15 February 2021

Dear Prof. Karl,

We are grateful to the editor and reviewers for their time and constructive comments on our manuscript. We have implemented their comments and suggestions and wish to submit a revised version of the manuscript for further consideration in the journal. Changes in the initial version of the manuscript are highlighted (with 'tracked changes') for added sentences or strikethrough for deleted sentences in the revised version. Below, we also provide a point-by-point response explaining how we have addressed each of the reviewers' comments. We look forward to the outcome of your assessment.

Yours sincerely,

On behalf of the co-authors

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"Identifying and Quantifying Source Contributions of Air Quality Contaminants during Unconventional Shale Gas Extraction" by Nur H. Orak et al.

**** Comments from Referees are in black, authors' responses are in green, changes are in red color.***

Comments from Reviewer #1

The author's made improvements to the article but many of the reviewer questions remain unanswered/unclear. This is a valuable dataset but the analysis and manuscript could still use improvement in my opinion. I completely understand that there will be future papers coming from this unique dataset, but it would be very helpful to the readers to know what is slated for future work to help put this analysis into better context.

A: We would like to thank Reviewer #1 for the detailed review and constructive comments. Our responses are under each comment and a revised manuscript is provided with marked-up changes.

Q1: Line 99-102: Hecobian specifically compares the emissions of distinct operation phases of natural gas extraction. The analysis presented here would be greatly improved by doing a more direct comparison of the emissions at each stage of well production with Hecobian (e.g., for lines 276-281). Both datasets are incredibly rare and valuable, so it would be really helpful to see if they compare for gas fields in different regions of the U.S. and would further help to put Orak measurements into context.

A: Thank you for the suggestion we have provided a more detailed comparison with the results of Hecobian, 2019. We have revised the results as follow (No Markup-Line 296):
"Hecobian et al. (2019) investigated the emissions during different well pad development phases to analyze emission rates in the Denver-Julesburg and Piceance basins in Colorado, US. They observed that emission rates of benzene and most VOCs were highest during flowback for both basins, on the other hand, they had much lower emission rates from the production phase, which can be related to the differences in duration of each phase (days to weeks). Light alkanes and benzene concentrations were higher during hydraulic fracturing. It is difficult to directly compare the VOCs concentrations of the two studies, because the proposed study is based on continuous data during each phase while Hecobian et al. (2019) collected 374 measurements from five drilling, eight fracking, nine flowback, one liquids load out, and 11 production sites to analyze emission rates."

Q2: The addition of the trailer location to Figure 1 is very helpful. This combined with Figure 2 highlights the fact that the wind was rarely from the SE sector where the majority of the drilling equipment, and presumably the drilling activity, was occurring. It would be really helpful to show what the air composition was as a function of wind direction. Was methane, ethane, etc. higher when wind was coming from the SE sector? Does Factor 1 of the PMF results have the highest contribution to the ambient measurements when wind is from this direction? This was one of the unaddressed questions (Reviewer 1, Q1). You should have a timeseries of each factor of your final PMF model that can then be analyzed as a function of any other variable that was NOT included in the PMF such as wind direction. If the Natural Gas factor is most prevalent in the SE sector, then it further adds confidence in the analysis.

A: Thank you for the suggestion. The trailer was situated at the northeastern corner of the MSEEL well pad (Figure 1) with wind direction at this location most frequently from the southwest (Figure 2). This position was optimized the occurrences of the laboratory being downwind of the well pad. We have prepared two new figures to show that SW dominated the higher overall concentrations (Figure S4) and added the following explanation on line 307 (No Markup):

"Figure S4 shows the dominant wind directions on overall concentrations, as well as giving information on the different concentration levels. Pollution roses show which wind directions contribute most to overall mean concentrations. For all air quality species, southwestern winds controlling the overall mean concentrations at the well pad. To explore the relationship between methane and ethane, we conditioned ethane by methane. Figure S5 indicates that higher ethane concentrations are associated with the SW and higher methane concentrations. The results also show that lower ethane and methane concentrations contributed from the east; the highest methane concentrations were obscured by a relatively high ethane background."

