Author note: *The long delay in addressing the Referee comments and revising this manuscript was so that we could conduct further data quality assurance. Although the re-analysis of the data has had little or no effect on the conclusions, the QA is more robust and the revised text and figures reflect this re-analysis.*

Interactive comment on **"Size-resolved aerosol water uptake and cloud condensation nuclei measurements as measured above a Southeast Asian rainforest during OP3"** ***by*** **M. Irwin et al.**

Anonymous Referee #1

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The paper by M. Irwin et al. is reporting on simultaneous measurements of size distribution (SMPS), chemical composition (AMS), hygroscopicity (HTDMA), and size-selected CCN properties of aerosol in the tropical Malaysian rainforest. It is a very dense data set of good quality, which enables to evaluate the consistency of the measurements and give some information on the degree of our understanding of aerosol- cloud interactions. The amount of information available in this work is very large, and it is sometimes frustrating that little space is left for a more in-depth analysis. Many results are shown without comments and the overall impression is of a rather descriptive paper. There might be some space for some synthetic figures (average size distribution diurnal variations?). On the contrary, some figures might be redundant with others (see detailed comments). It would have been convenient if some measurements such as the aerosol size distribution and chemical composition had been published separately, as the goal of this paper is clearly orientated towards aerosol water uptake.

We thank the referee for the comment, and have included average size distributions as recommended. Aerosol size distribution and chemical composition measurements have been published separately by Robinson et al. (2011; see cited reference in the main text)), which included detailed back-trajectory analysis and PMF, and the in-canopy to above canopy comparison of Whitehead et al. (2010; also cited in main text).

Detailed comments:

Page 3123, line 1: SMPS steeped discretely: a DMPS? Line 6: each mobility diameter is spanning around 12s (not scan)? Line 15: generally particles produced by a nebulizer are overcharged Line 20: if the lower limit is 45 nm, then this value should be used throughout the paper (abstract included)

Page 3123, Line1: We thank the referee for the comment. Since an SMPS becomes a DMPS system when discretely stepped, we have changed the sentence from: "The SMPS was stepped discretely through dry diameters from 20 nm to 300 nm, referred to hereafter as D-step." to "The TSI 3080 SMPS was operated as a differential mobility particle sizer (DMPS), stepping discretely through dry diameters from 20 nm to 300 nm, referred to hereafter as D-step (diameter-step)." We have changed Line 6 in accordance with the referee comment from "with each mobility scan spanning around 12 s to increase data yield." to "with the DMA voltage held constant for *periods of 12 s to increase data yield". Line 15: As the aerosol was passed through a charge neutralizer, the overcharging of the nebulizer doesn't matter. We have rephrased this sentence to read "The shape of the charge-neutralized size distribution from the nebulizer is such that multiple charging probabilities are extremely low, and as consequence no charge correction was needed." Line 20: We have amended the lower limit to 45nm throughout the manuscript.*

Page 3125, line 2: The CE is dependant on different parameters including the aerosol chemical composition so I am not sure that relying on measurements performed in the laboratory with another instrument (?) is useful for assessing the present CE.

Providing the sample air is dried (which it was), chemical composition is the main factor in determining the CE of the instrument (see the cited reference, Matthew et al., 2008). As stated in the manuscript, this was validated by a comparison with the collocated DMPS data.

Page 3126, line 10: Are the authors still referring to supermicron particles? Line 12: collapsing of the BL below the measurement site would lead to a decrease of the number concentrations during the night. From line 12: I am not convinced that the description of the time series is usefull as it is if no interpretation is given, nor any attempt to synthesize the observations. Is there a typical diurnal variation? Does it differ in marine and terrestrial air masses? Which processes might be the cause? Any noticeable event disturbing the eventual regular diurnal variations? It is not easy to get these information from figure 1c.

