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Parameterising Cloud Condensation Nuclei concentrations during HOPE

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Abstract.

An aerosol model was used to simulate the generation and transportation of aerosols over Germany during the HD(CP)^2 Observational Prototype Experiment (HOPE) field campaign of 2013. The aerosol number concentrations and size distributions were evaluated against observations, which shows satisfactory agreement in the magnitude and temporal variability of the main aerosol contributors to cloud condensation nuclei (CCN) concentrations. From the modelled aerosol number concentrations, number concentrations of CCN were calculated as a function of vertical velocity using a comprehensive aerosol activation scheme which takes into account the influence of aerosol chemical and physical properties on CCN formation. There is a large amount of spatial variability in aerosol concentrations, however the resulting CCN concentrations vary significantly less over the domain. Temporal variability is large in both aerosols and CCN. A parameterisation of the CCN number concentrations is developed for use in models. The technique involves defining a number of best fit functions to capture the dependence of CCN on vertical velocity at different pressure levels. In this way, aerosol chemical and physical properties as well as thermodynamic conditions are taken into account in the new CCN parameterisation. A comparison between the parameterisation and the CCN estimates from the model data shows excellent agreement. This parameterisation may be used in other regions and time periods with a similar aerosol load, and furthermore, this technique demonstrated here may be employed in regions dominated by different aerosol species.

1 Introduction

The influence that aerosols have on cloud microphysics is relatively well established, however clouds and aerosols continue to contribute the largest uncertainty to the Earth's energy budget in climate simulations (Boucher et al., 2013). In an effort to realistically capture aerosol cloud interactions, and hence reduce these uncertainties, cloud condensation nuclei (CCN) parameterisations have been developed for use in models. The ability of an aerosol to act as a CCN is determined by its size and composition, so accurately modelling CCN activation necessitates an understanding of these underlying physical and chemical properties.

The hygroscopicity parameter is now commonly used to characterise the chemical properties of a given aerosol species (Petters and Kreidenweis, 2007), however for the sake of simplicity, chemical composition can be neglected. Segal and Khain

(2006) state that aerosol chemical composition has a relatively small effect, and assume all aerosols are composed of NaCl. Some doubt does remain as to the relative importance of the aerosol physical and chemical properties in determining CCN concentrations (Hudson, 2007), however most evidence suggests the number concentration and size have the most significant effect (Dusek et al., 2006; Ervens et al., 2007; Feingold, 2003), since larger particles are more readily activated.

There are numerous possibilities for characterising the number concentration of aerosols. Early parameterisations, including the seminal work of Twomey (1959), used a power law to describe the size of aerosols. This approach combined with simple expressions for the number of nucleated drops has drawbacks, since anomalously high droplet number concentrations can be produced. A power law is also employed to define the aerosol size distribution (Khvorostyanov and Curry, 1999) in parameterisations of droplet activation (Morrison et al., 2005) employed by the WRF model.

Other parameterisations assume a prescribed uniform aerosol size distribution with only one, typically log-normal, mode (Abdul-Razzak et al., 1998; Segal and Khain, 2006). Several modes can be used to define the aerosol sizes (Abdul-Razzak and Ghan, 2000; Fountoukis and Nenes, 2005; Liu et al., 2012; Shipway and Abel, 2010), where the parameters of the size distribution are either calculated from an aerosol model, or derived from limited observations from a short time period (Rissler et al., 2004). If coupled to another suitable model, eg. the CAM-Oslo GCM, the aerosol modes can evolve over time, offering the next degree of complexity. Parameterisations can also employ a sectional representation of the aerosol size distribution (Abdul-Razzak and Ghan, 2002; Nenes and Seinfeld, 2003), which also allows the size distribution to evolve over time.

Perhaps unsurprisingly, a more complex representation of aerosol properties and processes leads to improvements in simulated aerosol forcing (Bellouin et al., 2013; Mann et al., 2012), as well as CCN concentrations (Weisenstein et al., 2007). However these approaches introduces a significant computational burden into simulations, which limits there applicability to short, limited area simulations.

Segal and Khain (2006) point out that an effective parameterisation should be as simple as possible, yet encompass all the governing factors affecting aerosol activation. This sentiment has also been echoed by Petters and Kreidenweis (2007). To this end, we present a parameterisation for estimating CCN concentrations which exploits the complexity of an aerosol model to accurately characterise chemical and physical properties of aerosols. All these detailed properties are then represented within a simple mathematical model, which is a function of the vertical velocity and atmospheric pressure. This represents a new approach for parameterising CCN for use in models. The parameterisation is developed for use in the ICON-LES model, from modelled aerosol data during the HOPE campaign. It is suggested the parameterisation is suitable for other time periods with a similar aerosol load.

2 Aerosol Data

The High Definition Clouds and Precipitation for advancing Climate Prediction (HD(CP)^2) project aims at improving our understanding of clouds and precipitation, by building and using a model capable of very high resolution simulations. A essential component of this project is the use of the ICOsahedral Nonhydrostatic (ICON) model to preform large eddy simulations (LES), as demonstrated by Dipankar et al. (2015). The ICON-LES model has no on-line aerosol scheme, which motivates the

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need for the new CCN parameterisation developed here. To achieve this, the COnsortium for Small-scale MOdelling (COSMO) meteorological model coupled to the MUlti-Scale Chemistry Aerosol Transport (MUSCAT) (Wolke et al., 2012) model was used to simulate the generation and transport of natural and anthropogenic aerosols to Europe. This time period covers the HD(CP)^2 Observational Prototype Experiment (HOPE) performed in Jülich, Germany, which will provide critical data for model evaluation.

The aerosol species simulated by COSMO-MUSCAT were: ammonium nitrate, ammonium sulfate, dust (5 sizes), elemental carbon, organic carbon, sea salt (2 sizes), and sulfate. Table 2 shows the chemical and physical properties of the aerosols simulated. The hygroscopicity parameter is κ, the mode standard deviation and mean radius are σ and r, respectively, and the density is given by ρ. Ghan et al. (2001) was used to define the hygroscopicity parameter for each species.

In COSMO-MUSCAT, the meteorological model COSMO, which is the operational forecast model of the German Weather Service (DWD), is coupled online with the chemistry transport model MUSCAT. Meteorological parameters such as humidity and temperature are interpolated and transferred from COSMO MUSCAT at each advection time step. This ensures that actual meteorological conditions are represented. MUSCAT computes atmospheric transport and chemical transformations of aerosol species and gas phase reactions. The transport processes include advection, turbulent diffusion, sedimentation, dry and wet deposition. In addition, size-resolved atmospheric particle number concentrations were simulated for Saharan dust aerosol. While the number distribution of secondary aerosol species are particularly important to determine cloud condensation nuclei concentrations, dust particles are efficient ice nuclei.

For the model results shown here, the horizontal grid spacing was 28 km, and 32 vertical layers were used. The domain considered in this study is between 48.25–54 °N, and 6–15 °E, shown in Figure 1. To ensure that the deviations in the modelled meteorological fields from the real atmosphere remain small, COSMO was reinitialised every 24 hours. COSMO ran for 48 hours at each cycle, and after 24 hours MUSCAT was restarted. Then, both models run parallel for 24 hours at each cycle. For the chemical compounds and aerosol species, MUSCAT computes total mass concentration. The model has been applied and tested for numerous case studies in Germany as well as annual simulations in the European domain (Wolke et al., 2012).

For the estimation of the aerosol number size distributions, the mode mean diameter, density and standard deviation of the lognormal mode have been predefined for each aerosol species. Dust size distributions have been described by Heinold et al. (2011). Sea salt modes are determined according to Gong (2003). The simulated mass concentrations were converted to total number concentrations by assuming spherical particles of a certain size and density individually for each component. Assuming a lognormal size distribution with a certain mean diameter and standard deviation, the total number concentration can then be used to estimate the number size distribution for each component. The sum of all individual size distributions results in the total particle size distribution, which can be compared to the observations.

