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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 #2

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The paper by Almeida et al. presents CCN concentrations measured in the Sao Paulo Metropolitan Area in Brazil. The measurements were compared with chemical composition data from an aerosol chemical ionization monitor leading to the conclusion that a size-resolved chemical composition is necessary to predict CCN concentrations reasonably well. The information given in this manuscript is relevant for the readers of ACP and of interest for the community in general. However, there are still quite some issues as detailed below that have to be addressed by the authors before I can recommend the paper for publication in ACP.

Major comments: 1. Calibration of the CCNC

You mention that "the factory calibration based on (NH4)2SO4 was used ("P. 32360, L. 7-8). However, DMT recommends calibrating the instrument's supersaturation regularly. And indeed, as shown by Rose et al. (2008) it is necessary to carefully calibrate the supersaturation in the CCNC, since the relation between the supersaturation and temperature gradient along the flow tube strongly depends on the operating conditions (air pressure, air temperature, flow rate). The factory calibration was performed in Boulder at about 1700 m asl. As your measurements were performed in Sao Paulo at 700 m asl there is about 100 hPa difference in air pressure between the two operating conditions. As demonstrated in Fig. 9 of Rose et al. (2008) an increase in pressure by 100 hPa leads to a relative increase in supersaturation of at least 10% (the increase is larger at small supersaturations). Taking this into account the actual supersaturation levels during your campaign were rather something like 0.22%, 0.44%, 0.66%, 0.88%, and 1.1%. Since the CCN concentration strongly depends on the supersaturation, this is an important fact if you want to compare your results with other measurements.

In the revised version of your manuscript you would definitely need to discuss this point. You should provide values of supersaturation that are corrected for differences between operating conditions during the campaign and during factory calibration. Maybe it is even possible to still calibrate your instrument under conditions similar as during the campaign.

Note that changing supersaturation values have a significant influence on the prediction of CCN concentrations. I assume that with supersaturation levels being actually higher you result in a smaller critical diameter of CCN activation, which would in turn yield higher CCN number concentrations. How much higher depends of course on the particle size distribution.

We address the issue and included two new paragraphs on the description of the CCNC. They read: “For this dataset, factory calibration using (NH4)2SO4 was applied, considering recommended corrections from recent literature. Lance et al. (2006) has provided the correction function in supersaturation for a given ambient pressure. Taking in account that the system was originally calibrated in Boulder, Colorado (820 mbar) and deployed in São Paulo (928 mbar), the correction factor is roughly 13%. As such, the supersaturation levels measurements during our campaign were estimated 0.23%, 0.45%, 0.68%, 0.90%, and 1.13%.”

Considering these two paragraph we made modifications on the text to adequate the values of concentrations to the new values of supersaturation used during observation. The activated fraction was corrected considering the actual values of supersaturation. The observed and modeled concentrations were compared considering 0.2%, 0.4%, 0.6%, 0.8% and 1.0% supersaturations. To obtain CCN concentration at these levels, we interpolated the observed CCN concentration between the actual supersaturations.

2. Relevance of measured mass composition for the size range of CCN measurements

As you point out correctly in Sect. 3.2.1 the mass of particles with larger size contributes much more to the bulk chemical mass than the mass of particles with smaller size. Therefore the question rises whether the bulk mass you measured with ACSM is at all relevant for the size range in which the particle activation happens. I wonder if there is a correlation between your CCN data and the bulk particle composition data. I would appreciate if you could test the correlation between kappa_ACSM and kappa_CCN, where kappa_ACSM = f_org * kappa_org + f_inor*kappa_inor.

kappa_CCN can be calculated by first looking for the diameter above which the integrated DMPS measured size distribution equals the measured CCN concentration, and secondly calculating kappa from your Eq. 3 inserting the supersaturation measured with CCNC and the just calculated diameter. Such a correlation has been made e.g. in Juranyi et al. 2010. From my experience this correlation is generally not very good but improves for decreasing supersaturation. Nevertheless, it may help you in the interpretation of your predicted CCN concentrations.

Just recently we received the information about the ion chromatography and, we are currently developing a study to determine the kappa variation as a function of the size for Sao Paulo. This suggestion made by reviewer #2 is one of the procedures we are planning to adopt. Despite that we include a few data on the answer to the next comment which gives an idea of kappa values during measurements on section 3.2.2 as is shown below.

