

**We thank the reviewer for their helpful comments. We have addressed every comment, and believe the result is an improved manuscript. Reviewer comments are below, with our author responses indented and in bold.**

The manuscript investigates ozone formation in Colorado, a region that consistently exceeds the 8-hour ozone standard. The authors' find that the region is transitioning to a NO<sub>x</sub>-limited regime, as well as observe temperature dependencies of ozone attributed to drought. Overall, I found this manuscript to be very informative and straightforward, and timely for a region that is relatively less-studied than other areas of the country. The manuscript is well-written and figures clear. Most of my comments are minor and relate to clarity. With minor revisions, I recommend this manuscript for publication in ACP.

General Comments (1) There is inconsistency in the statistics used. Figures 2 and 3 show 5th and 95th percentiles, while later figures show one sigma. Sometimes the standard deviation of the sample is shown (e.g., Figure 7) and other times the standard error of the mean (e.g., Figure 8). For clarity, I believe the authors should maintain consistency throughout the manuscript, and at a 95% confidence interval, needed to assess the statistical significance of results.

**Thank you to the reviewer for pointing out the inconsistencies in the error reporting. We agree that this should be improved, and the figures and references to figures or data have been updated in the revised manuscript as follows:**

**Figure 2b. The error bars are now the 95% confidence intervals around the reported ozone/year slopes.**

**Figure 3b. We included an additional figure similar to Figure 2b to show the NO<sub>2</sub>/year slopes for the 5<sup>th</sup>, 50<sup>th</sup>, and 95<sup>th</sup> percentiles with the error bars representing the 95% confidence intervals around the slopes.**

**Figure 5 was updated with suggestions from comment 7 to show the weekday and weekend averages with the 95% confidence intervals.**

**Figure 7a was updated and shows the average weekday minus weekend ozone for each year for the six sites. The solid grey line represents the aggregated average of the six sites with the shading representing the 95% confidence interval.**

**Figure 7b was updated and shows the average weekday minus weekend NO<sub>2</sub> for each year for the CAMP and Welby sites. The error bars now represent the 95% confidence interval of the averages.**

**Figures 8 and 9 were updated to include averages and 95% confidence intervals, and also to change the temperature binning approach as suggested by the second reviewer.**

**Figure 8a. was updated with the new equal bin size approach suggested by reviewer 2, and the averages of those temperature bins for each year are displayed. The 95% confidence intervals for the O<sub>3</sub> bin averages were not included in the figure for clarity purposes, but are typically <8 ppb.**

**Figure 9 was updated with the new equal bin size approach suggested by reviewer 2, and the 95% confidence intervals around the yearly O<sub>3</sub>/temperature slopes are included.**

#### Specific Comments

(2) Lines 78-31. Do the authors mean \*1980-1993\* instead of “1980-2008”? Also, the ratio of VOC/NO<sub>x</sub> emissions has evolved with time in cities (Parrish et al., 2011; McDonald et al., 2013), which could also affect ozone trends. Parrish, D. D., H. B. Singh, L. Molina, and S. Madronich (2011), Air quality progress in North American megacities: A review, *Atmos Environ*, 45, 7015-7025, doi:10.1016/j.atmosenv.2011.09.039. McDonald, B. C., D. R. Gentner, A. H. Goldstein, and R. A. Harley (2013), Long-term trends in motor vehicle emissions in U.S. urban areas, *Environ Sci Technol*, 47, 10022-10031, doi:10.1021/es401034z.

**We do mean “1980-2008”. Lefohn et. al (2010) compare trends at monitoring sites across the US for two overlapping time periods 1980-2008 and 1994-2008. They found that many sites had a decreasing O<sub>3</sub> trend for the longer 1980-2008 period, but most of the decreasing trends were not present during the 1994-2008 period indicating that O<sub>3</sub> decreases had slowed or stopped in the 1994-2008 period. We have revised the statement to try and clarify that point.**

Lefohn et al. (2010) found that when comparing O<sub>3</sub> at the same sites for a longer period of 1980-2008 and shorter period of 1994-2008, the predominant pattern was a change from a negative trend (decreasing O<sub>3</sub>) during the longer period to no trend (stagnant O<sub>3</sub>) in the shorter period, indicating that O<sub>3</sub> reductions had leveled off by the late 2000s.

