We appreciate having received detailed comments from the reviewers. We have revised the manuscript accordingly. Below, you will find our response and the summary of our approach, highlighted in *red,* with modifications to the manuscript highlighted ***in bold***:

Referee #1:

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 apply to ambient conditions).

The reviewer has raised a good point. We have estimated the RH in the CAPS-PM$_{ex}$ unit, using the measured ambient temperature and RH assuming aerosols had equilibrated to the temperature within the instrument. Our results indicate that on average the RH in the CAPS was 20 ± 7% with a range of 15-30% while ambient RH was on average 44±17%.

We have addressed this issue in section 2.2, paragraph 4 by adding the following sentences: ***"Based on the ambient RH and temperature and the temperature within the CAPS-PM$_{ex}$ extinction cell, and assuming that aerosols had equilibrated to the conditions within the measurement cell, the CAPS-PM$_{ex}$ measurements for the flights discussed here represent extinction values at an average RH of 20 ± 7 % (range of 15-30%)."***

-mismatch between AMS and extinction measured particle size ranges.

In order to determine whether the discrepancy between the aerosol size ranges being sampled by the CAPS-PM$_{ex}$ and AMS had a significant impact on our analysis or not, we have used the size distributions from the Passive Cavity Aerosol Spectrometer Probe (PCASP) instrument on board the C-130 to estimate the ambient scattering coefficients. By using a nominal refractive index of 1.5, estimated scattering (i.e., extinction, while assuming purely scattering aerosols) coefficients were calculated using the measured size distributions up to 800 nm (upper "true" size cut of the AMS) and 2000 nm (upper size cut of the inlet, and thus CAPS-PM$_{ex}$). The slopes of the scatter plots of the estimated scattering coefficients for PM$_{0.8}$ vs. PM$_2$ under the influence of urban, O&G, agricultural, and urban+O&G slopes were 0.95 ± 0.01, 1.0 ± 0.002, and 1.0 ± 0.01, 0.92± 0.01, respectively, indicating that the majority of the signal contribution to extinction originated from aerosols in the size range of the mAMS. We also note that the slope values mentioned above were not highly sensitive to the choice of the refractive index. Changing the refractive index from 1.48 to 1.52 changed the slope values by at most 4%.

The following has been added to the text in section 2.2, paragraph 3 to address this issue: ***"Ambient aerosol size distributions were measured on-board the C-130 by a Passive Cavity Aerosol Spectrometer Probe (PCASP). Estimated extinction values using Mie calculations with a nominal refractive index of 1.5 and the measured PCASP size distributions indicated that particles smaller than 800 nm captured >92% of PM$_2$ extinction values, confirming that the majority of the extinction signal originated from aerosols in the size range of the mAMS. We note that the calculated extinction coefficients were not highly sensitive to the choice of refractive index; only a 4% decrease in the slope of scattering coefficients from PM$_{0.8}$ vs. PM$_2$ was observed by increasing the refractive index from 1.48 to 1.52."***

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

We agree with the reviewer that it makes more sense to limit the data in Figure 2 to plume types where aerosol precursors are co-emitted with CO. Therefore, Figure 2 has now been updated to include data from urban emissions only, with the modified definition provided in Section 3.1 as ***"…plumes with enhancement of CO over the background (105 ppbv, as defined by the mode in the frequency distribution of CO in the Front Range boundary layer) while $\Delta C_2H_6/\Delta CO < 20$ pptv ppbv^{-1}****). We also had to remove data from July flights due to lack of optimum quantitative quality of CO data during those flights, that was reflected on the data archive site after initial submission of the paper. With these changes, aging categories needed to be updated to $NO_x:NO_y > 0.5$ and <0.5 to represent relatively fresh and aged plumes, respectively, in order to include enough data points in each category. Despite these changes, the conclusions remain the same that with the reduction in NO_x/NO_y and increase in photochemical aging, the enhancement ratio of $\Delta\beta_{ext}/\Delta CO$ increased significantly (by ~54%).*

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.

Since summertime extinction data from previous field studies in the Colorado Front Range are not available, we have used transmissometer extinction data, provided by the Colorado Department of Public Health, to consider monthly average values of extinction measured in Downtown Denver for the months of July and August during 2001-2014.

We have included the following sentences in section 3.3, paragraph 4 describing the observations: ***"In response to the wintertime haze episodes observed in the region, the State of Colorado has implemented a visibility standard based on total optical extinction of 76 Mm^{-1} at 550 nm, averaged during a 4-hr period when ambient RH is less than 70% (Ely et al. 1993). Total optical extinction measurements are provided by the Colorado Department of Public Health and Environment's transmissometer, installed in Downtown Denver. We have assessed the average monthly total extinction coefficients for August of 2001-2014 to examine the recent trend in summertime extinction and visibility in the region. Averaged monthly values varied from 40 to 80 Mm^{-1}, and no significant trend was observed since 2001."***

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?

