

Review report on Wu et al. (2021)

The authors present a very thorough investigation of the amount and chemical composition of organic impurities on aerosol particles produced from commercially available ammonium sulphate (AS). Atomised AS particles are probably the most used calibration and test particles in aerosol particle measurements. In many applications, organic impurities can bias the calibration results and thus the interpretation of measurements. Hence, this article is highly relevant for the community and should be published in AMT after some minor revisions which are mostly related to the presentation of the work.

Main comments

- 1) The authors build a very good case of why the amount of organic impurities is so important, e.g., for using AS in calibrations of instruments for hygroscopicity related measurements (HTDMA, CCN-Counter). It will strengthen this paper if they could give an estimation of how much such calibrations could be affected (i.e., be biased) if the used AS has the observed amount of impurities. The calibration of the true super saturation (SS) in a CCN-Counter or the RH in an HTDMA could serve as such an example. So, how much could the SS or RH be “wrong” if the user assumes 100% AS, but in reality, there is 1-4% organic with an average hygroscopicity parameter of, e.g., $\kappa=0.1$?
Such an estimation would be very helpful since it seems impossible to reduce the organic impurity content below 1-2 % even with the purest AS sample and the purest water.
- 2) Is it possible to quantify the amount of organic mass in the LC-MS measurements? If so, are the relative fraction of organic impurities ($[\text{Org}]/[\text{SO}_4]$) comparable to the one determined with AMS? Not every aerosol particle measurement group has access to an AMS to check for the amount of impurities of the AS they use. But it may be possible to send out samples for a LC-MS analysis to an external lab.
- 3) It is known in the AMS community that inorganic salts like AS or ammonium nitrate can trigger a CO_2^+ signal (m/z 44) due to reactions on the vaporiser of the AMS instrument (“Pieber effect”, Pieber et al. (2016)). This artefact is highly instrument specific and is strongly related to the measurement history of the AMS in question. Although it is mostly relevant for NO_3^- salts, it can occur for AS as well. Are the authors aware of this effect and have they checked if some of the observed CO_2^+ signal could be affected by this? It would be good to acknowledge the existence of this effect and point out that the observed organic signals in this study have a different source (i.e., the salt itself).

Minor and language related comments

- 30
- 1) line 11&16: “from 200 nm to 500 nm”: Please indicate which size is used here – aerodynamic or electromobility. As this paper is directed at different parts of the aerosol particle measuring community, you cannot be sure, what the reader will assume here.
 - 2) line 18: The authors have to be more precise when they use the term “density”. To my knowledge, the default definition is density = mass/volume. But here, the context suggests that the “surface density” is meant (mass/surface). The authors have to be clearer when they refer to density and when surface density is meant. See also later comment.
 - 35
 - 3) line 37: “it is well acknowledged...”: this phrase sounds odd to me. I suggest replacing it with “it is well established...” or “it is well known...”
 - 4) line 42ff: The water solubility of (di)carboxylic acids quickly decreases with carbon chain length towards very low solubility. Succinic acid (C4) is still somewhat soluble in water (58 g/L), but adipic acid (C6) is already barely soluble at room temperature (15 g/L). κ values derived from their HGF values are in the range of <0.006 (Petters and Kreidenweis, 2007 and therein). Please, specify that the authors are referring to small/short carbon chain (di)carboxylic acids in this sentence.
 - 40
 - 5) lines 47ff: This sentence over-simplifies the case of organic-inorganic mixtures. It is not simply the incomplete dissolution of the organic fraction that causes non-linear behaviour. Below the deliquescence point of AS, only small amounts of water are present in the particles due to the low hygroscopicity of the organic fraction. Consequently, the formed solution will be very concentrated and thus very much non-ideal. But the assumption of (dilute) ideal solution
 - 45
 - is used for most of the calculations around predicting hygroscopic growth behaviour (e.g. when calculating κ from HGF values).
 - 6) line 52ff: “Thirdly, several studies...” The sentence structure is difficult to follow. The double listing (marine organic compounds and ozonolysis products, a-pinene and monoterpene) is confusing. Is it the products of the reaction of O₃ with a-pinene & monoterpenes? Or the products of ozonolysis of a-pinene and the products of ozonolysis of
 - 50
 - monoterpenes? Also, it should be plural and not singular for monoterpenes.
 - 7) line 52: “marine organics under low concentration”. First, it should be “organic compounds” not “organics”. Second, it is “at low concentration” not “under”.
 - 8) line 53: “AS’s hygroscopicity” the “s” genitive form (possessive form) is usually not used for non-living things. Use “the hygroscopicity of AS” instead.
 - 55
 - 9) lines 52ff: Again, the authors oversimplify a rather complex matter. The conclusion that the presence of small amounts of moderately surface-active organic compounds results in a strong depression of the surface tension of the droplet is based on the interpretation of a specific modelling approach (Ovadnevaite et al., 2017). A recent comparison study for this and other thermodynamic models (Vepsäläinen et al., 2021) showed that the inferred behaviour of the surface tension of a droplet strongly depends on the selected modelling approach (e.g., if the surface tension could vary with

