Atmos. Chem. Phys. Discuss., 9, C11103–C11108, 2010
www.atmos-chem-phys-discuss.net/9/C11103/2010/

Atmospheric Chemistry and Physics Discussions

Interactive comment on "CCN predictions using simplified assumptions of organic aerosol composition and mixing state: a synthesis from six different locations" *by* B. Ervens et al.

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Received and published: 23 February 2010

Comment 1. "Although the manuscript deals with CCN closure, only few previous CCN closure studies are referenced. A comprehensive discussion on previous studies and how their results compare with the authors results would be useful."

Response: In the new version of the manuscript, we added a table (attached table = Table 3 in the revised manuscript) that summarizes results from various previous CCN studies at different locations, together with the assumptions that have been made about the hygroscopicity of organics and their mixing state. The table shows that with increasing distance from sources an assumed internal mixture of insoluble organics

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leads to reasonable CCN closure results. At very remote locations, where air masses did not have any influence of major pollution sources for several days, aerosol is sufficiently aged that organics can be considered soluble (kappa (org) >0). The range of kappa (org) that has been applied in these studies is 0.1 – 0.5, and, thus, the value we have assumed for hygroscopic organics (kappa (org) = 0.12) falls in this range. We added a Section 5 to the paper, entitled 'Comparison to previous CCN studies' where the findings from these cited studies are discussed in more detail.

Comment 2. As also the authors point out, the effect of using size-resolved chemical composition versus size-averaged is important when doing a CCN closure study (e.g., Broekhuizen et al., 2006; Stroud et al., 2006; Medina et al., 2007; Gunthe et al., 2009). Please, discuss the possible implications for your study given that you use a size-averaged composition.

Response: In the new table (Table 3), most of the studies have been performed by assuming bulk composition. However, the reviewer is right that in some studies the importance of considering size-resolved composition has been pointed out (e.g., Medina et al., 2007; Shantz et al., 2008; Kammermann et al., 2010). It has been shown that in many scenarios (both very close to pollution sources and more remote locations), CCN number concentrations tend to be overestimated if the size-resolution of composition is not considered because of relatively large fractions of insoluble material around the critical diameter. We have included the discussion of these impacts in the new Section 5 'Comparison to previous CCN studies'. We point out that we cannot perform a detailed analysis with the present data sets since size-resolved composition/hygroscopicity is only available in a few studies, and the goal of the current study is a synthesis of these data sets under uniform conditions.

Comment 3. Page 21248, lines 10-11 and last 3 lines of the abstract – Although the error in cloud droplet number concentrations from a factor of 2 error in CCN concentrations seems to be small there are other studies that estimate this error to be quit larger. For example, the study of Sotiropoulou et al., (2006) using size-resolved chem-

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ical composition found that the error in cloud droplet number concentrations is half the error in CCN concentrations that translates into a 0.5 Wm-2 uncertainty in indirect forcing (first order estimate) for a typical 10-25% error in cloud droplet number concentrations. Please rephrase appropriately.

Response: We changed the wording in the abstract and text and stated more carefully that the error in CCN number concentration translates on average in small changes in the radiative forcing but can have large absolute values in regions downwind of pollution sources (industry, biomass burning) as stated by Sotiropulou et al. They estimated the error in cloud drop number concentration to be at most 50% of the error in CCN number concentration in such regions which translates into a total change in radiative forcing of up to $\sim$ -10 W m-2. The calculated maximum uncertainty in radiative forcing in such regions is calculated in their study as $\sim$0.5 W m-2 which corresponds to a relative error in radiative forcing is $\sim$5%.

In order to give a more comprehensive analysis of the consequences of errors in CCN number concentration for drop number concentration Nd for conditions that are similar to the particular data sets in our current study, we add a Section 6 'Effects of uncertainty in CCN number concentration on drop number concentration' in the revised manuscript. In this section we discuss results from a cloud parcel model that considers the activation of a (lognormally distributed) aerosol population in a constant updraft (w).

