within nano-DMA2 is more likely due to the heat transfer between the inner electrode and air which flows around it by convection/conduction (Bezantakos et al., 2016). The plausible reason could be that when charged nanoparticles (similar to the electric current) hit the inner electrode, the inner electrode has some resistive heating from the electric current that flows. Such temperature difference/gradient within DMA was observed by previous studies (Biskos et al., 2007; Villani et al., 2008; Dupplissy et al., 2009; Bezantakos et al., 2016; Giamarelou et al., 2018). For example, a ±0.5 °C temperature difference within DMA was observed by Giamarelou et al. (2018) during the measurements. Except for the possibly slightly higher temperature of the inner electrode than the surrounding air, temperature gradient in DMA2 may also be caused by environmental disturbance or temperature difference between other parts of DMA and between sheath flow and aerosol flow.

Thus, we used the following heat capacity equation to calculate the change in heat of a nano-DMA2 system (Q) at a constant pressure.

$$Q = mdTC_{p,k} \quad (1)$$

Here, m is mass of air flow (sheath and aerosol air flow), dT is infinitesimal temperature change within a nano-DMA2 system, and $C_{p,k}$ is specific heat capacity of air. The heat produced from the inner electrode of nano-DMA2 is ~0.08 W.

Related changes included in the revised manuscript:

Page 16 line 363-369: we revised "Unlike previously reported by Bezantakos et al. (2016) that the RH at the outlet was higher than that the inlet of the sheath air, we monitored that the sheath flow temperature at the inlet of nano-DMA2 is slightly lower (less than ~0.2 K) than that at the outlet, i.e., the RH_s at the inlet of nano-DMA2 is slightly higher (~ 1%) than the RH of the excess air at the outlet. It may due to the heat produced from the inner electrode of nano-DMA2, which we estimated to be ~0.08 W ($Q = mdTC_{p,k}$) by considering the density and heating capacity of air, and aerosol and sheath air flow rate (ρ=1.2041kg/m^3; C_p= 1.859kJ/kg°C) (Atkins et al., 2006)." **as** "Unlike previously reported by Bezantakos et al. (2016) that the RH at the outlet was higher than that the inlet of the sheath air, we monitored that the sheath flow temperature at the inlet of nano-DMA2 is slightly lower (less than ~0.2 K) than that at the outlet (i.e., the RH_s at the inlet of nano-DMA2 is slightly higher (~ 1%) than the RH of the excess air at the outlet), while temperature of sheath flow is equal to that of aerosol flow at the inlet of nano-DMA2 during the measurements. A small temperature difference within nano-DMA2 is more likely due to the heat transfer between the inner electrode and air which flows around it by convection/conduction (Bezantakos et al., 2016). The plausible reason could be that when charged nanoparticles (similar to the electric current) hit the inner electrode, the inner electrode has some resistive heating from the electric current that flows. Such temperature difference/gradient within DMA was observed by previous studies (Biskos et al., 2007; Villani et al., 2008; Dupplissy et al., 2009; Bezantakos et al., 2016; Giamarelou et al., 2018). For example, a ±0.5 °C temperature difference within DMA was observed by Giamarelou et al. (2018) during the measurements. Except for the possibly slightly higher temperature of the inner electrode than the surrounding air, temperature gradient in DMA2 may also be caused by environmental disturbance or temperature difference between other parts of DMA and between sheath flow and aerosol flow. In this study, we calculate the change in heat (Q) of a nano-DMA2 system at a constant pressure, which estimates to be ~0.08 W ($Q = mdTC_{p,k}$) by considering the density and heating capacity of air, and aerosol and sheath air flow rate (ρ=1.2041kg/m^3; C_p= 1.859kJ/kg°C) (Atkins et al., 2006)."

2) Fig. 3 caption: The sentence regarding the black solid lines is repeated at the end of the caption.

