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CCN predictions using simplified assumptions of organic aerosol composition and mixing state: a synthesis from six different locations

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Abstract

An accurate but simple quantification of the fraction of aerosol particles that can act as cloud condensation nuclei (CCN) is needed for implementation in large-scale models. Data on aerosol size distribution, chemical composition, and CCN concentration from six different locations have been analyzed to explore the extent to which simple assumptions of composition and mixing state of the organic fraction can reproduce measured CCN number concentrations.

Fresher pollution aerosol as encountered in Riverside, CA, and the ship channel in Houston, TX, cannot be represented without knowledge of more complex (size-resolved) composition. For aerosol that has experienced processing (Mexico City, Holme Moss (UK), Point Reyes (CA), and Chebogue Point (Canada)), CCN can be predicted within a factor of two assuming either externally or internally mixed soluble organics although these simplified compositions/mixing states might not represent the actual properties of ambient aerosol populations. Under typical conditions, a factor of two uncertainty in CCN concentration translates to an uncertainty of ~15% in cloud drop concentration, which might be adequate for large-scale models given the much larger uncertainty in cloudiness.

1 Introduction

Aerosol-cloud interactions represent one of the largest uncertainties in estimating the effects of aerosol on radiative forcing. One key parameter for this estimate is the fraction of aerosol particles can act as cloud condensation nuclei (CCN) and form cloud droplets. The propensity of a particle to form CCN depends on its size, chemical composition and the supersaturation to which it is exposed. Whereas size distributions are routinely measured in field experiments, the full characterization of the chemical composition presents a major challenge since, in particular, the organic fraction of particles can be composed of hundreds of compounds with different physicochemical properties

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(e.g., surface tension, solubility, degree of dissociation, molecular weight). In laboratory and theoretical studies, it has been shown that these properties can enhance or reduce the CCN ability of organic particles as compared to better-characterized inorganic particles (Cruz and Pandis, 2000; Corrigan and Novakov, 1999). For ambient particle populations, it is not feasible to consider all individual compounds due to the far from complete characterization of the organic fraction at the molecular level, and the computational burden the description of hundreds of individual compounds represents in models.

There is no consensus about the importance of detailed knowledge of aerosol composition (including mixing state) in studies that compare measured and modeled CCN number concentrations ("CCN closure"). Some studies report that this information is of minor importance for successful CCN closure and that aerosol size distribution largely determines the fraction that can be activated at a given supersaturation (Dusek et al., 2006; Ervens et al., 2007; Conant et al., 2004). Other studies show that measured CCN number concentrations can only be reproduced if detailed organic properties/mixing state are taken into account (Mircea et al., 2005; Stroud et al., 2007; Cubison et al., 2008).

With increasing distance from emission sources, particles become mixed by both physical processes (e.g., coagulation or condensation of semivolatile and low-volatility organic and inorganic compounds) or chemical processes (e.g., oxidation of primary (organic) species yielding more water-soluble products, and particle-phase reactions). These ageing processes lead to growth of the particles, i.e. increasing their CCN ability, and/or to an increase in the hygroscopic fraction and, thus, to a reduction in the size threshold ("critical diameter") above which particles can be activated at a given supersaturation (Furutani et al., 2008; Petters et al., 2006).

Most CCN studies published to date have been limited to sampling at one site and, thus, it is not clear how to generalize their conclusions about CCN activation as a function of aerosol mixing state and organic solubility. In a global model study, it has been shown that different assumptions on mixing state and organic solubility might signifi-

cantly affect CCN number concentrations (Pierce et al., 2007). The motivation of the current study is to explore the applicability of these simplifying composition/mixing state assumptions in CCN closure studies, using a consistent model approach, based on data sets collected at very different locations and distances from sources. Unlike previous CCN studies (some of which analyzed in detail the same data sets as those considered here), it is not attempted to deduce exact size/composition/mixing parameters of the CCN-active particle population based on all measurements, but rather to evaluate the extent to which simple assumptions can reproduce measured CCN number concentrations across all data sets. A simple treatment of CCN composition/mixing state as a function of distance from sources would be highly useful for the description of aerosol-cloud interactions in large-scale models.

