

***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 #2**

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**General comments:**

In the present paper, Irwin et al., have reported the simultaneous measurements of aerosol size distribution, chemical composition, hygroscopicity, and CCN properties of aerosols using SMPS, AMS, HTDMA, and size resolved CCN measurements. The measurements were carried out, marking the first study, in the tropical rainforest in Borneo, Malaysia. The data appear to be of high quality and of high relevance for atmospheric science studies and manuscript is within the scope of Atmospheric Chemistry and Physics (ACP). And I congratulate authors for this achievement. However, manuscript appears to be, in parts, haphazardly written as there are some open questions, which I believe, authors should try to address before manuscript is finally accepted for publication in ACP. I echo the concerns raised by Referee#1 in his opening comments.

**Specific comments:**

1 Introduction Page 3119, Line 18: I believe the cataloguing of Amazonian aerosols as almost “marine” is derived based on mostly the numbers similar to marine regime and not on activation properties as they are markedly different from each other. Authors may consider rewriting the statement.

*Page 3119, Line 18: We thank the referee for bringing this point to our attention, and have corrected the sentence to read “with CCN concentrations lower than would be expected for continental regimes.”*

Page 3121, Line 14: I am not sure if Shinozuka et al., 2009 performed the size resolved CCN measurements, kindly double check and Pringle et al., 2010 was a model study. Recently Rose et al., 2010, 2011 have also reported the size resolved CCN measurements, although, from megacity.

*Page 3121, Line 14: We have changed the reference list and the sentence now reads: “In order to better understand the role of particle size and composition on particle water uptake ability and thus the CCN potential, size-resolved ambient measurements are*

*becoming increasingly common, though the relative importance of the aerosol number-size distribution and chemical composition is a topic of much debate (e.g., Wang et al., 2008; Good et al., 2010b; Dusek et al., 2010; Ervens et al., 2010; Jurányi et al., 2010; Rose et al., 2010a, 2011; Wex et al., 2010)."*

## 2 Methodologies

### 2.2 CCN measurements

This particular section is very important but appears little difficult to follow especially with the switching between how  $S_c$  and  $D_0$  were derived and used (as to derive a parameter one need to be kept constant and scan other one, is this the way it was carried out?). I did not find the values of the flows for the instruments used during campaign in the manuscript and same is with the values of set supersaturations in DMT CCNC. Authors have given standard errors of supersaturations derived from the fits but did not talk about errors associated with the model choice, as according to Rose et al., 2008 that may introduce errors up to >20% for lower supersaturations. Kindly comment.

*$S_c$  and  $D_{50}$  are merely products from different approaches to the analysis of identical data. DMA voltage is discretely increased every 12 seconds (and then reset) for each stepwise increase of supersaturation. In both analyses a sigmoid is fitted to the activated fraction vs either supersaturation or dry diameter, deriving  $S_c$  and  $D_{50}$  respectively. We have removed the use of  $s$ -step and  $d$ -step from the first paragraph on page 3123, as they are only analysis steps and not measurement procedures. We then introduce  $S$ -step and  $D$ -step on Page 3123 line 26: "Assuming the system to have a symmetrical transfer function, the data were fitted with an error-weighted sigmoidal function and the temperature gradient at which  $F_{A(S,D_0)} = 50\%$  was determined, and interpreted as the critical supersaturation if plotted against supersaturation setpoints ( $S_{set}$ ) or as the threshold dry diameter for activation,  $D_{50,S}$ , if plotted against  $D_0$  (hereafter referred to as  $S$ -step and  $D$ -step analysis respectively, explained in Sect. 1 of the Supplement)."*

*We are setting a temperature gradient in the instrument and have merely not adjusted the front-panel read out to the corrected values. We have changed page 3123 line 7 to: "The CCN counter was stepped through five different column temperature gradients ( $\Delta T$ ) of 2.56, 3.62, 6.52, 8.24 and 12.2 °C, calibrated to 0.11%, 0.18%, 0.37%, 0.48% and 0.73% as described below, repeatedly at 10 minute intervals."*

*The sample flow of both the CCNc and CPC instruments was  $0.490 \pm 0.005$  Lpm. The sample flow of the HTDMA was  $0.5 \pm 0.05$  Lpm. The DMA before the CCNc was operated with a sheath to aerosol ratio of 10:1 (at flow rates of 9:0.9 LPM). These flow rates have been added to the manuscript. In order to apply the data from the salt calibrations to the instrument supersaturations, activation models for inorganic model must be used. The Pitzer-Simonson-Clegg mole fraction based ion interaction model at the core of ADDEM has been clearly demonstrated as being extremely accurate across the mole fraction range at the point of activation (well within instrument precision); see Topping et al. (2005) and Rissman et al. (2008).*

## 2.4 Other measurements

This section is explicitly discussing about AMS measurements then why to call it other measurements and why not as AMS, or mass spectrometry measurements something like that? I do repeat Referee#1 comment about CE, unless authors compared submicron volume time series from the DMPS measurements “measured” during campaign. If that is the cases please say it explicitly that it was compared with DMPS measurements carried out in field.