Response: Many thanks. We have deleted this sentence and now they read as:

Page 43 line 918-924: "**Figure 3**. Sizing accuracy and sizing offset of nano-DMAs after calibration. **(a)** Normalized number size distribution scanned by the nano-DMA2 for 100-nm PSL nanoparticles (black solid square). Normalized number size distributions scanned by the nano-DMA2 for 100-nm PSL nanoparticles **(b)**, 60-nm **(c)**, and 10-nm **(d)** ammonium sulfate (AS) selected by the nano-DMA1 at RH below 5% at 298 K (black solid square). The dotted lines mark the diameters of the monodispersed nanoparticles selected by the nano-DMA1, i.e., 100 nm in **(b)**, 60 nm in **(c)** and 10 nm in **(d)**. The black solid lines mark the peak diameters from the Gaussian fits (red curve)."

3) Tables S3, S4, and S5: These tables have far more significant figures than are plausible. Please use an appropriate number of sig figs (maybe 3?).

Response: Many thanks. We have revised in the following tables accordingly.

Related changes included in the revised supplement:

Table S3. Average sizing offset between nano-DMAs in the nano-HTDMA system at RH below 10%

	Average sizing offset (nm)[a]	Size agreement between nano-DMA1 and nano-DMA2[b]
100-nm $(NH_4)_2SO_4$	0.619	0.619%
60-nm $(NH_4)_2SO_4$	0.299	0.498%
20-nm $(NH_4)_2SO_4$_	0.278	1.39%
10-nm $(NH_4)_2SO_4$	0.0896	0.897%
8-nm $(NH_4)_2SO_4$	-0.0160	-0.200%
6-nm $(NH_4)_2SO_4$_	0.0840	1.40%

[a] Calculation from $(\overline{D}_{measured\ by\ nano-DMA2} - D_{selected\ by\ nano-DMA1})$

[b] Calculation from $[(\overline{D}_{measured\ by\ nano-DMA2} - D_{selected\ by\ nano-DMA1})/\ D_{selected\ by\ nano-DMA1}]\times 100\%$

Table S4. The values of D_m, g_f, and D_m (< 5% RH) of 10-nm ammonium sulfate of Biskos et al. (2006b) system in the different RHs.

Relative humidity	D_m	g_f	D_m (<5 % RH)
25%	10.4	0.991	10.5
76%	10.4	1.02	10.2
78%	10.5	1.03	10.3
80%	13.3	1.29	10.3
44%	11.6	1.12	10.3
35%	11.2	1.08	10.4
34%	10.6	1.01	10.5
32%	10.2	1.00	10.2
31%	10.2	1.00	10.2
30%	10.4	1.00	10.4
29%	10.3	1.00	10.3
24%	10.3	0.997	10.3

Table S5. Uncertainties of nano-DMA voltage (V) and sheath flow rates (Q_{sh}), and calculated size uncertainty.

Size (nm)	Uncertainties in V and Q_{sh}	Uncertainty (Sizing accuracy)
100	2.65×10^3±0.0259 V, 10±0.0200 L/min	0.200%
60	1.06×10^3±0.0269 V, 10±0.0200 L/min	0.200%
20	1.31×10^2±0.0152 V, 10±0.0200 L/min	0.200%
10	3.37×10^1±0.0244 V, 10±0.0200 L/min	0.213%
8	2.16×10^1±0.0373 V, 10±0.0200 L/min	0.264%
6	1.22×10^1±0.0692 V, 10±0.0200 L/min	0.601%

4) References: The formatting is very uneven, a consequence of using EndNote-type software. Please consistently abbreviate journal names, do not capitalize article titles, and please follow the other Copernicus formatting requirements.

Response: Many thanks. We have checked and revised all reference formats according to the Copernicus formatting requirements, and revised references are marked with red in the manuscript.

5) Line 36: Change "interests" to "interest"

Response: Many thanks. We have revised in the following sentence and now they read as:

Page 2 line 36-37: "The climatic effects of aerosol nanoparticles have attracted increasing interest in recent years (Wang et al., 2016; Andreae et al., 2018; Fan et al., 2018)."