3. CCN prediction assuming size-dependent chemical composition (Sect. 3.2.2)

As far as I understood you did not have any size-resolved chemical composition data in the time period of your CCN measurements. Therefore you try to use the results of MOUDI samples taken during a previous campaign to account for the size dependence in the composition. However, I am not able to understand how exactly you did that. There is a lot of information missing. I suggest fully revising Sect. 3.2.2 to clearly describe the applied approach. The following questions may help you with that: Are the size distributions you show in Fig. 6 the results from the MOUDI samples? Where exactly were they taken? – The same location as your CCN measurements? If not, are the measurement sites comparable to each other? What was the average total mass concentration for the MOUDI samples and ACSM measurements? – Are they about the same? How exactly did you calculate the size-dependent composition during your CCN period? Did you fit a function through the size-resolved sulfate mass fraction and multiplied by the mass fraction measured by ACSM? Which size range did you assume to be relevant for particle activation, i.e., for which size range did you calculate kappa?

Which value did you assume for kappa_org? What is the purpose of the paragraph in P. 32368, L. 13-22? Where do the AMS measurements come from?

Following the reviewer suggestion, section 3.2.2 has been rewritten as such:

"Also as part of NUANCE-SP project, a measurement campaign was performed from 15th August to 5th September 2012, at the roof of the Institute of Astronomy, Geophysics and Atmospheric Science (IAG), to chemically characterize aerosols from SPMA. The building is about 150 meters from the place where CCN and aerosol measurements described above have taken place. During this measurement campaign aerosols were collected using a Micro-Orifice Uniform Deposit Impactor (MOUDI, model 100; MSP Corporation – Marple et al. 1986) once a day. The mass concentrations of the MOUDI samples were obtained gravimetrically using an electronic high precision microbalance with a sensitivity of 1 μg (Mettler-Toledo). Further analysis was performed using particle-induced X-ray emission (PIXE) and, more recently, ion chromatography, as described in Albuquerque et al. (2012), Vasconcellos et al. (2011) and Sánchez-Ccoyllo and Andrade (2002). Figure 6a illustrates the 24 hours mean mass distribution observed for the period. It is shown that most of the mass distribution is observed between 180 and 320 nm. Considering the four stages from 100 nm to 560 nm, the mean mass concentration sampled during the period of MOUDI operation was 10.9 $\pm$ 6.3 μg/m3, which was comparable to the one evaluated by the ACSM and MAAP described above (8.9$\pm$6.0 μg/m3). Values are in good agreements with the previous work (Albuquerque et al., 2012; Vasconcellos et al., 2011; and Sánchez-Ccoyllo and Andrade, 2002) that have shown a size dependency of inorganic matter in Sao Paulo. The work of Vasconcellos et al. (2011), for example, has shown that sulphate, nitrate, ammonium, calcium and sodium are the most abundant water-soluble ions in São Paulo. Analysis from 15th August to 5th September 2012 clearly show that sulphate is a major component of the accumulation mode (diameters larger than 180 nm), as is also shown in Figure 6a, but values are largely variable. At 100 nm the fraction of Sulphate (Figure 6b) varied from 5.8% to 17.4%, which depress the critical

