Dear Editor,

Please find our modifications to the manuscript and point by point responses to the reviewer. All edits to the text are marked with track changes. Despite the quite extensive language edits to the text, the results and conclusions of the manuscript remain the same. To Figure 3B dimensions of the inlet system were added, Figure 4 legend was corrected, the legend of Figure 5 has been moved to fit inside the graph, error bars were added to Figure 6 as asked by the reviewer, legend of Figure 11 corrected and data points are now presented in Figure 12 instead of line. The referred line numbers are line numbers of the edited word document with the track changes.

Regards

Juha Kangasluoma

Reviewer 1

Review of the AMT manuscript amtd-8-8483-2015

"Operation of the Airmodus A11 nano Condensation Nucleus Counter at various inlet pressures, various operation temperatures and design of a new inlet system" by J. Kangasluoma et al., 2015

The above manuscript deals with the characterization of a particle counter capable to measure aerosol particles in the sub-3-nm size regime. As this is highly relevant for the investigation of new particle formation events, such a paper is valuable for the scientific community and well suited to be published in AMT.

However …

The manuscript is NOT well written. I have the feeling that I read a draft, not the final manuscript, and I cannot imagine the well-known co-authors read the manuscript. The English is partly not good, the style is partly bad, the use of parameters and units is frequently wrong, partly wrong terminology… The manuscript might be worth being published, but only after major revision. And, to be honest, next time I would likely reject such a manuscript, not because of its content, but because of the bad presentation of the results.

General remarks:

Besides the criticism already made above, one major criticism is that almost only literature from the own (Helsinki) group was cited. When reading the paper, one gets the feeling that the principle instrument idea was developed there and is brand new (page 8484, lines 19&20). But that´s not true. The PSM goes back to Okuyama et al., AS&T, 1984, there are other important papers on this device, like for instance Kim et al., AS&T, 2003 or Gamero-Castaño and Fernández de la Mora, JAS, 2000, please provide more references to the underlying work principle.

We thank the reviewer for the kind words and the long list of edits. The specific remarks have been addressed as mentioned after each remark. We have also edited the text to be more accurate and scientific.

In page 1, lines 31-32 we referred to instruments which actually have been used in the measurements of sub-3 nm particle formation in atmospheric or chamber experiments, not for example ion induced nucleation inside the instrument. We, however, agree that there are not enough references to previous PSM technology, which we addressed in lines 47-55.

Specific remarks:

- p. 8484, l.7: There are four sentences in a row which start with "We …" The abstract is not bullet point list, please change.

Fixed

- p. 8484, l.9: please insert "lower" previous to "cut-off" to be more precise

Done

- p. 8484, l.12: which "background"? atmospheric background aerosol? electronic noise?

Background concentration of the homogeneously nucleated droplets, fixed

- p. 8485, l. 5: "It is crucial …" for what?

To understand the CPC operation, fixed

- p. 8485, l. 8-11: The "Kangasluoma 2013/12014" references for the given statement. This is again an example of the selective citation of the authors. Jiang et al., AS&T, 2011 have shown the same, but two years earlier, so why do you not cite this literature source?

So did Iida et al, AS&T 2009, but neither of them measured the chemical composition of the test particles. We agree that Iida et al, AS&T, 2009 showed the effect of relative humidity on the detection of particles with DEG CPCs, which was added.

- p. 8485, l. 26: the "heat conditioned inlet", what does this imply? When you heat or cool the sampled aerosol particle material might evaporate or condense thus changing the original particle size. How large is this effect?

We have not studied this effect extensively, which should be done with two high resolution DMAs operated in series for large variety of particle/vapor combinations. The heat conditioning in the inlet of this PSM is to minimize the effect of the sample temperature variations to the operation of the instrument (Figure 9).

- p. 8485, l. 28: 90 nm particles can be optically detected, see the UHSAS, hence the given statement is not true. But it is hard to detect them and needs some effort.

We were not aware of UHSAS. Clarified that they too small for CPC optics

- p. 8486, l. 1: what is a “size distribution of 1-2 nm” particles? Do you mean a scan over nominal about 1 nm diameter particles?

Corrected to “size distribution of particles in the size range between 1 and 2 nm”

- p. 8486, l. 3: “whereas” is wrong, the second part of the sentence is no contradiction to the first part

Yes it contradicts? Their theory says that ss maximum is at 0.1 mixing ratio, whereas experiments contradict with the theory.

