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A meta-analysis of particle water uptake reconciliation studies

J. D. Whitehead¹, M. Irwin^{1,*}, J. D. Allan¹, N. Good^{1,**}, and G. McFiggans¹

¹Centre for Atmospheric Science, SEAES, University of Manchester, Oxford Road, Manchester, M13 9PL, UK

*now at: Department of Earth and Planetary Science, Graduate School of Science, The University of Tokyo, Tokyo, Japan

**now at: Department of Environmental & Radiological Health Sciences, Colorado State University, Fort Collins, Colorado, USA

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Correspondence to: J. D. Whitehead (james.whitehead@manchester.ac.uk)

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Abstract

Reconciliation between measurements of sub-saturated and supersaturated aerosol particle water uptake was attempted at a wide range of locations between 2007 and 2013. The agreement in derived number of cloud condensation nuclei (N_{CCN}) or particle hygroscopicity was mixed across the projects, with some datasets showing poor agreement across all supersaturations and others agreeing within errors for at least some of the supersaturation range. The degree of reconciliation did not seem to depend on the environment in which the measurements were taken. The discrepancies can only be attributable to differences in the chemical behaviour of aerosols and gases in each instrument, leading to under- or overestimated growth factors and/or CCN counts, though poorer reconciliation at lower supersaturations can be attributed to uncertainties in the size distribution at the threshold diameter found at these supersaturations. From a single instrument, the variability in N_{CCN} calculated using particle hygroscopicity or size distribution averaged across a project demonstrates a greater sensitivity to variation in the size distribution than chemical composition in most of the experiments. However, the discrepancies between instruments indicate a strong requirement for reliable quantification of CCN in line with an improved understanding of the physical processes involved in their measurement.

1 Introduction

Changes to the number of Cloud Condensation Nuclei (CCN) will impact on cloud microphysical properties, with an increase in CCN resulting in more and smaller cloud droplets and in brighter clouds (Twomey, 1977) with longer lifetimes, higher liquid water content and increased cloud thickness (Stevens and Feingold, 2009). The net effect of these aerosol-cloud interactions is to cool the climate system, however significant uncertainties remain in predicting the magnitude of this impact (Boucher et al., 2013). A better understanding of these interactions is needed to improve climate predictions.

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The ability of aerosol particles to act as CCN depends upon their size and chemical composition. A number of regional and global models have been developed over recent years to predict CCN number concentrations based on these parameters (e.g. Spracklen et al., 2008; Kumar et al., 2009). In order to verify and improve these models, measurements of CCN properties from a wide range of locations around the world are needed.

CCN properties of aerosols can be measured in the subsaturated regime with a Hygroscopicity Tandem Differential Mobility Analyser (HTDMA; for a review, see Swietlicki et al., 2008) and in the supersaturated regime with a Cloud Condensation Nuclei counter (CCNc; Roberts and Nenes, 2005). Reconciliation studies between these two measurement techniques allows us to test our understanding of aerosol water uptake processes, and to investigate the suitability of applying simplifying assumptions to models.

Extrapolation between the sub- and supersaturation regime can be approximated through Köhler theory with hygroscopicity described by a single parameter. Several such parameters have been proposed (for a review, see Rissler et al., 2010), such as that of Petters and Kreidenweis (2007). Denoted κ , this has been widely used in recent years (e.g. Andreae and Rosenfeld, 2008; Kammermann et al., 2010; Irwin et al., 2010, 2011), particularly in reconciling sub-saturated particle hygroscopicity with CCN measurements. Several hygroscopicity-CCN reconciliation studies have been published over the past decade or so (e.g. Zhou et al., 2002; Rissler et al., 2004; Jurányi et al., 2010; Kammermann et al., 2010; Irwin et al., 2010, 2011; Fors et al., 2011), however all these studies concentrated on measurements at a single site, making it impossible to generalise the conclusions. Here we present, for the first time, a meta-analysis of reconciliation studies by our group at a large number of different locations including marine, tropical, background continental and urban environments. Such a comprehensive compilation of reconciliation studies allows us to explore instrumental discrepancies and whether reconciliation is affected by the environment in which the measurements were taken.