2 Data sets

CCN data sets at six locations that differ in proximity to pollution sources, aerosol loading and composition have been analyzed; details on these data sets and corresponding publications are summarized in Table 1. Three data sets are split into two subsets: During the first period of the MASE experiment at Point Reyes, CA (PYE), air masses were transported mainly from the west and did not have any land contact over the three days prior to sampling, whereas air masses during the remainder of the experiment came from the north and north east and thus had continental influence (Berkowitz, 2009). For some periods during the ICARTT experiment at Chebogue Point (CBG), analysis of the organic mass fraction (OMF) of the aerosol using the Aerodyne aerosol mass spectrometer (AMS) allowed their classification as either anthropogenically or biogenically influenced, respectively (Williams et al., 2007; Holzinger et al., 2007; Zhang, 2009). Note that not all data points could be unambiguously ascribed to such air masses. During GoMACCS, CCN data were collected on the NOAA research vessel Ronald H. Brown both in the Houston Ship Channel (HSC) and along the Houston Gulf Coast (HGC). It has been shown that aerosol composition and processing can

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differ significantly in these two areas due to different emissions (Bates et al., 2008; Bahreini et al., 2009). At Mexico City T0 (MEX) and Riverside (RVS), the sampling took place at ground sites that were located near and downwind of significant pollution sources. Emissions at the mountain-top at Holme Moss, UK, (HOM) are characterized by the plumes of Manchester, a conurbation of 2.5 million people (distance ~35 km), and nearby towns.

3 Model

The CCN model employed here is described in detail in previous studies (Cubison et al., 2008; Ervens et al., 2007). In brief, the model is initialized with measured size distributions and CCN number concentrations at a given supersaturation (S). In the current study, we compare calculated and measured CCN number concentrations at one S for each study in the range of $0.27\% \leq S \leq 0.44\%$ (Table 1). This S (range) is the only one available for all data sets. Depending on the data set, S is either a constant value for the whole time period or variations in the CCN counter are taken into account. At this relatively low S, CCN number concentrations are most sensitive when only a small fraction of the aerosol population is activated and, thus, any change in the number of activated particles (e.g., due to changes in hygroscopicity) could translate to a significant change in activated fraction (Ervens et al., 2007). Such low S is encountered in stratus clouds that have a significant influence on the global radiative forcing.

Aerosol composition in the model is constrained with time-dependent bulk (i.e., not size-resolved) mass fractions of sub-micron non-refractory inorganic ions (sulfate, nitrate, ammonium, chloride) and organics from Aerodyne AMSs (Canagaratna et al., 2007). The mass of black carbon (BC) has been derived based on absorption measurements for the studies in which this data was available.

The water uptake of the particles is calculated using Köhler theory and defining all inorganics as being fully dissolved. The organic fraction is considered as either

insoluble or composed of hygroscopic organics (e.g., fulvic acid and small dicarboxylic acids) which translates to hygroscopicity parameters κ_{org}=0 or κ_{org}=0.12, respectively (Petters and Kreidenweis, 2007). The carbonaceous (organics+BC) fraction is either internally or externally mixed with the inorganic fraction. The four resulting "composition assumptions" regarding the organic fraction are identical to those in a previous model study (Pierce et al., 2007):

1. Externally mixed, insoluble (κ_{org}=0) organics (EM-I)
2. Externally mixed, soluble (κ_{org}=0.12) organics (EM-S)
3. Internally mixed, insoluble (κ_{org}=0) organics (IM-I)
4. Internally mixed, soluble (κ_{org}=0.12) organics (IM-S)

The composition/mixing state of ambient aerosol populations is likely to be more complex than any of these assumptions. However, in this study, the extent to which these simplified composition models lead to reasonable closure for a variety of locations and aerosol types will be explored.

