Notes: Line numbers refer to the original document, and may have changed in formatting.

Comments from Referee #1

We thank the reviewer for their comments and recommendation of a revised manuscript. We have added additional text and figures to both the main text and supplementary material to define the derivation of terms such as $S_{c,D0}$ and to illustrate how the instrument uncertainties have been propagated.

The measurement 'cloud periods' and 'no cloud' periods have been adjusted throughout the figures for consistency. We have also developed the discussion of the figures to better improve their contextualisation in the paper.

Specific Comments

Abstract: *Because the AMS-derived organic:sulfate ratio is used for analyses and discussion repeatedly, the AMS measurement should be addressed more in the abstract*

Reference to the AMS measurements is now included in the abstract, which previously read:

"Aerosol physical, chemical and hygroscopic properties were measured in a range of airmasses during COPS (Convective and Orographically-induced Precipitation Study) ground-based in June and July of 2007 at the Hornisgrinde mountain site in the Black Forest, Southwest Germany. Hygroscopic growth factors at 86% relative humidity were measured and critical supersaturation simultaneously derived for particles of dry diameters 27 to 217 nm, both properties exhibiting substantial variability with time and with particle size."

and now reads:

"Aerosol physical, chemical and hygroscopic properties were measured in a range of airmasses during COPS (Convective and Orographically-induced Precipitation Study) ground-based in June and July of 2007 at the Hornisgrinde mountain site in the Black Forest, Southwest Germany. Non-refractory aerosol composition was measured with an Aerosol Mass Spectrometer, simultaneous to hygroscopic growth factors at 86% relative humidity and CCN activity measurements for particles of dry (< 20%) diameters 27 to 217 nm, with particle water uptake exhibiting substantial variability with time and with particle size.".

Experimental Methodology Section*: I do not find two references that seem be important to understand the methodology. Jones et al. (2010) is missing in the reference list. I could not find Irwin et al. (2010) although it is in the reference list.*

The study of Jones et al. (2010) is now in press and cited in the bibliography. Since the methodology does not rely on the detailed error propagation in Irwin et al. (2010) we have decided to make the paper more self-contained. We have therefore removed all references to Irwin et al. (2010) and included a description of the analysis method in the supplementary material.

Page 17077, line 24*: The definition of "dry" condition in this study should be provided.*

Definition of "dry" is now provided as < 20% RH

Page 17080, lines 1–4*: The definition and the calculation procedure of $S_{c,D0}$ is not clear, which should be provided more explicitly.*

A new section in the supplementary material has been added to better describe the calculation procedure for $S_{c,D0}$. The text has been changed from:

"Assuming that the transfer function is symmetrical about the selected dry mobility, the supersaturation at which 50% of the particles have activated into cloud droplets was directly interpreted as representing $S_{c,D0}$."

to

"Assuming that the DMA transfer function is symmetrical about the selected dry mobility, the supersaturation at which 50% of the particles have activated into cloud droplets for a given dry diameter, was directly interpreted as representing the critical supersaturation for activation, denoted $S_{c,D0}$ and derived by fitting a sigmoidal function to $F_A(S, D_0)$ vs S_{set}, as described in section 1.2 in the supplementary material, and hereafter referred to as S-step analysis.

Page 17080, line 17: *It seems that* $\kappa_{Sc,D0}$ *is the* κ *derived from* $S_{c,D0}$. *This definition of* $\kappa_{Sc,D0}$ *should be given more clearly.*

Section 2.3 has been largely rewritten to better describe the instrument operation and data analysis pathways. Page 17080, line 1 to Page 17080, line 17 has been rewritten, from:

"Assuming that the transfer function is symmetrical about the selected dry mobility, the supersaturation at which 50% of the particles have activated into cloud droplets was directly interpreted as representing $S_{c,D0}$. Variations in the hygroscopicity of particles of the same D_0 may lead to further spectral broadening, affecting the interpretation of $S_{c,D0}$. The impacts of such deviations from a perfectly internally-mixed particle population is discussed later. In addition to deriving $S_{c,D0}$, the diameter at which 50% of the particles activated into cloud droplets was derived at each supersaturation; $D_{50,S}$. The benefit of the $S_{c,D0}$ analysis is that the critical supersaturation for multiple dry diameters can be measured on an hourly basis. The $D_{50,S}$ analysis however typically has a smaller associated fitting error resulting from the increase in the number of data points used in each sigmoidal fit (~20 D_0 for each of the five supersaturations). The derivation of hygroscopicity parameter κ from $S_{c,D0}$ is described in Sect. 2.5 and an explanation of how measurement uncertainty is propagated through this derivation as standard errors is described by Irwin et al. (2010). The number of CCN that may potentially be activated as cloud droplets at any given size can be obtained from measured size distributions and either $\kappa_{Sc,D0}$ or $S_{c,D0}$ as described in Sect. 3.6."

