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Atmospheric Chemistry and Physics Discussions

Interactive comment on "Aerosol Optical Extinction during the Front Range Air Pollution and Photochemistry Éxperiment (FRAPPÉ) 2014 Summertime Field Campaign, Colorado U.S.A." *by* Justin H. Dingle et al.

Anonymous Referee #2

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Review of Aerosol Optical Extinction during the Front Range Air Pollution and Photochemistry Éxperiment (FRAPPÉ) 2014 Summertime Field Campaign, Colorado U.S.A.

Overall Comments This paper describes relationships between mass measurements from the mAMS and extinction measurements from a CAPS-PMex instrument in the Front Range of Colorado. The authors derive Mass Extinction Efficiencies for a number of different airmass types (urban, oil and gas, agricultural) based on gas-phase tracer measurements from the study. The paper presents some interesting, relevant results and is appropriate for publication in ACP after some significant revisions described

below. The major problems with the submitted manuscript are

1.) The error estimates for the CAPS-PMex are not explained or justified for this study, only a reference is given. The error estimate of 10% seems much too small for small extinction measurements (<10 Mm-1) given that the authors observed significant baseline shifts. 2.) The authors see poor correlation between mass measured by the AMS and extinction. This poor correlation is stated to be due to changes in size distribution. However, in some of the plume types, especially urban, there does not appear to be a dramatic difference in the size-distribution of low and high MEE plumes. If the authors are going to conclude that size distribution is responsible for widely varying MEE, this needs to be backed up with some Mie modeling that shows the observed shifts in size distribution can cause the observed shifts in MEE. It is also possible that varying size cutpoints between the mAMS and CAPS-PMex are significant an this needs to be discussed.

Additional comments are given below.

Comments on Introduction 1.) It is stated that, "it has been observed that the important contributors to light scattering in the Colorado Rocky Mountains are particulate matter from the urban emissions" Certainly there are dust and smoke events that cause visibility degradation too, in fact the authors show a smoke event. This statement is far too broad.

2.) The authors discuss the wintertime phenomenon of the Denver Brown Cloud but do not discuss summertime visibility issues. Some background on typical extinction, or at least PM2.5 mass in the summertime in Denver is needed.

3.) The statement "twofold increase in natural resource extraction" is vague. Is this an increase in wells, in gas production, in oil production? This is relevant because each has a different implication for air quality.

4.) The authors state that emissions from oil and gas include "emissions from industrial

equipment known to emit. . .particulate matter." Are they referring to BC from diesel engines? There needs to be clarification here because there are not typically large primary emissions of particulate matter from oil and gas operations.

5.) I don't see a basis for the authors to conclude that photochemical production of ozone is significantly impacted by oil and gas operations. There's a big jump from OH reactivity in the winter to ozone production in the summer and no evidence to back up this logical leap. This also has little to do with the paper, I would suggest removing the last line of page 2 and the first line of page 3.

Comments on Instrumentation and Methodology 1.) The description of the CAPS-PMex operating principles is fairly poor. It makes it seem that the extinction is detected by monitoring the phase shift that occurs during 1 transit of the light from the first mirror to the second. This is far from accurate, the entire point of the instrument is to create a very long effective pathlength.

2.) Line 4 page 4: An averaging time needs to be given for uncertainty and detection limit statements.

3.) Why were baseline values only interpolated when the shift was more than 5 Mm-1? I see no reason not to always interpolate them. It also needs to be explained why the baseline would shift that much. Given that most reported measurements are <20 Mm-1, this would seem to be a big issue.

4.) Error analysis for the CAPS seems to need significantly more attention in general. 10% is clearly not a correct estimate of error in the extinction coefficient when the average extinction is something around 15 inverse Mm-1 and you have a baseline shift of 5 Mm-1. The authors need to do a careful assessment of error in the extinction measurement.

5.) Why is particulate nitrate not considered in the NOx/NOy ratio? This seems odd given the authors are measuring particulate nitrate with the mAMS.

6.) A PCASP does not seem to be the best instrument for submicron aerosol analysis. Can the authors comment on why an SMPS or UHSAS was not used in addition to the PCASP? If these instruments were not on the C-130, this needs to be stated.

