

We appreciate reviewer's time and effort for providing us with comments and suggestions on our manuscript. We have made the necessary revisions to the manuscript. Below, you will find our response and the summary of our approach, highlighted in *blue*, with modifications to the manuscript highlighted *in bold*:

Referee #1

The discussion paper describes measurements of gas phase and aerosol species over Colorado, and relates them to the Denver Cyclone. In general the measurements and analysis are good and well-described. The clarity of the presentation could be improved in several ways before final publication.

General comments:

- 1. The abstract is extremely detailed and dense. Are all the numbers necessary? A number of abbreviations are used, which should not be necessary in an abstract.**

The abstract has been revised to reflect the reviewer's suggestions:

“We present airborne measurements made during the 2014 Front Range Air Pollution and Photochemistry Experiment (FRAPPÉ) project to investigate the impacts of the Denver Cyclone on regional air quality in the greater Denver area. Data on trace gases, non-refractory sub-micron aerosol chemical constituents, and aerosol optical extinction (β_{ext}) at $\lambda = 632$ nm were evaluated in the presence and absence of the surface mesoscale circulation in three distinct study regions of the Front Range: In-Flow, Northern Front Range, and the Denver Metropolitan. Pronounced increases in mass concentrations of organics, nitrate, and sulfate in Northern Front Range and the Denver Metropolitan were observed during the cyclone episodes (27–28 July) compared to the non-cyclonic days (26 July, 02–03 August). Organic aerosols dominated the mass concentrations on all evaluated days, with a 45 % increase in organics on cyclone days across all three regions while the increase during the cyclone episode was up to ~80 % over the Denver Metropolitan. In the most aged air masses ($NO_x/NO_y < 0.5$), background organic aerosols over the Denver Metropolitan increased by a factor of ~4 due to transport from Northern Front Range. Furthermore, enhanced partitioning of nitric acid to the aerosol phase was observed during the cyclone episodes, mainly due to increased abundance of gas phase ammonia. During the non-cyclone events, β_{ext} displayed strong correlations ($r = 0.71$) with organic and nitrate in the Northern Front Range and only with organics ($r = 0.70$) in the Denver Metropolitan, while correlation of β_{ext} during the cyclone was strongest ($r = 0.86$) with nitrate over Denver. Mass extinction efficiency (MEE) values in Denver Metropolitan were similar under cyclone and non-cyclone days despite the dominant influence of different aerosol species on β_{ext} . Our analysis showed that the meteorological patterns associated with the Denver Cyclone increased aerosol mass loadings in the Denver Metropolitan area mainly by transporting aerosols and/or aerosol precursors from the northern regions, leading to impaired visibility and air quality deterioration.”

- 2. The overall hypothesis and conclusion of the paper seems to be that the Denver Cyclone aerosol and/or aerosol precursors from the northern Front Range. This should be made much clearer in the abstract, introduction, and conclusions.**

We have integrated the overall hypothesis in the abstract, introduction, and conclusions of this manuscript. The following sentences have been added:

Abstract: “Our analysis showed that the meteorological patterns associated with the Denver Cyclone increased aerosol mass loadings in the Denver Metropolitan area mainly by transporting aerosols and/or aerosol precursors from the northern regions, leading to impaired visibility and air quality deterioration.”

Introduction “More importantly, limited studies have evaluated the summertime air quality implications of the Denver Cyclone that results in transport of pollutants from the Northern Front Range to the urban center.”

Conclusions: “The meteorological conditions during a Denver Cyclone promote transport of aerosol constituents and their precursors from the northern Front Range into the Denver Metropolitan area, increasing aerosol mass loadings and reducing visibility.”

- 3. Relative humidity is used throughout the paper. I understand that RH is very important for aerosol properties, but it has a number of drawbacks as a meteorological variable. A conserved humidity variable such as specific humidity or mixing ratio is more appropriate. RH varies strongly with height in a well-mixed boundary layer because the temperature varies, while potential temperature and mixing ratio may be more or less constant with height. RH can only be compared at the same temperature. For example, check section 3.5.**

Relative humidity is an important variable to consider for aerosols. Since flight segments were less than 2500 m in a well-mixed boundary layer, we believe it is appropriate to use surface RH as representative values for RH during the flight segments in the BL. However, we agree that the use of surface RH as a variable does not fully explain the meteorological changes in water content with changes in altitude. We have taken the recommendation of the referee to use a more conserved meteorological variable and have included a discussion on specific humidity in Section 3.1, along with associated plots referenced in the supplementary materials (Fig. S2-3) to understand the meteorological differences during the cyclone. The following has been added to Section 3.1: “As shown in Fig. S3b-d air masses with higher water content were advected westward by easterly winds, ahead of the intensifying low pressure system that was developed by 18:00 UTC (11:00 MST on 27 July).”