to:

"Assuming that the DMA transfer function is symmetrical about the selected dry mobility, the supersaturation at which 50% of the particles have activated into cloud droplets for a given dry diameter, was directly interpreted as representing the critical supersaturation for activation, denoted $S_{c,D0}$ and derived by fitting a sigmoidal function to $F_A(S,D_0)$ vs S_{set}, as described in section 1.2 in the supplementary material, and hereafter referred to as S-step analysis. Variations in the hygroscopicity of particles of the same D_0 may lead to further spectral broadening, affecting the interpretation of $S_{c,D0}$. The impacts of such deviations from an internally-mixed particle population is discussed later. In addition to deriving particle critical supersaturation through S-step analysis, fitting the sigmoid function to a plot of $F_A(S,D_0)$ vs D_0 defines D-step analysis, from which the diameter at which 50% of the particles activated into cloud droplets was derived at each supersaturation; $D_{50,S}$. The sigmoidal function is identical for both S-Step and D-step analyses, described in section 1.2 in the supplementary material.

The DMA supplying the CCNc performed a full cycle of sequential steps of values of D_0 every 7 minutes, with a 3 minute settling period before each 7 minute sequence (to allow temperatures in the CCNc column to stabilise), resulting in 10 minute size distributions. Simultaneously, the CCNc would step through 5 different values of S_{set} every hour, remaining at each supersaturation for 10 minutes, with lowest supersaturation scan repeated twice at the beginning of each hour to allow for the temperature to stabilise down the column.

Therefore, every 10 minutes, a full cycle of D_0 steps were performed and every hour, a full cycle of S_{set} steps were performed.

The benefit of S-step analysis is that the critical supersaturation for multiple dry diameters is measured on an hourly basis. However, D-step analysis typically has a smaller associated fitting error resulting from the increase in the number of data points used in each sigmoidal fit (~20 D_0 for each of the five supersaturations; see Section 1.2 in the supplementary material). The single hygroscopicity parameter κ may be derived from either S-step or D-step analysis. However, the error associated with the calculation of κ cannot be propagated as the the κ-model cannot be solved numerically or analytically, and as such the uncertainty in κ is simply propagated through as a standard error from the sigmoidal fit, resulting in maximum and minimum values of κ, described in Section 2.5. The number of CCN (N_{CCN}) for a given dry diameter and supersaturation can be calculated from both S-step and D-step approaches and the aerosol number-size distributions as described in section 3.6."

Page 17080, lines 28–29*: It is not clear which variables were affected only negligibly by multiple charging.*

The number fraction of particles with multiple charges at $F_A(S,D_0)$= 0.5 were found to be negligible due to the shape of the distribution. This has been added to the text.

Page 17082, line 14: *The definition of κ(GFD0) is not clearly given. The parenthesis in κ(GFD0) does not seem to be necessary.*

$\kappa_{(GF,D0)}$ has been changed to κ_{GF} to improve readability. Added to the text is "κ_{GF} (i.e. κ derived from HTDMA measurements of hygroscopic Growth Factor)"

Results and analysis section: *The measurement period, and the cloud and non-cloud periods are not defined consistently throughout the paper. Fig. S2 is from 25 June, although the measurement seems to have started earlier according to the text (P17083, L15). While the authors explain that a distinct cloud event was observed on 30 June (P17083, L16-17), the decrease in RH seems to have been greater on 1 July. The boundaries of cloud and non-cloud periods in Figs. 1–3 are different from those defined in Fig. S2. Moreover, the end of CP2 in Fig. 2 is different from those in Figs. 1 and 3.*

It is recognised that there were previous inconsistencies in the period boundaries displayed on the figures. The cloud and non-cloud periods are now consistently marked in the figures, and further clarified in Table 1. CP defines periods where the particles themselves are sampled from within a cloud (ambient RH > 100%). NP defines periods where the aerosol sampled is not from within a cloud (i.e. from ambient RH and Grimm measurements), as cloud base is above the inlet. Furthermore, measurements from different instruments started at different times, but only measurements from specific time periods are considered in the analysis (i.e. when all instruments were operational).

