Responses to reviewer 3

The changes in the manuscript are shown in italics here in the responses and they are marked in red in the revised manuscript.

Major comments:

I) Page 9 line 16 to 21: The Referee is familiar with both methods to calculate the growth rates (mode fitting and appearance time) and still had hard time to follow the Authors' explanation. Please rephrase in a way that can be obvious also to the non- experts. Consider to add a figure in the supplementary material if the wording doesn't get any easier.

Thanks to the reviewer for pointing this out. The description of the two growth rate determination methods is modified. No figures are added, because these methods have been presented in detail by Dal Maso et al. (2005) and Lehtipalo et al. (2014), respectively.

In the mode-fitting method, at each time stamp of the measurement, the representative size of the aerosol population is determined by fitting a normal distribution to the measured concentration distribution along the logarithm of sizes with a base of 10. The mode of the fitted curve is transcribed back to linear scale and taken as the representative size of the particle population measured at this moment (a more detailed description of the method has been presented by Dal Maso et al., 2015). In contrast, in the appearance time method, for each size (the geometric mean size of a measurement size bin), one determines the time (the appearance time) at which the particle population is considered to reach this size, based on the measured concentration evolvement in time (Lehtipalo et al., 2014).

II) Page 12 line 8 to 10: "Owing ... the ionisation of air molecules can be neglected". The Referee thinks that this statement is highly speculative and needs to be modified. Are there measurements in the literature that support this claim? If so, please cite them. Furthermore, even if there is a snow pack, radon could make its way through the snow crystals and gamma radiation would need at least several tens of centimeter of solid ice to be shielded effectively. In addition, there can be areas, such as vertical walls that can be snow free and made of rock that is rich in uranium. If this is not the case, please state it in the paragraph with, if possible some references. Otherwise, rephrase the statement signaling its speculative nature.

Our measurements were carried at Dome C on Antarctic plateau, which is the largest dissert on the earth covered by glaciers and snow. Solid ice and snow inhibit radon release at the South Pole (Maenhaut et al., 1979). The ice is even thicker at Dome C than at South Pole. The thickness of ice reported at Dome C by the European Project for Ice Coring in Antarctica (EPICA) is more than 3000 m (Augustin et al., 2004).

In continental air radon concentrations vary in the range of a few tens to hundreds of pCi m^{-3} (Wilkniss and Larson, 1984). In the early 1970s at the South Pole, radon concentrations were in the order of 0.5 pCi m^{-3} (Maenhaut et al., 1979), about the same or even less than over oceans (Wilkniss and Larson, 1984). The location of Dome C is much more inland than the South Pole back in the early 1970s. Therefore, characterised by the geographical location of Dome C, the contribution of radon or terrestrial gamma radiation to the ionisation in the air can be regarded non-existent. A slight modification is made to the sentence.

Owing to the presence of the thick ice and snowpack (over 3000 m in depth; Augustin et al., 2004), the contribution of radon exhalation or terrestrial gamma emissions from ground at Dome C to the ionisation of air molecules can be neglected.

III) Page 13 line 1 to 6: As CS tended to be higher during NPF can author exclude that NPF was generated by condensable vapors from "polluted" air masses from the continent? The Referee thinks that a look at back trajectories is needed in order to understand weather the source of vapors is local or from long range transport. This information would be very valuable and increase the relevance of the paper.
& IV) Page 13 line 24 and 25: The Referee thinks that a look at back trajectories might help the Authors to be less speculative about the origin of the air mass. Therefore, the Referee feels strongly that a back trajectory analysis should be added and discussed.

Response to III) & IV): The air entering Dome C originates almost entirely from subsidence from very high altitudes. Because of this, air mass back trajectories are not very helpful in tracking the sources of aerosols, or their precursors, at this site (air entering Dome C have plenty of ageing/mixing time during its transport from distant surface sources very difficult to capture with air mass trajectories). The situation would be totally different for coastal Antarctic sites, for which air mass back trajectories would be very helpful. We decided not to use back trajectories for estimating aerosol sources at Dome C, nor for estimating the reason for high values of CS during NPF events compared with event-free days during autumn.