- p. 8487, l. 5: Why do you use the word “PSM” for the combination of your PSM plus CPC? You already have an acronym for this combination “A11”, so please use this one and PSM only if PSM is really meant.

Fixed so that nCNC is for PSM+CPC, and PSM for PSM

- p. 8487, l. 7: “under pressure” is misleading, please use “low-pressure” throughout the manuscript

fixed

- p. 8487, l. 10: I do not know on which planet the author lives, but on Earth 50 kPa corresponds to about 5600 m altitude (US standard atmosphere) not 4000 m.

Very good. Fixed

- p. 8487, l. 15: “mass flows were calculated by Q/p” a volume flow (rate) divided by a pressure does not give a mass flow (rate). This is given when multiplying a volume flow rate with the density. Wrong units!

The equations were fixed, and also terminology related to flows

- p. 8487, l. 22, Eq. 3: The units on the right side of the equation are not the same (Q = Q/p ?)!

Fixed

- Fig 2: What is a “Herrmann DMA”? What instrument is the circle with the “x” inside? The pressure sensor, does it measure a pressure difference (as suggested by two lines)?

Added a reference to Herrmann DMA to line 184. Circle with x is a filter, added clarification to the figure. The pressure sensor was an absolute pressure meter, which was just connected to the line

- p. 8488, l. 2, the fill bottle was connected to the inlet line to equilibrate the different pressures (right), but in the next sentence (l. 3) you write that the fill bottle was not used. So what, if you did not use it, why do you describe the fill bottle connection?

We described it to show how we equilibrated the pressure in the saturator to the same pressure of the inlet line. We meant the connector of the PSM where the fill bottle is usually connected, not the bottle itself. Clarified

- p. 8489, l. 4: please explain “THABr” when you use it for the first time

Fixed

- p. 8489, l. 9: “maximum detectable size range” sounds for me that you checked the upper detection threshold, you probably mean the “lower detection threshold”

Yes exactly we were interested in the upper detection threshold when the lower detection threshold was set to 1.1 nm. Added clarification

- Fig. 3: “a” and “b” missing, the electric filter cannot be understood, just by looking at the figure

Figure updated

- p. 8489, l. 25: which concentrations were used to calibrate the inlet line? As you use two electrometers the concentrations should have been several thousand particles per cubic centimeter.

The concentrations were around 2000 – 11 000 cm-3, added to the text

- Fig. 4: This figure can only be understood if the reader knows how the flow of the four inlets and outlets of the PSM in Fig. 1 is controlled. But this is not described in the text. It is only stated the sample volume flow rate is constant because of the critical orifice in the CPC (right for the pressure range you investigated, at lower pressures also a critical orifice does not guarantee a constant volume flow rate anymore), but how is the flow controlled for the other two inlets and one outlet? Please describe and please do not refer to another paper, the reader should understand this just be reading this manuscript.

Added more explanation to lines 93-98

- Fig. 4: on the y-axis a volume flow rate is displayed but in the figure caption you speak of mass flow rates, again do not mix this and be correct concerning the units

Fixed

- p. 8490, l. 8: is it really the counting efficiency? In the literature, there are several examples which show that the detection efficiency of a CPC changes at low-pressure. This can also be caused by changes in the supersaturation profile, which is not related to the counting efficiency (as you use it). Same for Fig. 5 caption.

At least Hermann et al. 2001 JAS show that the cut-off diameter rather decreases with decreasing pressure. Also, the selected particles were 50 nm in size which we assume to be large enough to be activated at 100% throughout our pressure range. Of course we cannot be 100% sure, added the possibility of supersaturation profile change

- Fig. 6: the slope of the detection efficiency curve at 3 nm is not zero; the curve is still going up, although the detection efficiency is already 100%. Please explain this behavior.

The curves have been scaled to reach unity at 3 nm to explore the effect of the pressure on the cut-off diameter of the PSM. Added clarification

- Fig. 6: please provide uncertainty bars to the measurement points

Y error given as one standard deviation of the detection efficiency, x error given as the full width of half maximum of the DMA transfer function at the operation conditions, 4%.

- p. 8491, l. 16: "for reasons we cannot yet explain."´What does literature say to this shift, e.g. Ito et al., JAS, 2011?