Since RH is an important variable for aerosol partitioning and equilibrium, we kept Table 1 and the discussions related to ISORROPIA modeling in its original presentation.

- 4. The argument about relative increases in CO vs. ethane seems incorrect (section 3.3.1, last paragraph). CO has a small percentage increase because it has a large background, while ethane has no background. The correct comparison would be made by removing the CO background. The conclusion may not change. This propagates through to section 3.4, which should be checked for consistency.**

The discussion on relative changes in carbon monoxide and ethane has been omitted from the manuscript. We believe a stronger supporting evidence for transport of O&G-influenced air masses south to the urban corridor is the observed change in the ratio of i-pentane to n-pentane

over DM during the cyclone (added now as Fig.7e). The last paragraph in section 3.3.1 has been replaced with the following:

“Mean mixing ratios of CO over DM during the cyclone were 144 ± 23 ppbv compared to 110 ± 8.7 ppbv in In-Flow and 114 ± 12 ppbv in NFR. Additionally, mean values of CO and C_2H_6 in DM increased during the cyclone events compared to non-cyclone days (Fig. 7b,d). Since vehicular sources of CO are concentrated in DM, the slight increase in CO over DM during the cyclone was likely due to changes in the background CO in the region and a shallower morning boundary layer on 27-28 July. However, the increase in C_2H_6 could be due to release of emissions into a shallower morning boundary layer on cyclone days, the cyclonic mixing of air masses from northern latitudes with higher emissions of C_2H_6 from O&G operations, or a combination of these two phenomena. The observed increase in the mean C_2H_6 mixing ratio in DM during the cyclone compared to the non-cyclone days were 10.2 ± 6.2 ppbv vs. 6.0 ± 7.8 ppbv, respectively. To better understand the influence of O&G operations over DM during the cyclone, we examined the ratio of i-pentane to n-pentane since O&G emissions show a characteristic ratio in the range of 0.8 – 1.2 (Gilman et al., 2013; Swarthout et al., 2013; Thompson et al., 2014; Halliday et al., 2016) in contrast to urban sources predominately impacted by vehicular emissions, which typically have a higher ratio between 2-3 (Broderick and Marnane, 2002; Baker et al., 2008). Figure 7e represents the statistical analysis of i-pentane to n-pentane ratio in the three study regions. Non-cyclone days show a significant urban source of pentanes in DM compared to NFR. During the cyclone, a minor decrease in the ratio was observed in NFR, whereas the ratio decreased substantially in DM to values close to those in NFR. These observations suggest that the significant increase in C_2H_6 mixing ratio observed over DM during the cyclone cannot be solely explained by BL height differences, but rather driven by transport of O&G-impacted and C_2H_6 -rich air masses from NFR into the DM.”

Specific comments:

1. p.8, lines 27-30: The spatial contrast and separation are present but not "stark".

Then sentence has been modified as following: “Consistent with the meteorological conditions presented above, there is a contrast in the spatial distribution and separation of pollutants during the non-cyclone and cyclone situations.”

2. p.15, line 2: Again, "isolation" is too strong.

Then sentence has been modified as following: “Overall, particle formation and growth during the non-cyclonic episodes occurred predominantly downwind of the major point/area sources”

3. p.15, line 16: "Dramatically" is probably too strong. Can you make a quantitative estimate here?

Then sentence has been modified as following: “Based on these results, reduction in source strengths of aerosol precursors in NFR leading to OA and ammonium nitrate formation, including mitigation of NH_3 emissions from dairy and livestock farming, could effectively

reduce the impact of cyclone events on Denver's air quality by reducing the aerosol mass loadings by a factor of 2 (i.e., $\sim 11 \mu\text{g sm}^{-3}$ to $5 \mu\text{g sm}^{-3}$) and improving visibility by approximately 3 folds (i.e., $\sim 32 \text{Mm}^{-1}$ to 11Mm^{-1})."