- 60 droplet growth or how the surface-to-bulk partitioning was handled). I recommend that the authors add a sentence at
the end of this paragraph, pointing out that organic compounds can have a very complex impact on the hygroscopic
growth and CCN activity of aerosol particles which may go beyond the simple reduction of the value of a single
parameter in the Köhler equation.
- 65 10) line 65&69: what does “In (the) humidification process” mean in this context? Especially, the sentence in line 69f is
difficult to understand. Do you mean that the hygroscopic growth curve displays certain distinct features (like a one
or two sharp stepwise increases at the deliquescence points of the individual compounds?)
- 70 11) line 69: “internally mixing and/or organic coating”. If the particles are internally mixed, it means that the constituents
of the particle are truly mixed and not separated into a core and surface layer. Or do you mean that there is a bulk-to-
surface partitioning when the aqueous solution is formed with a part of the organic material in solution and another
part as a film/monolayer on the surface of the droplet?
- 12) line 75: “Ammonium cation (NH₄⁺) is in pH-dependent equilibrium with...” should be “Ammonium cations (NH₄⁺)
are in a pH-dependent equilibrium with...”
- 75 13) line 80: “... as they impact on both human health and climate.”. The use of “impact” as an intransitive verb has been
increasingly accepted. However, it still feels unprofessional in such a sentence. Using “... as they affect both human
health and climate.” or “... as they have an impact on both human health and climate.” would be a more elegant
phrase in this context.
- 80 14) line 86ff: This is just a note: Every scientist I know who works with AS (and other inorganic salt) particles for
instrument calibration purposes is aware that there is always a chance of contamination with organic compounds and
that that will bias the calibration results. Usually, the contamination is blamed on not very clean water (e.g. MiliQ
water sitting around in plastic containers for months before using) or an “old” AS supply (i.e., bottles of AS opened
years ago and everyone using a potentially contaminated spoon/spatula to get their sample). Scientists will check for
signs if this contamination affected their measurements (e.g., considerable water uptake below the deliquescence RH
for HTDMA measurements) and simply reject such measurements and try again to obtain a “better” calibration with
a cleaner AS solution. However, this procedure is rarely mentioned in publications as it is assume as “getting the
85 calibrations right”. It is commendable that finally a group of scientists decided to thoroughly investigate these
contaminations and their true origin. I hope that this study will trigger a more transparent and thorough approach to
the reporting of AS based calibration measurements.
- 90 15) Section 2: There are 3 different particle diameters used in this manuscript (d_m , d_a , and d_{ve}). Although these are standard
ways of describing the particle size and the authors do introduce them when they first occur in the text, it would be
nice to instead introduce them all at the same place and indicate their relation to each other and which
instrument/classifier is related to their measurement. The end of section 2.1.1 or 2.1.2 could be a place for this very
brief information.