7.) There needs to be a discussion of the aircraft inlets and size cutpoints for the various instruments. This is especially critical for the PCASP vs. CAPS-PMex vs. mAMS given that the instruments have different cut points around a micron.

8.) There needs to be a discussion of how the mAMS data was corrected for collection efficiency, especially given that agricultural plumes had significant nitrate.

Comments On Results and Discussion 1.) Figure S1 appears to show roads but this is not described in the caption

2.) Again, error analysis is a problem. The uncertainty in Bext being stated to be 10% when the authors state that the baseline shifts by over 5 Mm-1 and that the 1-sigma detection limit is 1 Mm-1 seems completely inconsistent. Values of Bext of less than 5 Mm-1 are shown and the authors are suggesting the error is < 0.5 Mm-1 for these measurements but this is half of the 1-sigma detection limit and does not account for baseline shifts. The authors make a major point that they are doing ODR because they want to weight according to the errors in the instruments, but the errors seem incorrect.

3.) Page 5 Line 17. The delta ext/co ratios have error bars associated with them, but the authors don't explain what they are. Are these 1-sigma intervals on the slope of the fitted line? For individual measurements the errors would be much larger than these ($\sim$11%), I am surprised at how small they are given the poor r of the fits. Also, the authors remove the error in the intercept from the discussion even though this is probably significant.

4.) The authors need to explain how they arrived at the criteria of 0.7, 0.3-0.7, and <0.3 for NOx:NOy ratios.

5.) Page 5 Line 29. The authors state that the "bulk" of the organic aerosol mass is

SOA because the enhancement ratio of organic aerosol increases significantly for aged aerosol. This is not adequate reasoning. The aerosol could be unrelated to CO (from non-combustion sources), as the authors themselves point out. The increasing Bext could be the result of increasing ammonium nitrate when plumes move farther from the city. The authors need to explain what fraction of aerosol mass is accounted for by the OA:CO enhancement or something in addition to the current argument if they are going to make this sweeping statement. Also, the word "bulk" needs to be quantified in some way.

6.) Page 6 Line 16. Correlation coefficients of .55 are not "strong" 7.) Page 6 Line 23. Nitric acid + aerosol nitrate is not "total" nitrate, this ignores organic nitrates. 8.) The relatively weak correlations in urban plumes between Bext and any of the aerosol species (all have r less than 0.55) mass concentrations suggests that there was poor correlation between total AMS mass and Bext. In fact, this is born out in figure 4. This is very confusing and needs to be addressed, especially given the very good correlation between Bext and nitrate in agricultural plumes. The authors suggest it may be caused by a change in size distribution (Figure S2 a-b), but it appears to me that the total mass distributions in these two figures are fairly similar, even though the organic distributions are a little different. Are the authors missing an important aerosol species? Alternaltively, is the mismatch in measured aerosol size between the CAPS and AMS significant (Figure S2 suggests this may be the case and that a careful application of the lens transmission is required)? If the authors are going to rest on the size-distribution argument then they need to implement a Mie model of the two distributions shown in S2 and demonstrate that they yield dramatically different Bext for the same total aerosol mass. The size distribution argument for urban + OG is more convincing.

9.) In Figure 4 the removal of the intercept and its associated error is again not mentioned, even though it is a significant issue given the lines don't go through 0,0 and the r values are fairly low. I am again of the opinion that the stated error in the slopes does

not accurately represent the error in this slope.

10.) Page 8 Line 7-8. If the authors are going to make this broad conclusion they need to some basic Mie modeling to convince the reader that these shifts in size distribution can generate MEE from 1.8 to 4.1, this does not seem obvious.

11.) In Figure 7a "total mass" needs to be changed to "total non-refractory PM-1 mass", though even the PM-1 designation is questionable with AMS data.

12.) In Figure 7a the fit to the non-biomass burning emissions does not seem to track the data. The authors need to discuss this. Perhaps it is due to including moderate HCN data points.

13.) Again in Figure 7 the intercepts are removed and not discussed, they need to be discussed because they are very different between biomass burning and non-biomass burning and this has physical meaning.

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