6) Line 68: Change "focus" to "focused"

Response: Many thanks. We have revised in the following sentence and now they read as:

Page 3 line 68-71: "Using these techniques, most of the early lab studies focused on the hygroscopic behavior of particles in accumulation modes and super-micron size range, including deliquescence, efflorescence of pure components and the effect of organics on the change or suppression of deliquescence and efflorescence of these inorganic components in mixtures."

7) Line 72: Change "attempting" to "that have attempted"

Response: Many thanks. We have revised in the following sentence and now they read as:

Page 3 line 72-75: "For nanoparticles with diameters down to sub-10 nm, there are, however, only very few studies that have attempted to investigate their interactions with water molecules, which mainly utilized the setup with humidified tandem DMAs (Hämeri et al., 2000, 2001; Sakurai et al., 2005; Biskos et al., 2006a, b, 2007; Giamarelou et al., 2018)."

8) Line 87: Change "pre-deliquesced" to "pre-deliquescence"

Response: Many thanks. We have revised in the following sentence and now they read as:

Page 4 line 85-87: "To accurately measure phase transition (e.g., DRH and ERH), a highly stable measurement condition is essential, especially maintaining a small temperature perturbation in the humidification system and inside the second DMA to prevent pre-deliquescence."

9) Line 85: Remove "the"

Response: Thank you for your comment. I cannot find "the" in line 85, but I checked "the" in the whole of manuscript. All revision is marked with red in the manuscript.

10) Line 98: Change to "we present the design of a nano-HTDMA. . . ."

Response: Many thanks. We have revised in the following sentence and now they read as:

Page 4 line 98-99: "In this study, we present the design of a nano-HTDMA setup that enables high accuracy and precision in hygroscopic growth measurements of aerosol nanoparticles with diameters less than 10 nm."

11) Line 109: Change to "We designed a nano-HTDMA system to. . . ."

Response: Many thanks. We have revised in the following sentence and now they read as:

Page 5 line 109-110: "We designed a nano-HTDMA system to measure the aerosol nanoparticle hygroscopic growth factor (g_f), especially aiming for accurate measurement of phase transition and hygroscopic growth factor for nanoparticles in the sub-10 nm size range."

12) Lines 126, 128, 130, 145: Please don't use abbreviations "Di." and "L.", and please use metric units instead of inches (").

Response: Many thanks. We have revised in the following sentences and now they read as:

Page 6 line 125-130: "In the deliquescence mode, dry nanoparticles are humidified by a Nafion humidifier (NH-1, TROPOS Model ND.070, Length 60 cm) to a target RH. In the efflorescence mode, nanoparticles are first exposed to a high RH condition (~97% RH) in a Nafion humidifier (NH-2, Perma Pure Model MH-110, Length 30 cm) and then dried to a target RH through NH-1. The humid flow in the outer tube of NH-1 is a mixture of high-humidity air produced with a custom-built Gore-Tex humidifier and heater (GTHH: TROPOS Model, Inner Radius 1.5 cm & Length 30 cm) and dry air in variable proportions."

Page 6 line 145-147: "In addition, we have tested a longer NH-2 (Perma Pure Model MH-110, Length 121 cm) in the efflorescence mode, and no significant difference in measured growth factors are found, indicating that the residence time in NH-1 and NH-2 should be sufficient."

13) Line 134: Missing a space between "NH-1" and "for"

Response: Many thanks. We have revised in the following sentence and now they read as:

Page 6 line 134: "The residence time is ~5.4 s in the NH-1 for both the deliquescence and the efflorescence modes."

