Interactive comment on “The mixing state of carbonaceous aerosol particles in Northern and Southern California measured during CARES and CalNex 2010” by J. F. Cahill et al.

Anonymous Referee #1

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The paper presents results from an A-ATOFMS during two 2010 field campaigns in California. Particle composition number fraction statistics in the accumulation mode are presented, as well as mixing state distributions in two-component detail for sulfate/nitrate and oc/soot.

The dataset presented is of considerable value, providing detailed quantification of aerosol types and their relative number concentration. Hence the paper certainly merits publication in ACP. However, there are a number of issues that should be addressed to enhance the value of these data outputs for the reader. For example, without any discussion of relative size ranges of different aerosol types, biases inherent in the limited detection range of the instrument are opaque, and are overlooked in the analysis. Hence I recommend some expansion of discussion of this and other issues (specified in the specific comments) that have not been clarified for the reader before publication in ACP.

Specific Comments:

1. The title suggests a more narrow treatment than was covered in the paper (due to inclusion of particle type statistics for CARES/CalNex, and mixing between nitrate/sulfate). I suggest that the authors broaden it.

2. “Soot” generally refers to the fresh emissions of various combustion processes; this is not as specific a term as “black carbon”, which refers only to the refractory component of soot. I suggest updating the terminology in the paper to reflect the more precise wording.

3. Section 2: I suggest adding discussion of the linearity of the calibrations over the wide particle size range, including dependence of sampling on particle size. Discussion, too, is needed highlighting the instrumental constraints on the particle size range relevant for total particle number and total particle mass. This type of information is fundamental to clear understanding of the value and limitations of the data set.

4. Section 3: The authors should include an estimate of uncertainty on the number fractions shown. Although >75,000 particles were sampled in CalNex, it is not clear how much statistical robustness is associated with the “sliced up” values presented to the reader. A note about the time resolution of the results, and perhaps a comment about variability in time/space within each mission would also be appreciated by a reader. The discussion on pg 18431 lines were 98% and 93% coverage were contrasted is a good example of where an uncertainty estimate is needed.

4b: The discussion leading up to this looks at SS and V-OC particles, but it looks like these make up only a small fraction of the total particles, so I wonder about the
statistical strength of these statements. Also, I wonder if these results are merely due
to size differences between the particle types, coupled with instrumental biases in size
sampling? Discussion of difference in size distribution of different particle types is
merited throughout the paper to the extent that differing sizes affect the interpretation
of the number fractions presented.

5. Throughout the manuscript, I suggest liberally adding the phrase “by number”, as I
often (e.g. 18428 line 23: sulfate (83%)) found myself thinking of mass fractions.

6. Section 3.3: Discussion on page 18429 lines 17-20: Without knowing the size
distribution of the soot at emission, which may well have extended into the A-ATOFMS
size range, the deduction that “soot” grew into the size range of the instrument is not
supported adequately.

7. Figures 4,5,6,7 do not show “ratios”. I suggest the authors adopt a less confusing
omenclature. Perhaps “mixing distributions”?

8. page 18432, line 28: “greater *relative* OC content”?

9. Page 18435: discussion centered around SI Figure 2 – Figure SI Figure 2 does not
show any significant size shift between NoCal-1 and -2. The broadening referred to in
the SI text appears to be predominantly an artifact of different particle statistics. The
related discussion in the SI and in the manuscript needs to amend for this misinter-
pretation. If the figure survives this, I suggest showing the number size distributions in
dN/dLog(D) space, as is normally done for aerosol populations.

10. I suggest including discussion in the context of the “ratio” figures of the physical
processes that lead to the fairly common minimum of “relative fraction” around equal
amounts of the different materials. Also, without context about the amount of “third
party” species, full understanding of these figures is not possible. Perhaps showing
additional lines for RPA > 10% and RPA > 20% would be sufficient for this.

Supplemental materials comments:

Discussion of “transmission efficiency” shift – this sounds more like simply a change
in flow rate. However, if this is the case, then particle statistics are biased towards the
time when flow was higher, no?

Line 68: missing space before “in” in “µmin”.

Interactive comment on Atmos. Chem. Phys. Discuss., 12, 18419, 2012.