V) Chapter 3.2.2: A discussion on the uncertainties of the GR is completely lacking. The Referee thinks that must be added and discussed in relation to the variability of the GRs calculated with the 2 different methods.

We are afraid that we have to point out that the uncertainties of GRs in relation to the procedures used in the two methods were discussed in the last paragraph in section 3.2.2.

…could be ascribed to the higher uncertainties associated with the mode-fitting method. The mode-fitting method tracks the mode concentration corresponding to sizes based on curve fitting for each measurement cycle, and it could be that the sizes at which mode concentrations were identified apart significantly in two adjacent measurement cycles, i.e. over a short time interval. A large size difference over a small time interval, therefore, would lead to a huge instantaneous GR. In contrast, the

appearance time method is based on looking for the time stamp when the concentration reaches 75% of its maximum in the concentration vs. time space for each size channel of the instrument. Owing to the fact that aerosol and ion data have a higher resolution in the time dimension than in the size dimension, the appearance time method could pick up the time stamp more precisely for each size than the mode-fitting method could do the sizes for each measurement cycle. Consequently, the appearance time method presents GRs with smaller uncertainties (Fig. 5 & Table 1) and yields more representative instantaneous GRs.

VI) Page 16 line 23 to 26: The Referee thinks that an estimate of the uncertainty of J2+ is necessary in order to better assess the significance of the statistical parameters included in the text. In other words, is the standard deviation of the J2+ comparable, larger or smaller with respect to the uncertainties? Please add the missing information and discuss.

The AIS typically underestimates the ion concentration by 15-30% in the size range of 2-3 nm (Wagner et al., 2016). We had no a separate CPC during the campaign measuring alongside the DMPS to estimate the uncertainty in the DMPS measurement. However, according to Wiedensohler et al. (2012), mobility particle size spectrometers of different design usually have an uncertainty range of around ± 10 % between 20 and 200 nm, and larger uncertainties are expected beyond this size range. In principle, the meteorological measurements at Concordia station follow WMO recommendations. According to WMO 2008 (WMO. Guide to meteorological instruments and methods of observation. Technical Report 8, World Meteorological Organisation, 2008.), the error for temperature measurement should be < 0.3 K in the range from -40 to +40 C and the uncertainty for pressure measurement should be about 0.1 hPa.

We made an estimation of uncertainties in J2+ by assuming an underestimation of 15-30% in our AIS measurement, an uncertainty of ± 10 % in our DMPS measurement in the whole size range, an error of ± 1 C in the temperature measurement and ± 1 hPa in the pressure measurement. We calculated the maximum and minimum estimates of J2+ based on these assumptions and evaluated the deviations of J2+ from the mean values. For more than 88% of the J2+ (26 cases in total), we obtained a deviation from the mean smaller than 0.020 cm-3s-1 (the standard deviation in all 26 J2+ values). More than 80% of the calculated deviation from the mean has a value smaller than 0.005 cm-3s-1.

An estimation of uncertainties in J_2^+ was made by assuming an underestimation of 15-30% in the AIS measurement (Wagner et al., 2016), an uncertainty of ± 10 % in the DMPS measurement in the whole size range (Wiedensohler et al., 2012), an error of ± 1 C in the temperature measurement and ± 1 hPa in the pressure measurement. We calculated the maximum and minimum estimates of J_2^+ based on these assumptions and evaluated the deviations of J_2^+ from the mean values of the maximum and minimum estimates. We found that this deviation was smaller than 0.005 $cm^{-3}s^{-1}$ (<0.020 $cm^{-3}s^{-1}$) for more than 80% (88%) of the values of J_2^+.

Minor comments:

Abstracts:
1) Page1, line 24: "One ... days." Rephrase. It is unclear if the Authors want to refer to event days or event free days.

We modified the sentence to deliver the message clearer that we refer to days with no sign of new particle formation, wind-induced ion formation, or ion production and loss associated with cloud/fog formation.

For the subset of days when none of these processes seemed to operate, the concentrations of cluster ions (0.9-1.9 nm) exhibited a clear seasonality, with high concentrations in the warm months and low concentrations in the cold.

