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Atmospheric Chemistry and Physics Discussions

Interactive comment on "Measured and modelled Cloud Condensation Nuclei (CCN) concentration in São Paulo, Brazil: the importance of aerosol size-resolved chemical composition on CCN concentration prediction" *by* G. P. Almeida et al.

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Interactive comment on "Measured and modelled Cloud Condensation Nuclei (CCN) concentration in São Paulo, Brazil: the importance of aerosol size-resolved chemical composition on CCN concentration prediction" by G. P. Almeida et al.

Anonymous Referee #1

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Major

This manuscript, as I see it, sets out to discuss two points; one of which is done better than the other. Firstly, the manuscript addresses measured and modelled CCN concentrations in Sao Paulo, Brazil. Secondly, the manuscript (as titled) stresses the importance of size-resolved chemical composition on the prediction of CCN concentrations. The first aspect is addressed quite nicely, and it no doubt of value to ACP and the scientific community at large. The second aspect is demonstrated neatly through the use of size-resolved chemical composition data, but I feel that a slightly different approach would have been more valuable, for the following reasons.

As stated in the manuscript a particle's CCN affinity is dependent on its size and composition. It is therefore a shame that the CCN measurements were not performed downstream of a DMA. It can be argued that size-resolved measurements are the best way of trying to measure the impact of a changing aerosol size distribution on CCN activity. Indeed, the authors acknowledge that size-resolved measurements of particle constituent matter via AMS measurements are beneficial in improving CCN closure studies. Though more complex, size resolved measurements of CCN activity will reveal the activated fraction for a given dry diameter - paired with size-resolved chemical composition; a valuable tool. Though it is widely accepted that chemical composition plays a larger role than particle size, this is only true within certain limits. These limits are best probed with size-resolved measurements of CCN activity. It is my hope that the authors consider this in future works. Some discussion related to this issue would be useful. Larger assumptions and consequently, uncertainties, are associated with the type of bulk measurements presented in this study (though no instrumental errors are propagated nor discussed in detail).

We acknowledged that measuring CCN activity downstream a DMA allows a more complete understanding on the role of chemical composition. Unfortunately, instrumental limitation at the time of the campaign prevented such setup to be deployed. Such explanation has been included in the manuscript (3rd paragraph of section 2.1):

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"Although originally planned, CCN measurements were not carried out downstream of a DMA due to instrumental issues. Such setup provides the activation fraction for a given aerosol dry diameter, allowing to better assess the role of the chemical composition." "

Though size-resolved CCN measurements should possibly have been used, the data seems to be of high quality, and it reasonably presented.

Yes, indeed extensive care has been taken to assure that the scientific questions were properly addressed, collecting data of the highest quality. The English throughout the paper needs to be carefully checked, as the numerous mistakes disrupted the flow from the otherwise nicely set-out paper. I have noted a few (not all) below in the "Misc" section.

We acknowledge the reviewer's comment and have thoroughly revised the text. Portions of the text were modified as well.

Minor

Page 32356, Line 26; What is "relative accuracy" in this context? It would be useful to include examples of studies where CCN concentrations were "not" predicted to within instrumental errors, or "relative accuracy", to give the reader some perspective on this difficult measurement and assumptions therein.

We decided to remove the expression "relative accuracy" and substitute it by a more appropriate denomination, "achievement of aerosol/ccn closure". We also followed reviewer suggestion and included examples of studies where CCN concentration were not predicted to within instrumental errors. The text now appears as: "Nevertheless, aerosol/CCN closure has been achieved assuming simplified composition and an internal mixture in some studies (e.g. Liu et al., 1996; Cantrell et al., 2001; Roberts et al., 2002; VanReken et al., 2003, Rissler et al., 2004; Conant et al., 2004; Gasparini et al., 2006; Broekhuizen et al., 2006; Ervens et al., 2007; Chang et al., 2007; Wang

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et al. 2008, Gunthe et al., 2009; Shinozuka et al., 2009), while previous studies were largely unsuccessful (Bigg, 1986; Quinn et al., 1993; and Martin et al., 1994). Given the complex. . .”

Page 32359, Line 25; It’s confusing to use S and then SS for supersaturation in consecutive sentences. Being as it is always the supersaturation with respect to water vapour that is considered, I recommend choosing one and then remain consistent in the text.

Following the reviewer, we adopted the usage of SS for supersaturation throughout the text.