supersaturation of particles with this size (from 0.5 to 0.3% considering only the contribution of (NH4)2SO4). For diameters smaller than 100 nm the fraction of sulphate decreases systematically, and one observes a value of about 2.5% at 20 nm. In some occasion, nevertheless, an increased sulphate fraction was seen at 50 nm, which produces a relatively large mean value for that size. Considering only the contribution of (NH4)2SO4, the variation of the critical supersaturation for particles of this size range would be from 0.7 ($\sim$ 7% of (NH4)2SO4) to 1.2% ($\sim$ 23% of (NH4)2SO4), which suggests that particles around 55 nm are the lower limit size range for activation on this study. Considering the large fraction of sulphate in the accumulation mode and the large fraction of organic compounds on total aerosol mass, one can argue that organic compound is predominant at smaller particles. One can also conclude that particles in the nucleation or Aitken size range were composedly mostly of organics. By observing that exist an aerosol chemical size dependency of inorganics, it is possible to improve the NCCN modeling. Considering that mass distribution observed by MOUDI was shown to be consistent with Aerosol Mass Spectrometer (AMS) measurements by MOUDI data (Zhang et al., 2005), we assume that the inorganic size fraction during the CCN measurement period takes the same mean size dependency as observed for sulphate during measurements taken from 15th August to 5th September 2012. It is worth to say that CCN closure utilizing AMS measurements tend to be more successful (typically within 20–50 %), due to its fast time resolution (1 Hz) and ability to resolve size-dependent composition. CCN closures in remote environments that use filter-based methods have nevertheless given good closure, on the order of a few percent (Bougiatioti et al., 2009, 2011). The time resolved mass fractions defined above can be used to feed the equation for κ considering a variation with size as a function of time. For this propose we distributed the total inorganic mass from ACSM at a given time through all sizes using a polynomial function fitted through the points that represent the size-resolved sulfate mass fraction (Figure 6b) and also ensure mass conservation. Strictly, the polynomial function defined above can only be applied from 75 nm to 650 nm. However, the application to particles with diameter smaller the 75 nm does not add large errors to the procedure, since usually there only a small amount of mass bellow this size range. For particles larger than about 250 nm, the procedure does not modify significantly the critical supersaturation, once at this diameter range the size is more important than chemical composition."

We also modified the mention to AMS (page 32368, L 13 – 22). In fact our intention was to justify our hypothesis to the use of size-dependency obtained using MOUDI data considering the accuracy obtained using AMS. Now we can read: “The implication of the aerosol chemical size dependency of inorganics shown above can be used to improve the modelling of NCCN. Considering that mass distribution observed by MOUDI was shown to be consistent with Aerosol Mass Spectrometer (AMS) measurements (Zhang et al., 2005), we assume that the inorganic size fraction during the CCN measurement period takes the same mean size dependency as observed for sulphate during measurements taken from 15th August to 5th September 2012. It is worth to say that CCN closure utilizing AMS measurements tend to be more successful (typically within 20–50 %), due to its fast time resolution (1 Hz) and ability to resolve size-dependent composition. CCN closures in remote environments that use filter-based methods have nevertheless given good closure, on the order of a few percent (Bougiatioti et al., 2009, 2011).”

4. Terminology: aerosol vs. particle

The term “aerosol” defines as “solid or liquid particles suspended in a gas”. So, you have to differentiate between the term “aerosol”, which means always the total population of particles in the air and the term “particles”. Instead of talking of e.g. “total aerosol concentration” you should write “total particle number concentration” since it is the number of particles per cubic centimeter of air. Or instead of “aerosol diameter” you should rather say “particle diameter”. I would really appreciate if you could check the whole manuscript with regard to the terminology and write “aerosol” only if you want to talk of the total population of particles. Otherwise you should always use the term “particle(s)” or “aerosol particle(s)”.

The terminology has been revised throughout the manuscript, as suggested by the reviewer.

5. English and captions

The manuscript needs major improvement in language. In some parts it is really difficult to read and understand. Also, please thoroughly revise your figure and table captions. It must be possible to understand the meaning of a figure or table from the caption alone!

We acknowledge the reviewer’s comment and have thoroughly revised the text. For more clarity, portions of the text were modified as well.

Minor comments:

P. 32356, L. 7-10: This sentence is not quite correct. The ability of a particle to act as CCN depends on its size (not size distribution) and chemical composition. The rest of the sentence is okay: Knowledge of particle number size distribution and chemical composition would suffice to predict ambient CCN concentrations.

We corrected the mistake. Now we can read: "The ability of a particle to act as a CCN depends strongly on its size and chemical composition, which implies that the knowledge of both parameters would suffice to provide an accurate prediction on ambient CCN concentrations."

P. 32360, Sect. 2.2.2: Which type of DMA did you use? A size range of 10 to 500 nm appears quite broad to me. At which flow rates did you operate the DMA? The size distributions you show in Fig. 3 reach only up to 400 nm. Why not up to 500 nm?

