Review of "Sources and Characteristics of Summertime Organic Aerosol in the Colorado Front Range: Perspective from Measurements and WRF-Chem Modeling" by Bahreini et al.

This study characterizes the impact of oil and gas (O&G) emissions on urban OA levels during the FRAPPE field campaign. A combination of measurements and modeling is used to better constrain O&G emissions and to estimate their contributions to primary and secondary OA. The study concludes that O&G emissions have a relatively minor (~5%) effect on OA concentrations during summer in the sampled domain (Colorado Front Range). The study also highlights the need to update O&G emissions inventories. These are both novel and important results. The topic is highly relevant for ACP - the manuscript is nicely organized and clearly written. I strongly recommend its publication after the below issues are addressed.

Specific Comments:

1. Overall, my biggest concern is with the predictions of biogenic SOA. There are several problems with the methodology and/or the interpretation of results. These are:

i) There is clearly a problem with the biogenic emissions inventory: Fig. 6b shows a large systematic difference in the measured and modeled monoterpene concentrations. The manuscript dismisses this as unimportant (Pg. 10, line 23-34), but I completely disagree. First, monoterpene emissions often peak at night and their lifetime against oxidation by nitrate radical and ozone is short. From my understanding, the aircraft measurements were mostly carried out during the daytime, so nighttime conditions are not represented here. Therefore, the difference in Fig. 6 does not actually show how large the bias in emissions (and hence biogenic SOA) may be.

ii) Overall, the predicted amount of biogenic SOA (54% and 40% in urban and O&G plumes, respectively) seems quite low. The present simulations do not seem at all consistent with the results of Schichtel et al. (2008) and Ridley et al. (2018), who found measured and predicted non-fossil OC fractions in Colorado during summer to be ~85%.

iii) Limiting biogenic VOC oxidation seems to provide an unrealistic constraint on SOA formation. This may improve predictions of OA mass concentrations, but my suspicion is that the predictions of carbon oxidation state or O:C will suffer in these model scenarios. Recent studies have shown that this is a critically important dimension to evaluate (Murphy et al., 2011; Chen et al., 2015). Otherwise, it is possible that the improvements in OA mass prediction are achieved for the wrong reasons. I strongly encourage adding this dimension to the analysis.

iv) The authors hypothesize that higher urban VOCs contribute to the overprediction of OA (Pg. 11, line 35-36), but the response is to limit aging of biogenics?

2. Perhaps related to the above point, but clarification is needed for how the model-measurement comparison has been carried out. The resolution of measurements is not always consistent with the 4 km x 4 km model resolution. For example, 15-s AMS measurements at a likely aircraft speed of ~200 – 300 m/s would provide AMS measurement

resolution of 3 – 4.5 km. Many of the (single) AMS measurements no-doubt cross grid lines. Other measurements will have the same issues. Pg. 10, lines 1-2 makes reference to the problem, but it is unclear how the actual averaging and comparison was carried out.

3. For the ΔOA/ΔCO analysis (Section 3.1), the CO background is given (105 ppb), but the corresponding OA background is given as a range (Pg. 7, line 35)? The choice of the OA background value has a direct (and profound) effect on the absolute ΔOA/ΔCO values: additional discussion is definitely needed.

4. Pg. 2 line 33-34: can the authors provide a brief explanation of what is meant by "liquid-rich" and "wet gas" (e.g., which liquid).

5. Pg. 3 line 30-34: can the authors clarify what are these altitudes in m AGL? It is also unclear which criteria were used to expand the altitude limit on plumes? How were the "recirculated air masses" identified, and how many were there relative to the total?

6. Pg. 11 line 14-16: this seems like it could easily be tested by comparing modeled and measured aerosol black carbon (or equivalent) concentrations.

7. While the "best" predictions are achieved using the non-volatile POA assumption, it is important to acknowledge that this assumption is inconsistent with many direct observations (see extensive body of work from A. L. Robinson). This may highlight other uncertainties (perhaps unspeciated emissions, IVOCs, etc.), but non-volatile POA is not consistent with actual emissions measurements. Therefore, I strongly suggest adding a brief disclaimer on this point, and re-wording some of the current discussion (e.g., Pg. 11 line 25: non-volatile POA is not "more realistic" even if we can't model it well).

8. Pg. 12 lines 36-37: what do the authors mean by ΔSOA/ΔCO and ΔSOA/ΔOx? Have the authors subtracted background values? If so, these should be stated clearly. If it is just the slope of OOA vs. CO, then I suggest stating this clearly and dropping the "Δ".

Technical Corrections:

1. Pg. 3 line 9: clarify that this 80% is not aerosol organic carbon?

2. Can the authors also clarify all of the various dates? Why is there not consistency between the field campaign time period (20 July – 18 Aug) and either the modeling period (24 July – 14 Aug ***and*** 27 July – 13 Aug) or the AMS PMF dates (26 July – 11 Aug)?

3. Pg. 3 line 36: delete " 's "

4. Pg. 4 line 4: define HAIPER

5. Pg. 10 line 19: clarify what is meant by "these species"

References:

Chen, Q., et al. (2015), Elemental composition of organic aerosol: The gap between ambient and laboratory measurements, Geophys. Res. Lett., 42, doi:10.1002/2015GL063693.

Murphy, B. N., et al. (2011), Simulating the oxygen content of ambient organic aerosol with the 2D volatility basis set, Atmos. Chem. Phys., 11, 7859–7873.

Ridley, D. A., et al. (2018), Causes and consequences of decreasing atmospheric organic aerosol in the United States, PNAS, 115 (2) 290-295.

Schichtel, B. A., et al. (2008), Fossil and contemporary fine particulate carbon fractions at 12 rural and urban sites in the United States, J. Geophys. Res., 113, D02311.