Reply to Reviewer # 1.

REVIEWER SUMMARY
Reviewer Comment:
I started reading this paper with great interest and expectation. Unfortunately, this turned out to be a summary of results from the CARES campaign and with no particular highlight.

Author Reply:
The CARES campaign highlights a transition from spring to summer conditions, from windy, lower temperature, primary aerosol driven optics to a steadily increasing summer condition with higher temperatures, lower wind speeds, and more secondary aerosol driven optics. The authors feel that nature provided an ideal case study for comparing and contrasting the impacts of primary versus secondary aerosol on aerosol optics.

Reviewer Comment:
The paper goes through measurements made with the PASS and a nephelometers in great detail and draws on other data sets collected during the experiment to support some of the analysis. There is no 'conclusion' as such and it is more appropriate to call this a summary of the data. The primary result, that the Angstrom exponent changed from 1.6 to 1.8 from an upstream site to a downstream site, separated by a distance of 40 km, as an indication of BrC in the air mass seems a little suspect.

Author Reply:
We felt that it was necessary to summarize the aerosol optics and chemistry measurements before drawing conclusions from it. We assembled data from a broad set of instruments.

As noted by the reviewer, one of the main conclusions in the paper was observation of a modest increase in Angstrom exponent. Note that the Angstrom exponent changed from 1.6 to 1.8. In other words, given that fresh diesel soot has an Angstrom exponent of closer to 1.0, the effects of coatings, aerosol morphology, and aerosol chemistry have already made an impact on the spectral aerosol light absorption compared with fresh emissions.

We found that the steady increase of secondary aerosol did not dramatically increase aerosol light absorption - that in this case, the secondary aerosol was not strongly brown. It is an important observation. While we all like to see dramatic Angstrom exponents of 3 or greater, as in the in case of primary emissions from biomass burning, that was clearly not the case in CARES. We had to pay careful attention to errors, and provide error bars for our measurements.

However, the reviewer missed some of the other important conclusions. We presented and analyzed the multispectral BC mass-normalized absorption cross-section (MAC) values ranging from ultraviolet to IR wavelengths for T0 and T1. We found MAC values for ultraviolet wavelengths at T1 increased by about 60 % compared to the relatively less aged urban emissions at the T0 site. Furthermore, we found that super micron aerosol contributed significantly to scattering, so much so that the 870 nm and beyond scattering was as if it were due to a different portion of the aerosol size distribution than the 532 nm and shorter wavelength scattering results.

In summary, our paper provides important insights into the impacts of aerosol on spectral light absorption, scattering, and extinction, and their connections, or not, with aerosol microphysics and chemical composition.

Reviewer Comment:
There is no discussion on the uncertainty in PASS measurements and given a range of approximately 20% uncertainty in other similar instruments, this difference of 10% seems fairly unremarkable and within the uncertainty range of the instrument. The primary problem may be that the data set is from a very clean environment and nothing much has happened during the study. The BC measurements are listed at about 0.06 ug/m3 to 0.16 ug/m3 on average. This is a fairly small signal and other numbers such as absorption are factors of 10 to 50 smaller than at moderately polluted sites. This introduces a large Signal/Noise problem in evaluating the results presented here and extracting any major conclusions.

Author Reply:
CARES was relatively clean compared with other campaigns we have been involved with. That is one overall, and interesting, conclusion of the study. Call it economic downturn and/ or environmental concern and/or air pollution meteorology; the relatively large metropolitan complex of Northern California is pleasantly clean considering its size. We presented our results in the context of instrument uncertainties and provided error bars.

Reviewer Comment:
I appreciate all the work put into explaining the correlation between the wind directions and measurements and the possibility of mixing with biogenic emissions and anthropogenic emissions creating larger particles to explain some of the results.

Author Reply:
We're glad that this message was conveyed to the reader.

Reviewer Comment:
Much of these results are similar to what was seen before and probably less obvious in this data set.

Author Reply:
We have not published our results elsewhere.

Reviewer Comment:
My primary issue is that there is no payback after spending several hours reading through this paper and the conclusions reads more like a summary than a conclusion.