- 95 16) Introduction to Section 2 and section 2.1: I find these sections displays overall a “sloppy” writing style. In many instances, the authors are not clear enough in the description of their setup and methods. A reader with strong background knowledge about atomisers, DMA, AAC, and AMS will draw the right conclusions. But the vagueness in almost all sentences will make it hard for a novice in this field to understand what exactly was done. The next 13 comments are all concerning section 2.1.1. I recommend that the authors carefully improve section 2.1 to get it to the same high quality that they show in section 2.2.
- 100 17) line 100: The term “suspended particles”. Why do the authors introduce this new term? It is ambiguous as it could mean particles suspended in air (= aerosol) or suspended in liquid (suspension or emulsion). The term “aerosol particle”, on the other hand, is clear and well established. Hence, it should be used throughout the manuscript if indeed aerosol particles are meant.
- 105 18) line 101 & 103: “previously employed”. It is absolutely unclear what this term is referring to: employed by whom? By the authors? In what study? I can only guess that the authors mean that these are solution concentrations, particle sizes, and aerosol particle concentrations typically used in AS calibration measurements. If that is the case, they have to write it that way.
- 110 19) line 102: “This procedure allowed to form...”. The grammatical structure for the verb allow used in this context must be: “to allow SOMEONE to verb something”. “allow” requires a direct object when used in the meaning of enable/make something possible. The authors must correct their usage of “allow to” here and throughout the manuscript to one of the following forms (using a shortened form of the sentence in line 102 as an example):
- a. This procedure allowed us to form AS aerosol particles. (adding a direct object)
 - b. This procedure allowed the formation of AS aerosol particles. (convert verb to noun)
 - c. This procedure allowed forming AS aerosol particles. (using gerund as direct object)
- 115 20) line 102: “monodisperse AS particles”. Since you point out later that there was a considerable amount of double charged particles when the DMA was used, the authors should change this to “quasi-monodispers”.
- 120 21) line 104: “Aqueous solutions of AS were prepared and analysed by a...” The structure of the sentence can be misunderstood as that preparation and analysis were conducted with an LC-MS. The prepared part is not necessary in this sentence and it can be simply “Additionally, aqueous solutions were analysed with a....”. Note that also In parallel is not the best choice of phrase here. The authors did not even use the same solution or even the same concentration for aerosol generation and LC-MS measurements.
- 125 22) line 104f: The employed method is called mass spectrometry. The employed instrument (LC-MS) is called mass spectrometer. So, it should be spectrometer here.
- 23) line 108: “Aerosol generation and classification” The term “classification” is also used with the meaning of characterisation. It is better to use “size classification” or “size selection” to clarify the content of this section.
- 24) line 109: “A scheme of the experimental setup...” should be “The schematic of the experimental setup...”