Page 3126, line 10: Here, we refer to both the submicron and total particle number concentrations (above 0.01µm) as measured by a 3010 CPC, and as described by Whitehead et al., (2010). We have changed "Particle number concentrations were typically higher across the entire size distribution" to "Particle number concentrations were typically higher across the submicron size distribution overnight (1800hrs - 0600hrs local time). Additionally, total particle number concentrations above 0.01 µm, as measured by a TSI 3010 CPC at the top of the GAW tower, were also highest overnight, as shown in Fig. 3 by Whitehead et al. (2010).". For consistency with subsequent day/night segregation, local times of 0600-1800 are now used for daylight hours (more representative of "day" than 0300 – 1800 LT); we have changed the averaging criterion accordingly. Line 12: The diurnal variation in submicron aerosol number concentration is more pronounced for terrestrial air masses than for marine. Markedly higher particle concentrations are observed below 70nm at night from free tropospheric air masses of terrestrial origin, possibly from nucleation.

Page 3127, line 4: the part may better belong to the previous paragraph. This means that the submicron aerosol concentration varies opposite to the supermicron aerosol concentration (page 3126, line1à)? Any comment? Lines 24-26: the methodology for screening for external mixing has not appeared very clear to me. If the mean growth factor is close to the peak growth factor (within 5%), this indicates an internal mixing state, it might be clearer to mention that you want to exclude external mixing from the data set.

Line4: We have moved this description to the previous section. There was a *typographical error – line 6 "day" should read "night" in line with the previous comment (both sub and supermicron aerosol concentrations are higher at night). Lines 24-26: We thank the referee for raising this point. We have changed this sentence to now read "As the HTDMA and CCNc data cannot be compared on a diameter-for-diameter basis, the CCNc data were binned into 5 discrete bins (with √[D0.D1] and √[D1.D2] for the lower and upper bins respectively). In order to probe the effects of aerosol particle composition on hygroscopicity, the data was screened for external mixing (which we attributed to data where the mean growth factor deviated by more than 5% from the peak growth factor bin)."*

Page 3128, lines 6-12: even when selecting periods when the aerosol is found as an internal mixture in a given air mass type, its hygroscopicity can be quite variable through these selected measurement period. Could this variability explain the non monotonic behavior of the average activation fraction distribution? I suggest further selecting for constant average HGF periods: 80 -90 nm particles activating more readily than 100-120 nm particles might just be an averaging artefact (these particles might never coincide in time).

Page 3128, lines 6-12: After further segregating the data into average HGF periods (chosen by meanGF+2sigma), no further deviation was found. Furthermore, averaging the p(GF) for each dry diameter shows the two smallest dry diameters (32nm and 53nm) to be internally mixed, with large sizes comprising an ever-increasing bi-modal externally mixed p(GF). This trend is seen for both day and night, and marine and terrestrial sectors, to different degrees. This figure is now in the supplementary material.

Page 3130, line 7: a quantification of the number fraction of aerosols belonging to each hygroscopic class would give a more precise idea of the degree of external mixing in each size class and help with the discussions later in the paper. An increasing contribution of the hydrophobic mode particles with increasing particle size is rather unusual, no? lines 8-11: this sentence is hard to follow; conclusions might change according to results obtained from the previous remarks

Page 3130, Line 7: Though an increasing hydrophobic contribution with increasing particle size is unusual, perhaps this less hygroscopic mode is present at all particle dry diameters but only apparent once the particle dry diameter is such that the majority of particles have a higher growth factor, leaving behind this mode. The source of this mode is unknown as size resolved composition data were not available. However, the point that this sentence tries to explain is that the variation (and thus standard deviation) within the activation data is much larger than the difference between screened and unscreened data (i.e. unscreen and screened data are not so different). Lines 8-11: "The external mixing shown for dry diameters below 100 nm (32 nm and 53 nm) cannot be said to account for the departure from the typical monotonic fraction activated pattern shown by Fig. 3, as the variation in activation data is much greater than the difference between screened and unscreened data." has been changed to "The departure from monotonic trend for particles of dry diameters 32 nm and 53 nm (shown in Fig. 3) cannot be solely attributed to external mixing, as the variation in the activated fraction data is greater than the difference in this variation between internally mixed and externally mixed data."