**We thank the reviewer for their comment and suggestion and have including the following reference as suggested;**

McDonald et al. (2013) report decreased VOC, CO, and NO<sub>x</sub> automobile emissions in major US urban centers, and more importantly decreasing VOC/NO<sub>x</sub> trends from 1990 to 2007 with a turnaround and small increase after 2007, which would affect local O<sub>3</sub> chemistry within the city and at downwind receptor sites

(3) Line 164. This is an example where I found the inconsistency in statistics confusing. The error bars shown would suggest that all these results are statistically significant, rather than only at the 95th percentile.

**In this section Figures 2 and 3 were referenced, both of which have been updated per comment 1. The statistical significance of the long-term O<sub>3</sub> and NO<sub>2</sub> trends were determine from both an F-test and from the 95% confidence intervals around the slope.**

(4) Lines 174 – 178. The authors’ qualify the AVOC emissions trend shown in Figure 4 as an inventory estimate. I think this paragraph could be strengthened by referencing studies that have assessed emission trends for key sectors of this analysis, e.g., motor vehicles (e.g., McDonald et al., 2013), and oil and gas (e.g., Duncan et al., 2016), as well as studies that have reported uncertainties in emissions (e.g., Petron et al., 2014). What explains the hump in VOC emissions from petroleum industries around 2011? Is this realistic, and comport with oil and natural gas production statistics from the Energy Information

Administration? Such a rapid increase and decrease in VOC emissions would likely have some influence on observed ozone, as many of the points shown in Figure 6 are still on the NO<sub>x</sub>-saturated side of the curve. Also, McDuffie et al. (2016) suggested that maximum O<sub>3</sub> was sensitive to NO<sub>x</sub> and reductions in VOCs in the Front Range.