The aerosol mixing state can influence aerosol size distribution and hygroscopicity, and hence CCN activity. Wang et al. (2010) shows that mixing state assumption is only important when primary organic aerosol and Black Carbon dominate aerosol volume. Here, aerosol compisition is mostly ammonium nitrate and ammonium sulfate, therefore mixing state assumption shouldn't affect results strongly. Furthermore, Ervens et al. (2010) shows that simple mixing state assumptions are not sufficient only very close to the pollution sources.

3 Aerosol Evaluation

Model simulations were performed for the period 26 March 2013 to 20 June 2013, which covers the period of the HOPE field campaign in Jülich. Furthermore, the model was evaluated for the site Melpitz near Leipzig (87 m a.s.l.; 51.53 N; 12.90 E) (Engler et al., 2007; Spindler et al., 2013), since no specific aerosol measurements were carried out during the campaign at the Jülich site. Therefore, comparisons of modelled and observed aerosol composition size distributions were performed at the Melpitz site. The station is situated in flat terrain, and no larger sources of pollution lie within close proximity to the station. Particle number size distributions at dry conditions were measured using a differential mobility particle sizer (TDMPS) (Birmili et al., 1999, 2015). The major ions and carbon species in the aerosol have been continuously measured from daily filter samples since 2003 (Spindler et al., 2013).

A comparison of modelled and observed chemical species is shown in Figure 2 for the time period of the HOPE Melpitz campaign. Only the results of modelled and observed concentrations of the species ammonium sulfate and ammonium nitrate as well as the small sea salt mode are shown, as these species dominate CCN concentrations over the model domain. The agreement between the model results and observations of the mass concentrations of secondary aerosol species as well as the sea salt is very good, both magnitude and temporal variability of the aerosol concentrations are well matched except for the first days of the time period, where the model underestimates the observed aerosol species. Total PM2.5, which is computed as the sum of all aerosol types excluding the supermicron size dust and sea salt fractions, is underestimated by the model by about a factor of 2. This underestimate can likely be tied to an underestimated submicron dust emission or secondary organic aerosol that is not considered by the model.

The modelled aerosol size distribution resulting from conversion of the simulated bulk aerosol concentration into size resolved aerosol concentration at the Jülich site is shown in Figure 3, for the example day 18 June 2013 at 12UTC. Here, ammonium sulfate contributes the main part to the modelled aerosol number concentrations. These results could not be verified at the Jülich site due to lack of observations. Therefore, comparisons of modelled and observed size distributions were performed at the Melpitz site (Figure 3b). While in the size range between 50 nm and 0.15 μm the model estimated number size distribution matches the observations well, the model underestimates the observations at smaller and larger particle sizes. This is also the case when comparing model results and observations for a full month (Figure 3c). The underestimation at smaller sizes is due to the fact that the nucleation mode, which is present in the measurements, is not taken into account in the model. However, at this size range such an underestimate is less important for diagnosing CCN concentrations. The underestimate of the model results at larger particle sizes (also reflected in underestimates of PM2.5 ad PM10 concentrations, not shown) may be more critical, however the number concentrations of the large particles are low. The model deficit may point to an aerosol type that is not included in the model, for example fugitive dust. Natural sea salt and desert dust aerosol are unlikely to be responsible for this deficit at the larger particle sizes, since the sea salt large mode was adjusted to observations, while independent comparisons of dust aerosol size distributions with observations during measurements at independent field campaigns have shown that simulated dust size distributions in the supermicron size range match well to ground and airborne dust size observations (Heinold et al., 2011).

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4 Aerosol and CCN concentrations during HOPE

Figure 4 shows the temporally averaged median and 85^{th} percentile vertical profiles of the number concentration of all aerosol species, and the resulting CCN number concentration at a prescribed vertical velocity of 0.5 ms^{-1}. To calculate the statistics, the domain wide median and 85^{th} percentile vertical profiles were first calculated at each time step. Then the mean of these profiles was taken over all time steps.

According to Figure 4, the dominant aerosols in the lower levels are ammonium nitrate and ammonium sulfate, and at higher levels the concentration of sulfate and elemental carbon become more significant. The concentrations of most aerosol species are constant at lower levels, and decrease at higher levels, with the rate of decrease varying between aerosol species. Sulfate is the exception, with concentrations increasing with altitude due to the nucleation of aitken mode particles in the upper troposphere. The median dust concentrations are constant with altitude, as already show by Hande et al. (2015) during a different time period.

The bottom panel of Figure 4 shows the temporally averaged 85^{th} percentile concentrations of aerosols and CCN at 0.5 ms^{-1}. Taking the ratio of the 85^{th} percentile concentrations to the median concentrations provides a rough measure of the spatial variability. For example, the 85^{th} percentile for ammonium nitrate is, on average, 5.6 times larger than the median. This increases to 44 times larger for dust, with the smallest difference being 2.1 times for organic carbon. In contrast to this, the 85^{th} percentile for CCN concentrations is only 1.8 times larger than the median concentrations, on average. This indicates, that while there may be significant spatial variability in aerosol concentrations, the spatial variability in CCN concentrations is significantly lower.

The spatial variability on shorter time scales, as well as the temporal variability over the course of the HOPE campaign are shown in Figure 5. The latitudinal and longitudinal median aerosol number concentrations and the resulting CCN number concentrations, averaged over all pressure levels and time steps for one day, 30 April 2013, are shown in the top and middle panel, and the bottom panel shows the temporal variability. For this specific day, ammonium nitrate shows a significant amount of variability in both latitude and longitude. Sea salt aerosols also have a large amount of variability with a strong north–south gradient, consistent with the source region being the oceans north of Germany. Indeed, at the north of the domain, sea salt becomes the dominant aerosol. The important point is that while there may be large variability in the spatial distribution of aerosols, the spatial variability in the resulting CCN is significantly lower.

The same conclusion regarding the horizontal homogeneity of CCN number concentrations over Germany can be obtained from analysing other days during the whole campaign time period. The spatial distribution of the aerosols can change significantly, particularly for ammonium nitrate, dust and sea salt. This is consistent with the findings from analysing the difference between the 85^{th} percentile and the median concentrations, as done above. However the resulting CCN number concentrations are much more homogeneous over the domain. Concentrations typically vary by around a factor of 2 over the domain. The temporal variability over the whole time period, on the other hand, is larger, as shown in the bottom panel of Figure 5. However, if shorter time periods of about one day are taken, this can be considered to be more constant.

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Boucher et al. (2013) also notes that there is low confidence in estimates of the anthropogenic fraction of CCN. In an effort to address this, the fraction that each individual aerosol species contributes to the calculated CCN concentrations is shown in Table 3. Here, CCN concentrations are calculated at a vertical velocity of 0.5 ms^{-1}, and according to Abdul-Razzak et al. (1998), this gives an activated fraction of approximately 0.1 for ammonium sulfate aerosols at 10 °C and 800 hPa. The same activated fraction is found at a supersaturation of approximately 0.2%, which is commonly used by other authors to calculate CCN concentrations (Pierce and Adams, 2009; Wang and Penner, 2009). Of course this depends on the particular aerosol species and thermodynamic conditions, however it indicates that the results in Table 3 are roughly comparable to those of other studies.

Table 3 indicates that the CCN number concentrations over Germany are dominated by CCN formed on anthropogenic aerosols. Specifically, ammonium sulfate and sulfate dominate CCN production in the upper levels, and ammonium nitrate and ammonium sulfate are the dominant aerosols in the lower levels. Although elemental carbon aerosols have a high concentration throughout the atmosphere, their contribution to CCN is negligible due to the very low hygroscopicity. The sea salt aerosols, which are the most hygroscopic, only play a minor role in CCN production over the continent. Organic carbon and desert dust aerosols have the same hygroscopicity, therefore the differences in CCN production are due to differences in the number concentrations of aerosols. These results are outside the upper bound of estimates of the global mean anthropogenic fraction of CCN. Boucher et al. (2013) combines numerous studies to suggest the anthropogenic fraction of CCN is between 0.25 and 0.66, however the authors did note the large uncertainties and large regional differences in these estimates. Furthermore, sea salt aerosols would be a larger contributor to the global mean, and this would act to reduce the anthropogenic fraction of CCN compared to the largely continental conditions over Germany.