4 Results and discussion

4.1 CCN closure results

In Table 2, the ratios "CCN(calculated)/CCN(measured)" are shown for the data sets in Table 1 and four composition cases. For aerosols sampled in source regions as encountered in RVS, MEX, and HSC, there is a high variability in the observed closure depending on the study. Cubison et al. (2008) showed for the RVS data set that freshly emitted, hydrocarbon-like organics at RVS comprise an externally mixed population with a significant contribution to the total number concentration of particles with a diameter of D~100 nm. A similar situation is encountered in Mexico City and the

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Houston Ship Channel where a significant fraction of the particles are fresh (especially from the late evening to the early morning) and have not yet undergone much physical or chemical transformation. In such scenarios, even the assumption of insoluble, externally mixed organics (EM-I) with bulk composition may lead to a significant overestimate of CCN number concentration. In those cases size-resolved composition and mixing state information is required to accurately predict CCN (Twohy and Anderson, 2008; Cubison et al., 2008). There is significant variability in the CCN prediction biases at these locations, which relate to differences in size-resolved composition and mixing state which are not captured by any of the simplified assumptions of this study. Whereas MEX and RVS resemble one another in terms of average organic fraction and distance from sources, single particle analysis at both locations has shown that the former location is significantly influenced by (aged) biomass burning particles (Moffet et al., 2008) whereas the latter one showed high fractions of elemental carbon in the diameter range below 200 nm, i.e. around the critical diameter for particles that activate at S=0.3% (Spencer et al., 2007).

In Fig. 1, a schematic size distribution is shown and sub-populations that are predicted to activate for each of the four composition assumptions are marked. If two composition/mixing state assumptions result in similar predicted CCN number concentrations, it does not imply that the same subset of the particle distribution is predicted to activate. It is evident that canceling effects leading to comparable predicted CCN numbers depend on the shape of the size distribution and the magnitude of the organic fraction. None of the four suggested assumptions represents the real composition/mixing state of an atmospheric particle population. There is an infinite number of other (more complex) composition/mixing states that can lead to the same predicted CCN number.

At locations with small organic fractions (HGC, PYE, HOM), the different composition/mixing state assumptions only lead to small changes in the overall quality of the CCN closure. At locations with higher organic fractions, internally and externally mixed insoluble organics or internally and externally mixed soluble organics, respectively, con-

sistently give similar predicted CCN number concentrations, i.e. the areas I and III, and the areas II and IV, respectively, in Fig. 1 are similar on average for all observed size distributions which shows that the assumed mixing state is less important than the assumed hygroscopicity of the organics. In some cases canceling effects of predicting different parts of the size distribution to activate (Fig. 1) can lead to similar predicted CCN number concentrations that are within a factor of two to the measured ones with several assumptions.

With increasing distance from pollution sources, the prediction of CCN number concentration under the assumption that all particles contain some hygroscopic material is improved, but no clear statement can be made whether physical or chemical mixing/ageing dominates as similar CCN ratios are predicted by applying cases EM-S or IM-I. The relative importance of mixing/ageing processes depends on factors that cannot be quantified in the present study, such as different photochemical activity and/or oxidant levels due to seasonal effects or concentration levels of precursors. Field studies and model simulations using results from laboratory experiments have suggested that chemical ageing of organic aerosols is too slow and limited to account for efficient hydrophobic-to-hydrophilic conversion as compared to physical mixing processes, especially condensation of hygroscopic secondary inorganic species across the whole particle population (Petters et al., 2006; DeGouw and Jimenez, 2009).

4.2 Spatial scale of ageing

In large-scale models, a time scale on the order of 1–2 days is assumed to convert particles from hydrophobic to hygroscopic, and, thus into potential CCN (Wilson et al., 2001). Aerosol age depends not only on distance from source but also on transport and processing time to the sampling site. However, wind data or highly time-resolved back trajectories that could allow an estimate of transport age are only available for a subset of the data sets investigated here. The relative age of air masses is often characterized by its "photochemical age", which is calculated based on the ratio of NO_x/NO_y, benzene/toluene or oxidized/total organic aerosol mass. This photochemical age is not an

absolute value that can be compared for different locations since it is also function of oxidant levels. Since none of these parameters is available for all the data sets used in this study, the present analysis explores spatial ageing scales by comparing distances from sources.