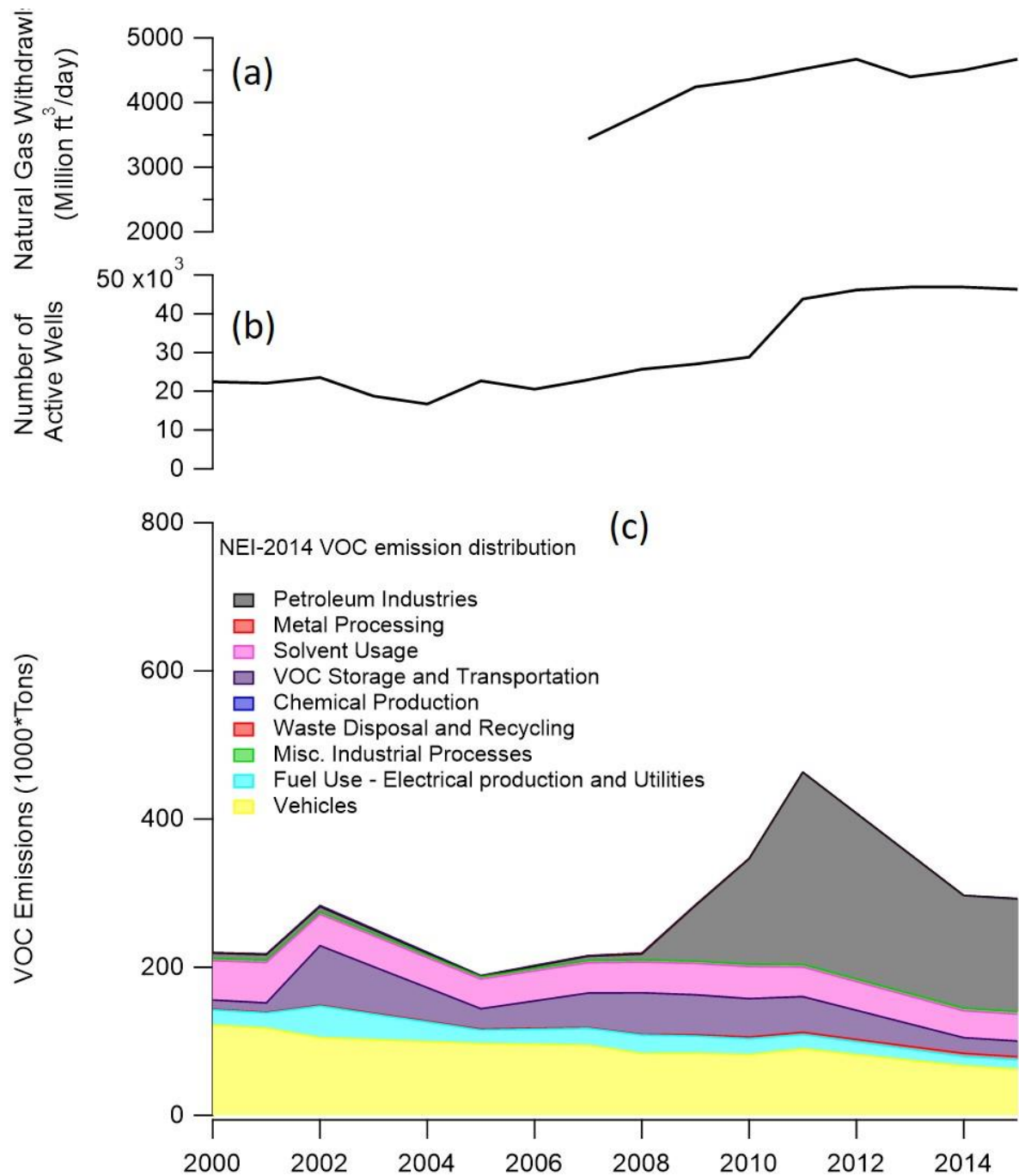
Duncan, B. N., L. N. Lamsal, A. M. Thompson, Y. Yoshida, Z. F. Lu, D. G. Streets, M. M. Hurwitz, and K. E. Pickering (2016), A space-based, high-resolution view of notable changes in urban NO<sub>x</sub> pollution around the world (2005-2014), *J Geophys Res-Atmos*, 121, 976-996, doi:10.1002/2015jd024121.

Petron, G., A. Karion, C. Sweeney, B. R. Miller, S. A. Montzka, G. J. Frost, M. Trainer, P. Tans, A. Andrews, J. Kofler, D. Helmig, D. Guenther, E. Dlugokencky, P. Lang, T. Newberger, S. Wolter, B. Hall, P. Novelli, A. Brewer, S. Conley, M. Hardesty, R. Banta, A. White, D. Noone, D. Wolfe, and R. Schnell (2014), A new look at methane and nonmethane hydrocarbon emissions from oil and natural gas operations in the Colorado Denver-Julesburg Basin, *J Geophys Res-Atmos*, 119, 6836-6852, doi:10.1002/2013jd021272.

McDuffie, E. E., P. M. Edwards, J. B. Gilman, B. M. Lerner, W. P. Dube, M. Trainer, D. E. Wolfe, W. M. Angevine, J. De Gouw, E. J. Williams, A. G. Tevlin, J. G. Murphy, E. V. Fischer, S. McKeen, T. B. Ryerson, J. Peischl, J. Holloway, K. Aikin, A. O. Langford, C. J. Senff, R. J. Alvarez II, S. R. Hall, K. Ullmann, K. O. Lantz, and S. S. Brown (2016), Influence of oil and gas emissions on summertime ozone in the Colorado Northern Front Range, *J Geophys Res-Atmos*, 121, doi:10.1002/2016JD025265.