2) Page 2 Line 4 and 5: The Authors use 2 time the expression "work better" which is very vague. Please rephrase using a more precise terminology.

The sentence is revised as follows

The former method seemed to have advantages in characterising NPF events with a fast GR, whereas the latter method is more suitable when the GR was slow.

3) Page 2 Line 7 and 8: "The ion production ... seemed a unique feature at Dome C ..." Please be more specific. A unique feature in Dome C relative to the whole world? Relative to the other data collected in Antarctica?

This refers to a feature that has not been reported for any other sites. The production of ions in the size range of 8-42 nm in relation to cloud formation therefore seems to be a unique feature at Dome C. The sentence is elaborated

The ion production in relation to cloud/fog formation in the size range of 8-42 nm seemed to be a unique feature at Dome C, which has not been reported elsewhere.

4**) Page 2 Line 10: "cleavage" replace with a word/rephrase using wording that is understandable to a wide audience.**

We replaced 'cleavage' with 'breakage'.

3) Page 2 Line 11 and 12 "our observation ... ice crystals." This is a very interesting result! The Referee thinks that it should be explained more thoroughly in the appropriate paragraph (see comment XX)

We believe that this result was presented and discussed in detail in section 3.3.2. Here in the abstract, we just provided a brief summary.

Introduction 4) Page 2 line 17: Please add the size/mobility ranges next to "primary ions" and "aerosol particles" for a more complete information for the reader that might be new to atmospheric ions.

Such information is added to the text.

Air ions, also known as atmospheric ions, are electric charge carriers in the atmosphere, ranging from primary ions (most likely have a mobility diameter smaller than 0.8-1 nm) to charged aerosol particles (with a mobility diameter up to several hundred nm).

5) Page 3 line 2:"Such charged nanoparticles ... are typically observed during new particle formation (NPF) events." Confusing. Stable air ions are observed also without NPF. Please rephrase.

The reviewer is right. This sentence is modified as
Charged nanoparticles in the mobility size range of 1.7-7 nm are typically observed during new particle formation (NPF) events.

6) Page 3 line 9 to 20: "Carslaw et al. (2013) ... years (Fiebig et al., 2014)." Confusing paragraph. Please add a closing sentence that explains why this work is important and how it is different from the cited previous work in this way the list of previous papers on the topic will make more sense to the reader.

Sorry for this confusion. This paragraph was meant to go together with the next paragraph. The information the reviewer looks for is actually contained in the following paragraph. These two paragraphs are combined into one in the revised manuscript.

Methods 7) Page 5 line 4 and 5. Please add the flow rate after "high sample flow rate", which is too vague to be useful. The Referee is aware that there is a flow rate some 20 lines below, but the use of "High" is not helpful and repetita iuvant.

The detailed flow rate information was given in the third paragraph of section 2.1.1. This first paragraph in section 2.1.1 was meant to serve an introductory purpose for section 2.1.1. However, since the review requested, we add the sample flow rate information as the reviewer suggested.

The AIS employs two cylindrical multi-channel aspiration-type analysers and a high sample flowrate (60 l/min).

8) Page 7 line 21 to 23: The Referee thinks that a plot of the shifted spectrum added to the supplementary material could be very useful to the readers and AIS/NAIS users. I recommend adding it.

Such a figure is added to the supplementary material.

9) Page 9 line 4: "growth and coagulation". Remove coagulation.

Coagulation can cause size increases of an aerosol population. Therefore, coagulation is not removed.

10) Page 9 line 10: "Air ion and total aerosol particle data are three dimensional: " Remove. It is confusing and unnecessary.

This sentence is removed as suggested.

11) Page 9 line 12 and 13: the Authors refer to " mode fitting method" and "appearance time method" throughout as "former" and "latter". Please spell them out each time. Readers will have easier times at understanding to which method the Authors are referring to.

These methods are spelt out as suggested.

12) Page 9 line 26: "assist". Confusing term. Please rephrase.

The word 'assist' is replaced by 'support'.

13) Page 3 line 3: "Sulphuric acid is considered as a key chemical species". remove "as".