Section 3.2*: Possible presence of refractory components, which are not detectable by the AMS, and its influence on the comparison of the AMS data to the hygroscopicity and the CCN activity should be addressed.*

Added the following; 'The AMS cannot detect refractory components and such there are no measurements of dust, black carbon or sea-salt. Variations in any of these components would alter particulate composition and thus hygroscopicity with less hygroscopic components such as dust and black carbon and more hygroscopic components such as sea-salt influencing particle growth factor and critical supersaturation for given dry diameters, though the AMS cannot be used to probe the magnitude of each component. Black carbon was measured by a Multi-Angle Absorption Photometer (MAAP), and analysis of the data showed no distinct correlation between the measured amounts of BC and aerosol hygroscopicity and derived products such as κ from HTDMA or CCNc as there was too little BC measured to affect the hygroscopic growth factor distrubutions. Dust and seasalt composition were not directly measured (though dust can be inferred from OPC measurements, aerosol hygroscopicity did not change greatly with the magnitude of the aerosol coarse mode and sea-salt is unlikely important in the COPS region). Aerosol composition during COPS is further discussed by Jones et al. (2010).'

Page 17083, lines 24–25*: This sentence is written as if activation of accumulation mode particles is a fact. However, it seems to be just an inference by the authors.*

"During cloud periods, accumulation mode particle concentrations were low, as the activated aerosol formed droplets larger than the inlet cyclone cut of 4μm."

has been changed to read:

"During cloud periods, accumulation mode particle concentrations were low. This would be expected because of the activation of particles larger than the threshold dry size for activation forming droplets larger than the inlet cyclone cut of 4 μm."

Page 17084, lines 12–14: *The mean organic:sulfate ratios in the cloud and non-cloud periods should be given, because the difference is not clear in Figure S2.*

The ratios for the different periods are now given in Table 1.

Page 17088, lines 10–17: *The model here seems to assume σ = 72.5 mNm−1. This point should be explained more clearly.*

"For dry sizes D_0=43 nm, 85 nm there is a clear tendency for the model to over-predict $S_{c(\kappa,GF_{D0,RH,mean,corr})}$"

Has been rephrased to:

"κ-Köhler evaluation of HTDMA growth factor and CCNc S-step data assuming a surface tension of water and a temperature of 298 K, shows that for D_0 = 43 nm and 85 nm there is a clear tendency for the model to over-predict $S_{c,GF}$, "

Page 17089, lines 20–24: Does this sentence mean that the variations in the hygroscopicity of organics and the surface tension were more important than the variation in the organic:sulfate ratios? More explanation may be necessary.

This sentence has been reworded, from: "Size-resolved AMS data (Fig. S3 in supplementary material) shows a markedly higher organic fraction at sizes greater than 100 nm in both periods of low and high organic fraction, possibly resulting in the lack of difference in water uptake between periods of high and low organic to sulphate ratio shown in Tables 2 and 3."

To:

"The total aerosol organic fraction is an average across all dry diameters, and the size-resolved AMS composition campaign average (Fig. S4 in supplementary material) show a markedly higher organic fraction at sizes greater than 100 nm in both periods of low and high organic fraction. This will most likely contribute to the lack of difference in aerosol hygroscopic growth factor during these periods (Tables 2 and 3), as the relative organic mass fraction at each dry diameter appears to influence their measured hygroscopicity and subsequently derived critical supersaturation, $S_{\kappa,GF}$ (Fig. 6).

Page 17089, lines 25–29*: Isn't there any possibility that the hygroscopic growth of particles was not in the equilibrium in the HTDMA, which resulted in the underestimation* *of $\kappa_{GFD0,RH,mean,corr}$?*

The reviewer is correct that our discussion of the potential for particles not reaching equilibrium could be enhanced and have added the following:

The aerosol sample airflow is dried through the use of a counterflow nafion drier, before entering both the CCNc and HTDMA. The RH of the aerosol sample flow after the drying process, will be between the RH before drying (ambient RH) and the RH of the counterflow in the drier (around RH = 2% from the compressed air supply). In the drying process, aerosol particles will lose water to the gaseous phase (and thus reduce in size) as the particle airflow attempts to reach equilibrium with the dry sheath air flow. In addition, any volatile or semi-volatile compounds can start to evaporate from the aqueous to the gaseous phase. The emerging aerosol sample flow is therefore a mix of relatively dry particulate matter and a gaseous mixture of water vapour and other semi-volatile compounds (inorganic and organic). Initially, this mixture will be further diluted through the first DMA, but after a few scans (circa 1 hr), the DMA sheath flow will have also reached equilibrium with the aerosol flow assuming the composition does not change in this time, with both containing similar amounts of components in the gaseous phase. In the HTDMA, after size-selection, the aerosol flow passes through the humidification system, which will grow the particle as water vapour and other gaseous components condense onto the particles. The second DMA (at the RH of the aerosol flow) then sequentially steps through voltages, determining the new particle diameters and thus the growth factor can be calculated. Since the initial drying procedure will not completely dry the particles, a small overestimation of D_0 will result, hence leading to an underestimation of the measured growth factor even if equilibrium is reached in the humidification section. This is in addition to any further underestimation in growth factor that would result from a perfectly dry aerosol not reaching equilibrium in the humidification section of the instrument (discussed further below).

For example, it has been shown that hygroscopic growth factor measurements of compounds containing nitrate are sensitive to instrument residence time (e.g. Gysel et al. 2007), and generally there was little nitrate seen during the COPS ground measurements. However, there were a few instances later on in the project when higher concentrations of NO_3^- were observed. The air mass trajectories for these events were different to the other periods (slower wind speeds and thus local influence from nearby valley), and a ZSR calculation showed that the HTDMA was most likely not seeing this NO_3^-. During COPS, the largest amounts of nitrate were measured on 19 July, incidentally a date marking the end of HTDMA measurements.

Further to the effects of evaporative equilibration in the particle pre-drying section, there are potential kinetic limitations to particle growth. After drying, any particles that are still in the aqueous phase may have different growth rates compared to those in a crystalline solid or glassy state, as a result of differences in the diffusion coefficient in the condensed phase. As the particle initially enters the humidification section, the high RH gaseous mixture within the sheath flow will start to condense onto the surface of the particle, which may not be exactly representative of the composition of the entire particle. As it takes longer for the molecules to diffuse through a solid particle compared to an aqueous one, any compositional influences of water uptake can encounter kinetic limitation to the rate of diffusion of the various compounds within the particle and of the diffusion of water into the particle. That is to say, that the particle will pass through the humidification system before all components of the particle in the solid phase are exposed to water molecules. Such limitation through increased viscosity and reduced diffusion in amorphous solid particles may be thought to be likely in the atmosphere following their discovery in chamber secondary organic aerosol systems (Virtanen et al., 2010).

Good et al. (2010b) showed how HTDMA humidification residence time could not be linked directly to aerosol hygroscopicity in multiple chamber experiments involving organic compounds. These finding may indicate that, if non-equilibrium behaviour is responsible for any variability, it would most likely be in the drying section for these particular aerosol systems.

It should be noted that similar drying effects and impacts of semi-volatile component evaporation and condensation can take place in the monodisperse CCNc configuration used here, providing further potential sources of discrepancies when reconciling HTDMA and CCNc data.

***Page 17090, lines 11–14**: As pointed out by a posted comment, the term "potential CDNC" is misleading and should be reworded.*

All references to potential CDNC were changed to N_{CCN} as per referee comments and comment from Ervens et al.

***Page 17090, line 15 – Page 17091, line 23:** The explanation in the paragraphs is difficult to follow because the definitions and the calculation procedures of the variables are not explained adequately. More detailed explanation should be added.*

We thank the reviewer for this comment and have rewritten this section to improve the clarity, with a more detailed explanation of definitions and calculation procedures. This section now reads as follows:

"The threshold dry diameter for activation were derived from CCNc measurement via three methods, two via S-step analysis and one from D-step analysis.
The threshold dry diameter for activation can be calculated from the S-step products $S_{c,D0}$ and κ_{Sc}. The first method does not require the hygroscopicity parameter and is found by plotting $S_{c,D0}$–S_{set} against D_0, defining the intercept (simply where $D_0 = S_{set}$) as the physical threshold dry diameter for activation. Connecting the two points either side of $S_{c,D0} - S_{set} = 0$ by a straight line, $D_{thres(Sc)}$ is determined, as shown in Fig. S5 in supplementary material. A clear benefit of this method is that fewer calculations are performed, thus not increasing the propagated errors further. However, a significant drawback is that this technique only works for interim supersaturations. Alternatively, the threshold diameter for activation can be derived from κ_{Sc} through the use of a κ lookup table at 1 nm and 0.01κ resolution. The κ value is then inserted into Eq. 3 and for a given S_{set} (effectively S_c), the threshold dry diameter for activation can be calculated, defined as $D_{thres(\kappa,Sc)}$. Additionally, using D-step analysis, the threshold dry diameter for activation has been derived by plotting $D_{50,S} - D_0$ vs D_0, where again, the intercept represents the physical threshold dry diameter for activation of the aerosol, denoted $D_{thres(D50)}$. The threshold dry diameter for activation can also be derived from κ_{GF}. As there are 7 dry diameters scanned by the HTDMA, each dry diameter has an associated $GF_{D0,RH,c}$ and thus κ. A threshold dry diameter is calculated for each κ_{GF} at each D_0 and S_{set}, and the intercept of $D_{thres} - D_0$ vs D_0 defines $D_{thres(\kappa,GF)}$ at each S_{set}.
The N_{CCN} calculation is most sensitive to the threshold diameter for activation around the peak of the aerosol size distribution, and is heavily dependent on the position and breadth of this peak. Out of cloud measurements were typically characterised by a monomodal distribution with a peak around 100 nm and with typical threshold dry activation diameters between 40 nm and 180 nm, the difference in N_{CCN} is affected greatly by this mode. Figure 7a shows the largest difference in single hygroscopicity parameter derived N_{CCN} occurs at the lowest supersaturation (0.11%). Even though the mean difference in the threshold dry diameter is 12% ($D_{thres(GF)}$ = 182 nm, $D_{thres(\kappa,Sc)}$ = 163 nm), the difference in N_{CCN} is 40%. For all supersaturations, the disagreement between the predictions of N_{CCN} is greater than 30%. In contrast, Figure 7b shows the smallest difference in N_{CCN} between $D_{thres(\kappa,GF)}$ and $D_{thres(D50,S)}$ at the lowest supersaturation. This is due to the increased proximity of $D_{thres(\kappa,GF)}$ = 182 nm, $D_{thres(D50,S)}$ = 184 nm. Excluding the measurement at the lowest supersaturation, the results follow a similar pattern as seen in Fig. 7. It can be seen that the discrepancies are usually outside the attributable errors and cannot be easily reconciled."

***Discussion section:** Possible reasons for the disagreements presented in Figs. 4, 5, and 7 (e.g., a possible presence of slightly-soluble materials) should be discussed more, in light of the simplifications assumed in the κ-Köhler model.*

The end of the discussion section has been rephrased, from:

"The lack of influence of the organic:sulphate ratio on particle hygroscopicity could suggest a predominantly internally mixed aerosol, or a main growth factor mode encapsulating a spectrum of compositions. An internally mixed aerosol will appear to be well represented by the mean growth factor, and a spectrum of compositions may appear as one broad mode. This would be lost within the CCNc data as the resolution of the instrument would not pick out these particles of varied composition. In theory, this broadens the ideal step function in activation space of the CCNc, yet with only 5 supersaturations for each D_0, the fitted sigmoid function is reasonably broad initially and a spread of compositions would not be resolved in this manner. It is also unlikely that a change in surface tension can explain the disagreement, as surfaces tensions as low as 50 mNm^{-1} are not expected."

to:

"As the AMS-derived composition is governed by the composition of relatively large particles, the fact that the hygroscopicity of relatively small particles (< 127 nm) did not correlate with the AMS-derived organic:sulphate ratio is not contradictory. Indeed, in the case of relatively large particles (> 127 nm), some relationship between the HTDMA growth factor data and organic:sulphate ratio was observed (Fig. 6, with HTDMA data represented by $S_{c,GF}$). It is worth noting that an internally mixed aerosol will appear to be well represented by the mean growth factor, and a spectrum of compositions may appear as one broad mode. This would be lost within the CCNc data as the resolution of the instrument would not pick out these particles of varied composition. In theory, this broadens the ideal step function in activation space of the CCNc, yet with only 5 supersaturations for each D_0, the fitted sigmoid function is reasonably broad initially and a spread of compositions would not be resolved in this manner.