5 Parameterisation Development

The previous section demonstrated that the median vertical profile of CCN number concentrations can be considered representative of the conditions over Germany during the HOPE campaign if short time periods are considered. Here, a parameterisation of CCN concentrations is constructed, which is a function of the atmospheric pressure and the vertical velocity. Using Abdul-Razzak and Ghan (2000), median CCN number concentrations were calculated at 40 different vertical velocities for each time step of modelled aerosol data. The average over all time steps in one day was computed, and this was used to define a series of best fit functions. In this way, aerosol physical and chemical properties are included through the use of Abdul-Razzak and Ghan (2000), as well as the important dependance on the vertical velocity at various pressure levels.

Figure 6 shows the CCN activation spectrum at each of the 32 pressure levels, for one day during HOPE, as well as the average of all data used in this study. Pressure levels closer to the ground have larger CCN number concentrations. The characteristic shape of this activation spectrum can be described, at each pressure level, by the following relation:

$$CCN(w) = A \times arctan(B \times log(w) + C) + D \quad (1)$$

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where *log(w)* is the natural logarithm of vertical velocity in ms^{-1}. This best fit function for each pressure level is shown as the red lines in Figure 6. This function provides a very good fit to the modelled activation spectrum, particularly in the range of vertical velocities between 0.01 ms^{-1} and 50 ms^{-1}. At very small vertical velocities, the function can produce negative CCN number concentrations, particularly for pressure levels closer to the ground. This is the largest source of discrepancies between the parameterisation and the modelled CCN data. The parameters *A*, *B*, *C*, and *D* act to control the scale, shape, and position of the curve at each pressure level, and themselves have a characteristic variation with pressure, as shown in Figure 7.

Curves of the following form can be fit to each of these parameters:

$$A(P) = a_1 \times arctan(b_1 \times P - c_1) + d_1 \tag{2}$$

$$B(P) = a_2 \times arctan(b_2 \times P - c_2) + d_2 \tag{3}$$

$$C(P) = a_3 \times arctan(b_3 \times P - c_3) + d_3 \tag{4}$$

$$D(P) = a_4 \times arctan(b_4 \times P - c_4) + d_4 \tag{5}$$

where *P* is pressure in Pascals. The shape of these curves is influenced by the structure of the atmosphere, and in some cases a different functional form may be more approapriate than Equations (2) to (5). The key to developing a parameterisation using this technique is that a function of any type can be fit to these parameters. In some examples examined during HOPE, the fit for the *B* parameter can be poor. This often occurs when there is a second increase in this parameter above about 700 hPa. However, the influence this parameter has on the final parameterised CCN concentrations is small, and it can be seen from the bottom panels of Figure 7, that on average the fit provided by Equations (2) to (5) is very good. Combining Equations (1) to (5), the CCN number concentrations (m^{-3}) are defined as:

$$CCN(w,P) = A(P) \times arctan(B(P) \times log(w) + C(P)) + D(P) \tag{6}$$

where the 16 parameters a_1 to d_4 must be defined for each time period. These parameters are provided in Table 1 of the Appendix for each day of HOPE, as well as the mean over the whole time period.

Figure 8 shows the parameterisation compared to CCN number concentrations calculated directly from the modelled aerosol data, at multiple vertical velocities. As it can be seen, discrepancies between the parameterised CCN concentrations and those calculated directly from the model data are most significant at vertical velocities less than about 0.02 ms^{-1}, and pressure levels lower than 800 hPa. However, at vertical velocities greater than about 0.1 ms^{-1}, the parameterisation derived above provides an excellent fit for the modelled CCN concentrations. These larger vertical velocities are most relevant for conditions within

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clouds. In order to prevent unrealistically low CCN number concentrations, it is recommended to implement a minimum CCN concentrations of 10^7 m^{-3}.

This approach to parameterising CCN concentrations has an advantage over other traditional methods. The CCN parameterisation developed by Segal and Khain (2006) assumes a constant CCN concentration up to a specified height, above which the concentration decreases exponentially. This would only be a suitable representation in the case of a well mixed boundary layer, with no aerosol, and hence CCN, production in the upper levels. The parameterisation developed above is flexible enough to account for an atypical vertical distribution of CCN. For example, the top panel of Figure 8 shows no well mixed region, instead an almost linear decrease in CCN concentrations from the surface through to the mid–troposphere. Furthermore, Figure 4 implies sulfate aerosols can be produced in the upper levels, therefore the rate of decrease in CCN number concentrations above the boundary layer may not be exponential. It is suggested that the vertical profile of CCN number concentration obtained through this new parameterisation provides a more accurate representation.

Finally, Figure 9 shows scatter diagrams of the modelled CCN number concentrations against the parameterised CCN number concentrations for 3 pressure levels, and 3 vertical velocities. Overall, there is no significant bias in the parameterised CCN number concentrations. At high pressures, the agreement between the parameterised and modelled number concentrations is excellent, but the differences between the two increase with decreasing pressure. At 1.14 ms^{-1}, the average absolute magnitude of the difference between the parameterised and modelled number concentrations is 8 %, 22 %, and 22 % of the modelled concentrations at 911, 715 and 516 hPa respectively. The three points at 516 hPa and a parameterised number concentrations of 1×10^7 m^{-3} would have very low number concentrations according to Equation (6), hence the values must be adjusted to a minimum of 1×10^7 m^{-3}. These profiles are from 9–11 April 2013.

6 Conclusions

The COSMO–MUSCAT model was used to simulate the generation and transport of aerosols to Europe during the HOPE campaign. An evaluation of the modelled aerosol concentrations with available observations shows good agreement. The mass concentrations and temporal variability of ammonium sulfate, ammonium nitrate, and sea salt concentrations are in very good agreement with observations from Melpitz. Total PM2.5 concentrations is underestimated by the model by a factor of about 2. The size distribution from the model also agrees well with observations, however there is a slight underestimate at smaller and larger particle sizes.

From these aerosol concentrations, CCN number concentrations were calculated. The analysis demonstrated that while there may be large variability in aerosol concentrations throughout the domain considered here, the spatial variability of the resulting CCN concentrations is significantly lower, typically only varying by a factor of 2. There is, on the other hand, a larger amount of temporal variability over the time period. This implies that the median vertical profile of CCN number concentrations is most representative of the conditions over Germany if short time periods are considered.

The anthropogenic fraction of CCN was found to be large over continental Germany, over 90% near the surface, decreasing to about 80% in the mid–troposphere. This is larger than estimates of the global mean, and demonstrates the significant impact that anthropogenic aerosols have on cloud properties.

A parameterisation of CCN number concentrations was developed, using a series of best fit functions to capture the dependency of CCN activation on vertical velocity at different pressure levels. In this parameterisation, the influence of aerosol physical and chemical properties on CCN are included through the prior use of a detailed aerosol activation scheme. The parameterised CCN number concentrations compare well to the number concentrations calculated directly from the modelled aerosol data, except at very low vertical velocities and pressure levels close to the ground. This represents a new approach for parameterising CCN for use in models, which to the authors knowledge, has not been demonstrated before. As long as the technique provides adequate fits of all the free parameters in equation (6), this technique can be employed to parameterise CCN in other regions and over other time periods.