The sentence is modified as

Sulphuric acid is a key chemical species in forming aerosol particles in the ambient air.

14) Page 11 line 14:"During the campaign period, there were nearly 300 days with valid air ion measurements". Please add what is the corresponding percentage of valid days as well. The Referee is aware that the Authors refer to Table 1 where this information can be retrieve, but thinks that would be of help to the reader to make it explicit in the text.

This information is given in the text.

During the campaign period (330 days in total), there were 287 days with valid air ion measurements, i.e. valid air ion data were collected on nearly 87% of the measurement days.

15) Page 11 line 18: "definite". Confusing adjective. Please rephrase.

The word 'definite' is replaced with 'certain'.

16) Page 12 Line 2: "high" please add a value e.g., median or a range of warm months to help the reader.

Typically, highest cluster ion concentrations were observed during warm months. Therefore, the sentence is modified as

The cluster ion concentration was the highest during the warm months, with a maximum in February.

17) Page 12 line 4: "natural ionising radiation" is a confusing term as cosmic rays are also natural and ionizing. Please change wording.

Natural ionizing radiation includes cosmic radiation. See the definition given by IAEA for example
https://www.iaea.org/newscenter/multimedia/photoessays/natural-and-artificial-ionizing-radiation

18) Page 12 line 24: remove "Markedly" it is unnecessary. Please reword the sentence to make it more understandable/intuitive. In addition, do the Authors have information about the boundary layer (BL) in February, March, October and November? Can the Authors exclude that some of those high cluster ion concentration are due to limited mixing due to shallow BL? Please elaborate and include in the text.

The word 'markedly' is removed as suggested and the sentence is modified.

The daily-median cluster ion concentration at Dome C was observed to be higher on NPF event days compared with event-free days.

Unfortunately, we have no information about the boundary layer height and we cannot exclude the shallow BL effect completely. However, the day length in February at Dome C is typically much longer compared with that in March and October. Therefore, a deeper mixed layer can be assumed in February than in March or October. However, the cluster ion concentration was found the highest in February. Therefore, even if the boundary layer height has an effect on the cluster ion concentration, this effect is likely to be very minor. A discussion about the boundary layer height effect is added in section 3.1 as follows

The development of the planetary boundary layer may additionally influence the concentration of cluster ions by imposing either a dilution or concentration effect. The longer day length in February than in March or October may result in the development of a deeper mixed layer, which could dilute the cluster ions within the mixing volume. However, the highest cluster ion concentration was found in February. Also polar nights would cause the formation of only a very shallow and stable boundary layer in winter months. The mixing volume in winter therefore is expected much smaller than in other seasons, but no concentration effect on cluster ion concentration can be identified. Consequently, even if the seasonal change of boundary layer heights has an influence on the seasonality in cluster ion concentrations, this effect is likely to be minor.

19) Page 13 Line 20: "Bumps ... NPF events (Fig. 3c)". Please rephrase being clearer. Consider replacing the word "bumps" with e.g., "sudden increase".

'bumps' is replaced with 'sudden increases' in the text as suggested by the reviewer.

20) Page 13 line 24 and 25: "Such differences result ... different origins". Replace "differences" with "different".

The sentence is modified as

Such differences result likely from the availability of vapours that sustain the growth.

21) Page 13 line 26: replace "perceptible" with "measurable"

The sentence is modified as

We could see slight concentration increases in the cluster ion size range at the time when NPF events were initiated, but…

22) Page 14 line 11: "Short after". Please be less vague, remove "short" and add temporal information in the text.

'Short after' is replace by 'About 4 hours after'

23) Page 14 line 18 to 21: "slowly growing" ... "slight growth". Please add next to this general expression the value of the GR. It will make the text more useful to the reader.

The growth rate and size range information is added in the text for clearer description. The wording 'slight' was a wrong interpretation. It is corrected in the revised version.