To us Ito et al. does not explain why in mixing mode the maximum detection efficiency of 1.2 nm particle is at saturator flow rate of 0.3 lpm, while for 1.7 nm particle it is at 0.6 lpm. The design of their instrument is different, and they did not conduct any experiments with varying the saturator flow rate or particle sizes below 2 nm. The only reason we can think of is that with increasing saturator flow rate the diffusion losses increase for some reason, lowering the detection efficiency of the smallest particles at higher saturator flow rates. Maybe the reviewer can help

- p. 8491, l. 20: I miss the discussion on Fig. 8

Added discussion to lines 322-325

- Fi.9: I do not understand the difference between the two curves of the same color. This is not related to the figure, which is clear, but rather to the description what was measured.

Added clarification to lines 219-221

- p. 8492, Sect. 3.3: the first part of this section is text book knowledge and has been applied to many field inlet systems before. Moreover, it is unlikely that other users will use exactly the same inlet set-up, hence they would have to do their own calibration measurements (anyway recommended). So the first part is of little new information and could be deleted or shortened. Same for the Conclusions. More interesting is the second part about the electrical filter, which should be described more in detail.

The several ambient aerosol sampling lines in some stations we have seen are not suitable for sub-3 nm particle sampling due to very high diffusion losses in the long tubes, even if there was core sampling from the long and thick tube. Moreover, we have plans of commercializing the whole inlet system, which is why the penetration efficiency curves can be of interest for others in the future. The same inlet system is also currently used in many of our sites and shorter field campaigns, for which the penetration plots give an important reference on the close to zero inlet line losses, which reduce the uncertainties of the measured number concentrations. Finally, we would be happy to see another core sampling system that has been characterized in the sub-3 nm size range.

More description of the electrical filter given in lines 233-237.

- p. 8492, l. 11: should be “smaller” instead of “larger”

fixed

Technical corrections:

Please …

- p. 8484, l. 3: should be “subject” not “subjected”

- p. 8484, l. 25: replace “are representative of” with “are still valid in”

- p. 8484, l. 26: replace “Subsequently” with “Thereby”

- p. 8485, l. 1: rephrase the first sentence

- p. 8485, l. 2: delete “of”

- p. 8485, l. 16: replace “big” with “large”

- p. 8485, l. 19-22: articles (“a” and “the”) are missing

- p. 8486, l. 8: “supersaturation fields” should probably read as “supersaturation regions”

- p. 8486, l. 12: delete “and understand”

- p. 8487, l. 9: “strangling” sounds for me to be the wrong word, please replace

- p. 8487, l. 17: replace “is.” with “occurs.” p. 8488?

- p. 8490, l. 11: replace “Figure” with “figure”

- Fig. 5: legend outside the graph, caption replace “lowering” with “lower”, insert “lower” previous to “pressure”, the green point at 55 kPa can only be reached if you used the fit curves of the CPC detection efficiency and for the dilution, but not the measurement points, please state this either in the legend or in the figure caption.

- p. 8490, l. 25: replace “cut-offs” with “lower threshold diameters”, also on the figure caption

- p. 8491, l. 8: replace “to onset supersaturation” with “to the onset”

- p. 8491, l. 14: insert “of” after “flows”

- p. 8491, l. 14: replace the “>” signs in the text with text

- Fig. 11: delete the second sentence of the figure caption; figure interpretation should be done

in the text not in the caption

All technical comments fixed

Reviewer 2

General Comments

The authors have obviously done an enormous amount of calibration work on the Airmodus A11 and have succeeded in their objective of elucidating the technical issues involved with operating it at different inlet pressures or operation temperatures. The inclusion of a revised, more efficient inlet system is also welcome. The trends noted from these calibrations leading to the final operational advice are certainly of some value to unwary users of this instrument. It is unfortunate that, as the authors rightly point out at the end of the Introduction, none of these calibrations may be considered as quantitatively applicable to the A11 system in general. This severely limits the overall value of this work.

In general, the experimental setups and the results of the measurements are adequately described, though dimensions for the revised inlet system should be given. However, the authors seem to suffer from a great deal of confusion with regard to volume flow versus mass flow, or more correctly, standardized volume flow. In a number of places, flows termed as mass flows in the text are actually volume flows. These will all be enumerated in the following section. In addition, to lessen the degree of confusion for the reader, mass flow (standardized volume flow) should be given a notation different from that of volume flow, Q. This will improve the clarity of, for instance, Eq. 3 enormously.