We used a bipolar charger, a medium-long Vienna type Differential Mobility Analyzer with a sample flow of 1.1 lpm and sheath flow of 6 lpm. Given that aerosol number concentration decreases drastically above 400nm, as is seen on Figures 3a and 3b, we limited the size distribution plot to that range.

P. 32383, Fig. 2: Could you please also plot the time series of the total CN concentration for comparison?

Total particle concentration was shown in Figure 3c.

P. 32363, L. 5: What is “aerosol mean diameter and volume”? Please define more exactly. I guess you mean the number and volume mean diameter. According to Seinfeld and Pandis (2006) the number mean diameter is the “average diameter of the population”, and the volume mean diameter is the “diameter of the particle whose volume equals the mean volume of the population”.

We acknowledge the mistake and modified the text according to the reviewer guess. Now we can read: "Fig. 3d shows the time series of aerosol number and volume mean diameter, ..." according to the Seinfel and Pandis (2006) definition.

P. 32362, L. 8-11: From the plots shown in Fig. 2 and from the numbers in Tab. 1 I think you are not able to draw the conclusion that the high variation in CCN number concentration is mostly due to variations in chemical composition rather than shape of the size distribution. In fact, I guess the opposite is true: A change in aerosol composition leads to changes of the critical diameter needed for cloud droplet activation but that in turn leads only to a small change of the CCN concentration (when the CN size distribution is kept constant). Instead, keeping the chemical composition constant (i.e., keeping the activation diameter constant) a change in the (shape of the) size distribution leads to a much higher relative change in the CCN number concentration. This has been shown intensely, e.g. by Dusek et al., 2006 and Juranyi et al., 2010. P. 32363, L. 5: What is “aerosol mean diameter and volume”? Please define more exactly. I guess you mean the number and volume mean diameter. According to Seinfeld and Pandis (2006) the number mean diameter is the “average diameter of the population”, and the volume mean diameter is the “diameter of the particle whose volume equals the mean volume of the population”.

In fact we cannot take the conclusion from Figure 2 alone. The conclusion was taking

observing Figures 2, 3 and 4 from where we observe that the activated fraction is more related to the inorganic fraction then to the number of particles. We move L. 8 - 11 of page t32362 to a new paragraph before the end of section 3.1 where now reads:

"Considering still the data from the 16th and 17th October, one can observe that activated fraction (Fig. 2b) decreases significantly when the inorganic fraction is reduced (Fig. 4a), even when the aerosol concentration remains relatively large (Fig. 3c). Data analysis also indicates that the CCN concentration is much better correlated to inorganic fraction than to aerosol concentration, which suggest that most of the high variability of CCN number concentration is due to the variations of the chemical composition, while a smaller part of it can be attributed to variability of the aerosol properties such as shape of the size distribution and the total particle number concentration." We do not really believe that the same analysis applied to Juranyi et al. 2010 can be done to our work. In Juranyi’s work the chemical composition is size-independent, while at our work we measure aerosol recently nucleated within a large urban center, implying in a strongly size-dependent chemical composition.

P. 32363, L. 6-8: I wouldn’t say that the total particle number concentration has a secondary peak at 18:00 LT. In Fig. 3e the value at 18:00 is only little higher than the values one hour before and after. For me the overall trend is rather a decreasing concentration from 11:00 to (even) 05:00. I suggest showing a so-called box or whisker plot of the whole statistics (median, quartiles, etc.) here. That would present a clearer picture of trends and peaks. Also, what kind of information do you have on the traffic emissions? Maybe statistics on the average diurnal variation of vehicles in a street? Could you provide a graph?

We corrected the mistake. Now we can read: "The mean aerosol number concentration attained a maximum values at noon (Figure 3e), and decrease continuously after that" . We went to the statistic as well. The text will be read as: “The scrutiny of the aerosol concentration data reveals that there is a lot of variation during the day as a function of meteorological condition, but the general trends shown in Figure 3e is maintained.

The second quartile approaches the mean values and the standard deviation is about the same all the day long."

Information about aerosols and traffic emission can be found at Backman et al. (2012).

P. 32363, L. 11-12: I don't understand the meaning and purpose of this sentence. It seems not to be related to anything you say before or after, nor can it be proven by any figure.

The sentence was removed.