Appendix A

Date	a1	b1	c1	d1
2013-03-26	257027511.629	0.000199979502434	17.6117849009	422546964.512
2013-03-27	257259587.478	0.000201760653742	17.0754515981	458225862.794
2013-03-28	248134036.277	0.000248771867112	19.8502078315	486429124.702
2013-03-29	293002066.039	0.000225778435403	17.3399883006	557584166.907
2013-03-30	252381141.108	0.000128527088931	10.0148105244	425988267.377
2013-03-31	315995778.379	0.000105411525029	8.52132974576	476826117.331
2013-04-01	383647486.647	9.53987511704e-05	8.32043216657	570523268.512
2013-04-02	324811271.228	0.000138642834665	12.2490672445	530652188.373
2013-04-03	223154550.008	0.000210211401757	18.2301131592	389572102.855
2013-04-04	185174471.514	0.00027916990502	24.9758011901	312654653.088
2013-04-05	123965671.328	0.000220481723183	20.2930529312	214063096.868
2013-04-06	186141937.179	0.000119207983665	10.6880203951	289075639.249
2013-04-07	197619469.896	0.000195683880524	16.6673454676	306111694.706
2013-04-08	228555620.453	0.000209968372046	18.0177227325	353752154.783
2013-04-09	165782282.315	0.000158862778002	13.3344716637	230965254.836
2013-04-10	149088253.239	0.000197799417401	16.8329323519	210898751.656
2013-04-11	188933606.377	0.000205926861534	18.7422637505	277465364.811
2013-04-12	151501378.329	0.000154496611164	13.3548084787	222893897.281
2013-04-13	182711318.548	0.000186202391181	15.4762408599	266493038.479
2013-04-14	165170320.037	0.000278508634965	26.9803029196	260454659.599
2013-04-15	195696398.475	0.000105519547855	9.51386317268	287279759.576
2013-04-16	224420005.659	0.000293961293006	23.3619212815	350287206.787
2013-04-17	132585467.481	0.00026097002081	22.2848137664	220276289.561
2013-04-18	66896808.8009	0.000622874606996	47.5770620781	149054005.269
2013-04-19	70902191.9176	0.000268682081376	22.3654923985	113755021.935

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2013-04-20	54046034.4806	0.000463840088873	39.6472498837	93778930.966
2013-04-21	237046050.889	0.000197584785429	15.7411031906	354983011.683
2013-04-22	350703557.487	0.000248652491121	19.1756371885	535342752.709
2013-04-23	277648946.464	0.000220951487302	18.756838732	417530444.244
2013-04-24	326536851.625	0.000476307032206	40.0176304001	520404849.951
2013-04-25	297051358.433	0.000160822913339	12.9411417647	437962207.588
2013-04-26	123186926.017	0.000195179520486	13.4949151996	183690869.78
2013-04-27	110210087.645	0.000239022945279	21.8073672723	169437053.325
2013-04-28	164323035.999	0.00015523807202	13.5145674577	236667368.805
2013-04-29	242468712.867	0.0001916705582	16.2115926716	355241545.175
2013-04-30	268062705.206	9.00228432568e-05	8.67002079484	369899109.621
2013-05-01	102511887.928	0.000421989534664	34.118007519	167341263.238
2013-05-02	188914931.006	0.000531227091679	41.6626456765	299914013.763
2013-05-03	166534415.574	0.000345442088574	27.3432677906	257750601.317
2013-05-04	170209024.489	0.000349594756448	27.9177073888	264146914.747
2013-05-05	169785924.886	0.000831242458893	66.8062397787	285125428.499
2013-05-06	278790529.473	0.000451534894898	34.9583814579	443180679.414
2013-05-07	242323023.311	0.000233685933182	17.7939128097	367463885.465
2013-05-08	274335739.495	0.000213306540004	17.0245331362	412727950.786
2013-05-09	113333109.973	0.000509886806569	38.659883261	195376416.716
2013-05-10	105744547.317	0.000538143118997	42.6695357287	172643018.723
2013-05-11	162385382.85	0.000332727679016	26.8845754909	251110679.598
2013-05-12	102248916.147	0.000541014965278	43.8361853001	175912999.174
2013-05-13	143779878.725	0.000155656177112	13.6193322737	227494184.849
2013-05-14	118394120.321	0.000336419574937	28.0940496016	211132273.023
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2013-05-17	177831147.397	0.000101960076261	9.76398595204	260133006.929
2013-05-18	104881119.861	0.000246405889005	21.9696591509	182569272.058
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2013-06-07	135064790.26	0.000388462942097	29.6327517089	263840022.66
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2013-06-14	101662155.522	0.00128461981711	104.950293772	217280192.35
2013-06-15	125114045.253	0.00164830616793	127.12314395	257013581.432
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2013-04-22	0.0527699977656	0.000248259433057	17.2667172986	0.72497660722
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2013-06-05	0.0121120858222	0.00074709092356	60.0364656402	0.676774244876
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2013-04-24	-0.465259558225	0.000237161116198	19.1049134773	1.08031121538
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2013-06-13	0.144601621176	-0.00045898983022	-39.7459325336	0.961554360906
2013-06-14	-0.173194732078	0.000971333233174	78.8987838138	1.10277014402
2013-06-15	-0.219682412065	0.000723465343821	55.7467841028	1.07610272578
2013-06-16	-0.169997837704	0.000748813288464	62.7335523194	1.13678591019
All Data	-0.436109722435	7.71432656612e-05	5.65244631785	1.22261713713

Date	a4	b4	c4	d4
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2013-03-27	309844331.918	0.000201553447317	17.0889369229	558425882.48
2013-03-28	303587929.431	0.000251184401777	20.0776058128	600415097.602
2013-03-29	358319870.576	0.000235307642443	18.109198242	689285340.24
2013-03-30	306411494.19	0.000130502738578	10.2047241636	525245208.824
2013-03-31	388602484.531	0.000106640142718	8.63617368737	591782562.502
2013-04-01	476072376.341	9.54062745542e-05	8.34686788976	712002557.477
2013-04-02	400738826.224	0.000137261994404	12.1441852379	657786306.277
2013-04-03	271154223.173	0.000211618554738	18.375924686	477963001.566
2013-04-04	222005914.307	0.000288026568537	25.7839773553	380103114.138
2013-04-05	146113683.536	0.000226728363976	20.8773726066	258303073.087
2013-04-06	227357309.834	0.000119261373742	10.6947244824	356992826.431
2013-04-07	241308439.109	0.000199317529083	16.9672587029	378439187.206
2013-04-08	280111919.968	0.000209214019672	17.9793642396	437741879.199
2013-04-09	201988263.429	0.000156175726257	13.1195924886	282584675.128
2013-04-10	181147956.533	0.000200223115362	17.0477941637	259133440.218
2013-04-11	228846926.496	0.000206978315709	18.8460886402	338431888.439
2013-04-12	183631089.844	0.000153283558203	13.226675174	272877803.21
2013-04-13	223513894.045	0.000185040047855	15.4004918451	328717289.549
2013-04-14	199272578.736	0.000279851195086	27.1229057226	318094162.339
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2013-04-16	275956307.198	0.000300567567101	23.9183353439	434779360.181
2013-04-17	162262731.983	0.000262094428674	22.3945772632	272763175.919
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2013-04-23	341478481.841	0.000224876807177	19.0903094313	517788768.446
2013-04-24	407017229.474	0.000487594856181	40.9747483446	653016436.865
2013-04-25	368817155.294	0.000162747488109	13.0976538218	546387196.913

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2013-04-26	149721272.911	0.000203000520319	14.0650317208	227118473.008
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2013-04-28	200772495.141	0.000157057847042	13.646482707	291386102.455
2013-04-29	298588454.619	0.000193175186475	16.3641258221	440534263.922
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2013-05-01	125985285.773	0.000423966625778	34.2688922351	207432107.013
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2013-05-03	203776163.79	0.0003437601979	27.2333299684	317651818.018
2013-05-04	208338967.959	0.000346456469493	27.6854907624	325702553.336
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2013-05-07	299783172.592	0.000235981220195	17.9925942326	457931647.005
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2013-05-14	143307785.834	0.000341916985123	28.5463023673	262248737.61
2013-05-15	171687143.506	0.000204960171235	16.8430743062	288242524.489
2013-05-16	201124120.263	0.000199727851722	16.4635685193	300436734.559
2013-05-17	216048762.633	0.000106256317825	10.1785101296	321330376.197
2013-05-18	127019310.246	0.000251878447586	22.4725980735	225359020.405
2013-05-19	216139229.118	0.00021017737448	18.8082043681	347954786.732
2013-05-20	142802725.584	0.00022408357457	18.9053267444	250315544.354
2013-05-21	198870491.145	0.000117988992593	10.139042474	307531288.823
2013-05-22	119432620.247	2.43039229589e-05	2.45632747881	157922601.653
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2013-05-24	84013680.5912	0.000185305147735	15.5745760304	167222406.923
2013-05-25	139565588.11	7.67575081862e-05	7.44485213716	214762700.992
2013-05-26	45132682.539	0.000361420216266	33.2187435764	91005818.8736
2013-05-27	125197132.219	0.000124622035228	10.5772038594	199654328.249
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2013-05-29	290645572.411	0.000151162246919	12.2349270666	420301468.84
2013-05-30	211052359.052	0.000220307547106	17.7401273148	355841319.637
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2013-06-02	17968652.1809	0.000996972925851	90.3011578173	144213180.306
2013-06-03	34016040.9651	0.000659872472514	58.6294118973	162640835.637
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2013-06-05	97909831.7666	0.000120067468126	9.79807570646	221959838.278
2013-06-06	101756470.014	0.000456646803813	35.3885995398	243262397.72
2013-06-07	161546476.68	0.00045407580782	34.799243064	334220452.75
2013-06-08	203720180.213	0.000482356961371	37.0202588174	405209487.11

