

***Interactive comment on* “The influence of chemical composition and mixing state of Los Angeles urban aerosol on CCN number and cloud properties” by M. J. Cubison et al.**

M. J. Cubison et al.

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We would like to thank Referee 2 for the positive remarks and constructive comments on our discussion paper, which are highly appreciated and will be taken into account upon manuscript revision. Responses to individual comments are given below.

- *The authors should use a term closure more cautiously. In the abstract a closure between the modeled and measured CCN number concentration is attempted, but the authors however, state in pp. 5641 that the aim is not to form a perfect closure but rather compare the measured and modeled CCN concentrations.*

The aim of the modeling study is to attempt closure with each model, but the conclusions focus on the differences rather than dwelling on the absolute degree

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of agreement. To clarify this, line 14 on pp5641 is changed to:

In addition, given the focus of this work to compare the degree of closure between various model schemes, any small errors induced through diurnal averaging are not critical to the overall conclusions.

- *How sensitive are the modeled concentrations to particle composition? As the composition data is averaged to a typical diurnal cycle, some of the variability is bound to be lost. Does this affect the conclusion that the chemical composition is only of a minor importance to the predicted CCN concentrations? By adding complexity to the model, slope is approaching one, but the correlation coefficient is still less than 0.46. Does the averaging cause this?*

As mentioned in the text, the size-resolved composition measurements do not have sufficient signal-to-noise at the 30-minute time resolution used in the model to satisfactorily reduce statistical noise. Running the model on the actual time series measurements, rather than the diurnal cycle, would introduce a great deal of noise into the size-resolved composition input and thus in itself grossly influence the r^2 value returned in the correlation plots. Thus, whilst it would seem likely that some correlation is lost by averaging to a diurnal cycle, in the absence of time series measurements with high signal-to-noise it is not possible to quantify this effect. In addition, the aerosol composition during SOAR-1 showed low variability from day to day (when comparing the same time of the day), and much of the variability was due to the diurnal cycle (Docherty et al., submitted EST 2008). Some additional variability occurs between weekdays and weekends, and for this reason weekends were excluded from the original analysis. Finally, the sensitivity of the modeled concentrations to composition are not too great, as demonstrated in Figs. 11 and 12, thus suggesting that the averaging process does not greatly reduce the correlation coefficient.

- *As presented in Figure 10 (Diurnal model output), during rush hour there is a lot of CN available, but the measured CCN remains quite stable. What is a diurnal*

cycle of e.g. $D_p > 100$ nm particle concentration?

The diurnal cycle of particles greater than 100 nm has now been included in Figure 10 to demonstrate their diurnal variability, which also follows a similar cycle to the overall CN (gr. than 10 nm) number concentration. As noted by the reviewer, the activated number (CCN) does not follow such a dynamic diurnal cycle and thus other factors than particle size distribution alone, namely composition and state of mixing, must influence the particle activation.

- *The most discrepancies take place during morning rush hour when a lot of the freshly emitted non-activating appear in the spectrum. How accurate is the estimated HOA concentration and how sensitive are the results to this parameter?*

Of order a few percent of the total organic mass is unaccounted for after the reconstruction into HOA and OOA mass, and the uncertainty in the HOA mass fraction (HOA/OA) (from analysis of the PMF output vs. number of factors and the FPEAK parameter) is a few percent of the total OA. For model case 5, where the HOA mass is treated as non-activating, a reduction (increase) in HOA/OA would directly translate to an increase (reduction) in modeled CCN as these particles are forcibly removed from the size-distribution to simulate a non-activating, externally-mixed component. However, the small changes in HOA that are possible within the uncertainty of HOA/OA cannot account for the over-prediction observed in model case 5.

- *Currently the cloud droplet model is slightly unconnected with the rest of the manuscript. I would suggest bridging the section 6 more closely with the excellent discussion in the previous section.*

Additional text has been introduced into the manuscript to better bridge the two sections. The first paragraph in section 6 now reads:

While the CCN calculations are performed for equilibrium conditions using the Koehler equation, i.e. for a prescribed supersaturation, in a parcel model the

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supersaturation in a cloud is represented by (i) a dynamic term that represents the source of supersaturation and (ii) the condensation term that depends on the size and composition of the particles, and the supply of water vapour. Thus it is not prudent to assume that conclusions drawn about particle activation properties from CCN counter studies can be used to infer the same for cloud droplet formation under real atmospheric conditions. The cloud droplet modelling study of Ervens et al., (2005) showed that the greatest impact of chemical composition on cloud droplet concentrations occurs at low supersaturation (i.e. low updraft and/or high aerosol concentrations), whereas at higher updrafts/lower aerosol concentrations, the composition effects are reduced. Further to their approach, we expand the CCN model results to consider the impact of the different composition and mixing state cases (C1-C5) on cloud droplet number concentrations.

- *pp. 5653 lines 12-14 Please give reasoning the selected values.*

In the cloud parcel model we assume a maximum liquid water content of 0.3 g m^{-3} as being typical for cloud-top liquid water content in stratocumulus clouds. The initial values of $T = 288 \text{ K}$ and $p = 919 \text{ mbar}$ are comparable to values that are predicted by Large Eddy simulations. Our simulations start at a height of 300 m and a relative humidity of 40 percent. Cloud base (i.e. $S = 1$) is reached around 1600 m, and the maximum liquid water content, i.e. cloud top lies around 1900 m. The liquid water path represents the vertically integrated liquid water content over the height of the cloud.

The asymmetry scattering factor of 0.85 is representative for cloud droplets at visible wavelengths. At these wavelengths the forward scattering of droplets greatly dominates over backward scattering (i.e., for g larger than zero) (Bohren, 1987). The cloud optical depth is calculated by: $\tau = \int_0^h \int_0^\pi r^2 Q_{ext} n(r) dr dh$ with $r =$ drop radius, $n =$ number concentration of droplets with respect to r , and $h =$ height.

The extinction efficiency factor Q_{ext} is a function of wavelength, r and refractive index. However for the sake of simplicity we have assumed a value of $Q_{ext} = 2$,

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a value that is approached over a wide range of wavelengths (e.g., Mitchell, J. Atmos. Sci., 59(7),1311, 2000).

- *Figures 5-9. S = 0.1 is not discussed in the text due to insufficient time to grow in the CCNC. I see no point showing the figures either.*

The authors accept that the plots for S = 0.1 percent may detract from the discussion; however they do neatly demonstrate one of the limitations of the instrumentation used in this (and many other) CCN studies. Without wishing to hide imperfections in the instruments, these plots have been shown. The text on p. 5643 is altered to better reflect this, for a more detailed discussion see the response to the first comment from reviewer 1.

- *Figure 1 and 10 trim the legend boxes. Figure 13 add unit for the updraft velocities. Figure 15 case 2 does not have a symbol.*

Figures corrected as noted.

- *Different model runs should be consistently marked as C1-C5 or M1-M5. pp. 5644 line 10, M1 pp. 5645 line 2, M1 pp. 5651 line 6, M5*

The letter C should have been used throughout. Typographic mistakes have been rectified.

- *Correlation coefficient should be consistently capital R or lower case r throughout the paper.*

Correlation coef. is now referred to as r^2 throughout.

- *pp. 5640 line 4, chemiscal? pp. 5648 line 5 ammonium sulphate pp. 5653 line 21 Ssc?*

These typos have been fixed.