Over the consecutive five days on 12-16 February, a slowly-growing (GR ≈ 1.4 nm/h) population of 40-200 nm particles could be observed in the background, with their initial formation traceable back to 06:00 UTC on 12 February. Interestingly, apart from the particles initiated at 10 nm and 40 nm, a third mode of particles with sizes larger than 100 nm was recognisable on the morning of 12 February. This particle mode grew approximately from 100 nm to 300 nm during 12-13 February, and then gradually merged with the mode initiated at 40 nm at the end of 16 February.

24) Page 14 line 24: Replace "owing to" with "because of"

We are thankful for he reviewer's suggestion. However, we decide to keep 'owing to' as it is to avoid repeated usage of 'of'.

25) Page 16 line 19: "... and yields more representative instantaneous GRs". The word "representative" should be reserved to statistical analysis. If such analysis was done to assess whether the calculated GRs were statistically representative please add a sentence about the method used, otherwise rephrase.

We are afraid that we do not agree with the reviewer. In our opinion, the word 'representative' is not reserved to statistical analysis only. Here we made no statistical evaluation, but we decide to keep the word 'representative'.

26) Page 16 line 20: The Referee thinks that a comparison between the GR for ions with Dp>10 nm and particles in the same size range should be discussed in this para- graph.

We add discussion at the end of the last paragraph in section 3.2.2 as the reviewer suggested.

At large sizes in the overlapping size range (10-42 nm) of AIS and DMPS, the instantaneous GRs derived from the AIS measurements tended to be larger than those from the DMPS measurements. This difference may result from the fact that the DMPS measures total particles, including both ions and neutral particles, whereas the AIS detects only charged particles. Also the AIS measurements at sizes larger than 20 nm are subject to the uncertainties brought by the detection of multiply charged particles as singly charged particles. At small sizes in the overlapping size range, the instantaneous GRs derived from the DMPS exhibited a decreasing trend with increasing sizes, which however was not shown by the instantaneous GRs derived from the AIS. This difference may again be attributed to the difference in the sampled particles targeted by the two instruments.

27) Page 17 line 17: "... 110 nm". Based on the data analyzed in this work and on literature is the activation in this size range typical? The Referee recommends, if possible, adding some discussion about this in this paragraph.

As suggested by the referee, we modified this paragraph into the following form:

This observation is well in line with the activation thresholds from <50 nm up to about 200-300 nm for the "dry" particle diameter observed in real atmospheric clouds (see Henning et al., 2002, and references therein; Anttila et al., 2009; Kyrö et al., 2013; Portin et al., 2014; Leaitch et al., 2016). Clusters ions were efficiently lost onto the cloud droplets at Dome C.

28) Page 18 line 11: "we observed wind-induced ion formation especially during the winter months". Please add some numbers to make this statement less vague. How many times in winter with respect to other months?

The sentence is modified by taking in to account the reviewer's comment as

We observed wind-induced ion formation especially during the dark months (15 cases during May-August).

29) Page 18 line 24: " Ionising radiation produces primary ions via ionisation". Please either remove "via ionization" or add what is ionized e.g., "via ionization of vapor molecules"

'via ionisation' is removed according to the reviewer's suggestion.

30) Page 19 line 8: "However … contribute to the ion burst captured by the AIS". The Referee thinks that this statement is highly speculative and unsubstantiated. The Authors should discuss more in length adding references.

The discussion on this point was actually given in the following up text in this paragraph. In this paragraph, we proposed a mechanism that we think might be the cause to the ion formation during strong wind episodes, which needs further experimental validation as mentioned in the text. The text in the beginning of the paragraph is modified to better show the speculative nature of the whole paragraph.

Turbulent conditions might enhance the collection of electric charges by the shattered snowflakes and ice particles via a charge transfer from initial charge carriers, contributing to the formation of an ion burst. In addition, the shattered particles might gain electric charges through friction charging. However, we think that these two pathways of ion formation are not likely to contribute to the ion burst captured by the AIS. In principle,...

31) Page 19 line 22: "similar" the wording makes the sentence too generic and vague, please discuss further, how those feature are similar.

The sentence is modified as

By putting together all the 36 wind-induced ion formation events, a linear correlation was identified between the logarithm of the ion concentration and wind speed (Fig. 9), like also found at Aboa.