**The reviewer's suggestion motivated us to include an updated Figure 4 to include the number of active oil and natural gas wells in Colorado from 2000 to 2015 and the yearly average natural gas withdrawal estimates from the Energy Information Administration, which show increases in both number of wells and the natural gas withdrawal in Colorado (see updated figure 4 below). We have included the following text for some more information regarding ONG in Colorado, changing VOC emissions around the country, and impacts on ozone in the Front Range.**

The US Energy Information Administration (EIA) report a 2-fold increase in active ONG wells from ~25000 to ~40000 from 2010 to 2012 (Fig. 4c) (US-EIA, 2017). A number of VOC studies in the NFRMA since 2011 report enhanced C<sub>2</sub>-C<sub>5</sub> alkanes relative to other urban/semi-urban regions (Abeleira et al., 2017; McDuffie et al., 2016; Pétron et al., 2012; Pétron et al., 2014; Swarthout et al., 2013). Pétron et al. (2014) reported that the state inventory for total VOCs emitted by ONG activities was at least 2x lower than May 2012, which indicates that the contribution of ONG related VOCs in figure 4 would increase substantially. McDonald et al. (2013) report decreases in both NO<sub>x</sub> and VOC emissions from automobiles, and a steady reduction in the VOC/NO<sub>x</sub> emission ratio in major cities from 1990 to 2008, with a possible trend reversal following 2008. McDuffie et al. (2016) reported that maximum O<sub>3</sub> at a site in the NFRMA was sensitive to NO<sub>x</sub> and VOC reductions.



(5) Line 172. The NEI is reported for a single year. I believe the authors mean the EPA Trends Report, which is now reported by state.

**The reviewer is correct, and this mistake was revised in the manuscript. Thank you.**

(6) Line 187. The weekday/weekend effect is really due to a drop-off in heavy-duty truck traffic (Marr et al., 2002; McDonald et al., 2014). Passenger cars drive similar amounts on weekends and weekdays.

Marr, L. C., and R. A. Harley (2002), , Environ Sci Technol, 36, 4099-4106, doi:10.1021/Es020629x.

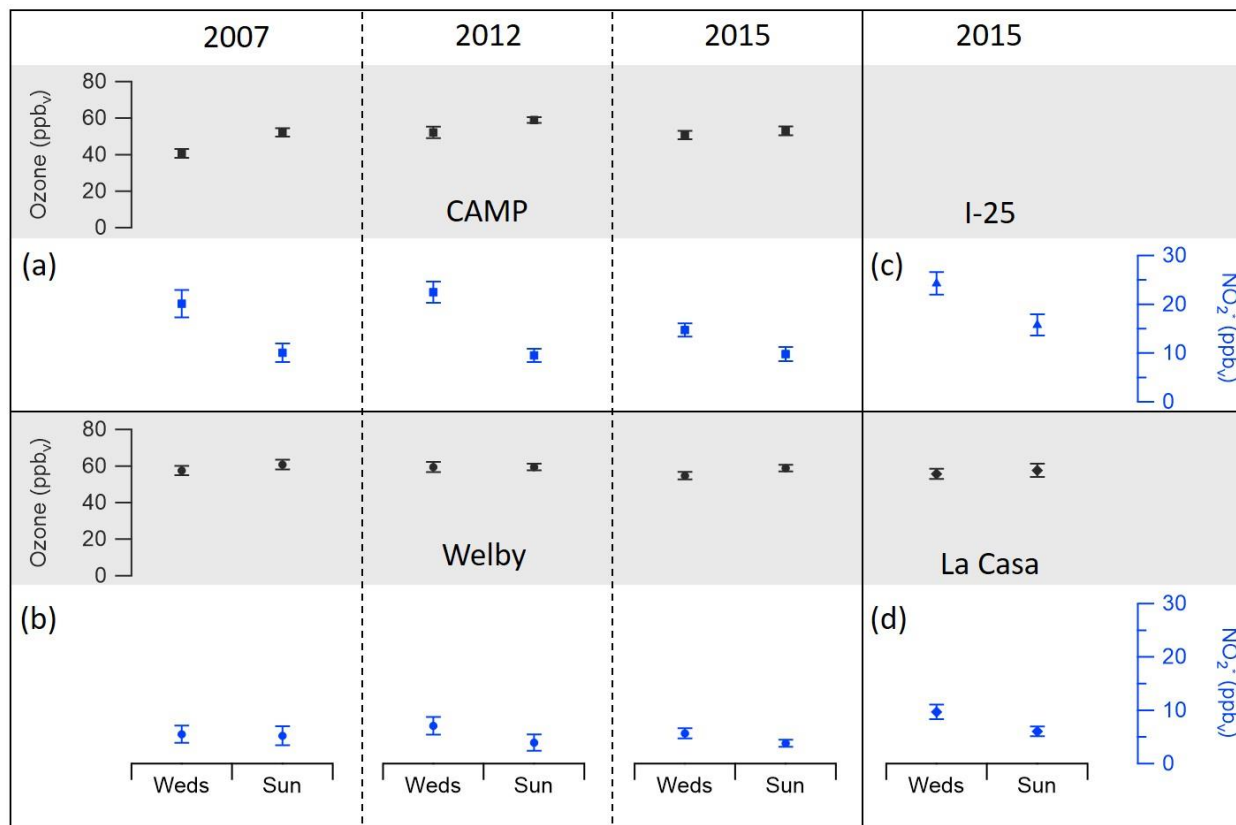
McDonald, B. C., Z. C. McBride, E. W. Martin, and R. A. Harley (2014), High-resolution mapping of motor vehicle carbon dioxide emissions, J Geophys Res-Atmos, 119, 5283- 5298, doi:10.1002/2013jd021219.