In Fig. 2, the range of CCN_{calc}/CCN_{meas} ratios, weighted by the frequency in the CCN closure studies, is shown for internally and externally mixed organics. No specific distance is ascribed to the sampling locations close to pollution sources (MEX, HSC, and RVS) because of the various pollution sources close to or within a few kilometers of the sampling location.

For studies downwind of but relatively close to major source areas (HOM and HGC), best CCN closure is achieved if all particles are assumed to be hygroscopic. This might be reasonable because of chemical and/or physical particle processing or due to the fact that the initial particle population dilutes and the resulting aerosol population is mostly determined by aged background aerosol. Applying IM-I (or EM-I) for remote locations as has been done in many global model applications will lead to an underestimate of CCN number concentrations. Our analysis indicates that, based on two studies (HGC and HOM), the ageing scale of particles is much shorter than several days and could be on the order of hours in agreement with a recent model study that explores the ageing time scale for soot particles (Riemer et al., 2009). This result for locations where photochemistry is active confirms studies that have shown that the addition of a few percent of soluble material to a hydrophobic particle significantly enhances its CCN ability (Bilde and Svenningsson, 2004).

4.3 Role of organic fraction

The data sets explored here, cover a wide range of OMF ranging from 7±4% to 79±10% (Table 1). For an aerosol with small OMF, the assumption of organic composition/mixing state is not crucial since the fractions of the size distributions that are predicted to activate due to hygroscopic and/or internally mixed organics are small (Fig. 1). For high OMF, however, predicted CCN number concentrations are quite sen-

sitive to the assumptions on mixing state/composition and quite different closure results are found.

In Fig. 3a–d, the ranges of predicted CCN(calc)/CCN(meas) ratio are shown as a function of OMF for all data sets and all composition cases. Despite much scatter around unity, the predicted CCN number agrees within a factor of two or better for small OMF under all assumptions (as also reflected in Table 2). As organic fractions increase above ~50% (MEX, CBG, HSC), it is evident that CCN number concentrations are increasingly underestimated if insoluble organics (EM-I or IM-I) are assumed (Fig. 3a and c), but reasonably well predicted for the EM-S and IM-S assumptions. This is in agreement with the study by Wang et al. (2008) who found that the best closure for aerosol with OMF>70% can be achieved if a hygroscopicity of κ_{org}=0.12 (IM-S) is assumed. The overestimate of CCN number concentration for some of the fresh aerosol (HSC, RVS), as discussed in Sect. 4.1, decreases slightly as OMF increases but this improvement may be a fortuitous result of compensating factors.

5 Summary and conclusions

CCN closure results for six different locations are compared using four simplified composition/mixing state assumptions for the carbonaceous (organics+BC) aerosol fraction (soluble/insoluble, internally/externally mixed with inorganic fraction). Despite very different locations and air masses, the following trends can be identified:

- Very close to pollution sources, simple assumptions of organic mixing state and solubility with bulk composition are not sufficient to predict CCN number concentrations. More complex assumptions about composition and mixing state (e.g., size-resolved) need to be made in order to predict CCN number concentration.

- Externally mixed, hydrophobic organic particles are likely to be sufficiently processed by chemical and/or physical ageing within a few tens of kilometers downwind of emission sources such that CCN composition can then be reasonably well

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represented by externally mixed, hygroscopic organics.

– Different assumptions for organic solubility and mixing state often lead to similar CCN number concentrations since different subsets of the aerosol population are predicted to activate. Thus reasonable CCN closure may be achieved due to such compensating factors even though the assumed composition/mixing state might not represent the true properties of the aerosol population.