32) Page 20 line 3: "unexpectedly" unnecessary adjective, please remove.

The word 'unexpectedly' is removed as suggested by the reviewer.

33) All figures of DMPS and AIS size distributions: add units to dN/dlogDp make the units of Dp consistent (all nm or all m) add thick labels so that are consistent and at least 2 in number e.g., 10 and 100 nm

units are added to dN/dlogdp and dp are shown in nm and tick labels are added as reviewer suggested to all DMPS and AIS contour plots.

34) Page 31 caption of figure 1: specify the polarity of the ions, add units to the y-axis, uniform the units of Dp (all nm or all m) and add a tick label near the cluster band.

The polarity is specified and the unit is added to the y-axis label. Dp are shown in nm and a tick label is added in the contour plot to point on the cluster band. The modified Figure 1 and its captions are
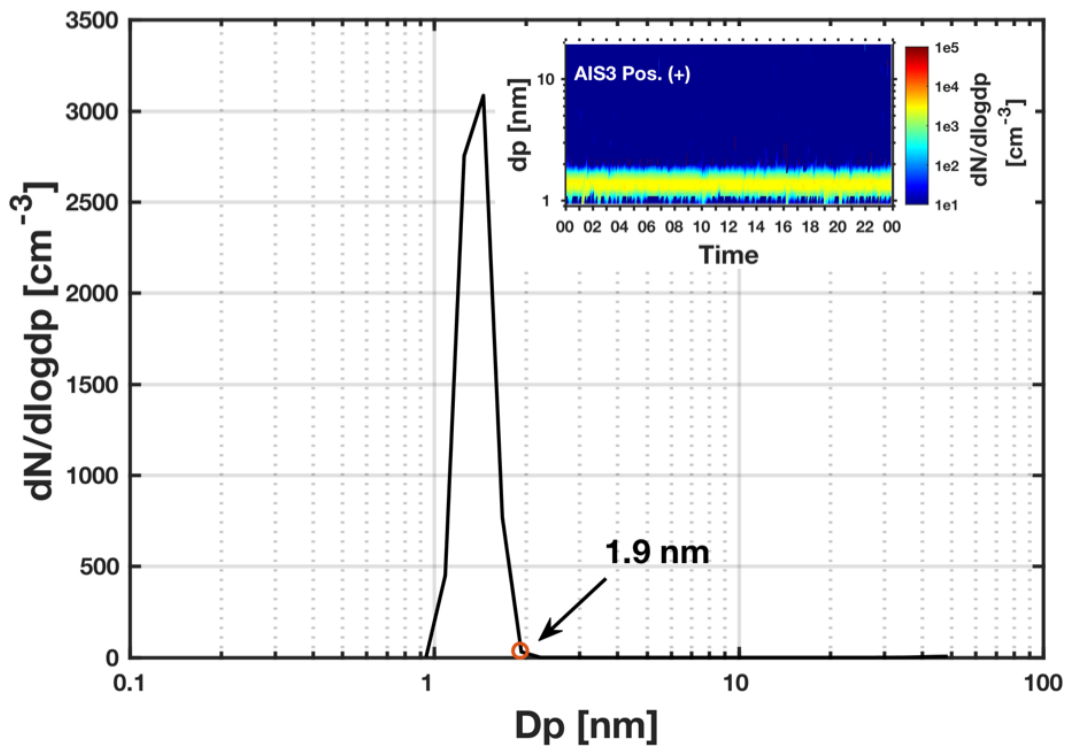

Figure 1. The median size distribution of positive ions measured by the AIS on an event-free day (16 January, 2011). The measured number size distribution of this day is shown in the contour plot.