14) Change from "Pure" to "Perma Pure"

Response: Many thanks. We have revised in the following sentence and now they read as:

Page 7 line 148-151: "The number size distribution of the humidified nanoparticles is measured with a combination of the second nano-DMA (nano-DMA2) and the ultrafine CPC. Similar to Biskos et al. (2016b), a multiple Nafion humidifier (NH-3, Perma Pure Model PD-100) is used in our nano-HTDMA system to rapidly adjust the RH of the sheath flow of nano-DMA2."

15) Line 161: Change to "a low flow resistance. . . ."

Response: Many thanks. We have revised in the following sentence and now they read as:

Page 7 line 161-162: “In order to minimize the pressure drop along the recirculating sheath flow loop, a low flow resistance MFM and hydrophobic filter (HF: Whatman Model 6702-3600) are used.”

16) Line 218: Change from "Thence" to "Thus"

Response: Many thanks. We have revised in the following sentence and now they read as:

Page 9-10 line 218-219: “Thus, accurate calibrations of sheath flow rates and HV are crucial for constraining the uncertainty associated with sizing of nanoparticles below 100 nm.”

17) Line 258: Change from "high" to "highly"

Response: Many thanks. We have revised in the following sentence and now they read as:

Page 11 line 258-262: “Since all our temperature sensors and the highly accurate DPM (EDGE TECH Model MIRROR-99) are installed in the aforementioned well-insulated chamber and the chamber temperature is maintained with air conditioner at about 292.15±0.1 K, we calibrate the temperature sensors and correct their systematic shift by comparing the record of temperature sensors and the DPM by keeping them in parallel inside the chamber over a 12-hour time period.”

18) Line 273: Change from "atomizing" to "atomized from a . . . ."

Response: Many thanks. We have revised in the following sentence and now they read as:

Page 12 line 272-274: “Also, 100-nm PSL nanoparticles were atomized from a PSL solution of mixing 3 drops of 100-nm PSL with 300 mL distilled and de-ionized milli-Q water.”

19) Line 298: Change from "select" to "to select"

Response: Many thanks. We have revised in the following sentence and now they read as:**Page 13 line 298-300:** “Afterwards, when using nano-DMA1 to select 100 nm PSL, the scanned size

distribution by nano-DMA2 has a peak diameter at 100.3 nm (Fig. 3b), indicating a good sizing accuracy of the nano-DMA1 too."

20) Line 302: Change from "to estimate" to "estimating"

Response: Many thanks. We have revised in the following sentence and now they read as:

Page 13 line 301-303: "Duplissy et al. (2009) and Wiedensohler et al. (2012) suggested estimating the sizing accuracy of sub-100 nm nanoparticles through the DMA transfer function."

21) Line 303: Place a "the" in front of "DMA"

Response: Many thanks. We have revised in the following sentence and now they read as:

Page 13 line 301-303: "Duplissy et al. (2009) and Wiedensohler et al. (2012) suggested estimating the sizing accuracy of sub-100 nm nanoparticles through the DMA transfer function."

22) Line 304: "HV" is already defined.

Response: Many thanks. We have revised in the following sentence and now they read as:

Page 13 line 303-305: "The theoretical DMA transfer function (see SI. S2. Eq. (S2-S4)) was proposed by Knutson and Whitby (1975) and they noted that sizing is crucially dependent on flow rates and HV applied to the DMA."

Page 9-10 line 218-219: "Thus, accurate calibrations of sheath flow rates and HV are crucial for constraining the uncertainty associated with sizing of nanoparticles below 100 nm."

23) Line 323: Change "to calibrate" to "the calibration of"

Response: Many thanks. We have revised in the following sentence and now they read as:

Page 14 line 323-324: “Note that, we also tested the calibration of the DMA voltage with a voltage meter with lower accuracy of ±1%, and the DMA voltages can only be kept within ±1% around the set value.”

24) Line 343: Change "deliquesce" to "deliquescence"

Response: Many thanks. We have revised in the following sentence and now they read as:

Page 15 line 341-344: “Since our RH sensors were all well calibrated and the uncertainty of RH measurement is ±1%, it is reasonable to hypothesize that the RH upstream of nano-DMA2 has already reached the deliquescence RH of ammonium sulfate nanoparticles.”