Our results provide a general framework that allows prediction of CCN number concentrations to better than a factor of two on average for a variety of scenarios where the aerosol has undergone some degree of ageing by making simple assumptions about the solubility and mixing state of the organic fraction. A factor of two error in CCN concentration will translate to an error of about 15% in cloud drop concentration (Cubison et al., 2008; Ervens et al., 2005). Given the rather poor representation of clouds in large-scale models, this error is relatively small compared to radiative forcing uncertainties associated with cloud fraction and depth.

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Table 1. Characteristics of the data sets that have been used for CCN closure in the current study.

	Dates	Approx distance to sources [km]	Mean total number concentration [cm^{-3}] (±1 std. dev.)	Mean submicron organic (BC) mass fraction (±1 std. dev.)	Super-saturation S [%] (±1 std. dev.)	CCN/CN [measured] (±1 std. dev.)	Detailed CCN data analysis
Riverside (RVS), CA [a]	16.7.–15.8.2005	close	15 058±3937	0.61±0.12 (0.06±0.06)	0.27±0.05	0.08±0.03	(Cubison et al., 2008)
Mexico T0 (MEX) [b]	3/2006	close	12 197±4712	0.44±0.15 (0.14±0.07)	0.28	0.41±0.15	Wang et al., in prep.
Houston, TX [c]	2.8.–11.9.2006						(Quinn et al., 2008)
Ship Channel (HSC)		close	17 867±14 702	0.65±0.12	0.44	0.45±0.25	
Gulf Coast (HGC)		10	1753±1258	0.14±0.11	0.44	0.70±0.18	
Holme Moss (HOM), UK	11–12/2006	35	790±360	0.23±0.08 (0.23±0.15)	0.30±0.03	0.47±0.15	(Corris, 2008)
Chebogue Point, Canada (CBG) [d]	1.7.–15.8.2004	several 100s	4041±4016	0.65±0.10	0.29±0.02	0.40±0.18	(Ervens et al., 2007)
CBG biogenic			3957±2660	0.79±0.10		0.27±0.14	
CBG anthropogenic			4930±3944	0.54±0.13		0.41±0.17	
	7/2005	several 100s	755±491	0.10±0.06	0.29±0.004	0.59±0.26	
PYE – sea			826±432	0.14±0.06		0.59±0.21	
PYE – land			694±557	0.07±0.04		0.59±0.30	

[a] Study of Organic Aerosols at Riverside, CA (SOAR-I);
[b] Megacity Initiative: Local and Global Research Observations (MILAGRO);
[c] Gulf of Mexico Atmospheric Composition and Climate Study (GoMACCS);
[d] International Consortium for Atmospheric Research on Transport and Transformation (ICARTT); measurement site located in Nova Scotia, Canada;
[e] Marine Stratus/Stratocumulus Experiment (MASE), off the coast of Monterey, CA

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Table 2. Average ratios (±1 std. dev.) of calculated to measured CCN number concentrations for several locations and four different assumptions of composition/mixing state (κ_{org}: hygroscopicity parameter of organic fraction). Best agreement is marked in gray shaded cells, poorest in bold for each location.

	ext. mixed κ_{org}=0 (EM-I)	ext. mixed κ_{org}=0.12 (EM-S)	int. mixed κ_{org}=0 (IM-I)	int. mixed κ_{org}=0.12 (IM-S)	# data points
RVS	4.4±1.8	4.9±1.7	4.6±1.7	**6.0±2.1**	306
MEX	0.5±0.2	1.1±0.1	0.5±0.2	**1.1±0.2**	198
HSC	2.4±1.9	4.0±3.0	4.0±3.0	**4.2±3.2**	120
HGC	1.9±1.1	**2.4±1.9**	1.7±1.0	2.3±1.9	123
HOM	1.0±0.5	**1.2±0.6**	0.8±0.5	0.9±0.5	769
CBG – all data	0.7±0.3	1.2±0.3	0.9±0.4	**1.4±0.4**	717
CBG anthrop.	0.8±0.3	1.2±0.4	1.2±0.4	**1.6±0.4**	194
CBG biogenic	0.4±0.2	1.2±0.3	0.6±0.3	**1.5±0.4**	80
PYE – all data	**1.3±0.6**	**1.3±0.6**	1.1±0.6	1.1±0.6	880
PYE land	**1.3±0.7**	**1.3±0.7**	1.2±0.6	1.2±0.6	448
PYE sea	1.3±0.5	**1.4±0.5**	0.8±0.4	0.9±0.4	432

