

We would like to thank Referee #3 for the constructive and thorough comments to help us to improve the manuscript. Below is our answer to the comments.

Answer to interactive comments by Referee #3 on our manuscript “Characterisation of corona-generated ions used in a Neutral cluster and Air Ion Spectrometer (NAIS)” by H. E. Manninen et al.

Specific comments:

2.2. How was the HDMA calibrated for the voltage range?

We'll add following sentence to p. 2104, line 9:

The HDMA was calibrated every day before starting the measurements, for establishing an accurate voltage to mobility conversion using mobility standards (Ude and Fernández de la Mora, 2005).

3.1. p. 2106: The mobility distributions presented in Figure 3 seem confusing to me. Are these solely single examples or averaged over several measurements? As they are the first distributions presented I got an impression they describe a “typical” case. However, the distributions in Figure 3 are different, in terms of peak mobility concentrations, than any of those presented in Figure 5. 1) Was something changed in the conditions or is this normal variation between different experiments? I understand. The experiments were demanding and probably, not too many repetitions were done per each measurement. 2) Still, if applicable, could the authors provide short comments on the repeatability of the measurements, since this is indeed an important point if the results are to be applied for wider use? 3) Based on the presented results, it seems the absolute concentrations remain not as stable as the locations of the major mobility peaks in the spectra. Is this a correct interpretation? 4) Another question on Figure 3 (and in fact, almost all the Figures after) relates to the scale of the y-axis: For me, it is always weird to see DMA results presented with raw concentrations in the y-axis, without any normalization. I understand, the HDMA provides nearly (but not fully) monomobile sizing. Since the characterized ions are not standards, I became wondering if the measured voltage bins are actually equal in width, and if not, can this have an effect on the shape of the mobility distributions. However, since I'm not familiar with HDMA in detail, here might be a misunderstanding from my side.

- 1) Yes, Fig. 3 presents the typical corona-generated ions size distribution. Different set-up were used in Fig. 3 and 5. We revise the manuscript chapter 2.4 Experimental set-up. We'll add more information which set-up was used and where the result from this set-up is presented.
- 2) True. Yes, these are all median size distribution. We'll this to the manuscript. Usually, we measured at least 3 full scans with same settings.
- 3) Yes, between the different experiments the total concentration of ions was different.
- 4) Yes, the size (mobility) bins are in linear scale. This has been taken into account.

3.1. p. 2107: In the end of 3.1. authors say: "In these experiments, the pre-filter was used to remove all sub-4 nm charged particles" Does this apply to all results presented in 3.1. and was there a reason for this additional filtering? I understood the incoming air was already filtered with HEPA so probably this was just double-checking?

Yes, the pre-filter was always on to make sure that no ions entered the main corona charger (excluding experiments in chapter 3.4). We'll add more detailed information to the manuscript:

In these experiments, the pre-filter, which is normally operated during offset measurements, was used to remove all sub-4 nm charged particles with a filter voltage ± 50 V before they reach the main charger region.

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In these experiments presented in chapters 3.1-3.3, the pre-filter, which is normally operated during offset measurements, was used to remove all sub-4 nm charged particles with a filter voltage ± 50 V before they reach the main charger region. As the carrier gas was already filtered to be particle-free air, this was only precaution.

3.2. p. 2108: The number of peaks and the shift in mobility with RH in Figure 5 are explained in very simplified manner. For example, I'm not convinced of the shift of negative ions towards smaller sizes with RH if the whole mobility distribution is considered. Also for positive ions, neglecting the RH 0%, for me the shift in mobility seems not so consistent towards larger sizes.

We were more focused on the location and height of the peak in the whole mobility distribution.

p. 2108 On lines 17-18 the conditions (dry compressed air with RH close to zero) for experiments presented in Figure 3 are mentioned. Could these be added on 3.1. already?

We'll modify sentence in p. 2107, line 1:

All spectra were measured with a fixed flow rate of filtered compressed air through the charger.

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All spectra were measured with a fixed flow rate of filtered compressed air (with RH close to zero) through the charger.

p. 2108 lines 20-24: This is an interesting result. Please check again the lines, legends and colors in Figure 6. No discussion on green line is given.