P. 32364, L. 2-5: What do you mean with this sentence? Something like that?: "The contribution of organics and BC to the total mass was much larger than observed in previous studies in this area. Among all components BC exhibited the largest variability".

We modified the sentence. Now it appears as: "Organic compounds and BC provide the largest contribution to the total mass of aerosols, in agreement with previous studies in the area (Ynoue et Andrade, 2004). Among the identified species, BC exhibited the largest variability".

P. 32364, L. 5-7: I do not agree with this statement. The diurnal variation of the organic mass concentration is not more than the diurnal variation of the inorganic components. Only the minima and maxima occur at different times.

Whereas organic aerosol concentration ranges from 3.5 up to 6.5 μgm-3, (3.0 μgm-3, or 45% of peak value), SO4 concentrations are observed to range from 0.45 and 0.65 μgm-3 (0.2 μgm-3, or 30% of peak value). As such, the reviewer is correct considering relative variation, while the original meaning of the text was on absolute concentration. The text has modified as such:

"In absolute terms, the mean hourly concentration of inorganics does not exhibit a remarkable variation during the day. SO4, for example, ranges from 0.45 and 0.65 μgm-3. Organics, however, given the much higher ambient concentration, was observed to

span over a wider range of values throughout the day (from 3.5 up to 6.5 μgm-3). In relative terms, however, both organics and inorganics present a comparable variation during the day (45% and 30% relative to peak value, respectively)."

P. 32364, L. 7-9: I also do not see that there is agreement in the diurnal variation between organic mass and total particle concentration. What about the diurnal variation of BC? Is it related to the number of vehicles?

The reviewer is correct as the correlation between total particle concentration and organic mass is not readily observed. This is assumed to occur mainly from two reasons: The marked change in aerosol sizes throughout the day by, e.g., condensation of processed organic vapors or evaporation of semi-volatiles, which strongly impacts aerosol mass loading but not aerosol number. Possible strong contribution of aerosol number and mass outside the ACSM measurement range, e.g., nucleation mode aerosols.

The BC does correlate with traffic, being the vehicles (in particular heavy-duty) the dominant source of soot in the area (Backman et al., 2012). For the period studied here, BC diurnal variation correlates with the first rush hour peak, extending close to noon (local time). The second rush hour peak is not observed, potentially due to a wind direction shift.

A manuscript is currently being prepared dealing with aerosol chemistry and life cycle in the São Paulo Metropolitan Area with longer time series as detailed in this manuscript, which shall address this topic.

P. 32365, L. 21-22: Which measured parameters did you use for the calculation of f_org and f_inor? Since you talk of "ACMS/MAAP derived kappa", is the BC mass measured by the MAAP somehow used in this calculation? You cite Dusek et al. (2010) and Rose et al. (2011) here, but they calculated only AMS derived kappa values.

We considered Organic, inorganic and BC for the calculation of the derived kappa. We included the information on line 19 of page 32366. There now we can read: "...(κ=0.1

for organics and κ=0.7 for inorganics, κ is considered zero for BC)."

P. 32366, L. 2-3: I do not understand this sentence. Do you mean that the calculated kappa value experienced large variations because of the strong variation in f_inor?

Yes. The largest variation of κ was due to variation of the inorganic fraction. To make the text clear, we modified this sentence and moved it to the end of the sentence beginning at line 3 of the same page. It reads as: "The mean ACSM /MAAP derived κ value for the period studied was 0.15±0.04, from this values 0.10±0.03 can be attributed to the inorganic fraction, which imply that the largest variation experienced by κ is due to the variation of the inorganic fraction."

P. 32366, L. 4: "0.10 +/- 0.03 for inorganic fraction": This value is meaningless.

We modified the text. Now it reads: "The mean ACSM /MAAP derived κ value for the period studied was 0.15±0.04, from this value 0.10±0.03 is due to the inorganic fraction"

P. 32366, L. 13-18: Please revise this paragraph and write exactly how you calculated the total CCN concentration. I guess what you did is the following: 1. calculating kappa from f_org and f_inor; 2. calculating the critical diameter at the supersaturation under consideration by Eq. 3 using kappa; 3. integrating the DMPS derived particle number size distribution above the critical diameter. Note that since you have only 22 size channels for the whole size distribution it is necessary to interpolate it in order to be able to integrate properly.