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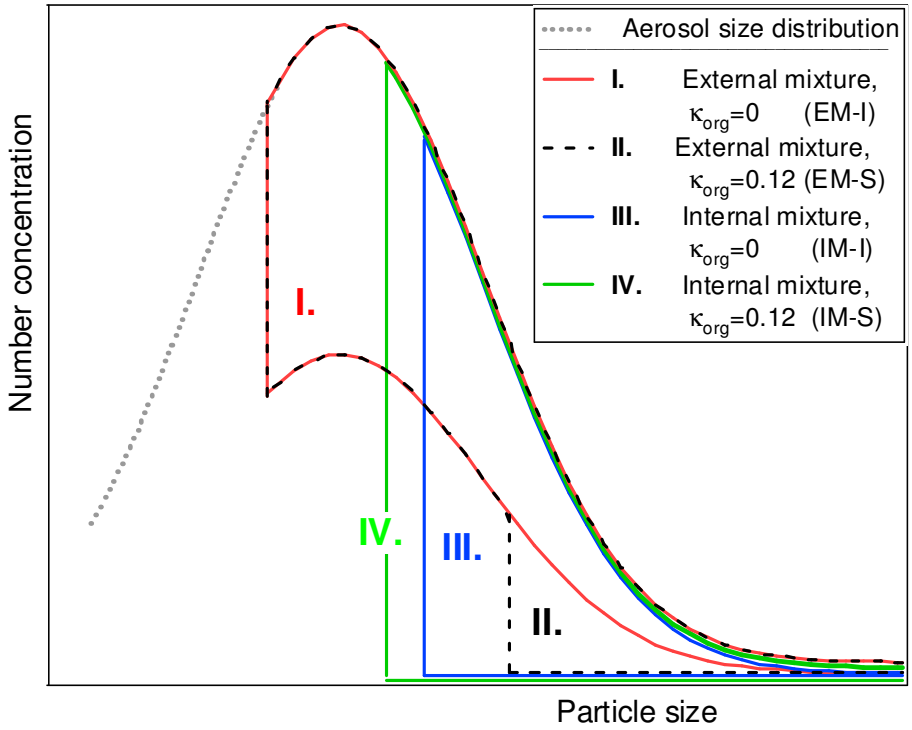

Fig. 1. Schematic of areas of activated number concentrations for different composition assumptions.