The κ-Köhler model takes a snapshot of the aerosol behaviour at a specific RH, assuming the system to be in equilibrium, and extrapolates these properties to the maximum point in the Kohler curve. The application of the same κ assumes the same degree of non-ideality (same activity coefficient of water) at the point of sub-saturated measurement and the critical point of the Köhler curve. Departure from this and from the assumptions of equilibrium (no kinetic limitations to evaporation on drying or growth by re-condensation of material at high RH), will manifest themselves as disagreement in reconciliation studies. As previously discussed, the residence time of the sample aerosol on drying and humidification within the HTDMA will impact on measured particle growth factor. Similarly for the CCNc, the residence time of the sample aerosol in the drying prior to the DMA size selection may impact on the derived critical supersaturation. Though the role of surface-active compounds at the point of activation remains unclear, we have illustrated that an unrealistically low surface tension of 50 mNm_{-1} is required for HTDMA and CCNc measurements to agree. It is possible that kinetic limitations to condensation of water in humidification and evaporative departure from equilibrium in drying may substantially contribute to the disagreements shown in Figures 4, 5 and 7.

Page 17091, lines 25–27: *Is this statement based on the result in Fig. 7? If the degree of the agreement depends on the analytical methods for the CCNc data, how does the difference affect the discussion in this paper? For example, results in Fig. 5 is based on $\kappa_{Sc,D0}$, with which the closure of potential CDNC in Fig. 7 seems to show poorer agreement than those from other approaches.*

We thank the reviewer for raising this point, and have added a paragraph at the beginning of the discussion to better explain analysis methods and reconciliation:

"The degree of success of each reconciliation method is shown in Figure 7, represented by the slopes of the linear regression fits to the data. The degree of agreement is dependent on the analytical methods for CCNc data, with the *κ*-model showing the weakest overall agreement (most notably at lower supersaturations, as shown by Figure 5) compared to the better agreement shown for data analysed without invoking the κ-model. Though it is useful to calculate κ values from both CCNc and HTDMA data for comparison, the more direct analysis (i.e. D-step analysis) allows the threshold dry diameter for activation to be known directly from measurement and is therefore not subjected to model approximations in the same manner. Furthermore, as the threshold diameter for activation cannot be calculated from S-step analysis (i.e. $S_{c,D0}$) data for the highest and lowest supersaturations, the best overall reconciliation between HTDMA data and CCNc data, are for CCNc products derived via D-Step analysis, without the use of the κ-model (i.e. $D_{50,S}$), as shown by Figure 7. "

Page 17093, lines 1–6: *The point of the discussion in the two sentences is not very clear.*

Rephrased from:

"It is not clear that any atmospherically reasonable organic compounds at the concentration found at the point of activation can lead to the larger surface tension suppressions. Furthermore, one might assume that a clear relationship between organic:sulphate ratio and critical supersaturation would emerge if the suppression of surface tension greatly influenced the cloud activation behaviour."

to:

"It is not clear that any atmospherically reasonable organic compounds at the concentration found at the point of activation can be responsible for such large surface tension suppressions as required to reconcile the data. Furthermore, if the influences of organic compounds on surface tension were important, the critical supersaturation would be lowered with an increasing amount of organic matter, offsetting the increase of $S_{c,D0}$ expected from a reduction of inorganic matter, potentially leading to a cancellation of effects and no clear relationship.

Page 17093, lines 27–29: *I do not see the point of this explanation. Because the AMS-derived composition is governed by the composition of relatively large particles, the fact that the hygroscopicity of small particles (<127 nm) did not correlate with the AMS- derived organic:sulfate ratio is not contradictory. In the case of relatively large particles (≥127 nm), some relationship between the HTDMA data and the organic:sulfate ratio (Fig 6a) was seen.*

We have rephrased this paragraph to read 'As the AMS-derived composition is governed by the composition of relatively large particles, the fact that the hygroscopicity of relatively small particles (< 127 nm) did not correlate with the AMS-derived organic:sulphate ratio is not contradictory. Indeed, in the case of relatively large particles (> 127 nm), some relationship between the HTDMA growth factor data and organic:sulphate ratio was observed (as previously shown in Fig. 6, with HTDMA data represented by $S_{c,GF}$).'