Based on wintertime optical extinction measurements in 1978, (Groblicki et al. 1981)estimated that gaseous scattering and NO_2 absorption each contributed to 7% of total extinction at 550 nm. As described in Section 2.2, CAPS-PM$_{ex}$ provides only measurements of aerosol optical extinction since frequent filtered-air samples are collected during normal operation to subtract the background gaseous contributions to extinction. Following Groblicki's derivation of absorption coefficient at 550 nm using NO_2 mixing ratios and since NO_2 absorption cross section at 632 nm is about 10× lower than at 550 nm (Schneider et al, 1987), estimated average NO_2 absorption at 632 nm in the Front Range was less than 0.1 Mm^{-1}. Therefore, although the reported measurements of extinction are for aerosol particles, contribution of anthropogenic gases to total extinction at 632 nm in the Front Range is negligible.

The following has been added to Section 2.2: ***"It is worth reiterating that anthropogenic gases such as nitrogen dioxide have minimal effect on the measured β_{ext} at 632nm since regular baseline***

corrections based on sampled filtered air were applied to the data. Given the average mixing ratio of NO_2, the parameterization by Groblicki et al. (1981) for estimating NO_2 absorption at 550 nm, and the factor of 10 smaller value of NO_2 absorption cross section at 632 nm compared to 550 nm (Schneider et al, 1987), we estimated the average absorption of NO_2 to be ~0.1 Mm^{-1}, indicating a minor contribution of NO_2 to total extinction at 632 nm."

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

In section 2.1, we have now noted that altitude is above sea level.

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.

Since the data presented in the manuscript were limited to the boundary layer, variations in the transmission efficiency of the inlet were really minor. We have calculated transmission efficiency of the inlet given a range of ambient pressures (760-860 mbar) and ambient temperatures (15-30 °C) representative of the BL; the 50% size cut for these conditions was 2.05± 0.05 um. As further discussed below, most of the signal contribution to aerosol extinction was from much smaller particles (<800 nm), so minor variations in the transmission of ~2 um particles could not pose significant uncertainties in the measurements.

In order to determine whether the discrepancy between the aerosol size ranges being sampled by the CAPS-PM_{ex} and AMS had a significant impact on our analysis or not, we have used the size distributions from the Passive Cavity Aerosol Spectrometer Probe (PCASP) instrument on board the C-130 to estimate the ambient scattering coefficients. By using a nominal refractive index of 1.5, estimated scattering (i.e., extinction, while assuming purely scattering aerosols) coefficients were calculated using the measured size distributions up to 800 nm (upper "true" size cut of the AMS) and 2000 nm (upper size cut of the inlet, and thus CAPS-PM_{ex}). The slopes of the scatter plots of the estimated scattering coefficients for $PM_{0.8}$ vs. PM_2 under the influence of urban, O&G, agricultural, and urban+O&G slopes were 0.95 ± 0.01, 1.0 ± 0.002, and 1.0 ± 0.01, 0.92± 0.01, respectively, indicating that the majority of the signal contribution to extinction originated from aerosols in the size range of the mAMS. We also note that the slope values mentioned above were not highly sensitive to the choice of the refractive index. Changing the refractive index from 1.48 to 1.52 changed the slope values by at most 4%.

The following has been added to the text in section 2.2, paragraph 3 to address this issue: ***"Ambient aerosol size distributions were measured on-board the C-130 by a Passive Cavity Aerosol Spectrometer Probe (PCASP). Estimated extinction values using Mie calculations with a nominal refractive index of 1.5 and the measured PCASP size distributions indicated that particles smaller than 800 nm captured >92% of PM_2 extinction values, confirming that the majority of the extinction signal originated from aerosols in the size range of the mAMS. We note that the calculated extinction coefficients were not highly sensitive to the choice of refractive index; only a 4% decrease in the slope of scattering coefficients from $PM_{0.8}$ vs. PM_2 was observed by increasing the refractive index from 1.48 to 1.52."***

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.

The reviewer has raised a good point. We have estimated the RH in the CAPS-PM$_{ex}$ unit, using the measured ambient temperature and RH assuming aerosols had equilibrated to the temperature within the instrument. Our results indicate that on average the RH in the CAPS was 20 ± 7% with range of 15-30% while ambient RH was on average 44±17%.

We have addressed the issue on RH in section 2.2 paragraph 4 by adding the following sentences: ***"Based on the ambient RH and temperature and the temperature within the CAPS-PM$_{ex}$ extinction cell, and assuming that aerosols had equilibrated to the conditions within the measurement cell, the CAPS-PM$_{ex}$ data discussed here represent extinction values at an average RH of 20 ± 7 % (range of 15-30%)".***

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

The sentence has been rephrased.