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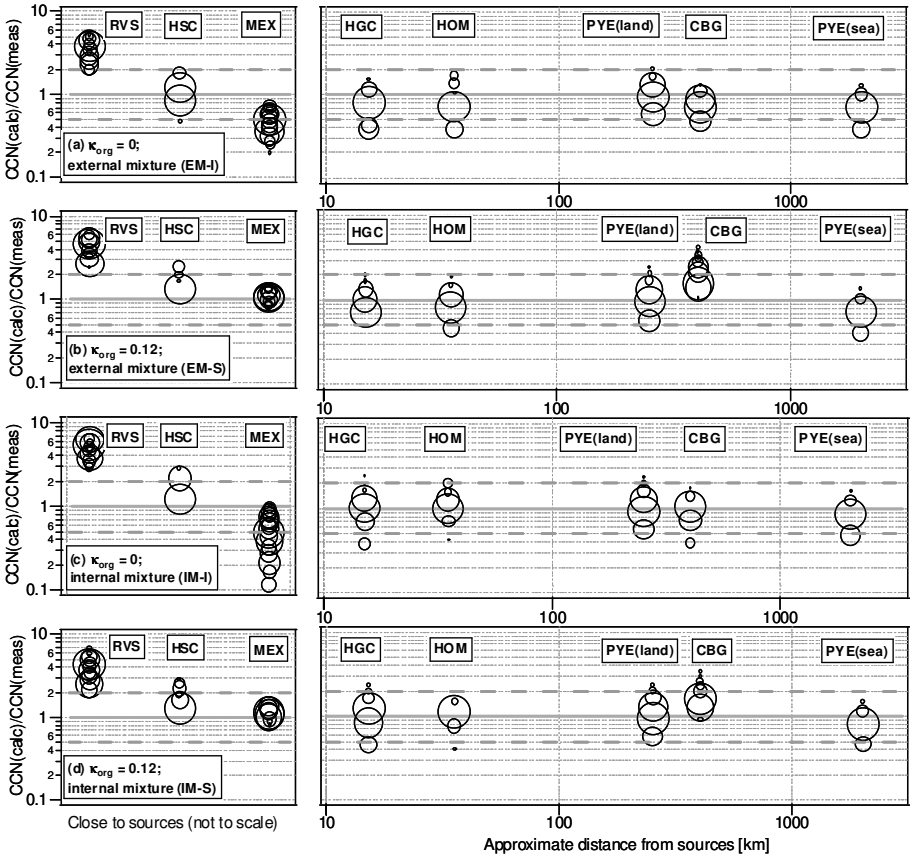

Fig. 2. Ratio of calculated to measured CCN number concentration for seven different data sets. The symbol size corresponds to the frequency of the respective ratio in the CCN closure.

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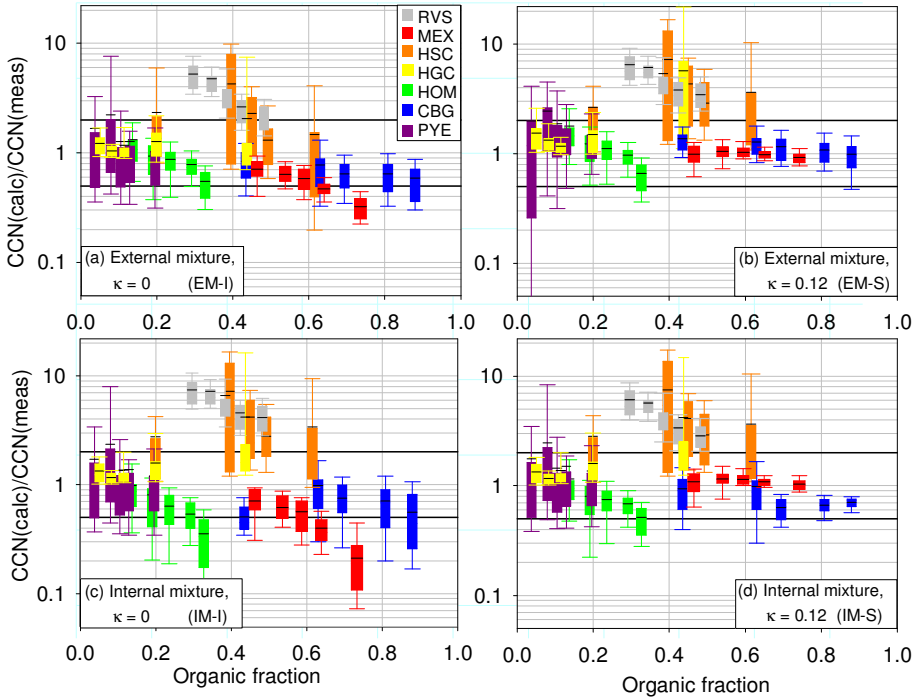

Fig. 3. Ratio of calculated to measured CCN number concentrations as a function of organic mass fraction for seven different data sets and different assumptions of hygroscopicity and mixing state of organic fraction. Each bar represents 20% of the respective data set, error bars show ±one standard deviation; mean value is marked in each bar. Horizontal black lines represent CCN(calc)/CCN(meas)=0.5 and 2, respectively.

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