We thanks this comment. In fact the approach was a little bit different and we included it on the text. Now the text appears as follows: "Initially, the size-independent solubility value, κsi, is determined using the values of Dusek et al. (2010) (κ=0.1 for organics and κ=0.7 for inorganics, κ is considered zero for BC). We then calculate the critical supersaturation for each diameter in the DMPS using eq. (3) and κsi. The total modelled CCN concentration for a given supersaturation is determined integrating the DMPS derived particle number size distribution considering those classes of diameters whose critical supersaturation are lower or equals to the supersaturation under consideration. Since we have only 22 size channel for the whole size distribution it is necessary to interpolate it in order to be able to integrate properly. "

P. 32370, L. 5-6: "…presenting peaks that alternate with low N_CCN values": This phrase is meaningless.

We removed this sentence.

P. 32370, L. 22: None of your graphs is showing "an increase in aerosol hygroscopicity in the afternoon". For this you would have to plot the diurnal variation of kappa (kappa_CCN as explained above), which would actually be interesting to see. Technical corrections: Figures: Can you please plot all graphs showing average diurnal cycles (e.g., Fig. 2c) from 0 to 24 LT.

Although we have not shown results from the calculated hygroscopicity we included a small description on the aerosols properties during the studied period. A description of kappa for SPMA is currently in progress and will be submitted to publication in the near future. All graph will be provided showing average diurnal cycles from 0 to 24 LT.

P. 32355, L. 10-11: mention what the numbers are, e.g.: (arithmetic mean _ standard deviation)

Done.

P. 32355, L. 14: define SS

Done.

P. 32355, L. 22: define N_CCN

Done.

P. 32356, L.12-14: "…of droplets in clouds and decrease their sizes…"

Done.

P. 32356, L. 24: define N_CCN

Done.

P. 32357, L. 7: “. . .to act as CCN is a difficult task.”

Done.

P. 32357, L. 8: “. . . is often . . .”

Done.

P. 32357, L. 9: “. . .to be internally mixed. . .”

Done.

P. 32359, L. 25: do you want to name the supersaturation as “SS” or “S”? Please choose only one symbol.

Done.

P. 32359, L. 27: define SS_crit

Done.

P. 32360, L. 1: “Droplets leaving the column. . .” instead of “particles”

Done.

P. 32360, L. 2: “CCN” without “s”

Done.

P. 32363, L. 8: Fig. 3e shows no figure on traffic emissions

We move the reference to Fig 3 to a more appropriated point. Now we can read: " ...but has a secondary peak at 18 hours (Fig. 3e), as a function of traffic emissions. . ."

P. 32366, L. 7: “This is a result of the relatively low inorganic mass fraction.”

Done.

P. 32366, L. 19-20: “The modeled results indicate an overestimation of N_CCN, which is increasing with increasing supersaturation.” It is not the “critical supersaturation”.

Done.

P. 32369, L. 12: “CCN concentration” instead of “CCN spectra”

Done.

P. 32370, L. 11: “higher mass fractions” instead of “volume fractions” since you talk of mass fractions throughout the paper

Done

P. 32370, L. 12: What do you mean with “charge” of aerosols?

We substitute "charge” by concentration

P. 32382, Fig. 1: “Time series” instead of “diurnal variation”.

Done.

References

Backman J., Rizzo L. V., Hakala J., Nieminen T, Manninen, H. E., Morais F., Aalto P. P., Siivola E., Carbone S., Hillamo R., Artaxo P., Virkkula A., T. Petäjä, and Kulmala M. On the diurnal cycle of urban aerosols, black carbon and the occurrence of new particle formation events in springtime São Paulo, Brazil. Atmos. Chem. Phys., 12, 11733–11751, 2012

References:

Dusek, U., Frank, G. P., Hildebrandt, L., Curtius, J., Schneider, J., Walter, S., Chand, D., Drewnick, F., Hings, S., Jung, D., Borrmann, S., and Andreae, M. O.: Size matters more than chemistry for cloud nucleating ability of aerosol particles, Science, 312, 1375–1378, 2006.