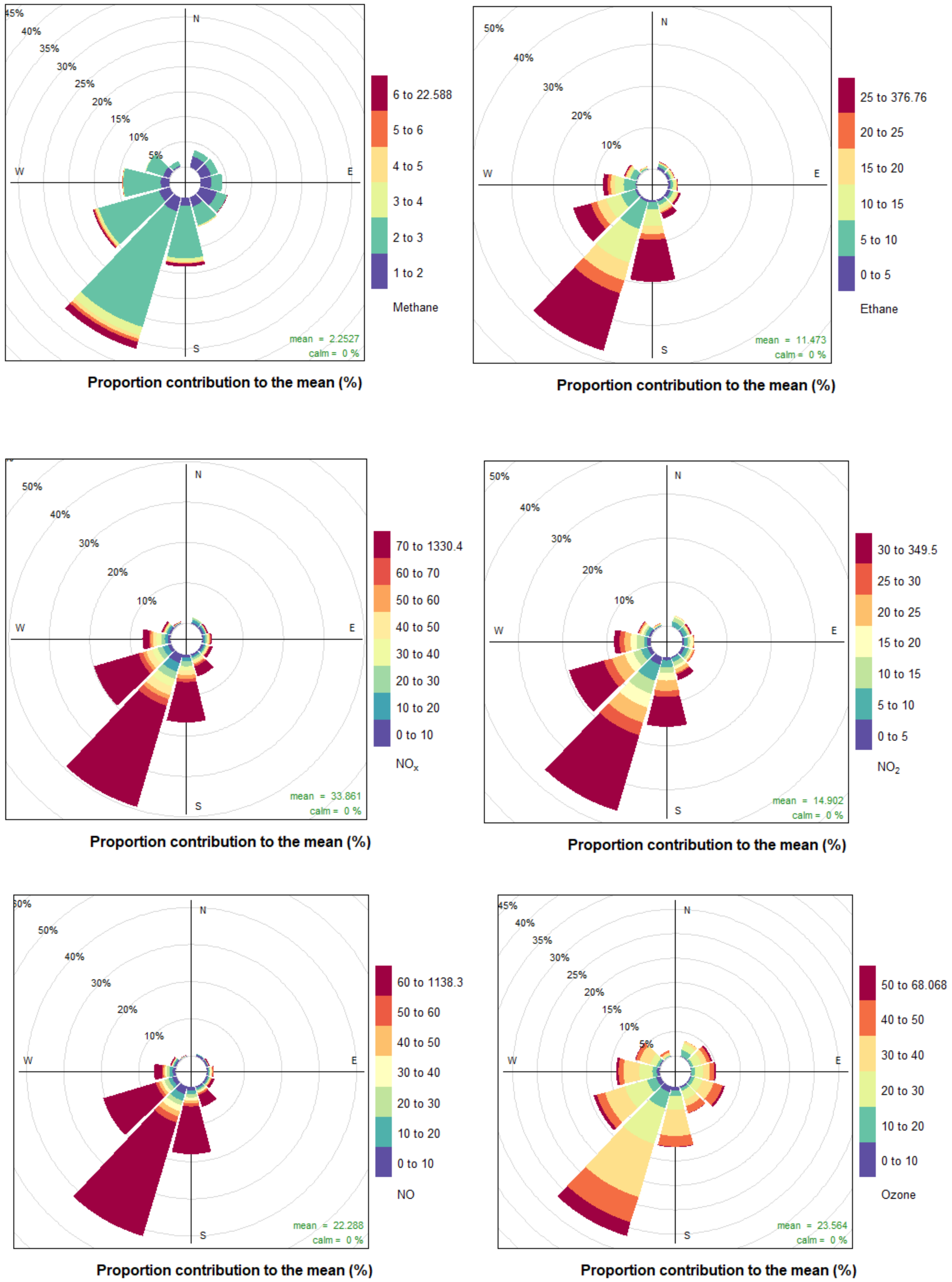


Figure S4. Methane, Ethane, NOx, NO_2, NO, and Ozone pollution roses

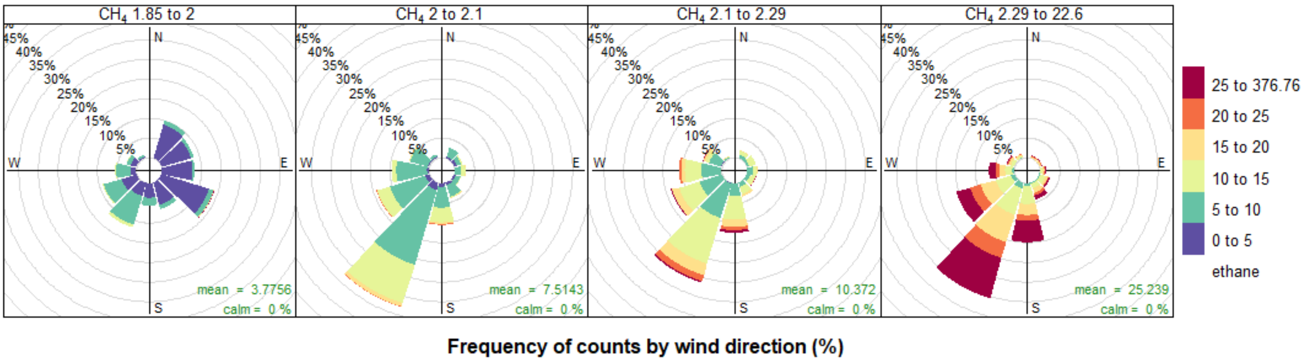
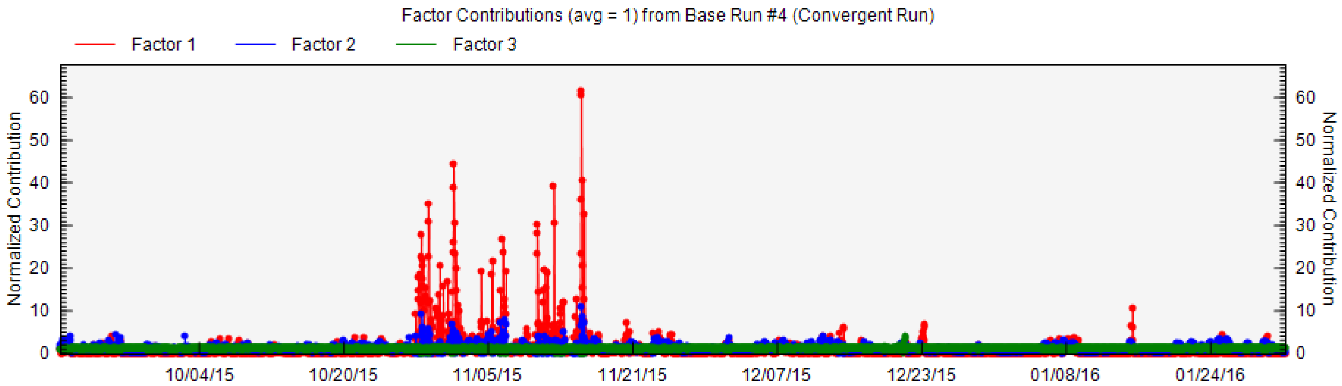


Figure S5. Ethane pollution rose conditioned by Methane concentration.

In terms of time series of each factor, as you can see below the trend of factor contribution does not provide any meaningful knowledge by itself, therefore, we think it would be confusing to share this figure with the audience. Instead, we prepared Figure S6 to show the contribution of wind direction to each PMF factor and added the following sentence on line 315 (No Markup) "The highest contribution to the factors were provided from the SW data (Figure S6)."



