CCN Closure Studies at Urban and Background Locations

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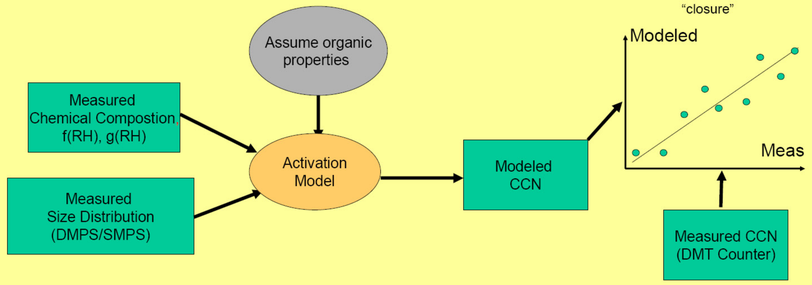
INTRODUCTION

The indirect influence of aerosol particles on the radiative balance of the atmosphere through changes in droplet number and persistence of clouds, known as the "aerosol indirect effects", causes the largest uncertainty amongst the presently known causes of change in the radiative forcing of climate (IPCC, 2001). Further analysis of the physical and chemical parameters influencing cloud condensation nuclei (CCN) activation is required in order to quantify these effects and allow parameterizations of these processes in large scale models. We use results from two recent field campaigns in both marine and urban environments in order to explore the relationship of the physico-chemical properties of aerosol particles and their CCN activity. Based on measured aerosol size distributions, composition, and/or growth characteristics, we show model approaches that link these aerosol properties to their CCN activity.

CCN Activation Model - Summary



• Standard Köhler equilibrium theory - considers mixtures of different solutes (inorganic, organic) and their appropriate physico-chemical properties.

• No treatment of surface tension or solubility (lack of experimental information)

• **Internally-mixed Model:** All aerosol composed of a soluble and insoluble fraction with same composition for all sizes.

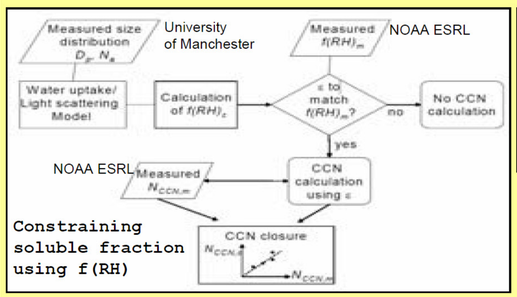
• **Size-Resolved Composition Model:** The insoluble fraction varies over the size range, but at each size, the particles are all identical and still consist of a soluble and insoluble fraction.

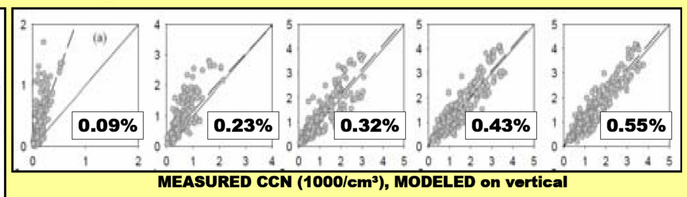
• **Externally-mixed Model:** As per the size-resolved case, but in the case of observing an apparently externally-mixed small diameter mode, this is treated as totally non-activating in the model.

• **Constraining soluble fraction:** The soluble fraction in the model is constrained either 1) through iteration until the calculated f(RH) or g(RH) matches the observed values, or 2) through direct parameterisation from size-resolved composition measurements.

• **Measurements:** Composition from the Aerodyne Aerosol Mass Spectrometer (AMS, Jayne *et al* 2000), f(RH) from 40 to 85% from TSI integrating nephelometers, CCN from the Droplet Measurement Technologies (DMT) CCN spectrometer.

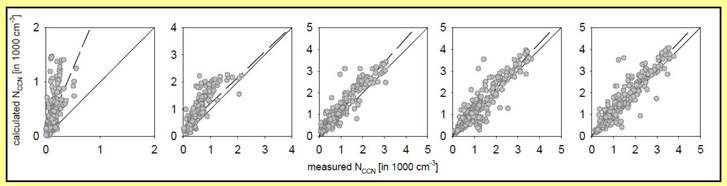
Marine Case, Chebogue Point, ICARRT 2004





MEASURED CCN (1000/cm³), MODELED on vertical

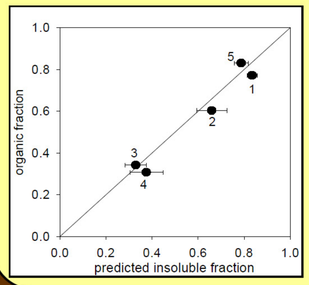
• Using f(RH) measurements to constrain the soluble fraction in the internally-mixed model achieves a good degree of closure with the CCN measurements.



• But a similar level of closure can be achieved simply by assuming a soluble fraction of 0.37.

• Note- measurements were used to guide this guess for the soluble fraction so it cannot be applied ad hoc.

• Clearly at Chebogue Pt, the size distribution is the driving force behind the activated CCN number.