Author Reply:
It is a bit like going to a restaurant. Some people are satisfied by steak. Vegetarians would rather graze in the backyard than eat steak. The facts of the study are as we portray them. The drama associated with increasing secondary aerosol contribution throughout the campaign provided us with much enthusiasm for analysis. The conclusions can not be over-hyped - they are what they are - we did not end up with a huge feast of absorption Angstrom exponent changes, but instead had to settle with strong increases in the short wavelength scattering coefficient. By nature the Angstrom exponent for absorption is much more subtle than the scattering exponent as the former is due to a complex interplay between aerosol morphology and composition whereas the latter is mostly just a function of aerosol size. Sometimes the lessons learned from an exercise can only be appreciated through careful reflection.

Reviewer Comment:
I am not entirely sure as to the purpose of Mie calculations. The refractive index is unconstrained (reasonable value used), the size ranges are prescribed and shape is assumed.

Essentially there is a single constraint on what is a probably three parameter model and the results are in reasonable agreement with measurement are reasonable given the constraints imposed on the model. I am guessing, we have to conclude from the calculation that the shapes are closer to spherical.

Author Comment:
The reviewer did get our purpose for including the scattering calculations based on Mie theory. Choice of a modestly common, single refractive index resulted in a calculated scattering that is in reasonable accord with measurements. The manifestation of our ability to model aerosol impacts is exemplified by our ability to display closure. Scattering calculations are also very helpful for providing a 'second opinion' for direct measurements given that instruments have real world issues like aerosol transmission efficiency as a function of particle size and nephelometer truncation angle realities.

Some specific Reviewer comments:
1. The use of AEA (angstrom exponent of absorption) is confusing. The general use in literature is AAE and may be helpful to stick with that. This field is already pretty confusing.

Reply:
AE, SE, and EE would be the most concise representation of these exponents, leaving out the tribute to 'Angstrom' and the confusion provided by having 'A' represent both 'Angstrom' and 'Absorption'. It could be that in the future that the community would find a reasonable standardization. However, the most important thing for now to is to provide consistency with a definition and its use throughout the paper.

2. Why not use the same wavelength pairs for making the AEA calculations? The AEA does seem to depend on the choice of wavelength pair as noted by Russel et al., 2010 and others.

Reply:
The AEA of course does depend on wavelength choice. We tried to use as many wavelength pairs as possible, especially highlighting the 355 nm results since we know least about the optics of aerosol at this wavelength.

3. A value of 1.8 for AEA seems much closer to dust type aerosols than BrC (Bergstorm et al., 2007). I guess this being a point measurement as opposed to column values in Bergstorm makes a difference?

Reply:
The reviewer makes a good point that the AEA of dust need be considered as an explanation for the observed AEA of 1.6 or 1.8. The AEA is of course weighted by the strongest absorber, typically submicron carbonaceous aerosol in an urban environment. Chemical measurements seem to point toward sea salt as the primary super micron aerosol composition. However, column measurements may very well have different optics than surface values. Our treatment of super micron aerosol optics and chemistry is coarse at best, as is our assessment of column versus surface properties, even in conditions when the entire column is dry.

4. Line 74; line 82; line 84: The use of cooling the climate and warming the climate is probably not common. Climate change or global warming or cooling may be more appropriate.

Reply:
Our intention here is to provide only a useful rough guideline for aerosol impact on climate. Our hope is that these results help constrain models for aerosol light absorption and scattering so that we may collectively do a good job of understanding the role of aerosol in climate, weather, and ecosystem health.

5. Line 88: have identified OA as a major absorber - This is a big claim. I would modified it to say something like 'a small fraction of OA was shown to absorb'.

REPLY:
Agree as will appear in our revision.

6. Line 188/189: Why use different wavelength pairs for calculating AEA at T1 and T0?

REPLY:
Our choice of wavelength pairs was driven by our best measurements over the longest time period during the campaign. Not all instruments were functioning for the entire campaign.

7. Line 317-319: I can't follow the logic here - why can't fresh or local emissions account for this change and why is a transport and particle size increase needed to explain this observation?

REPLY:
We only had access to page number and line number on a particular page, so unfortunately can not reply to this comment.

8. The AEA variation from 1.3 to 2.3:the higher values seem more like dust, have there been any measurements of dust aerosols during this time?

REPLY:
The AEA is driven by the strongest absorber present, carbonaceous aerosol in CARES. Supermicron aerosol chemical composition has not been measured with the same intensity as sub micron aerosol.