*This section has been renamed “Measurements of Aerosol Composition”. Further to the response given to Referee #1, we have now explicitly confirmed in the text that the AMS was compared directly to the DMPS measurements carried out in the field.*

2.5 Air mass classification Page 3125, Line 17: Did authors mean 850 hPa instead 950 hPa????

*Page 3125, Line 17: We mean 950 hPa; as described by Robinson et al. (2011; please see citation in main text).*

## Results 3.1 Aerosol composition and size distribution

Is it possible for authors to mention the local season when the measurements were carried out? It would be meaningful to show the average number size distribution of aerosols (diameter vs. concentration) along with what is show in Fig. 1c, at least in supplement. For the complete result section I recall the concern that some figures and associated Tab. 1 appears to be redundant and need to be modified. In addition it is not clear what is the reason behind the diurnal variation is? Does it mean that the aerosol properties were dominated by some local sources and not by wind change as evident from back trajectory analysis? Please comment.

*We have added “The measurements were carried out during Borneo's dry season (the Sabah region has a dry season from April to October), though year-round rainfall varies only slightly.” to the text at page. We are unsure what the referee means regarding Figures and Table 1. The diurnal variation is most likely due to the contraction of the boundary layer at night, exposing the site to the free troposphere, or at least higher in the boundary layer. Furthermore, nocturnal fogs would influence aerosol properties.*

## 3.4 Aerosol sub-saturated water uptake

Page 3130, Line18: With refereeing to Figs. 6a, 2, and 1c authors claim that number distribution does not influence the water uptake behaviour substantially. Did authors mean number – size distribution or only numbers? What is water uptake behaviour; did authors mean GF or kappa? Please be specific as water uptake does not have any definition. I have major concern about the claim here: First of all Fig. 6a humidogram is from 26th June to 27th June where as on page 3122, line 6 authors explicitly mentioned that results during 3rd to 20th July are discussed, I am confused here and secondly Fig. 2 and 1c do not cover the data period shown in Fig. 6a humidogram (i.e., 26th to 27th June). Hence by comparing two different “regime” time data sets I am not convinced

with this claim. Kindly address or take out the sentence, mainly because authors not only do not have CCN data but also any other data except HTDMA (humidogram).

*Page 3130, Line 18: We mean that the number of aerosol particles in each size bin, i.e. the number-size distribution, does not influence the hygroscopic behaviour substantially. By “water uptake behavior” we mean “hygroscopic behavior”, though this paragraph has been removed as follows: We have removed the first humidogram as only HTDMA and DMPS data exist for this time period and as such, we have removed the corresponding paragraph and changed the paragraph on page 3131 line 1 to:*

*An aerosol humidogram was performed between 5 July and 6 July; a terrestrial period with higher variability in the DMPS number-size distribution for the duration of the humidogram (see inset). It should be noted that no measurements were possible below 45% RH. The majority of the data from the humidogram corresponds to the morning and early afternoon of 6 July, which is characterised by low particle concentrations above 60nm, but high concentrations between 20nm and 60nm in the late morning. The humidogram was started at 90% RH and brought down to 45 % RH, whence the RH was increased again up to 85%. Contrasting the efflorescence and deliquescence branches of the humidogram (denoted by solid and dashed lines respectively), reveals the late-afternoon/evening aerosol particles below 258 nm to be more hygroscopic at the same RH, than particles of the same dry diameter the following morning. This could be due to the increasing ambient RH as the boundary layer (and moisture) lifts up the side of the valley, scavenging the most hygroscopic particles. The lack of a well-defined deliquescence point is consistent with aerosol comprising multiple components able to attract liquid water to the particles below the deliquescence point of commonly expected inorganic salts (Marcolli et al., 2004). The two smallest sizes (32 nm and 53 nm) do not achieve the same high growth factor of 1.4–1.5 seen by the larger particles; consistent with activation data (Figs. 2 and 3).*

### 3.5 and 3.6 Hygroscopicity

I am partly not convinced and sure with the reason behind the incongruities in the kappa values of kappa\_sc and kappa\_gf. I request authors to please mention this incongruity explicitly in the abstract and may be elaborate what do authors mean by “instrumental difference” so as to support that other data is not affected.

*We have changed a sentence in the abstract from: “The derived hygroscopicity parameter  $\kappa$  ranged from between 0.05–0.37 for the supersaturation range 0.11–0.73% compared to those between 0.17–0.37 for measurements performed at a relative humidity of 90%.”*

*to:*

*“The derived hygroscopicity parameter  $\kappa$  ranged from between 0.05–0.37 for the supersaturation range 0.11–0.73%, inconsistent with measurements performed at a relative humidity of 90%, with derived  $\kappa$  between 0.17–0.37.”*

*By “instrumental difference”, we mean the differences in the aerosol particles measured in each device; i.e. differences in RH in drying or humidification sections will change by different amounts the equilibrium points for each particle being sampled. In the context of this study, it could mean that the particles comprise different amounts of organic material whether in the CCN or HTDMA. This has now been addressed in the text, as a response to reviewer 1.*

**References:**

*Rissman, T. A., V. Varutbangkul, J. D. Surratt, D. O. Topping, G. McFiggans, R. C. Flagan and J. H. Seinfeld, Cloud condensation nucleus (CCN) behavior of organic aerosol particles generated by atomization of water and methanol solutions, Atmos. Chem. Phys., 7, 2949-2971, 2007*

*Topping, D. O., G. B. McFiggans and H. Coe, A curved multi-component aerosol hygroscopicity model framework: Part 1 – Inorganic compounds, Atmos. Chem. Phys., 5, 1205-1222, 2005*