- 25) line 113: "...controlling AS concentrations". This is another example of the sloppy writing style in this section. Yes since this is now about the particles, the reader can infer that this is the particle concentration. But why not be precise in the description?
- 26) line 114: "it consists in" should be "consists of"
- 130 27) line 115: "an electrostatic classifier combined with a differential mobility analyser" What the authors describe here is a tandem DMA: An electrostatic classifier (=DMA) and a DMA. Someone who has worked with these platforms can guess that they used an TSI 3080 platform as a size selector. But again, without this detailed background knowledge, the sentence is misleading.
- 135 28) line 116f: "To regulate the flow rate at the exhaust..." What exhaust is meant in this sentence? The only part in Fig 1 labelled with "exhaust" is at the inlet of the dilution system. There is no MFC added there (which is what the sentence implies). Having worked with similar setups, I am very sure that the authors try to describe that they regulated the total sample flow through the classifier by adding a variable additional flow controlled with an MFC. Then the sample flow through (not before) the classifier was 0.4 – 1.4 L min⁻¹.
- 140 29) line 116f: follow-up question: was a variable sample flow only used for the AAC or also for the DMA setup? From the results section it seems that this was only done for testing the AAC behaviour. The current sentence implies that both classifiers had variable sample flow.
- 30) line 135: which water was used for the recrystallisation?
- 31) line 136f and Table 1: How do the 5 d_m sizes selected with the DMA relate to the 4 d_a sizes selected with AAC?
- 145 32) line 141f: "the smaller size, the higher rotational speed and the larger σ_{geo} ". First, it will be easier to understand if the cause (the rotational speed) is put first in this and the effects last (smaller size and larger σ_{geo}). Second, this needs the defined articles (the): "The higher the rotational speed, the smaller the selected size and the larger the σ_{geo} ".
- 33) line 155f: Please add which vaporiser was used in the AMS – normal or capture.
- 34) line 162: "The data treatment has been performed by AMS Analysis toolkit..." It is not the toolkit that performs the data analysis, it is the user. Change "by" to "with" to clarify this.
- 150 35) line 164ff: The authors make it sound as if the presence of air ions was unexpected or exceptional. The detection of the named ions are the normal behaviour of any AMS. Their absence or lower abundance than the ions stemming from the aerosol sample would indicate instrumental issues or require special treatment (e.g., for the air beam correction to account for sensitivity variations).
- 155 36) line 166f: As an experienced AMS user, I am confused by this sentence. They calculated the organic mass from CHO^+ and CO_2^+ only? That would be very strange. OR are there referring to the correction of gas-phase contributions to signal of these two ions and using the established fragmentation table (Frag Table) approach? If so, the method is to adjust the amount of subtracted gas-phase CO_2^+ signal to reach a signal of 0 of the particle-phase CO_2^+ for $C_2H_3O^+$ signals of 0 (i.e., when no particles are present in the sample air). The standard setting for the Frag Table approach is to set the CO^+ signal (which cannot be determined next to N_2^+) to the same value as CO_2^+ . If this was done, the

160 estimated CO⁺ also contributes to the calculated organic mass. If indeed these things were done in this way (which is the standard procedure for AMS data analysis), these confusing sentences starting with “The measured mass spectra...” should be changed to something like:

“CHO⁺ and CO₂⁺ ions were corrected for the influence of the remaining gas phase in the AMS (Aiken, Canagaratna). Other air ions which also might have small contributions from particulate organic matter were omitted to avoid any over-estimation of the organic fraction.”

As a final note on this matter: Since a HR-AMS was used, the only problematic ions for organic mass calculation are CO⁺ (masked by N₂⁺), CHO⁺ (masked by ¹⁵NN⁺) and CO₂⁺ (gas-phase contribution). The other air ions do not overlap with any strong organic signals. The calculation of particulate water is a whole other matter but not relevant for this study.

170 37) line 169: “In parallel...” If the LCMS measurements were not truly in parallel, it should be “In addition...”

38) line 172: Same as Comment 22: The instruments are a Liquid Chromatograph and a Mass Spectrometer.

39) line 173: “...the mobile phase consisted in two eluents” should be “consisted of”

40) line 205: “...three extraction cycles were operated.” “conducted” is a better word than “operated” in this context.

175 41) line 213ff & Figure 2: How were these background experiments conducted? Was the pure water atomised in the same way as the solutions? Is this still size selected or was the polydisperse sample used.

The issue with spraying pure water on its own is that even if contaminations are present, their total amount is probably low and the formed particles are typical much smaller than the AMS inlet cutoff (~60nm Dva with standard PM1 inlet). Hence, these impurities are potentially “invisible” to the AMS. With AS (or any other salt) solutions, the AS will form large particles. Any contaminations of the water itself will then be on these larger particles and thus be detectable with the AMS.

180 42) line 216: “quantity of organic” should be “quantity of organic compounds”

43) line 220f: “The same observations were made...”

44) line 222: “the presence of organic” should be “the presence of organic compounds”

185 45) line 222f: “respectively” is used incorrectly here. In this context, it needs to follow the items it is referring to and be separated with a comma: “Reference A and reference B, respectively”. However, it would be easier for the reader to understand which study had the higher/lower values if this sentence was restructured.