35) Page 32 figure 2: the x-axis is and its label are confusing. Please make the x-axis so that have the same label and take the same space in this way the reader will be able to easily compare CS and cluster ions during the same month.
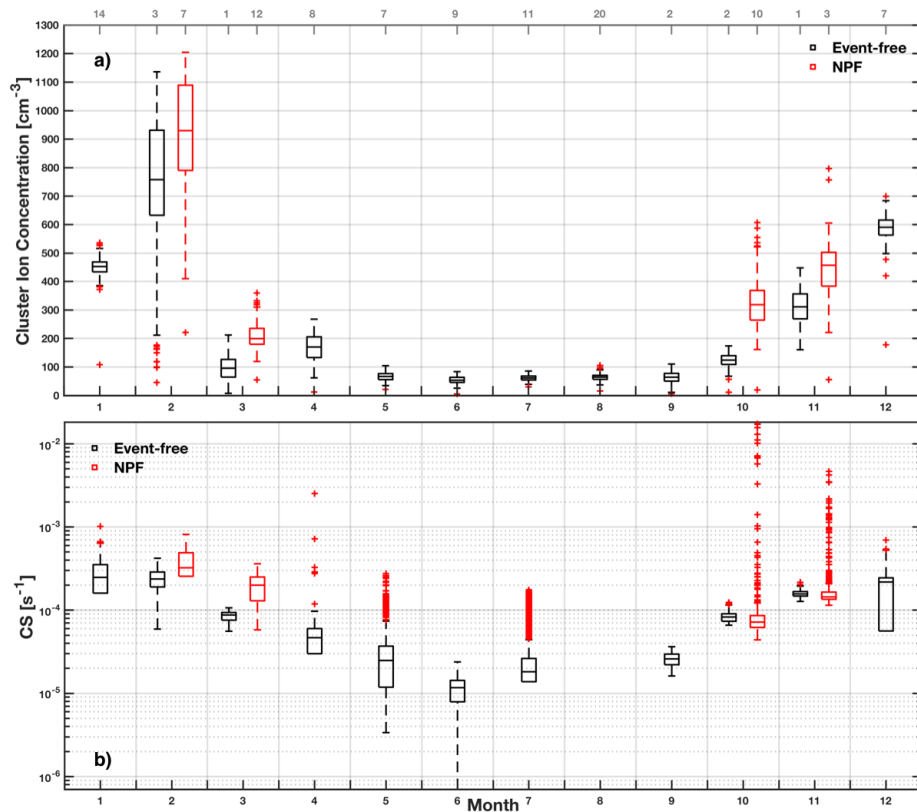
Figure 2 is fixed according to the reviewer's suggestion.

Figure 2. Seasonality in the median a) cluster ion (0.9-1.9 nm) concentration and b) condensation sink (CS). Tops and bottoms of the boxes are the 75th and 25th percentiles of the median daily values in 10 min time resolution, with bars in the middle showing the 50th percentiles. Whiskers represent spans of the interquartile ranges multiplied by 1.5. Cluster ion concentrations or CS on new particle formation (NPF) days shown in red and on event-free days in black. Event-free conditions were restricted to days, on which no NPF, cloud activation, wind-induced events or contamination as well as other anomalies altering the ion concentration in the cluster band. The numbers of days classified as either event-free or NPF are displayed on the top of the panel a) in grey colour. No CS was obtained in August due to the lack of measured temperature and pressure data from the station database.

36) Figure 3, all panels: adding a visual indicator for the fog would help the reader to identify the fog period. Please consider adding it.

No fog was observed during the time shown in Figure 3.