• The insoluble fraction required in the model to force it to match the measurements is correlated to the measured organic fraction in the AMS.

• The organic fraction at Chebogue, despite being aged and oxygenated, does not contribute to water uptake.

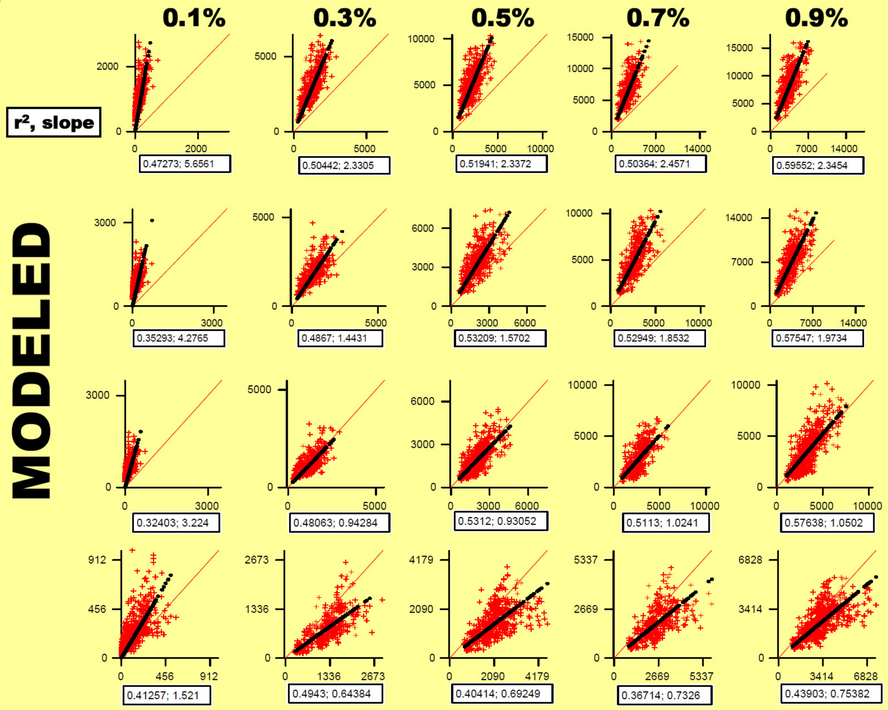
! The overprediction in the model at low supersaturation is believed to be an instrumental issue ...

• Introducing size-resolved composition did not improve closure on the Chebogue dataset.

• Temporal variation in composition does not appear to be of great importance, even during organic-dominated periods.

• The most important information for CCN closure is the measured aerosol size distribution and the supersaturation in the CCN spectrometer.

Urban Case, Riverside CA, SOAR 2005



• **Internally-mixed model**

• As for Chebogue Pt, but constraining the insoluble fraction directly from AMS measurements of the organic fraction.

• **Size resolved, internally-mixed model**

• Using the size-resolved composition from the AMS to directly constrain the soluble fraction for each point in the size distribution.

•**Size resolved, externally-mixed model**

• Using the size-resolved composition from the AMS to directly constrain the soluble fraction for each point in the size distribution.

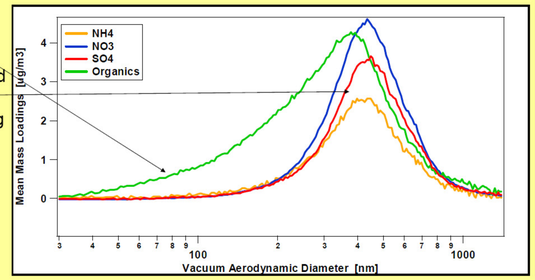
• Also treating the small-mode organics in the AMS as completely non-activating

• **Completely externally-mixed model**

• As for the internally-mixed size-resolved case, ***but*** particles are treated as separate modes of either completely organic OR completely Ammonium Sulphate.

MEASURED

• Four different model parameterisations...
• Strong evidence of external mixing of the small diameter organic mode (from fresh local emissions) with the background mixed inorganic/organic mode.
• Broekhuizen et al 2005 showed that using size-resolved composition in a CCN model could achieve closure in an urban environment (case 2 above).
• At Riverside, the fraction of (assumed) insoluble small mode organics is sufficient that further complication in the model is required for successful closure (case 3).



• Assuming internal mixing at Riverside leads to soluble material being placed on particles which, in the real atmosphere, are extremely insoluble → overpredict CCN.

• Using size-resolved composition does improve CCN prediction, but the soluble material in the small particles still leads to an overestimation in the model.

• To achieve closure, treatment of the mixing state is crucial!

CONCLUSIONS

• In a location ~2 days from emission sources, an internally-mixed parameterisation of the aerosol population is sufficient, with good size distribution data, to achieve CCN closure.

• At an urban location, the small-mode organics from local emissions must be treated as externally-mixed in order to predict CCN activation. Treating this mode as entirely non-activating is shown to be a sufficient parameterisation for Riverside.

• **Future work...** how do these conclusions apply to other similar locations?? Data sets from other marine and urban areas are being used to test the validity of our conclusions.

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