**We agree with the reviewer, and have included the following revision;**

**Traffic patterns in urban regions are different between weekends and weekdays with a decrease in heavy-duty truck traffic on weekends (Marr and Harley, 2002). VOCs are expected to be stable across the week (Marr and Harley, 2002) as major VOC sources do not vary by day-of-week.**

(7) Line 209. I found the variability in concentrations across days, as shown in Figure 5, distracting for discerning weekday-weekend effects. I think this figure could be made clearer by showing a mean and confidence interval of weekdays (Mon-Friday), and of weekend days (Sa/Su) combined. Also, I think 95th confidence intervals should be shown, to make it easier for the reader to discern statistical significance.

**The suggestion from the reviewer was used to clarify the data presented on Figure 5. Figure 5 was remade with average +/- 95% confidence interval for the same sites and years as the original figure. See updated figure below.**



(8) Lines 212-213. Are these 24 hour averages or daytime averages? If it is the former, could nighttime chemistry affect the weekday-weekend effect?

**All data presented in the manuscript is constrained to daytime (10:00am – 4:00pm local) values.**

(9) Line 226-227. This sentence is confusing. Suggest revising.

**This section was updated with new insight provided by updating the figures, and includes the following revisions.**

**Measured NO<sub>2</sub>\* decreased at both CAMP and Welby between 2001 and 2015 (Fig. 3b), but with larger decreases at the CAMP site. The ΔNO<sub>2</sub>\* at Welby remained stable with an average value of  $-1.7 \pm 0.9$  ppb<sub>v</sub>, while ΔNO<sub>2</sub>\* at the CAMP site exhibited a statistically significant decrease of  $0.6 \pm 0.4$  ppb<sub>v</sub>/yr. The decreasing ΔNO<sub>2</sub>\* at the CAMP site appears to be converging with ΔNO<sub>2</sub>\* at the Welby site. It is unlikely that traffic patterns are assimilating between the two sites, and a more plausible explanation is that emission control technologies on heavy duty commercial fleet vehicles are reducing the impact on emissions of those specific vehicles, and thus reducing the measurable ΔNO<sub>2</sub>\* (Bishop et al., 2015).**

(10) Lines 281-287. On Line 283, I believe the authors mean \*2002-03\* instead of "2001-02". To my eye in Figure 9, it is clear that 2008 and 2011-12 are suppressed, but I found it harder to see for 2002-2003. For 2002-2003, it only looks like the Fort Collins and Welby sites are suppressed, and not the other locations.