2013-06-09	158442495.012	0.000269794457705	20.8081300188	314318566.708
2013-06-10	141865122.829	0.000156947950278	13.0695209013	263075585.99
2013-06-11	235733177.228	0.000230751828926	18.1591128028	410288514.094
2013-06-12	415210885.836	0.000306663251406	24.3201029806	674658738.154
2013-06-13	147073267.639	0.000309682098903	25.9637042653	288869265.912
2013-06-14	120316710.214	0.00156899264184	128.236691148	269620027.277
2013-06-15	150145578.989	0.00201136712971	155.146937742	320889638.795
2013-06-16	102540538.867	0.000827094676632	67.9962750683	224807397.25
All Data	199788909.439	0.000182424683277	14.8932330043	328676886.493

Table 1: Parameters defining the CCN parameterisation.

Author contributions. Christa Engler, working with Ina Tegen, ran the COSMO-MUSCAT model, and performed the aerosol evaluation. Luke B. Hande, working with Corinna Hoose, developed the CCN parameterisation. Luke B. Hande prepared the manuscript with contributions from all co-authors.

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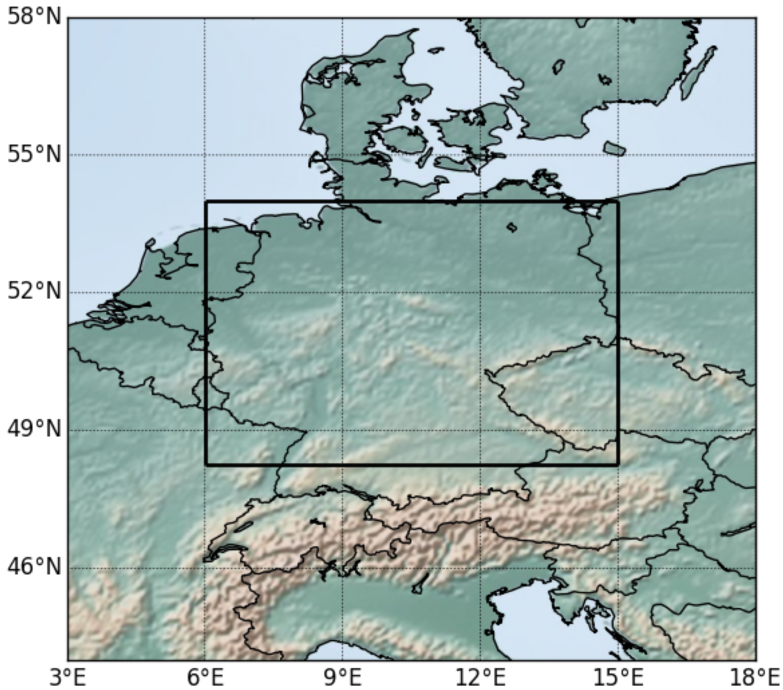

Figure 1. Domain over Germany used in this study.

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Species	κ	σ (μm)	r (μm)	ρ (kg m^{-3})
Amm Nitrate	0.54	1.6	0.05	1.725
Amm Sulfate	0.51	1.6	0.05	1.77
Dust 1	0.14	2.0	0.2	2.65
Dust 2	0.14	2.0	0.6	2.65
Dust 3	0.14	2.0	1.75	2.65
Dust 4	0.14	2.0	5.25	2.65
Dust 5	0.14	2.0	15.95	2.65
Elemental C	5×10^{-7}	1.8	0.03	1.8
Organic C	0.14	1.8	0.055	1.0
Sea Salt 1	1.16	1.8	0.065	2.2
Sea Salt 2	1.16	1.7	0.645	2.2
Sulfate	0.236	1.6	0.05	1.8

Table 2. Aerosol physical and chemical properties.

	Amm Nitr (A)	Amm Sulf (A)	Elem C (A)	Sulf (A)	Total A
507 hPa	3.55	42.96	1.57e-5	32.58	79.09

	Dust (N)	Org C (N)	SS (N)		Total N
507 hPa	2.23e-2	16.89	3.99		20.91

	Amm Nitr (A)	Amm Sulf (A)	Elem C (A)	Sulf (A)	Total A
906 hPa	46.18	46.53	1.26e-7	2.66e-2	92.74

	Dust (N)	Org C (N)	SS (N)		Total N
906 hPa	1.21e-3	5.24	2.02		7.26

Table 3. Percentage contribution of each aerosol species to total CCN number concentrations at 0.5 ms^{-1} for 507 hPa and 906 hPa. Each aerosol species is indicated as either anthropogenic (A), or natural (N).

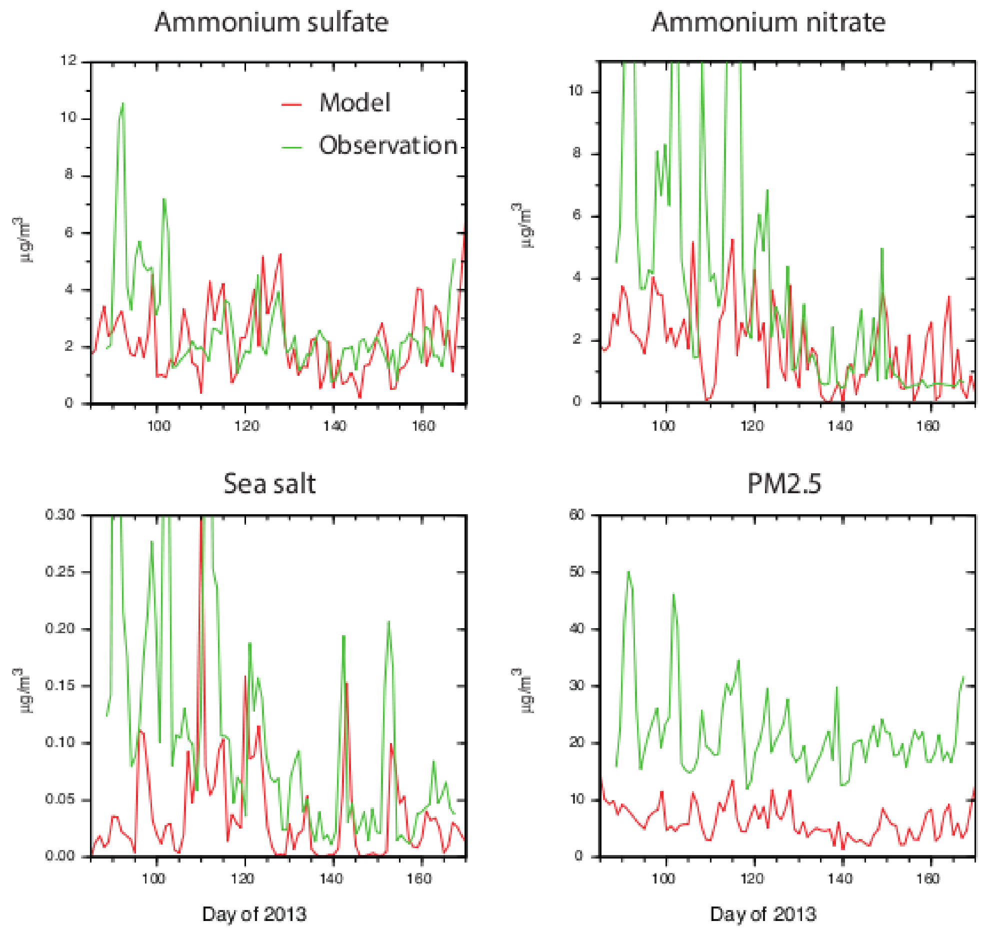

Figure 2. Comparison of the modelled and observed concentrations of aerosol species ammonium sulfate, ammonium nitrate, sea salt and total PM2.5 at the Melpitz site for the HOPE simulation period in spring 2013.