We thank the reviewer for the comments and good review of the paper.

The dimensions of the inlet system added to Figure 3b. The equations and terminology related to the flows fixed

Specific Comments – major and minor

Page 8484, line 24: "… laboratory calibrations are representative of field conditions performance."

Fixed

p 8487, ln 9: Though "strangling" a flow presents quite an amusing picture, a more appropriate word here would be simply "restricting".

Fixed

p 8487, lns 10-11: 50 kPa corresponds roughly to 5600m altitude in a standard atmosphere.

Fixed

p 8487, ln 15: It should be made clear here which flows the mass flow controllers control. I believe it is the saturator and excess flows. This would also be a good place to give the values for those flows as well as additional information about normal operation. For instance, when the saturator flow is scanned, is the excess flow simultaneously scanned so as to keep the inlet flow constant? Though this may be described more fully in Vanhanen et al., it is not much to include these few extra details of operation here.

More description added to lines 93-98

p 8487, lns 15-16: "The volume flows were calculated as Q/p, where Q is mass flow and p is pressure.

Flow terminology and equations fixed

p 8487, lns 16-18: Please give the actual volume flow for the CPC here. 1 L min-1?

Fixed

p 8487, ln 13: "… the inlet volume flow …" makes this clearer.

Fixed

p 8487, Eqs (2)-(3): In these two equations, QCPC, Qinlet and Qinlet_low_pressure are volume flows while Qexcess and Qsaturator are mass or standardized volume flows. This would be so much clearer if a different symbol were used for the standardized volume flows. It could be as simple as Q'. Also, Eq. (3) is generally good for all pressures, not just low ones.

Equations fixed

p 8488, ln 1: "… strangling restricting the flow …"

Fixed

p 8488, lns 21-22: The use of "hydrogen sulfate" to describe these particles is at best ambiguous, if not actually wrong. I believe these are particles composed primarily of sulfuric acid molecules, H2SO4, with perhaps one hydrogen sulfate ion, HSO4 - , to account for the charge. Thus, "sulfuric acid" should suffice here.

Hydrogen sulfate was asked by the publisher. From previous experiments (Kangasluoma et al. 2013 AS&T) we know that the composition is $(HSO_4)_x(NH_3)_ySO_4^-$ (which maybe should be more correctly noted as $(H_2S_2O_8)_x(NH_3)_ySO_4^-$ and $(H_2S_2O_8)_x(NH_3)_yHS_2O_8^-$ due low probability of multiple HSO_4 coexisting in one cluster) where the amount of NH_3 is low. Therefore it is not sulfuric acid. Fixed as "particles formed during heating of ammonium sulfate (Kangasluoma et al., 2013)"

p 8489, lns 8-13: The use of the phrase "maximum detectable size range" is very misleading. Though it is the range that is to be maximized, it is more readily interpreted as the size being maximized, which makes no sense in this context. "Detectable sizes" are everything greater

than the cutpoint size. Instead of "detectable size", which apparently is meant to refer to the "minimum detectable size", it would be better to refer to the "cutpoint size", which is commonly understood in reference to CPC efficiencies. The phrase "tested the range of cutpoint sizes" would then seem to more precisely describe what was done.

Fixed as suggested

p 8490, lns11-12: The scaling in Fig. 5 needs a little more explanation. What other effects are being scaled out? And as long as you are repeating all of the legend information in the main text, please include “The blue circles are the uncorrected data.”

Added “The scaling was done due to the fact that the CPC usually detects 90-95% of the electrometer reading, and that a fraction of the 50 nm particles were multiply charged, which lowered the apparent detection efficiency at normal pressure.” Removed legend information from the text

p 8490, ln 21-22: Earlier it was noted that the CPC flow is controlled by a critical orifice, thereby fixing the volume flow at a constant value. So clearly it cannot be decreasing here with decreasing pressure. Furthermore, there is no obvious connection between the flow rate and the “aerosol volume concentration in the optics.” So just what is it that is being corrected for here? Certainly when the pressure drops by a factor of 2 through the sample flow restricting valve, the aerosol volume concentration, barring losses, also drops by a factor of 2. But since the electrometer measures the same reduced concentration, this effect is already accounted for.