Page 3131, line 16-18: again, the small diameters seem less externally mixed than the large diameter, so external mixing can not explain the variability in the activation fraction; however, a higher variability in the mean HGF itself might introduce a higher variability in the activated fraction?

We agree with the reviewer's comment. The sentence has been changed from: "The largest variability in critical supersaturation is shown to be at the smallest dry diameters, which may be attributed to the influences of an externally mixed aerosol." to "The critical supersaturation is most variable for the smallest dry diameters, where a higher variability in the mean hygroscopic growth factor could have introduced higher variability in the activated fraction and subsequently critical supersaturation, as at these dry diameters, the aerosol is mostly internally mixed (or comprises external mixtures of a narrow hygroscopic range)."

Page 3133, lines 15-20: from Figure 10, the same cocnlusions are given than from figure 9. Is there any use of the figure if the differences between marine and terrestrial conditions are not commented? Discussion on discrepancies: upon volatilization, the dry diameter might decrease, but the resulting HGF might either increase or decrease according to which type of compound has volatilized (NO3, Organics..). In the present data set, organics would be preferentially volatilized, and preferentially from the largest particles?

We have added the following to the discussions on the differences seen in κ: "Furthermore, any volatile or semi-volatile compounds can start to evaporate from the condensed phase to the gaseous phase. Therefore, the resultant aerosol sample flow is a mix of relatively dry particulate matter, water vapour and other semi-volatile compounds (inorganic and organic). After some time (circa 1 hr; and assuming the composition does not change with time), this aerosol sample flow will reach equilibrium with the sheath flow of the DMA, with similar amounts of components in the gaseous phase. Differences in both aerosol composition (from marine and terrestrial air masses) and instrumental sampling (differing rates and degrees of drying/wetting) could increase this incongruity."

Page 3135, The authors have now a very good knowledge and experience of reconciliation exercises (Good et al. 2010a, 2010b, Irwin et al. 2010 for the most recent). Here, the authors point different features than in previous studies. Any explanation?

It could stem from a difference in the organic composition, as we know from the AMS data (from Good et al. 2010a, 2010b, Irwin et al. 2010) that they're not the same. Further data points are required to be sure.

Page 3136, line 18-19: "possible external mixing": the degree of external mixing is exactly known from HTDMA measurements. Line 21-22 : this should have been quantified before ; again, quite unusual.

Page 3136, line 18-19: The reviewer is correct; the degree of external mixing is known from HTDMA measurements. There is an overall increase in the amount of external mixing for terrestrial periods over marine periods. Consequently, the observation has been changed from "possible external mixing" to "external mixing".

Line 21-22: This external mixing is now highlighted earlier in the text. The less hygroscopic mode around 1.1 in the p(GF) is most likely a regional non-hygroscopic mode (perhaps not local as it is present in each back-trajectory classification).

Page 3138, line2-6 : this has not been quantified neither before, by how much the Nccn differ one from the other using different instruments/methods ?

Page 3138, lines 2-6: The slopes and R^2*correlation are now given in Table 2. On Page 3135 line 19: we have changed "Fig. 11" to "Fig. 11c", and further expanded how* N_{CCN} *differs between the different instruments and methods, at line 27:*

"Data were fitted with an ODR line function, the results of which are shown in Table 2. Of the two different N_{CCN} *comparisons between instruments, the* $D_{(thres,\kappa,GF)}$ *vs* $D_{(thres,D50)}$ *method (shown in Fig. 11c) shows considerably less deviation from the 1:1 line, though with increased scatter (denoted by* R^2*), compared to data derived from* $D_{(thres,Sc)}$*.*