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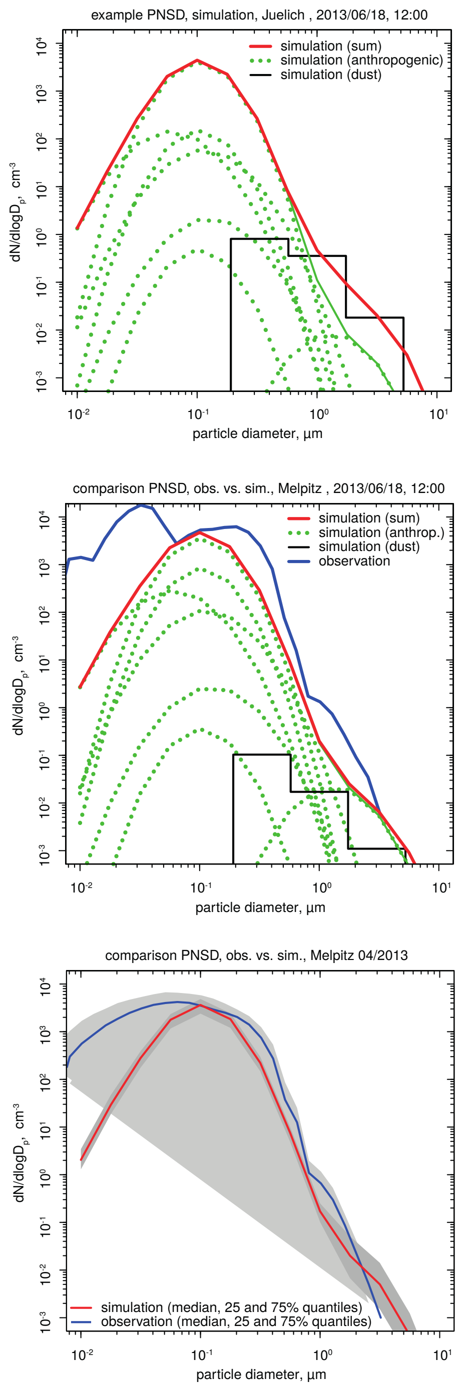

Figure 3. Aerosol particle number size distribution for 18 June 2013 at the sites Jülich (a) and Melpitz (b), and for the month April 2013 at Melpitz (c). The red lined mark the resulting simulated aerosol size distributions for the sum of the individual species (dotted green lines). Black lines represent modelled number size distribution of dust transported from the Sahara desert to the sites Jülich and Melpitz, respectively.

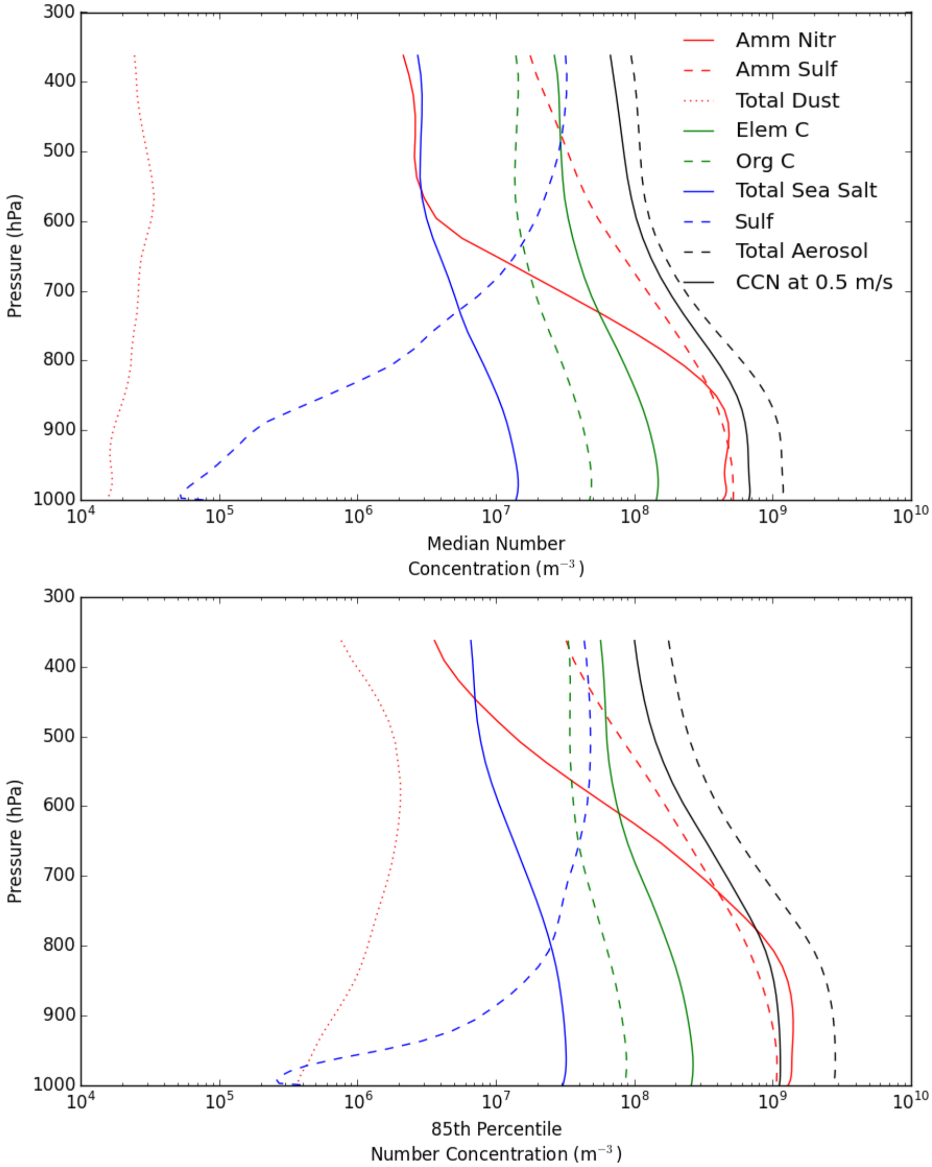

Figure 4. Temporally averaged median (top) and 85th percentile (bottom) number concentration for aerosols and CCN at 0.5 ms^{-1} from 25 March 2013–16 June 1013