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?

We agree with the reviewer that it makes more sense to limit the data in Figure 2 to plume types where aerosol precursors are co-emitted with CO. Therefore, Figure 2 has now been updated to include data from urban emissions only, with the modified definition provided in Section 3.1 as ***"…plumes with enhancement of CO over the background (105 ppbv, as defined by the mode in the frequency distribution of CO in the Front Range boundary layer) while $\Delta C_2H_6/\Delta CO$ < 20 pptv ppbv^{-1}****). We also had to remove data from July flights due to lack of optimum quantitative quality of CO data during those flights, that was reflected on the data archive site after initial submission of the paper. With these changes, aging categories needed to be updated to NO_x:NO_y > 0.5 and <0.5 to represent relatively fresh and aged plumes, respectively, in order to include enough data points in each category.*

Carrying out PMF analysis is outside the scope of this paper.

Indeed the correlation coefficients improved from r ~0.6-0.7 to r ~0.85-0.9, when excluding the non-urban plumes from this plot, confirming that some species that contributed to β_{ext} were not co-emitted with CO.

The different intercepts observed when considering all plume types would have suggested different background levels of β_{ext} due to inclusion of all aerosol source types in the plot. With the current *modification of including data from only the urban plumes, the fresh and aged fitted lines cross similar β_{ext} values (6.0-6.7 Mm^{-1}) at the background CO level of 105 ppbv.*

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, : : :).

On average, less than 0.5 $\mu g/m^3$ of Ca^{2+} plus Mg^{2+} (Na^+ concentrations were not reported) was present in the PM_1 aerosols as measured by a PILS aboard the C130; therefore, contribution of refractory salts is not expected to be significant.

Why is there so much OA associated with agri emissions?

The organics that were associated with aerosols observed in agricultural plumes were not originating from agriculture emissions since no significant enhancement in OA was observed while crossing such plumes. Therefore, the organics merely represent the composition of the background aerosol onto which agricultural emissions were superimposed.

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.

*It is true that formation of ammonium nitrate depends on aerosol pH and other components of aerosol. As mentioned above, dust components of aerosol based on PILS data were minor. Additionally, AMS composition indicates that chloride and sulfate levels were very uniform in different air masses. The most variable parameter that could have an impact on aerosol composition was NH_3 levels, with average values of 1.41 ± 1.2 ppbv, 2.75 ± 1.88 ppbv, 8.21 ± 2.06 ppbv, and 5.47 ± 1.81 ppbv in urban, O&G, agriculture, and urban+O&G plumes, respectively. The following has been added in Section 3.2 to support our hypothesis: "**Aerosol nitrate formation depends on ambient conditions (temperature and relative humidity), relative mixing ratios of nitric acid and ammonia, as well as aerosol composition and pH (Seinfeld and Pandis 2006, Weber et al. 2016). With uniform concentrations of sulfate aerosol and small contribution of chloride and dust components to the Front Range fine aerosol mass, variability in aerosol pH was not expected to be high. Furthermore, there was no specific trend in temperature or relative humidity in different plume types. On the other hand, mixing ratios of ammonia were observed to be variable in the different air masses, with average values of 1.41 ± 1.2 ppbv, 2.75 ± 1.88 ppbv, 8.21 ± 2.06 ppbv, and 5.47 ± 1.81 ppbv in urban, O&G, agriculture, and urban+O&G plumes, respectively.**"*

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.

The reviewer might have misread the horizontal axis in Fig 6. The axis represents the number of aerosol particles in 300-2000 nm size range and not the total number of fine aerosols. The low number of aerosols in the larger size bins just indicates that the majority of ambient particles were at sizes smaller than 300 nm, which is typically the case. This figure is no longer included in the manuscript.

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)?

We have updated this Figure to include data with masks designating the 4 plume types that have been examined in detail in the paper. This has greatly improved the correlations of b_{ext} and NR-PM_1 mass, with r values ~0.75, as well as the trends of the weighted ODR fits. Please note that because BB also contributes to atmospheric CO, we decided the conclusions drawn from the scatter plot of β_{ext} vs. CO in the presence and absence of BB could not be as robust as desired and have therefore deleted panel b.

The following text in Section 3.4 has been updated accordingly: ***"MEE values were analyzed for days with and without the BB influence, using weighted linear ODR fit analysis, as explained previously. As seen in Figure 8, average MEE on Aug. 11-12 was ~70% greater compared to days without the influence of BB (3.65±1.16 m^2/g vs. 2.24±0.71 m^2/g). Additionally, during Aug. 11-12, background value of airborne β_{ext} was higher at 4.00 ± 0.71 Mm^{-1} compared to 0.25± 0.11 Mm^{-1} on days without the BB influence, suggesting the additional contribution to β_{ext} from the wildfires."***

References

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