Factor 1- Engine Emissions Factor

Factor 2- Natural Gas Factor

Factor 3- Regional Transport/Photochemistry Factor

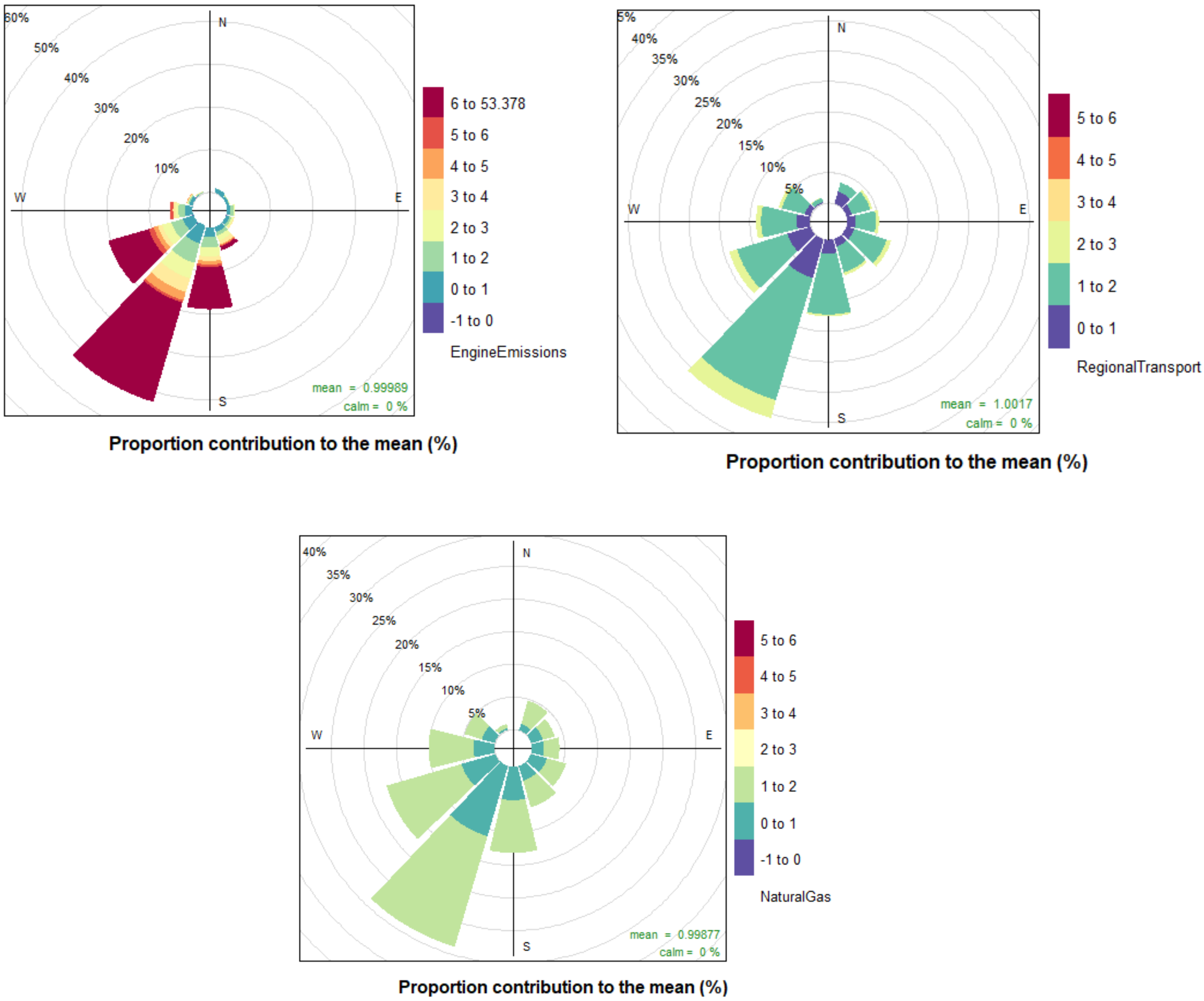


Figure S6. The PMF factor contribution roses for Engine Emissions factor, Regional Transport/ Photochemistry factor, and Natural Gas factor.

Q3: Figure 3 is the key figure in this manuscript, in my opinion. This clearly shows the different chemical composition of the air measured during each of the important drilling/hydraulic fracturing/production steps. Why doesn't PMF pick up these differences and lump all into a single generic factor? How do these results compare to other studies?