2 Measurements and methods

The measurements that are included in this study are labelled in bold text in Fig. 1. These covered a range of different ambient environments including marine (Discovery cruise, Mace Head, Weybourne), tropical (Borneo, Amazonia), continental background (Hornisgrinde, Chilbolton) and urban (London). These datasets were selected out of all those in Fig. 1 for the quality of the data and suitability of measurement configuration for reconciliation study.

For each experiment, CCN activity was measured as a function of supersaturation and particle dry size using a Droplet Measurement Technologies Cloud Condensation Nuclei counter (CCNc; Roberts and Nenes, 2005). The calibration and operation of the CCNc is described fully by Good et al. (2010a, b) and Irwin et al. (2010, 2011), with the same methods used in all projects. Briefly, a dried ($< 20\%$ RH) monodisperse aerosol sample was supplied by a Differential Mobility Analyser (DMA) stepping discretely through a range of sizes. The sample was then split between the CCNc and a Condensation Particle Counter (CPC; TSI model 3010). The ratio of the number of CCN (N_{CCN}) to the total number concentration of aerosol particles (N_{CN}) is the fraction of particles activated ($F_A(S, D_0)$) at a given supersaturation, S , and dry diameter, D_0 . The resulting activation spectra ($F_A(S, D_0)$) as a function of dry diameter, D_0 can be used to derive the diameter at which 50% of the particles activate (D_{50}) by fitting a sigmoid curve function. The hygroscopicity parameter, κ , can then be derived using the κ -Köhler model, and will be denoted by $\kappa_{D_{50}}$.

Hygroscopic growth factor distributions were measured during each experiment using a Hygroscopicity Tandem Differential Mobility Analyser (HTDMA). Two different instruments were used: the first (HTDMA1), developed by Cubison et al. (2005), was used during the Discovery, Hornisgrinde, Borneo, and Amazonia experiments, while the second (HTDMA2), developed by Good (2009), was used in the remaining experiments. In all cases, calibrations were conducted as discussed by Good et al. (2010a), and the data were processed using the TDMAinv software described by Gysel et al.

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(2009). In the HTDMA, a dry aerosol sample is size-selected with the first DMA and then humidified to 90 % RH (except at Hornisgrinde where 86 % RH was used; Irwin et al., 2010). The second DMA is then used to measure the size distribution of the humidified aerosol, to give the distribution of Growth Factor (defined as the ratio of wet to dry aerosol diameter) as a function of RH and dry diameter (GF_{RH,D_0}). For most of the studies, 5 to 7 dry sizes were scanned in this way, ranging from 24 to 300 nm. Values of κ can be calculated from the mean growth factor measurements as described by Eq. (1):

$$S = \frac{\overline{GF}^3 - 1}{\overline{GF}^3 - (1 - \kappa)} \exp\left(\frac{4\sigma_w M_w}{RT\rho_w D_0 \overline{GF}}\right) \quad (1)$$

where S is the supersaturation (RH/100%), \overline{GF} is the mean growth factor, κ is the hygroscopicity parameter, σ is the surface tension of water, M_w is the molar mass of water, R is the universal gas constant, T is the temperature, ρ_w is the density of water and D_0 is the dry diameter. The κ derived from the growth factor data will be denoted by κ_{GF} .

A number of approaches can be taken to calculate total N_{CCN} as a function of supersaturation. From the CCNc data, the simplest way is to integrate $N_{CCN}(S, D_0)$ as a function of D_0 for each set supersaturation, S_{set} . Alternatively, the aerosol number size distribution (as measured by the CPC on the DMA) can be integrated from the largest size down to a threshold diameter (in this case, the D_{50} derived from the activation spectra). For the HTDMA data, a threshold diameter can be derived from the calculated κ_{GF} values for a given supersaturation, and from this, N_{CCN} can be calculated as before. By using the S_{set} from the CCNc in deriving N_{CCN} from the HTDMA, a direct comparison between the instruments can be made. In this study, N_{CCN} from the CCNc data is derived from the aerosol size distribution and D_{50} and compared with N_{CCN} derived from the HTDMA data. κ values are also compared between the instruments using the methods described above.