**We have updated the manuscript to reflect this observation.**

Minor Comments (11) Line 211. Terminology switches from “weekday-weekend” to “weekend-weekday”. Suggest choosing one word ordering and sticking with it.

**The terminology throughout the manuscript has been updated to “weekend-weekday”.**

Abeleira, A., Pollack, I., Sive, B. C., Zhou, Y., Fischer, E. V., and Farmer, D.: Source Characterization of Volatile Organic Compounds in the Colorado Northern Front Range Metropolitan Area during Spring and Summer 2015, *Journal of Geophysical Research*, In Press, 2017.

Bishop, G. A., Hottor-Raguindin, R., Stedman, D. H., McClintock, P., Theobald, E., Johnson, J. D., Lee, D.-W., Zietsman, J., and Misra, C.: On-road Heavy-duty Vehicle Emissions Monitoring System, *Environmental Science & Technology*, 49, 1639-1645, 10.1021/es505534e, 2015.

Lefohn, A. S., Shadwick, D., and Oltmans, S. J.: Characterizing changes in surface ozone levels in metropolitan and rural areas in the United States for 1980–2008 and 1994–2008, *Atmospheric Environment*, 44, 5199-5210, 2010.

Marr, L. C., and Harley, R. A.: Modeling the Effect of Weekday– Weekend Differences in Motor Vehicle Emissions on Photochemical Air Pollution in Central California, *Environmental science & technology*, 36, 4099-4106, 2002.

McDonald, B. C., Gentner, D. R., Goldstein, A. H., and Harley, R. A.: Long-term trends in motor vehicle emissions in US urban areas, *Environmental science & technology*, 47, 10022-10031, 2013.

McDuffie, E. E., Edwards, P. M., Gilman, J. B., Lerner, B. M., Dubé, W. P., Trainer, M., Wolfe, D. E., Angevine, W. M., deGouw, J., and Williams, E. J.: Influence of oil and gas emissions on summertime ozone in the Colorado Northern Front Range, *Journal of Geophysical Research: Atmospheres*, 121, 8712-8729, 2016.

Pétron, G., Frost, G., Miller, B. R., Hirsch, A. I., Montzka, S. A., Karion, A., Trainer, M., Sweeney, C., Andrews, A. E., Miller, L., Kofler, J., Bar-Ilan, A., Dlugokencky, E. J., Patrick, L., Moore, C. T., Ryerson, T. B., Siso, C., Kolodzey, W., Lang, P. M., Conway, T., Novelli, P., Masarie, K., Hall, B., Guenther, D., Kitzis, D., Miller, J., Welsh, D., Wolfe, D., Neff, W., and Tans, P.: Hydrocarbon emissions characterization in the Colorado Front Range: A pilot study, *Journal of Geophysical Research: Atmospheres*, 117, n/a-n/a, 10.1029/2011jd016360, 2012.

Pétron, G., Karion, A., Sweeney, C., Miller, B. R., Montzka, S. A., Frost, G. J., Trainer, M., Tans, P., Andrews, A., and Kofler, J.: A new look at methane and nonmethane hydrocarbon emissions from oil and natural gas operations in the Colorado Denver-Julesburg Basin, *Journal of Geophysical Research: Atmospheres*, 119, 6836-6852, 2014.

Swarthout, R. F., Russo, R. S., Zhou, Y., Hart, A. H., and Sive, B. C.: Volatile organic compound distributions during the NACHTT campaign at the Boulder Atmospheric Observatory: Influence of urban and natural gas sources, *Journal of Geophysical Research: Atmospheres*, 118, 614-610,637, 10.1002/jgrd.50722, 2013.

Natural Gas - Data: <https://www.eia.gov/>, access: 4/15, 2017.