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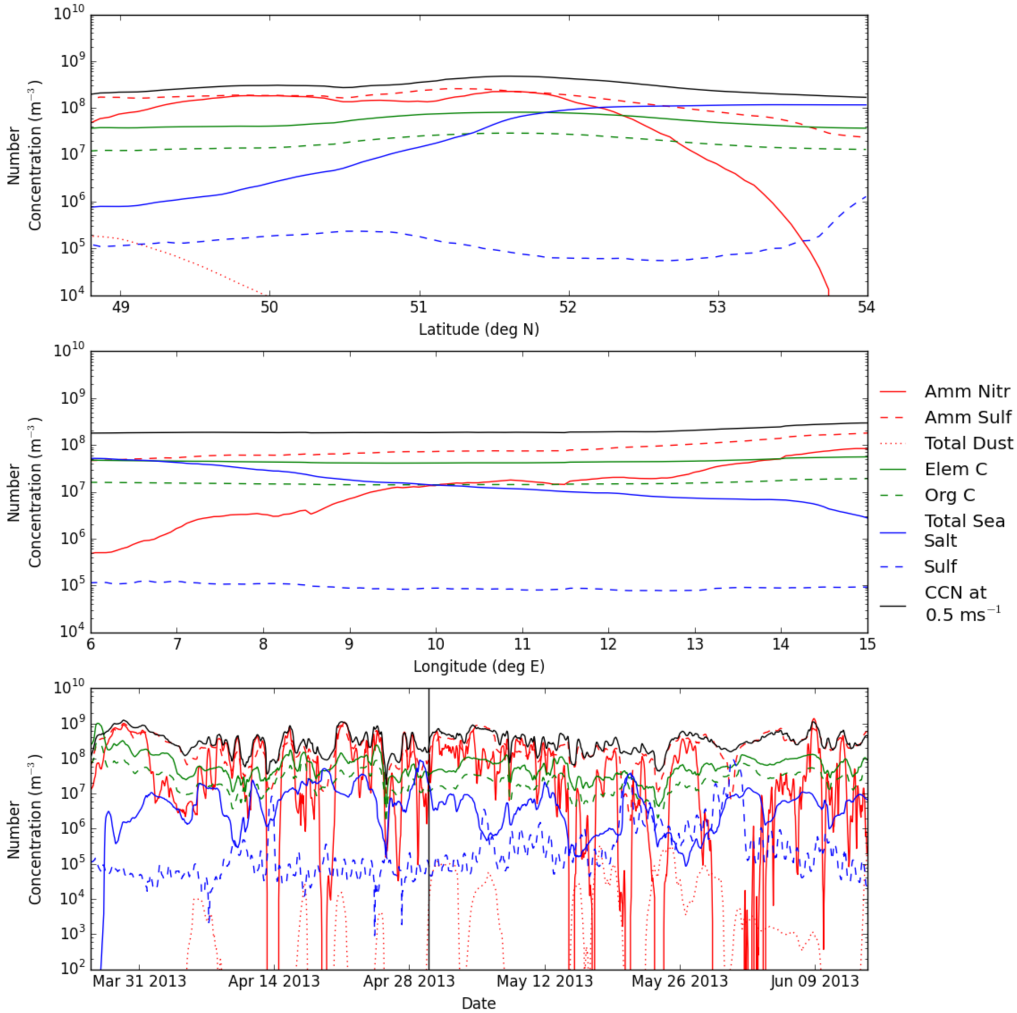

Figure 5. Latitudinal (top) and longitudinal (middle) median number concentration for aerosols and CCN at 0.5 ms^{-1} for 30 April 2013. Domain wide median number concentration (bottom) for aerosols and CCN at 0.5 ms^{-1} for the HOPE campaign time period. The solid vertical line indicates the time period of the two upper panels.

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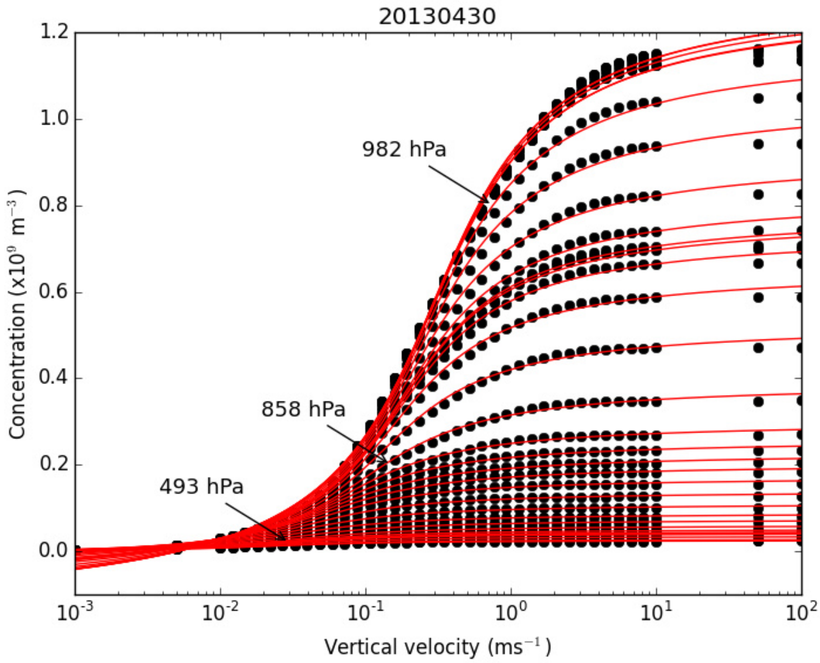

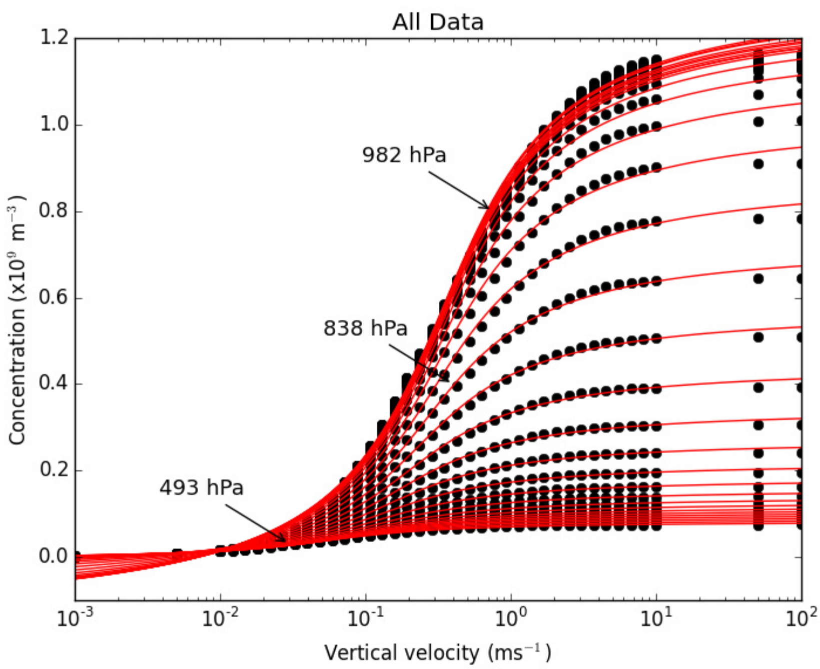

Figure 6. CCN activation spectrum for 30 April 2013 (top) and all data (bottom). Black circles represent the model data, red lines are the best fit functions.

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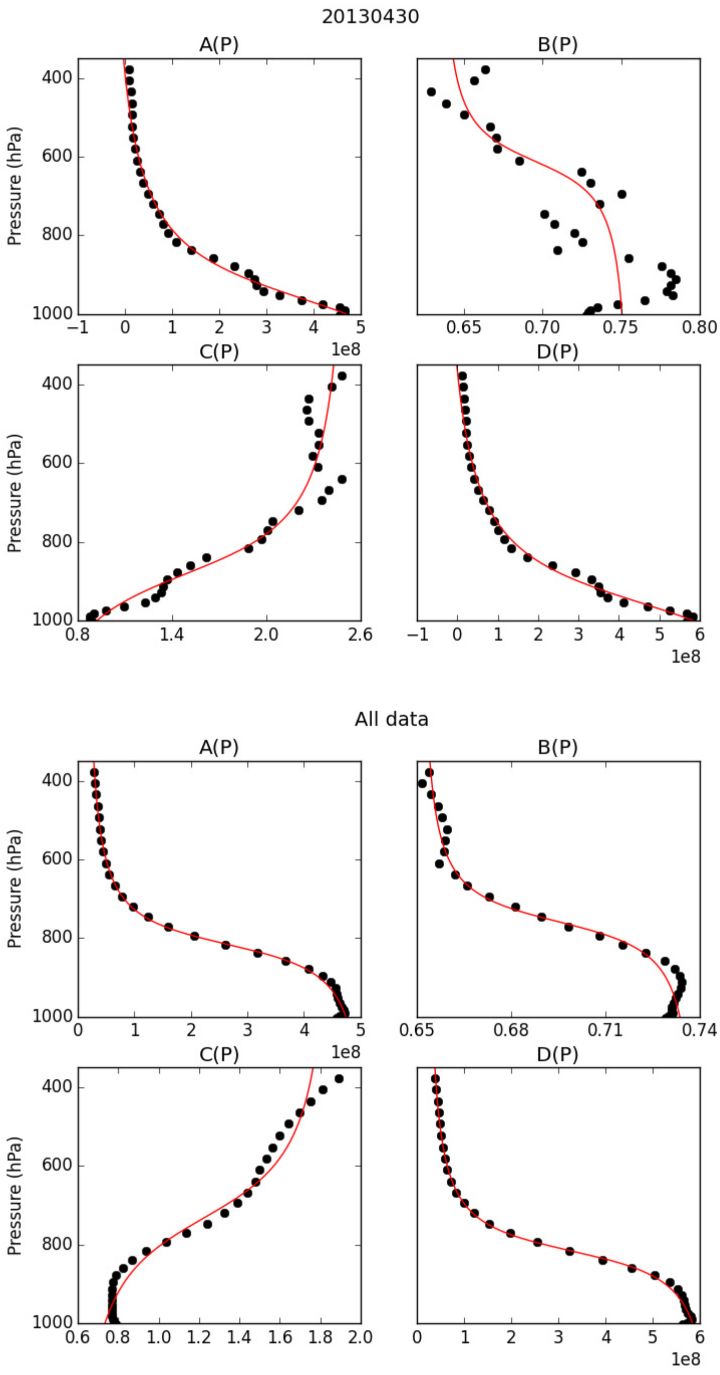