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3 Results and discussion

For each measurement campaign, the mean values of N_{CCN} and κ derived from D_{50} and GF were found for each S_{set} , and the ratios of these means are plotted in Fig. 2. The error bars represent the standard deviation of these ratios, and hence show the variation throughout a given experiment. In some campaigns, where two calibrations disagreed, both were applied and the spread is illustrated in Fig. 2 as shaded areas. The graphs show that the level of reconciliation varied greatly between the different experiments, generally varying with supersaturation. Poorest agreement between the HTDMA and CCNc across the range of supersaturations was found in the measurements from Hornisgrinde, Borneo and the Discovery cruise. The other experiments largely showed agreement within the error bars for at least some of the supersaturation range, although there is a tendency for the ratios to decline with increasing supersaturation in these datasets.

Possible reasons for discrepancies between CCNc and HTDMA derived κ and N_{CCN} for the Discovery, Hornisgrinde and Borneo datasets have been discussed at length by Good et al. (2010b) and Irwin et al. (2010, 2011), respectively, and they are likely to apply in varying degrees to the other datasets. The discrepancies are described as being due either to instrumental differences or assumptions made in the model. Previously, discrepancies between measured and modelled CCN behaviour have been attributed variously to particle surface tension at the point of activation, changes in the kinetics of uptake in the instruments, or external mixing.

When using the κ -Köhler model, the surface tension is often assumed to be that of pure water, $\sigma = 0.072 \text{ J m}^{-2}$ (Petters and Kreidenweis, 2007; Rissler et al., 2010). In reality, surface active compounds may concentrate at the water-air interface of a deliquesced particle, suppressing surface tension and affecting the CCN activity of the particle. A number of studies have explored the effect of this assumption on reconciliation, and have found either that reducing surface tension in the calculations would not improve closure (Jurányi et al., 2010), or that unrealistic values of surface tension

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would be required to account for discrepancies (Irwin et al., 2010; Good et al., 2010b). Moreover, more recent work has demonstrated that bulk-to-surface partitioning offsets most of the influence of any surface tension reduction (Li et al., 1998; Sorjamaa et al., 2004; Kokkola et al., 2006; McFiggans et al., 2006; Topping et al., 2007; Topping, 2010; Frosch et al., 2010, 2011; Romakkaniemi et al., 2011; Prisle et al., 2012).

Instrumental differences mainly relate to the chemical behaviour of aerosols and gases in the respective instruments. Growth factor may be underestimated in the HTDMA if the residence time following humidification is too short to reach equilibrium before sizing in the second DMA (Duplissy et al., 2009), leading to an underestimate of hygroscopicity. In addition, volatile and semi-volatile compounds can evaporate during the drying process. While the HTDMA and CCNc use the same dryer in all these measurements, the subsequent behaviour of the volatilised gases in the different conditions of each instrument can lead to further discrepancies (Irwin et al., 2010). For example, the saturation column of the CCNc can act as a mist chamber, where droplets take on soluble material from the gas phase, potentially increasing the N_{CCN} count. Indeed, the possible influences of semi-volatile components on droplet activation and on reconciliation between sub- and supersaturated measurements has been discussed by Topping and McFiggans (2012) and the impacts of semi-volatile co-condensation expanded by Topping et al. (2013).