This is correct, and a mistake from the author. The point was supposed to be that the electrometer needs to in equal pressure to directly compare the concentrations. This was the case in our experiments, and therefore the data in the figures is correct. The statement removed.

p 8492, ln 13: To explain the observed phenomenon, it is hardly necessary to postulate some mysterious mechanism for homogeneous nucleation in the absence of any significant supersaturation. There is a far more plausible explanation for why adding aerosol does not increase the measured concentration over that of background at a saturator flow of 0.3 L min-1 . The concentrations involved are getting quite high, about 105 cm-3 , no doubt leading to a notable degree of vapor depletion as well as latent heat release. Either of these or both, in concert, are limiting the maximum concentration of DEG droplets formed which can grow to a size detectable in the CPC. This asymptotic limit is independent of whether the droplets are formed via homogeneous or heterogeneous nucleation. The effects of this limit are also visible to some degree at a saturator flow of 1 L min-1 where it is evident that the curves are again converging to the same maximum value as the measured concentration increases.

We suggest that the homogeneously formed particles at saturator flow rate 0.3 lpm can form before the mixing of the aerosol flow between the saturator and mixing zone. Vapor depletion between the saturator and the mixing zone has been shown for example by Gamero and de la Mora (2000, JAS). The convergence of the curves at 1 lpm is due to the fact that the

concentration was constant throughout the experiments, and with lower inlet temperature the temperature of the flow in the growth tube becomes lower due to not perfect temperature conditioning. Thus the supersaturation increases with decreasing inlet temperature, and the detected concentrations converge to 100% detection efficiency. Also, at 0.3 lpm saturator flow rate and inlet T 40, and 1 lpm saturator flow rate and inlet T 10 the concentration of homogeneously nucleated droplets is the same, about 80 cm-3. This suggests that the supersaturation at those conditions were identical, however, in the latter conditions particles are activated but not in the former. How does vapor depletion or latent heat release explain that?

In any case we agree that we do not understand this properly, and removed the discussion on the probable explanation.

p 8492, lns 19-20: Again, the use of "maximum detectable size" is very confusing here. An alternative wording might be "We found it possible to vary the cutpoint size from 1 nm to 6 nm."

Fixed as suggested

p 8493, ln 2: Is 10 mm the tube ID? Please make that clear. For that matter, the general dimensions of the revised inlet system should be noted, including the tube IDs as well as the extraction tube OD along with any relevant lengths. This would more appropriately fit in section 2.3 on page 8489.

Dimensions given, moved the sentence to experimental section.

p 8493, ln 11: "… ions larger smaller than 4.5 nm." There should be a reference to Fig 12 here.

Reference added

p 8493, ln 18: The mass flow controllers are keeping the mass flows fixed, so it is the volume flows that are changing at reduced pressures.

Fixed

p 8493, ln 26: As the saturator temperature is important in determining the cutpoint size and activation efficiency, one cannot simply "reduce the saturator temperature." The concept is more clearly stated in the second Solution entry of Table 1: "Adjust saturator (temperature) for suitable activation efficiency."

Fixed as "to achieve the required supersaturation with as low as possible saturator temperature"

p 8494, lns 4-5: "… transmission efficiency close to 100% … ." Please qualify this statement with a phrase such as "for particles greater than 1.xx nm."

Fixed

Figure 4: Excess and saturator mass flows are fixed by the mass flow controllers, independent of pressure. What is being plotted here are the varying volume flows as a function of pressure. The only mass or standardized volume flows on the plot are those designated “at std conditions”. But then if it is already designated as a mass or standardized volume flow, there is no need to add “at std conditions.” Also, why are the measured points above the calculated line for the “inlet mass flow” but below the line for the “inlet mass flow at std conditions”? Both should be converted to standard conditions with the same formula.

Fixed, small deviance between the measured and calculated points are probably due to the measurement error of the TSI mass flow meters.

Figure 9: The reference to “squares” in the caption needs updating. It should read “Curves with no symbol represent the background concentration and triangles the concentration …”.

Fixed

Figure 11: The legend refers to “box”. Please explain that reference.

Changed box to inlet system

Figure 12: Is this a fit to measured data, in which case show the points, or from theory?

Measured data. Fixed by putting points instead of line