Page 17095, line 25 – Page 17096, line 2: *I do not understand the explanation in the sentences. The variables mentioned here should be given more clearly.*

Reworded from:

"What is clear, is that the instrumentation shows the best agreement when the derived products are able to be measured wholly from either instrument, i.e. the best agreement between CCNc and HTDMA occurs when the range of supersaturations measured by the CCNc are such that they allow for the entire S_c range of a particular size. Should particles of a given diameter have critical super that cannot be derived by the CCNc but can be predicted by the HTDMA, results will surely diverge."

to:

"It is clear that the predicted critical supersaturation of the HTDMA ($S_{c,GF}$) shows best agreement with the derived critical supersaturation from the CCNc ($S_{c,D0}$) at a dry diameter range that activates mostly within the measured S_{set} range; a trend that follows for all derived products.".

***Figs. 1-3**: Some data seem to be omitted. What are the criteria of the data screening?*

Data are missing in Fig. 1 due to low number concentrations in the HTDMA instrument, and from 10/07/07 to 12/0/07 a calibration was performed on the HTDMA. Data are missing in Fig. 2 due to the sensitivity of $F_A(S,D_0)$ to low number concentrations. $F_A(S,D_0)$ data are screened for the range $0 \leq F_A \leq 1.4$ as activated fractions outside of this range indicate an issue with the instrument measurement. Fig. 3 is screened for critical supersaturations within the S_{set} range (0.11% – 0.80%) as it is possible for the sigmoidal function to predict $S_{c,D0}$ outside of the measurement range if the fit extrapolates beyond the data.

***Fig. 7:** The blue marker whose vertical axis value is about 20 nm in Figs. 7a and 7b is missing in Fig. 7c.*

The blue marker whose vertical axis value is 20 nm, is actually underneath a green data point in Fig. 7c; the thickness of the error bar has been increased.

***Fig. S2:** Why were the aerosol number concentrations very high on 27 June? Is there any possibility of the failure of the measurement?*

We thank the referee for bringing this to our attention. The high number concentrations from the DMPS during this period have been removed. This was a graphing error, as the data during this time corresponds to CCN and HTDMA calibrations. No data from this period were used in any analysis procedures.

Technical Corrections

***Page 17081, line 22**: "wasmeasured" → "was measured"*

changed from 'wasmeasured' to 'was measured'

***Page 17082, line 11:** "$GF_{D,RH}$ " → "$GF_{D0,RH}$ "*

Changed from '$GF_{D,RH}$' to '$GF_{D0,RH}$'

***Page 17087, line 27:** Should "20 nm ≤ D0 ≥ 200 nm" be changed to "20 nm ≤ D0 ≤ 200 nm?"*

Changed from '20 nm ≤ D_0 ≥ 200 nm' to '20 nm ≤ D_0 ≤ 200 nm'

***Page 17087, line 27:** "Fig. 4)" → "Fig. 4"*

Changed from 'Fig. 4)' to 'Fig. 4'

***Page 17095, line 21:** "instrument instrument" → "instrument"*

Changed from 'instrument instrument' to 'instrument'

***Tables 2 and 3:** The expression of κ with parenthesis in the suffix is different from the expresssion in the text.*

κ suffixes have been addressed throughout the manuscript.

***Fig. 1:** Because both the average growth factor and the upper end of the normalized counts are drawn in red, they are not distinguishable.*

The mean growth factor is now a dashed black line to aid the reader.

Fig. 1: *A space should be added before “nm.”*

A space has been added before nm.

Fig. 5: *A space should be added before “nm.” The letter k in the axis labels should be corrected to κ. The expression of the horizontal axis label (with parenthesis) is different from that in other parts of the manuscript.*

A space has been added before nm. The ‘k’ is now κ and the other expressions are now consistent.

Fig. 7: *The letter k in the axis labels should be corrected to κ. The suffix of κ (k) in Fig. 7b and that of S in Fig. 7c are different from those in the text.*

‘k’ is now ‘κ’. The suffixes have been changed for consistency.

Figs. S1-S3: *A space should be added before “m,” “m/s,” and “nm.”*

Spaces have been added before units.

Fig. S4: *The unit of “1.92” in the annotation should be added. Two alternate expressions, “$S_{c,D0}$” and “$S_{c(D0)}$,” seem to be used for the same* variable.

Suffixes fixed and unit of ’70.94 ± 1.92 nm’ added.