The aerosol mixing state might also affect agreement, since the methods commonly used to derive hygroscopicity and N_{CCN} with the HTDMA and CCNc do not account for externally mixed aerosol, which can have different effects in the two instruments. A number of studies have considered this, using different methods to account for external mixing. Most of these (e.g. Rissler et al., 2004; Kammermann et al., 2010; Irwin et al., 2010) found that mixing state has no effect on measurement reconciliation, however Wex et al. (2010) found that it is important in obtaining agreement between HTDMA and chemical composition derived hygroscopicity. For this study the mixing state was parameterised, using the HTDMA growth factor distribution, by the absolute value of the mean growth factor subtracted from the peak growth factor. Strong

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external mixing could be seen in the HTDMA measurements at Chilbolton and London (Summer and Winter), and the mixing state parameter ranged 0.12–0.20. For measurements that showed a lesser degree of external mixing (i.e. a weaker secondary mode in the growth factor distribution; e.g. Borneo, Mace Head), the mixing state parameter ranged between 0.06–0.12, and was less than 0.05 for measurements showing a largely internally mixed aerosol sample (e.g. Discovery cruise). Kammermann et al. (2010) accounted for external mixing in their reconciliation study by defining a critical growth factor at each dry diameter, above which particles activate at a given supersaturation. The fraction of particles above this growth factor is the activated fraction, thus providing an activation spectrum ($F_A(S, D_0)$) from which to calculate D_{50} and hence N_{CCN} as described above. For the CCNc, external mixing can be taken into account by integrating $N_{CCN}(S, D_0)$ as a function of D_0 for each S_{set} . The ratios of the mean values of N_{CCN} derived from each method was calculated and compared to those shown in Fig. 2. No improvement was seen in reconciliation in any of the data sets, suggesting that mixing state does not affect hygroscopicity-CCN reconciliation, even when the degree of external mixing is high.

As already mentioned, there is a tendency in some of the datasets shown in Fig. 2 for the ratios to decline with supersaturation. A similar trend has also been observed in other studies (e.g. Kammermann et al., 2010; Fors et al., 2011), and has been explained as resulting from greater uncertainties in the instrument at lower supersaturations. The threshold diameter at these supersaturations is higher up in the tail of the particle number size distribution, and so predictions are more sensitive to the counting statistics in the size distribution. This is consistent with the wide variation in the measurements (shown as large error bars) that can be seen here. This trend is not observed in the datasets that show the poorest agreement (Discovery, Hornsgrinde and Borneo), and it is noted that all these measurements were conducted with the same HTDMA (HTDMA1). However the measurements in Amazonia also employed HTDMA1 and these show relatively good reconciliation, plus the trend of higher ratios at lower supersaturations. The two HTDMAs were operated side-by-side, sampling

ambient air in Manchester, UK, along with a CCNc, in order to compare reconciliation results. The derived N_{CCN} and κ ratios are shown in Fig. 3. Better agreement is seen using HTDMA2, but importantly, both exhibit the trend of increased ratios at lower supersaturations. This information shows that differences between campaigns in the relationship between ratios of N_{CCN} or κ and supersaturation cannot be attributed to different instruments.

The wide range of locations from which the studies presented here derive make it possible to explore whether different environments (characterised by different aerosol populations) result in different degrees of reconciliation in water uptake measurements. No common patterns could be seen in Fig. 2 for measurements from similar environments, distinct from others, so there appears to be no such dependency.

For each dataset, N_{CCN} was also calculated from both the HTDMA and CCNc data using campaign averages of either κ (or D_{50} in the case of the CCNc) or size distribution. The results are shown in the supplementary material as box plots of N_{CCN} as a function of supersaturation for each method. In most of the datasets, averaging κ does not lead to a significant change in mean $N_{\text{CCN}}(S)$ from either instrument, whereas $N_{\text{CCN}}(S)$ derived using the mean size distribution shows a much reduced variability. Taken in isolation, the data from a single instrument may imply that N_{CCN} is rather insensitive to κ , and hence chemical composition and that, unsurprisingly, size distribution is more important for predicting N_{CCN} , in agreement with previous studies (e.g. Dusek et al., 2006; Jurányi et al., 2010). However, that N_{CCN} derived from different instruments frequently differs markedly indicates a strong requirement to understand the processes leading to the discrepancies and thereafter to define a protocol for reliable N_{CCN} quantification in line with our most informed understanding of the physical processes involved in their measurement.

