

CCN predictions using simplified assumptions of organic aerosol composition and mixing state

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INTRODUCTION

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.

Seven data sets on aerosol size distribution, chemical composition, and CCN concentration (N_{CCN}) have been analyzed to explore the extent to which simple assumptions of organic composition and mixing state can reproduce measured CCN number concentrations. In addition, effects of these assumptions on cloud drop number concentration (N_d) are investigated.

DATA SETS

Data sets differ in distance from and nature of sources and pollution level.

- Riverside, CA (RVS) ^(1*)
- Mexico City, TO (MEX) ^(2*)
- Houston Ship Channel (HSC) ^(3*)
- Houston Gulf Coast (HGC) ^(3*)
- Point Reyes (PYE) ^(2*)
- Holme Moss, UK (HOM) ^(2*)
- Chebogue Point (CBG) ^(4*)

(* detailed CCN analysis in previous study)

CCN MODEL

INITIALIZATION

- Measured size distributions
- N_{CCN} at one supersaturation in the range $0.27\% \leq S \leq 0.44\%$ (depending on data set); if available, temperature gradients in the in the CCN counter are taken into account.
- Aerosol composition:
 - SO_4^{2-} , NO_3^- , NH_4^+ , (Cl), organics (Aerosol mass spectrometer)
 - Black carbon (if available)
- Time-dependent bulk measurements; not size-resolved

ASSUMPTIONS OF ORGANIC COMPOSITION AND MIXING STATE

- Externally mixed, insoluble organics
- Externally mixed, soluble organics ($\kappa_{\text{org}} = 0.12$)
- Internally mixed, insoluble organics
- Internally mixed, soluble organics ($\kappa_{\text{org}} = 0.12$)



REFERENCES

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- 3 Ervens, B., et al., J. Geophys. Res., 112, doi: 10.1029/2006JD007426, 2007.
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CCN CLOSURE

... AS A FUNCTION OF THE DISTANCE FROM SOURCES

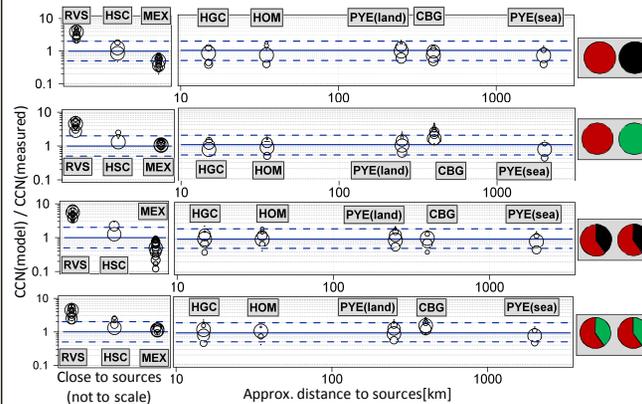


Figure 1: CCN closure results as a function of distance from sources; symbol size: Frequency of this ratio ' CCN_{model}/CCN_{meas} ' in CCN closure

- Close to emission sources, particles are externally mixed. Assumption of bulk composition is not sufficient and size-resolved composition has to be taken into account (e.g., hydrophobic organics ~100 nm)
- ~10's km downwind of sources, particles are sufficiently mixed and the assumption of hygroscopic organics gives best closure

... AS A FUNCTION OF THE ORGANIC FRACTION

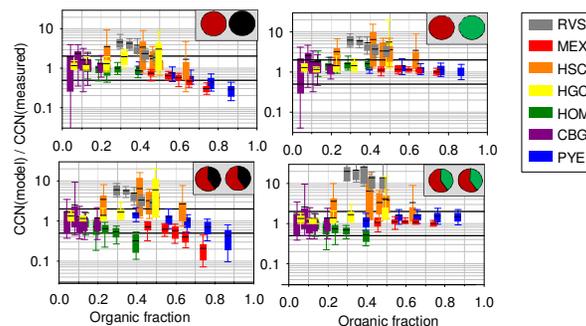


Figure 2: CCN closure results as a function of organic fraction

- N_{CCN} are underestimated if organics are assumed insoluble at organic fractions > ~70%
- In all other cases, CCN number concentrations in aged aerosol are reproduced within a factor of 2

EFFECTS ON CLOUD DROP NUMBER CONCENTRATION

CLOUD MODEL

- The model describes the activation of an internally mixed aerosol population into cloud droplets in a constant updraft (w)
- A reference composition (κ_1) is chosen that translates into a critical diameter D_1 (equilibrium diameter at $S = 0.3\%$; Köhler equation)
- In order to simulate an error of 2 in N_{CCN} ($\Delta N_{CCN} = 100\%$), two 'D, κ ' pairs are defined (D_{05} , κ_{05} and D_2 , κ_{02}) that define $0.5 \cdot N_{CCN}$ and $2 \cdot N_{CCN}$ (Fig. 3)

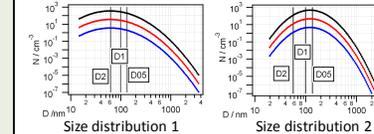


Figure 3: Size distributions: D_2 (D_{05}) are critical diameters above which twice (half) as many particles are activated as compared to D_1

RESULTS

- At low w , the error in N_d (ΔN_d) can be as high as ΔN_{CCN} for high particle number concentrations ('polluted conditions')
- At $w > \sim 100 \text{ cm s}^{-1}$, $\Delta N_{CCN} = 100\%$ translates into $\Delta N_d \leq 20\%$ due to the relatively larger impact of w on S as compared to that of aerosol composition
- Errors are largest if the smallest activated particles are at the steepest part of the particle number concentration

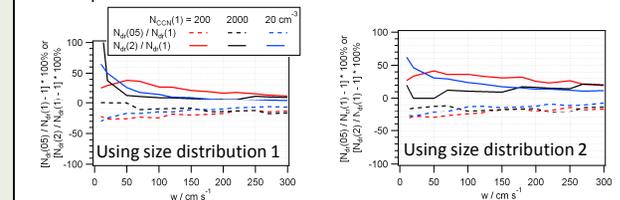


Figure 4: Effect of different compositions on N_d as a function of w

CONCLUSIONS

- Close to pollution sources, complex composition and mixing state (size-resolved) assumptions need to be made in order to predict N_{CCN} .
- Externally mixed, hydrophobic organic particles seem to be sufficiently processed by chemical/physical ageing within 10s km downwind of emission sources and can be represented as an internal mixture.
- Different composition/mixing state assumptions lead to similar N_{CCN} as different subsets of the population are predicted to activate. Such compensating factors might lead to good CCN closure even though the assumed and actual composition/mixing state are not identical.
- $\Delta N_{CCN} = 100\%$ translates into $\Delta N_d \sim 15\%$ under most conditions which is relatively small compared to radiative forcing uncertainties associated with cloud fraction and depth.