We thank the reviewer for their helpful comments. The manuscript has been revised to address the reviewers' recommendations. Specifically, significant changes have been made to Sections 2, 3.1, 3.5, and the Supplementary Section. References (Zaveri et. al. 2012) and (Setyan et. al. 2012) have been updated. Additional responses to specific reviewer comments (repeated in italic) are given below.

Reviewer 1 comments:

1) *"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.*
    - Designating soot as BC is not appropriate, since BC implies a measurement of the absorbing component of the aerosol which includes brown carbon (EC + BrC = BC). The single particle mass spectra contain dominant carbon cluster ions at 12, 24, 36 and beyond. The particles labeled as soot in this study rarely contain peaks indicative of brown carbon such as aromatics or other UV/Vis absorbing OC species. We have re-labeled the types as soot and aged soot. The distinction between soot and aged soot is appropriate, as both types arise from incomplete combustion but differ in the relative abundance of secondary species present on the particle. 'Soot-OC' has been replaced with 'Aged Soot' in the manuscript in addition to extra clarifying text in Section 3.1.

2) *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.*
    - Additional information has been added to the main text and supplementary regarding A-ATOFMS transmission efficiency and aircraft inlet parameters. To be clear, A-ATOFMS does not measure "total particle mass", but instead the number or number fraction of the different particle types. Number fraction is not affected by transmission biases as the transmission biases are solely size dependent not chemistry dependent.

3) *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.*
    - We state "Calculated standard errors of number fractions were small, <1%, hence were not included in this discussion." This also includes the fractions stated on line 18431. Since the reported fractions are calculated from a combination of many flights in each study, which results in tens of thousands of particles, so the

calculated errors were small for all reported values as stated in the text. Once particle counts are greater than 3000 assuming simple random sampling with replacement, the calculated error (one standard deviation) is <1%. Hence, the reported fractions on pg 18431 '98% and 93%' are significant as well as the shorter time period values, since they both are calculated using well over 3000 total particles. Additional text has been added regarding variability in time and space. Though some variability was seen flight-to-flight during CalNex, generally number fractions for the main particle types did not deviate greatly from the reported values for each study as a whole.

4) *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.*
    - While both types represent a small fraction of total aerosol, the statistical errors in the reported fractions are derived using the total number of particles sampled. As stated above, if counts are greater than 3000 the error in the reported fractions will be less than 1%. 75969 and 60230 particles were measured for CalNex and CARES, respectively, and thus are statistically robust. In terms of size, additional text has been added throughout the manuscript and supplemental discussing the effect of size on the number fractions presented, as well as an additional supplemental figure featuring the size-chemistry relationship of the particles sampled for both studies. In general, number fractions of most particle types were consistent across the relatively narrow (100-1000 nm) A-ATOFMS size range and so size did not significantly influence the reported results. In both regions most carbonaceous particle types with the lone exception of HP particles (i.e, OC, BB, HP, Aged Soot and Soot) have increased fractions at lower sizes (< 250 nm), though the confidence in these fractions is weak due to low particle counts within this size range. Of all the particle types SS is the only type to have a significant dependence on size, with its fraction greatly increasing as size increases; consistent with typical supermicron size of SS particles.

5) *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.*
    - Typical 'pure' soot diameters measured during CARES were < 100 nm (Zaveri et al., 2012), and are below the A-ATOFMS optical detection limit. The particle type labeled in text as "soot" represents relatively fresh soot measured by the A-ATOFMS. The only way we could detect soot is if it extended into the A-ATOFMS size range. On days with very little soot (2 June 2010 – 19 June 2010), soot was still evident in the smallest sizes (<100 nm) based on other measurements. During the final days of the study (21 June 2010 – 28 June 2010),

there was more secondary pollution, and soot was detected in the larger sizes detected by A-ATOFMS (> 100 nm). These larger particles were mixed with OC, nitrate, and sulfate. The 11 to 27% increase of Soot-OC from NoCal-1 to NoCal-2, respectively, indicates that fresh soot aged and grew in size to the peak transmission of the A-ATOFMS. In addition, UHSAS, CPC, and UFCPC number concentrations discussed in Section 3.5 support the fact that soot (and other) particles grew in size from NoCal-1 to NoCal-2. As stated above, the term 'Soot-OC' has been replaced by 'Aged Soot' to avoid confusion.

6) *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 misinterpretation. If the figure survives this, I suggest showing the number size distributions in dN/dLog(D) space, as is normally done for aerosol populations.*
   - We agree with the reviewer. SI Figure 2 and associated discussion has been removed from the manuscript. The A-ATOFMS detected only a narrow size range during the CARES study. This is why the shift between NoCal-1 and NoCal-2 was very small.

7) *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.*
   - It is not clear what the reviewer thinks is too narrow. The title covers the main points of the manuscript, and thus we did not change it.

8) *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.*
   - Thank you for the suggestion. We have added these clarifications where appropriate.

9) *Figures 4,5,6,7 do not show "ratios". I suggest the authors adopt a less confusing nomenclature. Perhaps "mixing distributions"?*
   - Where appropriate 'ion ratio distribution' has been used when referring to these figures, though we would like to emphasize that every bar in the graph is a collection of single particle ion peak ratios.

10) *page 18432, line 28: "greater *relative* OC content"?*
    - This has been corrected.

11) *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.*

- During both studies, sulfate and nitrate far outweighed the influence of additional "third party" species, and did not have a significant influence on RPA. We had included additional lines for RPA > 10% and RPA > 20% in a previous version of the manuscript but found that (1) it did not provide any additional insight into the dominance of the species in question, (2) that inclusion of additional RPA lines sufficiently complicated the graphs making understanding more difficult, and (3) because the RPA fraction is looked at in bulk it can lead to misinterpretation, which is precisely why single particle ratios were used. For these plots the fairly common minimum can be attributed to an increased number of bins for ratios between 2:1 and 1:2 as compared to higher ratios. Due to the exponentially increasing nature of the ratios, bins approaching -1 or +1 cover a larger range of ratios than those bins near 0. Additional text has been added to clarify this.

12) *SI 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?*
    - The shift discussed was a result of a slightly altered operating pressure of the aerodynamic lens. This can potentially have an effect on particle transmission. However, this had a negligible effect on both particle statistics and size dependent transmission. For completeness we included it in the manuscript, but upon further inspection we removed this discussion from the manuscript.

13) *SI Line 68: missing space before "in" in "umin".*
    - This has been corrected.

References

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