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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 #1

Received and published: 10 May 2016

This paper discusses observed light extinction of nominally PM2 particles measured over the Colorado front range during the FRAPPE aircraft study. The authors assert that this paper provides an updated assessment on the Denver Brown Cloud. This is a worthwhile topic and the paper is suitable for publication in ACP. However, there are number logical inconsistencies, important missing information, and other issues that must be addressed, including:

-particle size range and RH of the extinction measurement is not well characterized making the data of questionable value (ie, how to compare to other studies and how to

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apply to ambient conditions).

-mismatch between AMS and extinction measured particle size ranges.

-the justification for the use of extinction versus CO to compare extinction versus photochemical age for all combined sources.

Also, given the discussion in the Introduction that the motivation of this work was to take a new look at the Denver Brown cloud, it is rather odd that this is never done. It would be insightful to add a section on comparing/contrasting these results to earlier studies; has visibility improved, have sources that contribute to visibility reduction changed, etc.

Specific comments:

Why is there no discussion of any anthropogenic gases that may contribute to the Denver Brown Cloud, either in past studies or this study? Are they not important (give numbers to support). Are they included in the reported extinction measurement, or subtracted out with the blank correction?

Page 3 and throughout; specifically note that the altitudes give are above sea level (I assume), not surface?

Page 4, line 18; the CAPs(ext) did not have a size selective inlet; apparently upper size limit is controlled by only inlet/sample line transmission efficiencies? Discuss in more detail, specifically how well is the size range of particles contributing to the measured extinction really known (give the uncertainty, my suspicion is that it is large of it is bases solely on calculated inlet and sample line transmissions). What are the implications of this uncertainty (the size distribution was measured so a quantitative estimate should be possible).

How does one handle the mismatch in particle sizes sampled with the AMS and CAPs? This could have impacts on much of the reported data, depending on the shape of the size distribution. Add a discussion.

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No discussion on RH (or T) of sample in the CAPS? RH variability could have a large effect on extinction. In the paper it is referred to as dry extinction, but RH is never given? It appears that the authors are just assuming the particles are dry since the ambient RH was low and the particles heated in the inlet/sample line. Much more detail, along with possible differences in LWC of sampled and ambient aerosol, should be considered. Note, at the least one could estimate the RH in the CAPS assuming the aerosol has reached cabin T, if one knows the ambient RH and T. Claiming a dry extinction measurement really requires reporting actual RH in the CAPs.

Page 4 line 22, typo, intends or just tends?

Re. Fig 2 and the general idea of looking at extinction vs CO: The logic behind the graph and more details may be needed. First, is this data just for well defined plumes or include all data, except biomass burning (ie, it includes urban and agri, urban+O&G, and O&G)? Second, this plot is predicted on a correlation between extinction and CO; that is that the components driving extinction and CO are co-emitted in all sources included in this plot. This appears to be the case, but it is curious why this is so if it includes all these various sources. That is, if this plot is for all sources combined, why do they all have similar Ext/CO ratios (ie, only a function of age)? Maybe this plot is mainly driven by urban emissions. This would also mean that most of the aging is just due to OA aging. Fig 3 would support this, in a general sense. Why not use a PMF analysis and look at evolution of specific AMS OA factors?

Why lump all the data together in this plot since it is more valid for a plume from a specific source; wouldn't graphs like this for each specific source make more sense, or maybe just focus on the urban data? Also, one would expect that some components that contribute to extinction, such as sulfate and nitrate would not be correlated with CO and so not appropriate to include sources with high emissions of these components in this analysis. Maybe this accounts for much of the scatter? One might also give the overall r2 between extinction and CO (ie not segregated by age) in Fig 2, and finally, why the different intercepts in Fig 2?

Fig 3, any estimates on potential bias in the composition data due to sampling only submicron non-refractory aerosol with the AMS? In some sources this could lead to substantial bias, eg, the AMS would not measure more refractory nitrate salts that could be present in some of the sources (eg, NaNO3, Ca(NO3)2, . . .).

Why is there so much OA associated with agri emissions?

Page 6 last line, the assumption is being made that nitrate formation is controlled by NH3 concentrations through partitioning of nitric acid. What is the justification for this? The process is actually likely to be much more complicated as it depends on the pH of the aerosol, which in turn depends on the amount of mineral dust and sulfate also present; it doesn't just depend on NH3 concentration. Also, given that NH3 was measured, one could be more specific and quantify the differences in NH3 levels in the various source regions.

Fig 6, how can there be so few particles (generally less than 40 or so particles per cm3 of air, get mass concentrations are up to 15 to 20 ug/m3? Seems very odd.

Fig 8, the correlations are not that good, total mass explains only 25 to 35% of the extinction variability (r2), so are the regressions really meaningful (comparisons of slopes for each plot)?

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