37) Figure 3, panel c): The secondary axis have a weird grey halo, please consider fixing it.
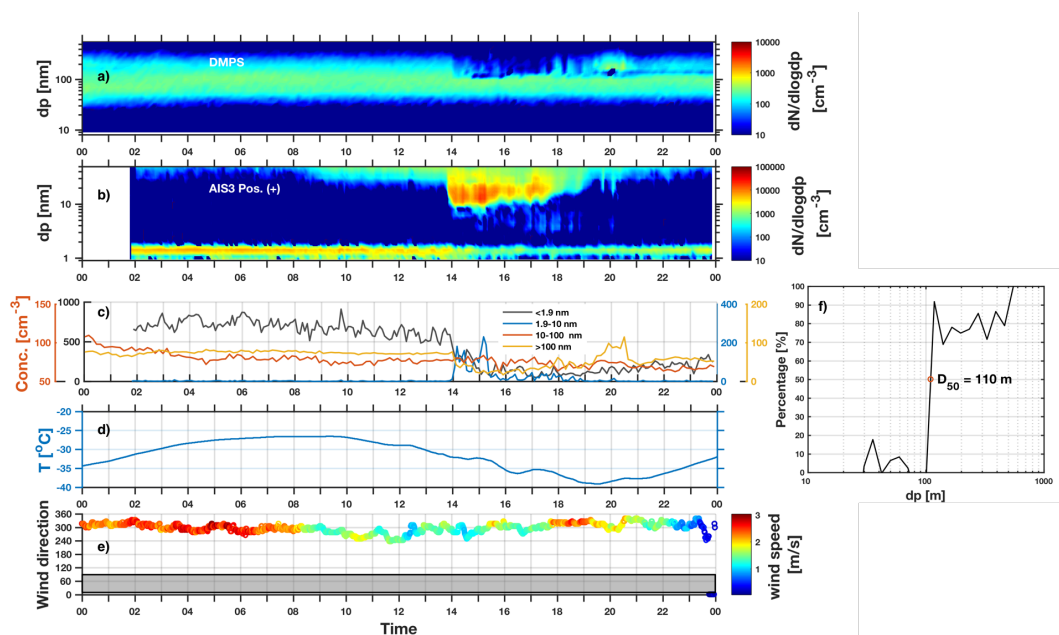
The grey halo is removed.

38) Figure 5, caption: "envolvement" pleases reword, this might not be English. Maybe "evolution"?

We used 'evolvement', which is an English word. However, it is changed to 'evolution' as the reviewer suggested.

39) Figure 7, panel f): The Referee thinks that adding a label "D50 = 1.1e-7 m", or even better "D50 = 110 nm" instead of the number alone would make the figure easier to read.

Figure 7f is modified according to the reviewer's suggestion.

40) Figure 9 and S3, all panels: please give only the significant digits for the fit.

The fitting parameters are rounded to 2 significant digits and are presented in Table S1 in the revised manuscript.

Table S1. Coefficients for the fittings shown in Figs. 9 and S4. R^2 is the coefficient of determination measuring the goodness of fit, which denotes the fraction of the total variation in the data can be explained by the fit. For Dome C data shown in Fig. 9, fits 1 and 2 are obtained based on all data below or above the wind speed threshold (7 m/s), respectively. The grey data points in Fig. 9 are used in determining the fitting coefficients for fits 3 and 4. For Aboa data shown in Fig. S4, a wind speed threshold of 17 m/s is used.

				Cluster (0.9-1.9 nm) ion concentrations vs. wind speeds		
		Fits	a	b	95% conference interval for a	95% conference interval for b	R^2
DOME C (Fig. 9)		1	0.69	26.34	[0.65 0.73]	[21.62 32.10]	0.24
		2	0.51	68.64	[0.41 0.60]	[29.88 157.67]	0.12
		3	0.73	21.83	[0.69 0.77]	[18.02 26.44]	0.28
		4	0.44	327	[0.40 0.47]	[244.95 436.53]	0.52
					1.9-10 nm ion concentrations vs. wind speeds		
		Fits	a	b	95% conference interval for a	95% conference interval for b	R^2
		1	1.14	0.07	[1.08 1.2]	[0.05 0.09]	0.29
		2	0.61	2.1	[0.71 0.88]	[0.88 5.01]	0.15
		3	1.19	0.05	[1.25 0.04]	[0.04 0.07]	0.31
		4	0.54	9.87	[0.58 7.18]	[7.18 13.58]	0.58
ABOA (Fig. S4)					0.9-2.2 nm ion concentrations vs. wind speeds		
		Fits	a	b	95% conference interval for a	95% conference interval for b	R^2
		6	0.17	14.98	[0.15 0.19]	[9.02 24 88]	0.66
					2.2-9.5 nm ion concentrations vs. wind speeds		
		Fits	a	b	95% conference interval for a	95% conference interval for b	R^2
		5	0.24	1.45	[0.22 0.26]	[1.26 1.68]	0.35
		6	0.06	63.24	[0.04 0.09]	[34.72 115.18]	0.17

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