25) Line 384: Change "undergo" to "undergone"

Response: Many thanks. We have revised in the following sentence and now they read as:

Page 16 line 381-384: “Biskos et al. (2006b, 2007) attributed these two modes to the co-existence of solid and liquid phase nanoparticles at RH close to the DRH of ammonium sulfate, due to the slight inhomogeneity of RH in the second nano-DMA, i.e., some nanoparticles have already undergone deliquescence (liquid state) and some are not (solid).”

26) Line 389: Change to "As RH further increases, the peak. . . ."

Response: Many thanks. We have revised in the following sentence and now they read as:

Page 17 line 389-391: “As RH further increases, the peak diameter of normalized number size distribution of the blue mode increases, indicating the continuous growth the nanoparticles after deliquescence.”

27) Line 445: Change to "in the hygroscopic growth factor. . . . "

Response: Many thanks. We have revised in the following sentence and now they read as:

Page 19 line 444-446: "For example, a slight increase in the hygroscopic growth factor of 6-nm ammonium sulfate nanoparticles is observed in the RH range from 65 to 79% RH before deliquescence."

28) Line 454: Change "a" to "an"

Response: Many thanks. We have revised in the following sentence and now they read as:

Page 19 line 450-454: "Note that, the ammonium sulfate hygroscopic data from Biskos et al. (2006b) shown here are all generated by an electrospray, but in our experiments, only the ammonium sulfate nanoparticles with diameters smaller than 20 nm (i.e., 10, 8, and 6 nm) were generated by an electrospray, while the larger nanoparticles (i.e., 20, 60, and 100 nm) were generated by an atomizer."

29) Lines 460, 461: Remove hyphen before "nm"

Response: Many thanks. We have revised in the following sentence and now they read as:

Page 20 line 461-464: "Figure S12a shows a ~ 0.1 higher growth factor of 20 nm ammonium sulfate generated by the electrospray than that using the atomizer in the RH range from 55% to 82%, which is similar to the difference in hygroscopic growth factor of 20 nm NaCl aerosol nanoparticles using the different generation method as observed in Fig S12b in Biskos et al. (2006a)."

30) Line 472: Change to "the hygroscopicity of. . . ."

Response: Many thanks. We have revised in the following sentence and now they read as:

Page 20 line 471-473: "As a common constituent of atmospheric aerosol particles (Tang and Munkelwitz, 1993, 1994; Tang 1996; Tang et al., 2007), the hygroscopicity of sodium sulfate with diameters above 20 nm particles has been investigated by a few groups (Tang et al., 2007; Xu and Schweiger, 1999; Hu et al., 2010)."

31) Line 482: Change to "an external mixture of . . . ."

Response: Many thanks. We have revised in the following sentence and now they read as:

Page 20-21 line 479-482: "Two intersecting modes in the measured number size distribution of humidified sodium sulfate nanoparticles are observed at RH close to the DRH (Fig. S9 and S10 in the Supplementary Information) and ERH, suggesting an external mixture of aqueous and solid nanoparticles."

32) Line 488: Change to "RH=84%"

Response: Many thanks. We have revised in the following sentence and now they read as:

Page 21 line 488-492: "For example, at RH=84%, the hygroscopic growth factor of 6 nm sodium sulfate is only ~ 1.3 (in efflorescence mode), while the respective growth factors are about 1.5 and 1.8 for 20 nm and 14-16 μm particles."

33) Line 515: Change "underline" to "underlying".

Response: Many thanks. We have revised in the following sentence and now they read as:

Page 22 line 513-516: "As different hydrates of sodium sulfate may exist during the deliquescence and efflorescence processes (Xu and Schweiger, 1999), to explain the underlying mechanism of the size dependent hygroscopicity of sodium sulfate particles can be challenging."