We define a reference case with a composition kappa1 that translates into a critical diameter D1 above which all particles are activated at a supersaturation S = 0.3% according to Köhler theory (equilibrium) if an internal mixture is assumed. In order to simulate an 'error of factor 2 (0.5)' in CCN number concentration at the given S, we determined the critical diameter D2 (D05) above which the number concentration is twice (half of) the number concentration associated with D1. Each of these critical diameters is unambiguously associated with a composition parameter (kappa 2, kappa 05). In Figure 4 a and b, two size distributions together with D1, D2, and D05 are shown. Depending on the shape of the size distribution, the distances of the critical

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diameters vary. Since it has been discussed previously (Ervens et al., 2005) that the number concentration Na can have a large impact on the relative effect of composition on drop number concentration, three different number concentrations for each size distribution are assumed. Thus, in total nine different initializations (3ïĂă kappa, 3 Na) of the cloud parcel model are defined using these size distributions and compositions. The resulting cloud drop number concentrations (at a cloud liquid water content of LWC = 0.3 g m-3) from these simulations using kappa2 and kappa05 are compared to the results using kappa1 for the same size distribution (and Na). While in an equilibrium state, by definition, these ratios equal 2 (0.5) (Figure 4 a and b), in a simulated cloud, they differ from this constant value due to the non equilibrium growth associated with supersaturation production (proportional to updraft) and depletion (proportional to condensation). The ratios of the resulting Nd are shown in Figure 4c and d. When the ordinate (Nd(2)/Nd(1) – 1) = 0 the composition does not have any impact on Nd while when (Nd(2)/Nd(1) – 1) = 100%, the error in Nd is the same as for CCN concentration. Thus, the percentages shown in Figure 4 give a direct comparison of the effect of composition on changes in Nd as compared to NCCN. In agreement with prior studies, we show that at low updraft these terms might be as high as $\pm$0.5, i.e., indeed the error in cloud drop number prediction is half of that of CCN number prediction. At updraft velocities w > $\sim$ 100 cm/s, the error reduced to $\sim$ 20%. We add in the abstract and in the conclusions the findings from this analysis and change the text accordingly.

Comment4. Page 21255, Figure 2 - Please state what the dashed lines represent.

Response: The horizontal lines represent the ratios CCN(model)/CCN(measured) = 2 and 0.5, respectively. We added this information to the figure legend.

Interactive comment on Atmos. Chem. Phys. Discuss., 9, 21237, 2009.