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4 Conclusions

This paper presents a meta-analysis of particle water uptake reconciliation studies from measurements taken at a large number of locations in a wide range of environments over the period from 2007 to 2013. By examining such a range of datasets, it is possible to produce more general conclusions, particularly regarding instrumental discrepancies or different environments. Reconciliation between HTDMA and CCNc measurements was examined by comparing N_{CCN} and κ as a function of supersaturation, derived from the threshold diameter seen with the CCNc and the mean growth factor measured by the HTDMA. Many of the datasets showed agreement within the variability of the measurements throughout the supersaturation range and some disagreed at all supersaturations. There did not appear to be any clear dependence of the degree of measurement reconciliation on whether the dataset was collected in a marine, tropical, background continental or urban environment, and neither could differences between campaigns be attributed to the use of different instruments. There was generally poorer reconciliation at the lower supersaturations, likely resulting from greater relative uncertainties in the size distribution at the threshold diameters that are observed at these low supersaturations. Discrepancies at other supersaturations are attributed to differences in the chemical behaviour of vapours and/or particles in the different instruments. Aerosol mixing state did not appear to affect reconciliation, even when strong external mixing was observed. Calculating N_{CCN} from any one instrument using campaign averages of either κ or size distribution would indicate relative insensitivity of N_{CCN} to the chemical composition with more of the variability in N_{CCN} arising from the size distribution.

Supplementary material related to this article is available online at
**[http://www.atmos-chem-phys-discuss.net/14/9783/2014/
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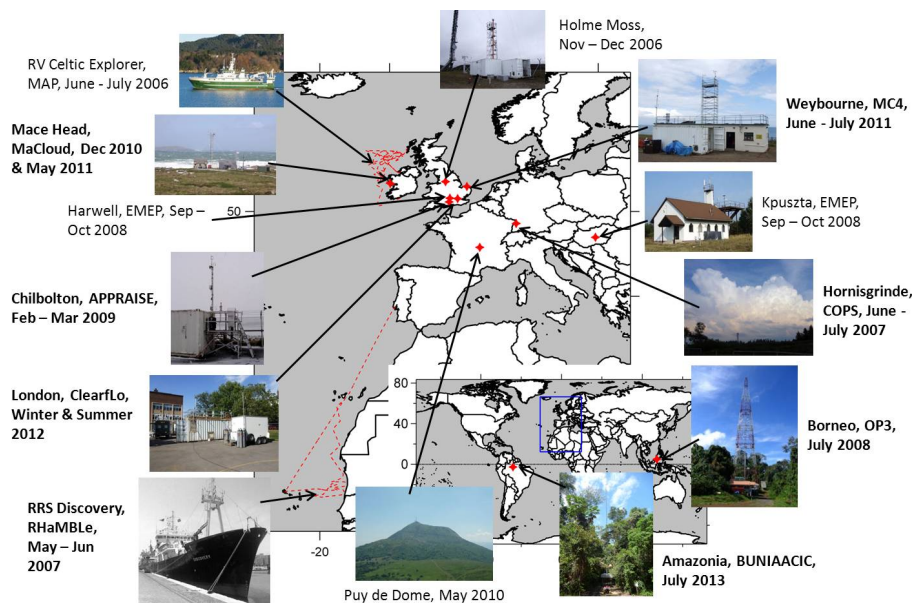


Fig. 1. Map showing the locations of measurements. The labels name the locations, projects and dates of the experiments. The datasets used in this study are labelled in bold print.