Page 32360, Line 1; Not all particles are activated within the column, and not all will grow to supermicron sizes. This is even confirmed in the following sentence, whereby it is stated that the CCN counter identifies particles larger than 0.75μm in diameter as being CCN. Consider revising this sentence for consistency.

We believe the reviewer misunderstood the sentence, probably because our text was not clear enough. It was said: "The particles having lower critical supersaturation (SScrit) than the SS in the column are activated and grow into the supermicron size-range. Particles leaving the column are sized by an optical particle counter (OPC) and counted as CCN IF their diameter is larger than a threshold size of 0.75 μm.". Although we did not identify the mistake the reviewer is advising, we modified the sentence and now we can read: "Only particles having lower critical supersaturation (SScrit) than the SS in the column are activated and can grow into the supermicron size-range. Droplets leaving the column are sized by an optical particle counter (OPC) and counted as CCN if their diameter is larger than a threshold size of 0.75 μm." We believe that the small modification we included is enough to avoid any doubt on the sentence.

Page 32360, Line 5; ratio should be written as 10:1

Done

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Page 32360, Line 6; I do not fully understand what “Data reported here include only under supersaturation steady state" exactly means. Furthermore, after changing supersaturation (e.g. from 1

It seems that something is missing at the end at the comment. We modified the sentence and now we can read: "...we evaluated only the data produced after completely adjustment to the supersaturation level." We believe that with this modification we makes clear what we mean.

Page 32360, Line7; I have concerns regarding “”the factory calibration. . . was used”. How was the instrument verified to be working correctly? In general, it is good scientific practice to perform one’s own calibrations, especially in the field under varying conditions. A calibration using inorganics before and after the project would have been optimal, and would have verified the instrument performance.

We acknowledge that it is good scientific practice to perform one’s own calibrations, especially in the field under varying conditions. However, technical limitation prevents that. We included this comment on the text. “To determine that the instrument was working correctly we considered the temperatures presented by the instrument, the variation in CCN concentration according to the related supersaturation, the amount of mass determined by ACSM, and the DMPS aerosol spectra. In our analysis those factors were good enough for ensure the reliability on the presented data.”

Page 32360, Line 20; In what sense were the DMPS data “corrected”? Is this “corrected” data used for the subsequent integrated number calculations? (the latter is not clear)

Yes, integrated number concentrations were corrected according to a CPC running in parallel. The text has been changed as such: “Based on such intercomparison, DMPS integrated number concentration has been corrected by a factor of 1.12”

Page 32364, Line 23; Only “Köhler, 1936” should be used here. The authors later

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present the simplified Köhler equation (eq. 1), which was described by McFiggans et al., 2006.

We followed reviewer suggestion

Page 32365, Line 6; It should be stated that this is the surface tension of water.

We followed the reviewer suggestion and now the text reads: “considered constant and equals to the surface tension of water on this study, 0.072 J.m-2”

Misc

Page 32355, Line 9; “internal mixture” should be “internal mixtures” or “internal mixing”

Done

Page 32355, Line 13; do the authors mean “with a dominant nucleation mode”?

Yes, we mean it. The reviewer suggestion was adopted.

Page 32355, Line 21; I think it would be helpful to say in what regard (i.e. compared to what) the CCN results were overpredicted (e.g. from Köhler model)

We followed the reviewer suggestion and now we can read: “Considering the bulk composition alone, observed CCN concentrations were substantially overpredicted when compared with Köhler theory”

Page 32356, Line 2; “on a site” should be “at a site”

Done

Page 32356, Line 5; “enables” should be “enable”

Done

Page 32356, Line 8; delete “distribution”

Done

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Page 32356, Line 13; “its sizes” should be “their size”

Done

Page 32357, Line 9; “an internally” should be “internally”

Done

Page 32357, Line 10; “the identical” should be “an identical”

Done

Page 32357, Line 21; there should be a comma after “processing”

Done

Page 32359, Line 19; following section? following text?

We adopt following section.

Page 32366, Line 13; “Initially, the κ size independent solubility value” would read better as “Initially, the size-independent solubility value, κ,”

We adopted the reviewer suggestion and now we can read: “Initially, the size-independent solubility value, κsi, is determined using the values of Dusek et al. (2010)...”

Page 32367, Line 9; “for the assumptions of size averaged” is not clear

We modified the paragraph and now we can read: “Considering the assumptions of size-averaged chemical composition, particles smaller than 40 nm do not affect the calculated CCN number concentration because D0,crit at the 1.0% supersaturation was always above 40 nm.” Page 32370, Line 8; “chemistry composition” should be “chemical composition”

Done

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