A: Thank you for pointing out your concerns. We think Figure 6 is the main figure that serves to the objective of this study. As we mentioned in our first response, the research team is conducting several analysis that has different objectives and methods. We examined several factors to capture the optimal number and analyzed different fPeak values to explore the robustness of the selected PMF solution. We explained the rotational ambiguity of the factors to justify the selection. However, there are several limitations of PMF model. To be able to answer your questions, we have added the following explanation (No Markup-Line 370):

“ PMF models have several limitations. First, it needs large datasets. In this study, the number of data varies based on the duration of the activity (Figure S2). Therefore, the contribution to the factors is not same for each phase. This is the main reason behind the uncertainty of defined factors. Second, the accuracy and precision of measured species limit the analysis. The determination of the number and character of factors is based on an expert’s interpretation. Comprehensive information is needed on source profiles to verify the defined source profiles. Finally, the pre-set parameters are playing an important role on the model results.”

Q4: Lines 288-292: Be sure to specify the units as you are comparing the ethane (ppb) to methane (ppm) ratioor else add 10^-3 to the ratios. How does the ethane to methane ratio compare to other ONG emissions (i.e., Yakovitch et al. and may other papers)?

A: We regret for the mistake, we have added the unit on Figure 4. For the comparison please see our response to Q1.

Q5: Lines 328: Source Profiles. Another useful reference regarding PMF analysis in an oil and gas field to add to the discussion could be:
Source characterization of volatile organic compounds in the Colorado Northern Front Range Metropolitan Area during spring and summer 2015. A. Abeleira, I. B. Pollack, B. Sive, Y. Zhou, E. V. Fischer and D. K. Farmer. 122(6), 3595-3613, doi:https://doi.org/10.1002/2016JD026227, 2017

A: Thank you for the suggestion. We had added the suggested reference.

Q6: Figure 6: I’m still trying to figure out why CO2 and methane have virtually the same PMF factor fingerprints. Clearly, the natural gas factor isn’t just natural gas as raw natural gas does NOT contain NOx and I would not expect it to be composed primarily of CO2. Also, the engine emissions factor doesn’t contain an appreciable amount of CO2. Why? It seems to me that the PMF factors aren’t fully resolving in a meaningful way. Also, why do hexane and benzene not have any attribution to “regional transport” as these two species are sufficiently long-lived in the atmosphere to have a significant background, much more so than toluene that has ~25% attributed to transport?

A: We do understand your concerns. There are several limitations of the study. Please see our response to Q3. PMF has limitations and the factors are not usually perfectly resolved. The signal for each measured species can have something to do with it, too. So although they are valid data points, they maybe do not get resolved into factors as well as if the signal was stronger.

Q7: The last, “big picture”, piece of this analysis that is missing is how the natural gas drilling activities actually affected air quality, which is stated as being the motivation for this paper. Since you have the PMF factors for each species, then you should be able to answer the question of how the air quality would be different if the “natural gas” factor was removed or how it compared to the other factors by calculating an Air Quality Index or OH-reactivity, or some other metric.

A: Thank you for the suggestion. It would be a separate scenario to analyze the impacts on air quality if we remove the natural gas factor. We think the proposed analysis is beyond the scope of this study. We have explained the big picture on line 394 (No Markup):

“As determined by the PMF analysis, a measurable increase in natural gas-related pollutant concentrations and the associated natural gas factor contribution from different stages of active phase was not observed. At the downwind distance of 600m from the well pad center to the air monitoring laboratory, the emissions from the well pad were not easily distinguishable from typical variations in ambient background concentrations. West Virginia has many natural gas wells that contribute to the ambient background, as evidenced by ethane concentrations that are higher than typical global background (Rinsland et al. 1987; Rudolph et al. 1996). Short-lived peak events that were observed when the wind direction was coming from the well pad show that emissions can be dispersed downwind and detected at this distance, but when concentrations are averaged and analyzed with a PMF analysis the peak events were not significant enough to result in a measurable impact of the well pad emissions at the receptor location. Understanding the air quality impacts of operational phases is important since it has potential to help inform future decision-making and constrain cumulative impact assessments.”