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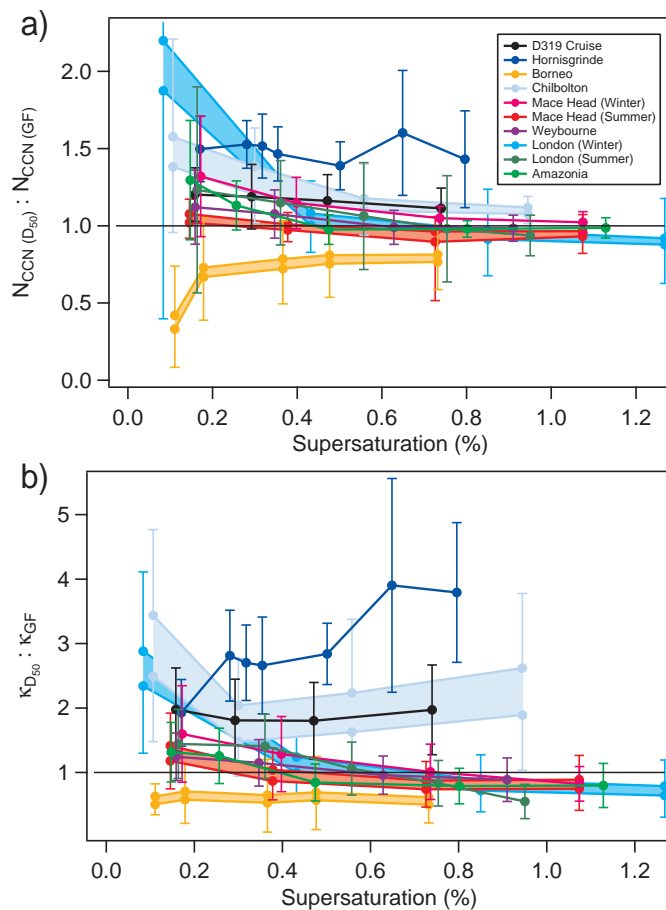


Fig. 2. Ratios of mean D_{50} and GF derived (a) N_{CCN} and (b) κ values as a function of S_{set} for each measurement campaign. Error bars represent one standard deviation.

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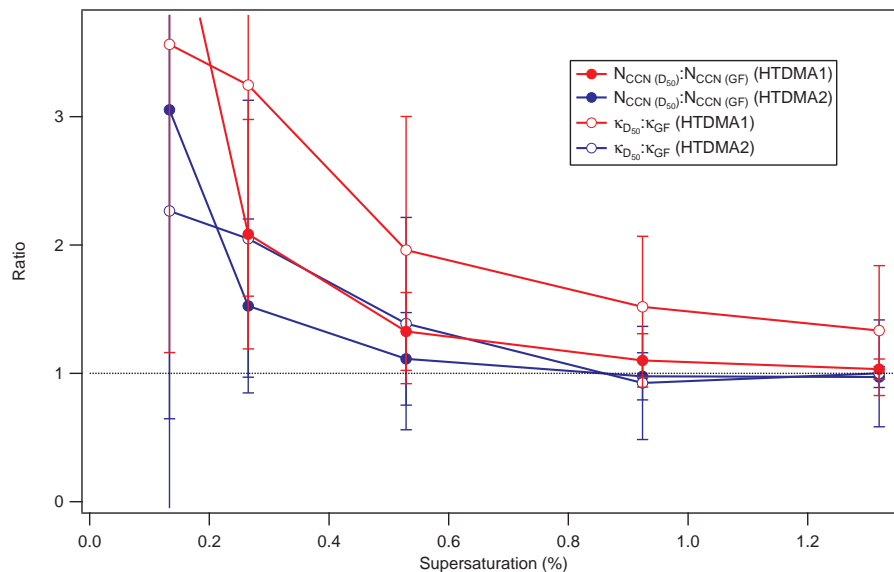


Fig. 3. Ratios of mean D_{50} and GF derived (a) N_{CCN} and (b) κ values as a function of S_{set} for each HTDMA, run side-by-side with a single CCNc, sampling ambient air in Manchester. Error bars represent one standard deviation.

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