Dusek, U., Frank, G. P., Curtius, J., Drewnick, F., Schneider, J., Kürten, A., Rose, D., Andreae, M. O., Borrmann, S., and Pöschl, U.: Enhanced organic mass fraction and decreased hygroscopicity of cloud condensation nuclei (CCN) during new particle formation events, Geophys. Res. Lett., 37, L03804, doi:10.1029/2009GL040930, 2010.

Jurányi, Z., Gysel, M., Weingartner, E., DeCarlo, P. F., Kammermann, L., and Baltensperger, U.: Measured and modelled cloud condensation nuclei number concentration at the high alpine site Jungfraujoch, Atmos. Chem. Phys., 10, 7891-7906, doi:10.5194/acp-10-7891-2010, 2010.

Rose, D., Gunthe, S. S., Mikhailov, E., Frank, G. P., Dusek, U., Andreae, M. O., and Pöschl, U.: Calibration and measurement uncertainties of a continuous-flow cloud condensation nuclei counter (DMT-CCNC): CCN activation of ammonium sulfate and sodium chloride aerosol particles in theory and experiment, Atmos. Chem. Phys., 8, 1153-1179, doi:10.5194/acp-8-1153-2008, 2008.

Rose, D., Gunthe, S. S., Su, H., Garland, R. M., Yang, H., Berghof, M., Cheng, Y. F., Wehner, B., Achtert, P., Nowak, A., Wiedensohler, A., Takegawa, N., Kondo, Y., Hu, M., Zhang, Y., Andreae, M. O., and Pöschl, U.: Cloud condensation nuclei in polluted air and biomass burning smoke near the mega-city Guangzhou, China – Part 2: Size resolved aerosol chemical composition, diurnal cycles, and externally mixed weakly CCN-active soot particles, Atmos. Chem. Phys., 11, 2817-2836, doi:10.5194/acp-11-2817-2011, 2011.

Seinfeld, J. H. and Pandis, S. N.: Atmospheric Chemistry and Physics: From Air Pollution to Climate Change, John Wiley and Sons, Inc., New York, 2006.

Interactive comment on Atmos. Chem. Phys. Discuss., 13, 32353, 2013.

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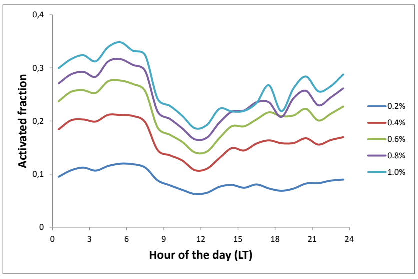

Fig. 1. Figure 2c. The mean hourly averaged activated fraction over the whole period. The different colours represent the different supersaturations (SS)

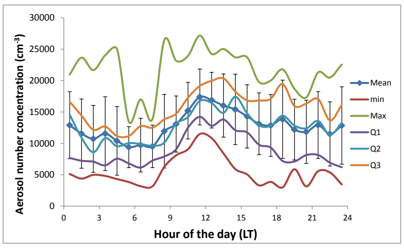

Fig. 2. Fig 3d. Mean hourly aerosol total concentration; Bars refers to stadard deviation; Min and max refers to the lowest a largest concentration; Q1, Q2 and Q3 refers to quartiles

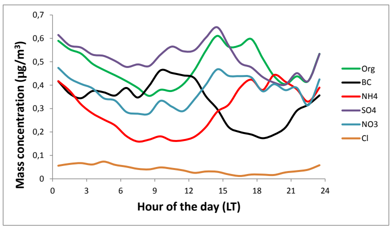

Fig. 3. Fig 4d. The mean aerosol chemical composition as a function of local time.

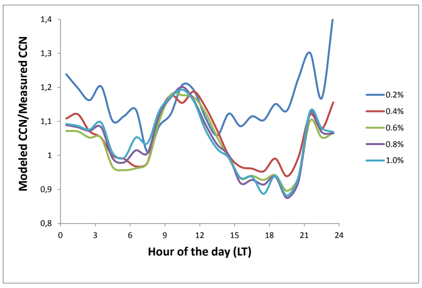

Fig. 4. Fig 8. Mean value of modelled NCCN/Observed NCCN as a function of local time.