46) Figure 2: “two pure waters in different qualities”. This just feels wrong. It should be: “two types of water with different purity”

190 47) line 230: “Unit-mass HR-ToF-AMS spectrum” It should be “unit-mass resolution spectrum”. However, it would be even better to explain that these were not UMR measurements but rather that the HR data was converted to UMR to enhance readability of the mass spectrum.

48) Caption of Figure 3: What are 12 measurement runs? The term “run” is not defined anywhere.

49) Caption of Figure 3: “Unit-mass spectra” should be “unit- mass resolution spectrum”

195 50) line 235: "...SO_x⁺ and NH_x⁺ are the major fragments." The term SO_x⁺ or NH_x⁺ fragments is not commonly in the context of AMS data. Instead, related ions (e.g., with only C and H atoms; or with only S, H, and O atoms) are grouped into "families" (e.g., the C_xH_y family, representative of hydrocarbons, or the SO_x family, representative of sulphate or sulphuric acid). The names of these families are spelled without the "+". Carefully check each instance in the manuscript to clarify when these families are meant and use the correct terms to differentiate them from the cases when individual ions ("fragments") are meant. E.g., in line 304, the term clearly refers to the families and not individual ions.

200 One of the reasons not to use the term "fragment" in this context is that some of the ions in the families can indeed be the "parent ion" of a compound. This is especially the case for inorganic salt ions, e.g. the SO_x family which includes: SO₄⁺, HSO₄⁺, H₂SO₄⁺. The corresponding compounds are sulphates, bisulphate, and sulphuric acid.

205 51) line 236: I do not understand the comment about the low signal inhibiting the calculation of inorganic/organic nitrate contribution? The first Figure in SI3 shows that NO⁺ could be fitted nicely. What was the problem? The authors should at least provide an estimation of the upper limit of organic nitrates assuming that all detected NO⁺ and NO₂⁺ stem from organic compounds. Also, what is the upper limit of inorganic nitrate mass calculated from these values assuming that all of the signal stems from an inorganic compound? Inorganic nitrate also constitutes a contamination of the AS which potentially alters its properties.

210 52) Figure 4: The y-axis in this figure seems to be logarithmic which is not easily visible at first glance since the axis range is from 1.5 to 4 and the ticks on the right-hand side are equidistant. Why was a logarithmic scale chosen for such a small range? It makes it harder to interpret the spread of points.

215 53) Figure 4: The unit in the legend is m³ for both density types. Also, the abbreviations are not very helpful since they were not introduced elsewhere. The legend would be more readable if the authors introduced the surface density properly in the text and used a single symbol throughout.

54) line 255: What does "multi-characterisation" mean?

55) line 258: This sentence is broken. Should it read: "The calculations of the organic-inorganic ratio using each of these two hypotheses are detailed hereafter."?

220 56) line 261: The relation of d_a and d_{ve} is not described in Appendix A. Appendix A introduces the shape factor and explains the relation between mobility and vacuum aerodynamic diameter. The relation between d_a and d_{va} is missing. This is even more confusing since Appendix A uses only equations with d_{va} and then provides values calculated from ACC data while explicitly stating that that instruments selects by d_a.

225 All this confusion could be avoided by properly introducing all used diameter types together as early as possible and clearly stating which instrument provides which diameter type and how they are related. A logical point for this would be a section in the methods part after all instruments were introduced.

57) line 259: "size-independent density". I assume this is supposed the same as "surface density" in the rest of the text?

58) line 265: Which diameter type is used in the calculation of the Knudsen number?

