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 2 comments

1) *The authors never provide much information on the size dependency of the mixing state. Given that there are more than 75,000 particles analyzed in the southern California data set and 60,000 particles from the northern California flights, surely there are sufficient number of particles to show the mixing state as a function of size. This does not have to be stratified in that many bins, but given that the instrument should capture a large portion of the accumulation mode and at least part of the coarse mode, it might at least shed some light on the processes that led to the various internal mixtures.*
    - See comments above. Additional information has been added to the supplementary concerning A-ATOFMS transmission efficiency and the dependence of chemistry on size. For both regions the size dependent chemistry was remarkably uniform across the entire measured size range of the A-ATOFMS, with a few exceptions. 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.

2) *The second detail that should be addressed has to do with the temporal evolution of the mixing states during the day, particularly during the morning flights. Although little is said about photochemical processes that are likely very important in the growth and mixing of the particles, the morning flights that started at 8 am in the morning and extended until around noon, all in the same general area, offer the opportunity to see if the particle chemistry is changing as the boundary layer grows and the sun rises. Likewise, why not also compare the average properties in the morning with those in the afternoon.*
    - There was no consistent difference in chemistry between morning and afternoon flights, with number fractions in the afternoon lying within 5% of the morning fractions. A more detailed Lagrangian analysis of the dataset beyond the scope of this paper is likely needed to resolve chemical differences. Chemical evolution is being investigated in more detail for a future manuscript so that this important topic can be covered adequately.

3) *The detection limits for mass need defining. At one point reference is made to the fact that "soot" is below the detection limit of the mass spectrometer when it is freshly emitted, but this was referring to its aerodynamic diameter. The paper goes on to explain*

that the soot grows into the size range of the instrument but what isn't clear to me is the minimum mass that can be detected. The growth of the soot is not the soot itself actually growing, if it is indeed EC (unless it is growth by coagulation with other EC particle), but is soot that is acquiring a coating of OC or some inorganic. If the core of the particle is EC, but its mass is below the detection limit of the mass spectrometer, then a fair amount of soot mass is possibly being missed.

- To clarify, detection limit in the A-ATOFMS is referring to the lower size cutoff based on optical scattering in the instrument rather than any mass dependent limit. By "growing" we refer to the growth of small soot particles with coatings of OC, sulfate, and nitrate above the lower size detection limit (>100 nm) of the instrument. These mixtures of soot and coatings are represented by the newly termed Aged Soot particle type in the revised paper. Additional text has been added to clarify the appropriate sections.

4) *Nothing is said about the counting statistics and how representative these measurements are. There are over 75,000 particles in the one case and 60,000 particles in the other. This sounds like a lot, but given the approximately 115,000 seconds of sampling time in the 75,000 particle case (8 flights at approximately 4 hours per flight) and 288,000 seconds in the 60,000 particle case, this works out to be 7 particles and 2 particles per kilometer, respectively, assuming a cruise velocity of 100 ms-1. This is an uncertainty that needs addressing with respect to how representative the data are.*

- While the reviewer is correct in stating that there should be more uncertainty when resolving down to a single kilometer, no attempt of this sort is made in this manuscript. Instead we wish to comment on the region as a whole, which all particles measured in these studies contribute to (75,969 and 60,230 particles for CalNex and CARES, respectively), and hence provide additional statistical robustness. The calculated error in reported fractions using these counting statistics assuming simple random sampling with replacement are <1%.

5) *The term soot needs defining at the very beginning since I started reading thinking that the authors were talking about the mixture of EC and OC such as is usually the case when talking about soot; however, I later realized that they were using soot as another name for elemental carbon. This was rather confusing throughout.*

Additional text has been added to clarify the discussion of soot, as discussed in response to reviewer 1 comments. 'Soot-OC' has been replaced with 'Aged Soot' in the manuscript in addition to extra clarifying text in Section 3.1.

6) *The other uncertainty not addressed concerns the particle losses in the inlet system. Is in isokinetic, is there dynamic heating that would lead to volatilization, how long is the distance between inlet and spectrometer, etc.?*

- Additional information has been added to the supplementary regarding aircraft inlet parameters. The Twin Otter aircraft (CalNex) inlet transmitted ~100% of particles up to 3500 nm (Hegg et al., 2005), with the Gulfstream-1 (CARES) transmitted near unity up to 5000 nm (Zaveri et al., 2012). The Twin Otter inlet is sub-isokinetic while the Gulfstream-1 inlet is isokinetic (leading to the lower size

cutoff compared to the Gulfstream-1). However, the transmission of both inlets is near unity within the A-ATOFMS size range (100-1000nm). In both, aircraft sampling lines were reasonably similar, ~2 m long and unheated, so no further corrections are warranted.

7) *Does the TSI 3010 really have a lower cut-size of 10 nm? I thought it is usually reported as > 20 nm.*
    - Yes, TSI 3010 manual states the lower cut-size of CPC 3010 is 10 nm.

References

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