Figure 7. Parameters A, B, C, and D as a function of pressure for 30 April 2013 (top) and all data (bottom). Black circles are represent the model data, red lines are the best fit functions.

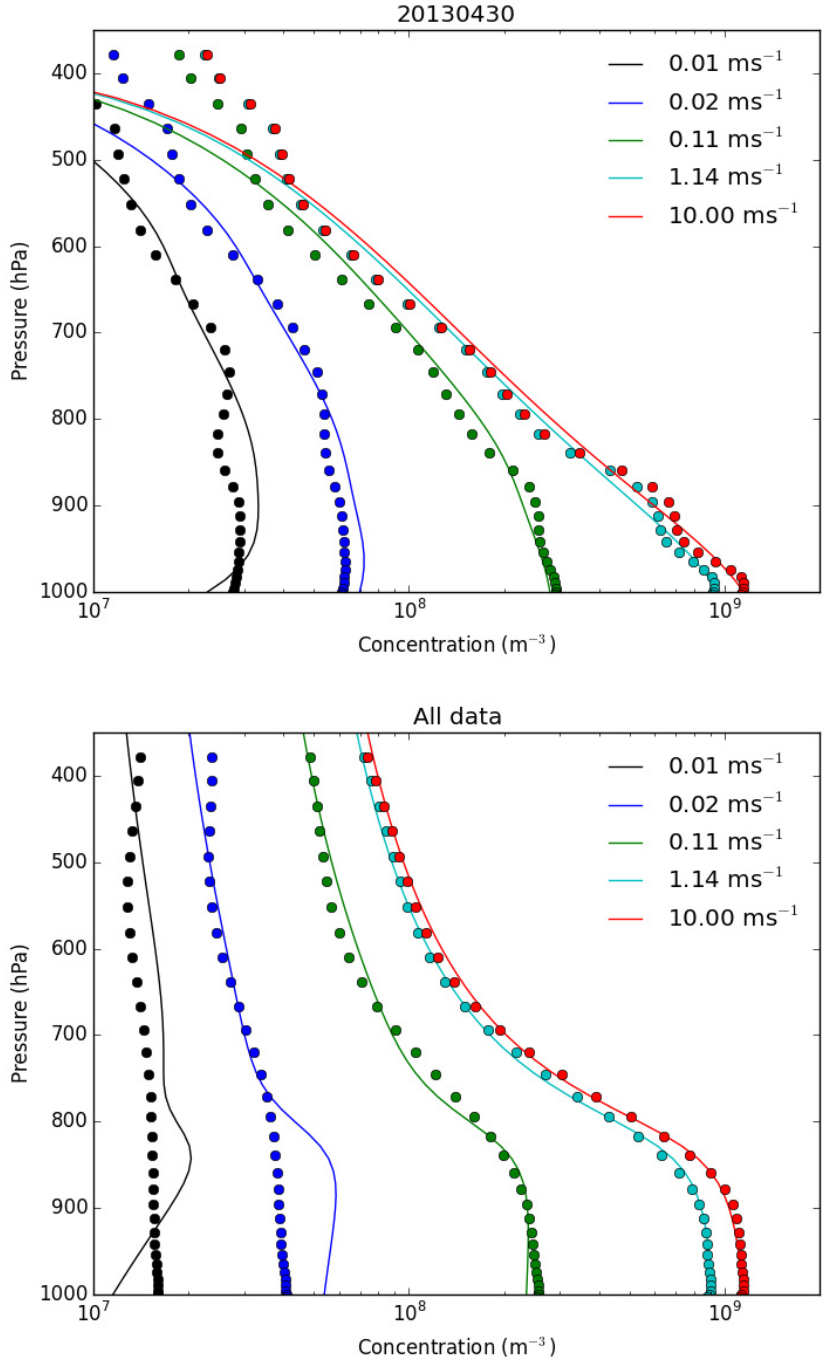

Figure 8. Modelled (circles) and parameterised (lines) CCN concentrations at multiple vertical velocities for 30 April 2013 (top) and all data (bottom).

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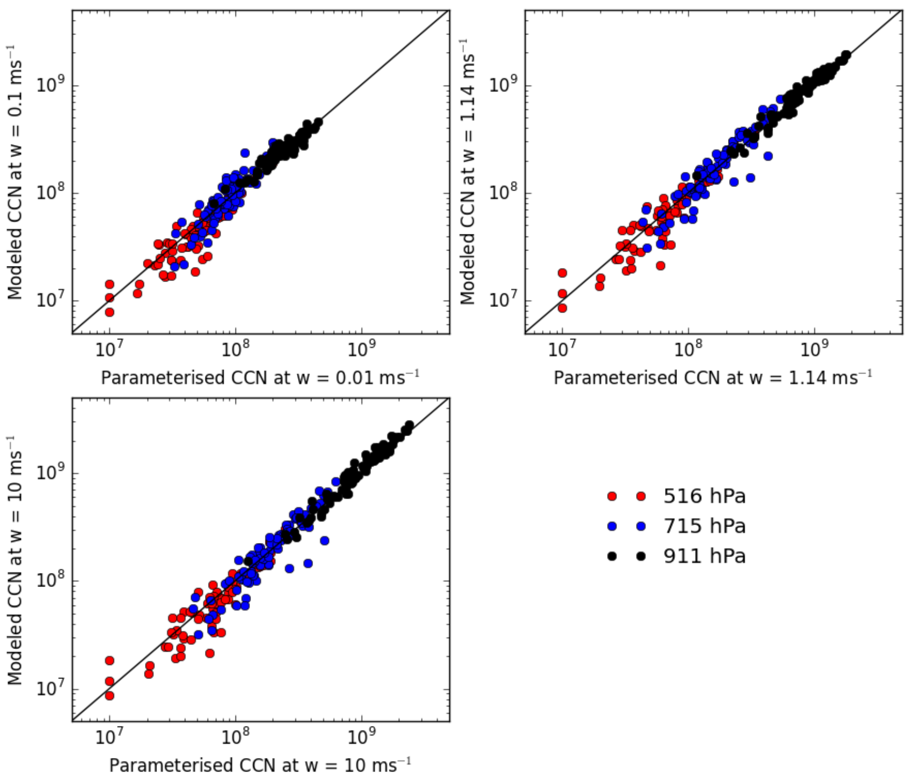

Figure 9. Scatter diagrams of modelled CCN number concentrations against parameterised CCN number concentrations, for w = 0.01 (upper left), 1.14 (upper right), and 10 ms^{-1} (lower left), and for 516 (red), 715 (blue), and 911 hPa (black).