- 59) line 268 f: “Therefore,...” It is not clear how Eq. 3 is connected to the Eq. 1 and 2 or the content of Appendix A.
Also, M_{SO_4} and M_{AS} are not introduced.
- 230 60) Eq. 4: what is $\rho_{N,intern}$?
- 61) line 275: “...independent to particle size.” should be “... independent of the particle size.”
- 62) line 279f: Did the authors expect that the Nafion drier should be efficient in removing the organic compounds?
Usually, only semi-volatile compounds or very polar ones are assumed to be lost significantly in Nafion driers.
- 235 63) line 287: ”... the DMA selects particles bearing a given electrical mobility size...” This should be “... the DMA selects particles by their electromobility.” Or “...due to their mobility in an electrical field.”
- 64) line 287f: The rest of this sentence is also not strictly correct and should be split into a separate sentence to enhance readability “Thus, the selected distribution is not truly monodisperse but also contains double- and triple-charged particles at the corresponding sizes.”
- 240 65) line 290: The authors should clearly state, why the presence of multi-charged particles of larger sizes is an issue in their analysis. For an SMPS (same selection method and size resolved measurement), the correction for multi-charged particles is routine and no issue at all. The problem arises from using the “bulk” mass concentrations measured by the AMS over all sizes because typically the pToF mode (size resolved measurement) is not sensitive enough to reliably detect the small amounts that this study is investigating.
- 245 66) Figure 5: The logarithmic y-axis makes it difficult to correctly judge the differences between the black and white markers and the red line. This would be much easier with a linear scale.
- 67) Figure 5: What are the y-direction error bars? Standard deviation over from averaging multiple measurements? Or are they an estimation of the true uncertainty?
- 68) line 297: SI5 is not very helpful for understanding what was done here. Too many steps are skipped or not well described. How were Eq. 5.1 and 5.2 combined to obtain 5.3? What diameter type is “d” in these equations?
- 250 69) line 297ff: From how I understand the corrections lined in SI5, $[Org]/[Sulphate]_{corr}$ is not the total amount of organic compounds and sulphate but rather only the amount attributed to the first mode. The reason that this value is higher is that there is a size dependence with smaller values for larger particle sizes. Thus by removing the contribution of the larger multi-charged mode, the corrected values are higher.
- 70) line 299: “From the result, using the DMA selection...” delete “from the result”
- 255 71) line 302: “...selection by ACC leads to lower uncertainties...” What are the errors/uncertainties in x direction? I.e., what are the uncertainties for the size selection in the DMA and ACC? Only if this information is given can such a statement be made.
- 72) line 313: “Further identification was investigated LC-MS on liquid AS solutions.” Should be “Further identification was conducted by LC-MS with liquid AS solutions.”
- 260 73) line 325f: This sentence is broken. Probably needs to be split in two.
- 74) line 336: “ ...molecules marked as *...” Should be “...molecules marked with *...”