Location	Dist [km]	κ	Mixing state	'slope'	S [%]	Comment	Reference
Riverside	close	$\kappa_{org} = 0$	int	2.8-7.1	0.1-0.9		(Cubison et al., 2008)
		$\kappa_{org} = 0$	ext	0.79-4.1	0.1-0.9		
		$\kappa_{org} = 0$	int/ext	1-3.7	0.1-0.9	hydrophobic org at ~100 nm (ext. mixed) size-resolved composition	
Houston (ship)	close	$\kappa_{org} = 0$	ext	0.85 – 1.2	0.22-1	CCN prediction of particles ~ 200 nm	(Quinn et al., 2008)
Houston (aircraft)	close	$\kappa_{all} = 0.6$	int	1.36	0.3-1.1		(Lance et al., 2009)
		$\kappa_{org} = 0$	int	1.03		Hydrophobic org at ~100 nm	
Toronto	close	$\kappa_{org} = 0$	int	1.12	0.56-0.6		(Broekhuizen et al., 2005)
		$\kappa_{org} = 0$	ext	1.03	0.56-0.6		
		$\kappa_{ore} = 0.096$	int	1.16	0.56-0.6	assumption: 10% of org fraction soluble	
New Hampshire (Thompson Farm)		$\kappa_{org} = 0$	int	1.22	0.3		(Medina et al., 2007)
		$\kappa_{org} = 0$	int	1.052	0.3	size-resolved composition	
Vancouver	45	$0.001 < \kappa_{org} < 0.11$	int	~0.8-1	0.19-0.5	$\kappa_{all} = 0.16$, size-resovled	(Shantz et al., 2008)
Guangzhou (China)	60	$\kappa_{all} = 0.32 \pm 0.1$	int	1.0 ± 0.07	0.27	κ derived based on HTDMA	(Rose et al., 2008)
Toronto (rural)	70	$\kappa_{org} = 0$	int	0.89 –1.14	0.42		(Chang et al., 2009)
		$\kappa_{ox} = 0.2$; $\kappa_{non\text{-}ox} = 0$	int	1.23	0.42		
Duke Forest (polluted)	10s	$\kappa_{org} = 0.13$	int	1.7 - 2.1	0.2		(Stroud et al., 2007)
		$\kappa_{ore} = 0$		1.4 - 1.65			
Monterey		$\kappa_{org} = 0$	int	0.94-0.95	0.2	boundary layer and free troposphere	(Wang et al., 2008)
		$\kappa_{org} = 0.25$	int	1.1-1.15	0.2		
		$\kappa_{org} = 0.1$	int	1.17	0.2	above clouds	
		$\kappa_{org} = 0.1$	ext	0.89	0.2	above clouds	
Californian Coast		$\kappa_{all} = 0.13$	int	~ 1	0.6	κ derived based on $D_{crit,mean}$ (66.7 nm)	(Furutani et al., 2008)
Jeju Island	100s	$\kappa_{all} = 0.17$	int	0.73	0.1-1		(Kuwata et al., 2007)
		$\kappa_{all} = 0.6$	int	1.16			
North Sweden	100s	$\kappa_{org} = 0.09$	int	1.12	0.6	size-dependent κ derived based on HTDMA	(Kammermann et al., 2010)
N American Coast Free troposphere Central Valley	80-1000s	$\kappa_{all} = 0.6$	int	~1-1.5	0.3	CCN closure results reported for whole data set (aircraft data) Assumption: pure $(NH_4)_2SO_4$	(Roberts et al., 2010)
Northeast Atlantic		$\kappa_{org} = 0$	int	0.34	0.1	instrumental errors (?)	(Chuang et al., 2000)
Tasmania		$\kappa_{org} = 0$	int	1.26 (0.99)	0.5	Better agreement in air masses with low aerosol loading and Rn	(Covert et al., 1998)
North Pacific		$0 < \kappa_{org} < 0.5$	int	0.6 – 1.15	0.34	size-resolved	(Shantz et al., 2008)
Remote, marine		$\kappa_{org} = 0$	int	0.92	0.38		(Bougiatoti et al., 2009)
		$\kappa_{org} = 0.158$	int	0.98			
Eastern Pacific		$\kappa_{all} = 0.6$	int	1.78	0.3		(Roberts et al., 2006)
Amazon		$\kappa_{org} = 0$	int	0.2-0.3	0.2 - 1		(Mircea et al., 2002)
		$\kappa_{org} \sim 0.1$	int	0.5-1			
Amazon		$\kappa_{org} = 0.1$	int	1.17	0.1–0.82	size-resolved composition	(Gunthe et al., 2009)
Amazon		$\kappa_{org1} = 0.03$; $\kappa_{org2} = 0.1$	int ext	1.11 1.06	0.3-1	2 internally modes of different sizes	(Rissler et al., 2005)

Fig. 1.

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Figure 4:
a) and b): Example size distributions that are used as input data to the cloud parcel model. D1 denotes the critical diameter for particles that are activated at S = 0.3% κ = 0.18 ('base case'). D05 and D2 denote the critical diameters for particles with a composition that results in 50% or 200%, respectively, of the activated CCN number concentration as compared to the base case.
c) and d): Relative difference between predicted drop number concentration for the compositions as specified by D05 and D2 to drop number concentration predicted by composition associated with D1 as a function of updraft velocity in the cloud parcel model.

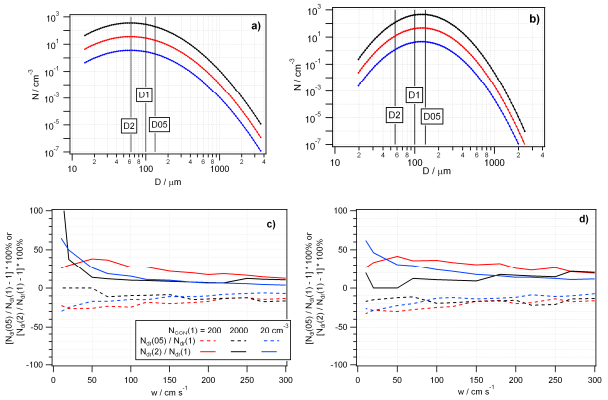

Fig. 2.

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