Yes, we'll correct this. We're sorry about this ambiguous expression. We will add following sentences to p. 2108, line 23, to clear the matter:

"It should be noted that the data points represented by red line was measured when carrier gas was additionally 'dried' with a silica gel. The compressed air used in this study has a dew point of -36 °C, which in room temperature (25 °C) correspond to very low RH of $\sim 0.6\%$.

When the silica gel was added to the set-up, it actually increased the RH as the extremely dry compressed air was drying the silica gel – quite the contrary to what was supposed to.”

We also will modify legend of the figure (please, see the modified figure from the attachments to referee #2 comment):

('Dry compressed air', 'Comp.+Filt.+Silicagel','Nitrogen')

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('Dry compressed air', 'Humidified compressed air','Nitrogen')

3.3. Were there any remarkable differences in mass spectra of 0.98, 1.06 and 1.33 nm negative ions?

Unfortunately, we don't know. We have not had resources to have a look at the mass spectra as a function of ion size. This data exist but it's not analyzed so far. We'll look back into it later but it will not happen in near future.

3.4. p. 2111 lines 7-8: "Clearly, the figure indicates that the electrical filtering is size dependent". This was at first glance slightly confusing, comparing with the statement in p. 2107 lines 19-20, where it is said: "increasing the filter voltage . . . decreased the concentration of the ions equally for all the sizes". Maybe this sentence could be reformulated to indicate more precisely that here the increase of the NAIS cut-size with voltage is meant, instead of purely the corona ion filtering?

Yes, very much true. We'll revise the sentence in p. 2111 line 7-8:

Clearly, the figure indicates that the electrical filtering is size dependent.

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Clearly, the figure indicates that here the electrical filtering is size dependent as it increases the lowest detection limit of the NAIS with increasing filtering voltage.

p. 2111 lines 19-22: "The electrical filtering of the corona-generated ions seemed to be more sensitive to the particle size than to the absolute concentrations. These results suggest that if we would have generated WOx particles at 2-3 nm size range the post-filtering with lower voltages would have been more efficient." It was not easy to understand the meaning of this sentence and I'm not convinced the results in Figures 8 and 9 prove it. 1) Could the authors explain more carefully how this conclusion is deduced from the results (and also to which concentrations, corona ion or generated particle they refer)? 2) If I understood correctly, the conclusion was drawn by comparing negative and positive polarities showing different corona ion concentrations but equally effective filtering? 3) Does the statement in conclusions p. 2112 lines 20-25 also refer to this particular result? If not, to what?

- 1) According to Fig. 9, both the negative and positive ions are filtered with the same voltage although the original concentrations between the polarities were very different. Therefore, we did the conclusion that the electrical filtering of the corona-generated ions seemed to be more sensitive to the particle size than to the absolute concentrations.

We'll revise the manuscript.

Although the positive ion concentrations were six times higher at the beginning of the experiment, both negative and positive corona-generated ions were filtered at approximately the same filter voltage. With 50 V only a portion of the WO_x particles was removed. According to these two observations, the electrical filtering of the corona-generated ions seemed to be more sensitive to the particle size (mobility) than to the absolute concentration.

- 2) Yes, according to Fig. 9 the positive ion concentrations were six times higher at the beginning of the experiment, both negative and positive corona-generated ions were filtered at approximately the same filter voltage.
- 3) No, it refers to the size distribution of corona generated ions presented in Fig. 3. We will modify the manuscript in p. 2112 lines 20-22 as following:

According to our results negative corona generated ions were < 1.6 nm and positive ions < 1.7 nm, the lowest detection limit of the NAIS can be set between 2 to 3 nm, being smaller in the negative and larger in the positive polarity.

The sentence in line 23-25 “The limit could be further decreased if the measured small particle concentration is high (when an even larger fraction of the corona-generated ions is used to charge the sample particles)” means that if the ambient small particle concentrations are high, more corona ions will be removed as they charge the sample (ambient) particles. In this case we don’t need to filter as much to reach as low corona-generated ion concentrations after the charger.

We’ll add sentence “In this case we don’t need to filter as much to reach as low corona-generated ion concentrations after the main charger.” to the manuscript.

p. 2111 line 24: Could the authors repeat here the NAIS normal filtering voltage range (it was mentioned to be 70-100 V earlier, I think)?

Yes, we’ll modified the manuscript and add this information.