- 75) line 337: "...another mark of AS..." should be "... another brand of AS..." Also correct this in line 340 and other places.
- 76) line 345f: $C_5H_9^+$ is a typical CH type ion in AMS mass spectra data which occurs in many samples of organic aerosol. It can have many different parent molecules. Hence, the co-occurrence in LC-MA and AMS spectra should not be over interpreted.
- 265
- 77) Figure 9: It is impossible to see the red squares for most sizes which makes it hard to follow the discussion in the text. The authors could think about changing the marker shape to try make the P2 data more visible. Another option is to apply a small offset to the true d_m values for each set of data points (eg., plot the dark blue ones at 245nm, the green one at 250nm, and the red ones at 255nm. Then add a note to the Figure caption that markers were spread around the true d_m value to enhance visibility).
- 270
- 78) line 383f: "... a wide range of commercial AS products..." The study contained 3 different AS products. I would not call that a "wide range".
- 79) Line 389: "Regarding to the particle size..." Should be "Regarding the particle size..."
- 275
- 80) line 391: "that their structures are independent to the particle size" First, it is "independent of". Second, the conducted MS measurements did not reveal any structural information – only the "raw formulas" were determined. Thus, structure is the wrong term here. Third, the only comparison of composition data between particle sizes was Figure 6. Frankly, that is not convincing to support such a statement. LC-MS suggests highly functionalised molecules with nitrate, amin, and/or sulphate groups. These are compounds which show strong fragmentation with mostly loss of the functional group. Thus, the AMS may not be sensitive to changes in composition between these compounds.
- 280
- 81) line 394: "...confirming their low volatility." Deriving volatility from sum formulas is not as straight forward as it may seem. The presence of O and N in a molecule may mean that there is 1 NO_2 or NO_3 group. Depending on the size of the carbon backbone, this may lead to anything between intermediate and extremely low volatility. Since no volatility measurements were conducted, it may be better to speak of "... suggesting low volatility".
- 285
- 82) line 394f: "As these molecules..." This sentence is broken.
- 83) SI has different formatting for section headings than main manuscript. Some of the table have a different font. This is sloppy. The authors must ensure that the style of the SI meets the standard of the main manuscript text.
- 84) SI3: The quality of the figures in SI3 is not acceptable. These are literally low-quality screenshots of the HR fitting panel in PIKA with blurry y-axis labels, no legends, the residual y-axis is cut off, no Figure numbers, no caption.
- 290
- I do not see the point of adding these Figures. They only show that some $C_xH_yN_x$ ions could be separated from other potentially overlapping ions. The main message I can derive from these figures as an AMS user is that the majority of organic compounds containing also N are detected as NO^+ & NO_2^+ (NO^+ signal is one order of magnitude larger than $C_3H_8N^+$ and $C_4H_{10}N^+$). No inorganic nitrate should be present in the sample. Hence, a method using the NO^+ and NO_2^+ signals should be used to determine an upper limit for the contribution of "organic nitrate" mass.

- 295 85) SI4: The first 3 sentences in SI4 are identical to the main manuscript text in 2.1.2. This repetition does not seem to serve any purpose. The one additional piece of information (the exact speeds) could be added to the main text in section 2.1.2 and the exact values of the result (average +/- stdev) stated in section 3.3 with a reference to the Figure in the SI. Note that “CN/sulfate” is not introduced anywhere. The name of the family is $C_xH_yN_x$ in the main text.
- 300 86) I do not understand why Appendix A and SI5 are split. Things will be easier to follow if these two parts are joined (either as Appendix or as SI section). However, more information is still needed (about the skipped steps in the equation development, see comments above) to make this part usable.

References

- Ovadnevaite, J., Zuend, A., Laaksonen, A., Sanchez, K. J., Roberts, G., Ceburnis, D., Decesari, S., Rinaldi, M., Hodas, N., Facchini, M. C., Seinfeld, J. H. and O’ Dowd, C.: Surface tension prevails over solute effect in organic-influenced cloud droplet activation, *Nature*, 546(7660), 637–641, doi:10.1038/nature22806, 2017.
- Petters, M. D. and Kreidenweis, S. M.: A single parameter representation of hygroscopic growth and cloud condensation nucleus activity, *Atmos. Chem. Phys.*, 7(8), 1961–1971, doi:10.5194/acp-7-1961-2007, 2007.
- Pieber, S. M., El Haddad, I., Slowik, J. G., Canagaratna, M. R., Jayne, J. T., Platt, S. M., Bozzetti, C., Daellenbach, K. R., Fröhlich, R., Vlachou, A., Klein, F., Dommen, J., Miljevic, B., Jiménez, J. L., Worsnop, D. R., Baltensperger, U. and Prévôt, A. S. H.: Inorganic Salt Interference on CO_2^+ in Aerodyne AMS and ACSM Organic Aerosol Composition Studies, *Environ. Sci. Technol.*, 50(19), 10494–10503, doi:10.1021/acs.est.6b01035, 2016.
- Vepsäläinen, S., Calderón, S., Malila, J. and Prisle, N.: Droplet activation of moderately surface active organic aerosol predicted with six approaches to surface activity, *Atmos. Chem. Phys.*, 1–25, doi:10.5194/acp-2021-561, 2021.