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Atmospheric Chemistry and Physics Discussions

Identifying and Quantifying Source Contributions of Air Quality Contaminants during Unconventional Shale Gas Extraction

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Abstract

The United States experienced a sharp increase in unconventional natural gas (UNG) development due to the technological development of hydraulic fracturing ("fracking"). The objective of this study is to investigate the effect of unconventional natural gas development activities on local air quality as observed at an active Marcellus Shale well pad at the Marcellus Shale Energy and Environment Laboratory (MSEEL) in Morgantown, Western Virginia, USA. Using an ambient air monitoring laboratory, continuous sampling started in September 2015 during horizontal drilling and ended in February 2016 when wells were in production. High resolution data were collected for the following air quality contaminants: volatile organic compounds (VOCs), ozone (O_3), methane (CH_4), nitrogen oxides (NO and NO_2), carbon dioxide, (CO_2), as well as typical meteorological parameters (wind speed/direction, temperature, relative humidity, and barometric pressure). Positive Matrix Factorization (PMF), a multivariate factor analysis tool, was used to identify possible sources of these pollutants (factor profiles) and determine the contribution of those sources to the air quality at the site. The results of the PMF analysis for well pad development phases indicate that there are three potential factor profiles impacting air quality at the site: *natural gas*, *regional transport/photochemistry*, and *engine emissions*. There is a significant contribution of pollutants during horizontal drilling stage to *natural gas* factor. The model outcomes show that there is an increasing contribution to *engine emission* factor over different well pad drilling through production phases. Moreover, model results suggest that the major contributions to the *regional transport/photochemistry* factor occurred during horizontal drilling and drillout stages.

Keywords: ambient monitoring; natural gas; air pollution; source apportionment

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Introduction

There is a rapid increase in unconventional natural gas exploration by recent technological advances (USEIA 2020). The success of the US in exploiting unconventional natural gas has stimulated other countries. As a result, there is a growing attention by public for the potential public health impacts of UNG extraction. In response to emerging public concern regarding the process of fracking for UNG extraction, several studies have investigated the potential public health risks of UNG development (Adgate et al. 2014; Hays et al. 2015; Hays et al. 2017; Werner et al. 2015). A part of adverse health effects are related to exposure of environmental pollution (Elliott et al. 2017; Elsner and Hoelzer 2016; Paulik et al. 2016). The majority of environmental impact studies focus on water quality impacts of unconventional natural gas development (Annevelink et al. 2016; Butkovskyi et al. 2017; Jackson et al. 2015; Torres et al. 2016). However, relatively fewer studies focus on air quality impacts (Islam et al. 2016; Ren et al. 2019; Swarthout et al. 2015; Williams et al. 2018). Some studies focus on collecting and analyzing data for pre-operational phase of fields to provide baseline dataset for future work that operational shale gas activities can be later evaluated (Purvis et al. 2019). Non-methane hydrocarbons (NMHC) and nitrogen oxides (NO_x) are of most interest as some NMHC can be toxic (such as benzene) (P. M. Edwards et al. 2014), therefore, several studies focuses on increases in methane, NHMC, and ozone in oil and gas producing regions (Pacsi et al. 2015; Roest and Schade 2017). Another study explored the importance of the deployment autonomy of portable measurement systems by measuring exposure upwind, within and downwind of operation of hydraulic fracturing equipment to protect workers (Ezani et al. 2018). There are also more comprehensive studies for data collection. Swarthout et al. (2015) conducted a field campaign to investigate the impact of UNG production operations on regional air quality. Highest density of methane, carbon dioxide, and volatile organic carbons

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(VOCs) were observed closer to UNG wells. A limited number of studies available on source apportionment for major air pollutants (Majid et al. 2017; Prenni et al. 2016). These studies have lacked a comparison of the effects during distinct operational phases of natural gas extraction: well pad construction, drilling (vertical and horizontal), well stimulation (hydraulic fracturing followed by flowback), and production.

Several compounds are associated with emissions from each phase of well installation and development, depending on the activity and equipment in use for each phase. Activities that require the use of off-road diesel construction vehicles have emissions of coarse particulate matter (PM_{10} aerodynamic diameter $\leq$10 μm) from the suspension of dust from vehicle traffic on dirt and gravel roads, as well as volatile organic compounds (VOCs), nitrogen oxides (NO_x) and fine particulate matter smaller than 2.5 μm in aerodynamic diameter ($PM_{2.5}$) from the vehicle exhaust. During vertical and horizontal drilling, there are emissions of NO_x, $PM_{2.5}$, and VOCs from diesel powered drilling rigs, and fugitive emissions of natural gas (methane (CH_4) and other hydrocarbons). Hydraulic fracturing activities add emissions from truck traffic and diesel-powered compressors (NO_x, PM_{10}, $PM_{2.5}$, VOCs). Emissions of VOCs and CH_4 from water separation tanks, venting, and degassing of produced waters occur during flowback operations. In addition to these primary sources of emissions at the site, secondary production of ozone (O_3) and $PM_{2.5}$ from photochemistry can result from emissions during any of the operational phases.

This is the first study, to our knowledge, to collect high time resolution ambient concentrations of compounds emitted from well pad activity during various phases of operation such that the relative air quality effect of each phase of development can be investigated. This detailed information about the distribution of emission sources'

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impact through a well pad's development phases is needed to manage the associated risks from emissions.

Methods

Monitoring Location: Marcellus Shale Energy and Environment Laboratory

The Marcellus Shale formation covers an area of approximately 240,000 km^2 across several states: New York, Pennsylvania, Ohio, West Virginia, Maryland, and Virginia (Kargbo et al. 2010)(Figure S1). The Marcellus Shale Energy and Environment Laboratory (MSEEL) is an approximately 14,000 m^2 study well pad in Morgantown, WV, USA (39.602^0 N, 79.976^0 W) (MSEEL 2019). The MSEEL is a multi-institutional, long-term collaborative field site where integrated geoscience, engineering, and environmental research have been conducted to assess environmental impacts and develop new technology to improve recovery efficiency as well as reduce environmental footprint of shale gas operations (MSEEL 2019).

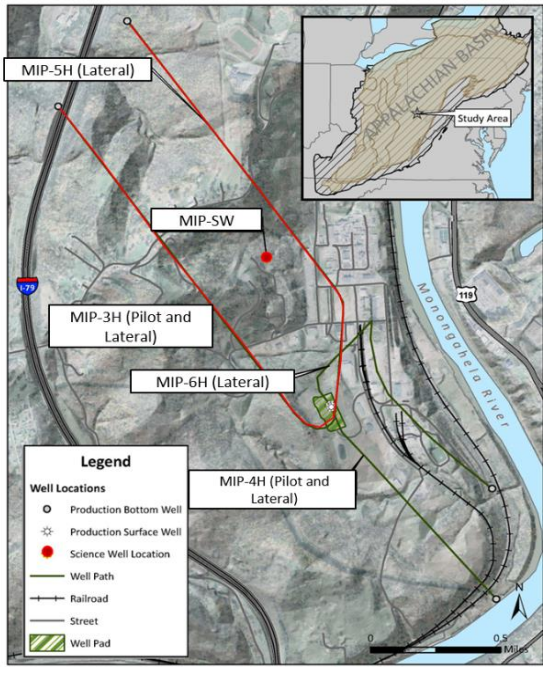

Figure 1. Location of the Marcellus Shale Energy and Environment Laboratory and the four production wells.

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The MSEEL is the site of two horizontal production wells completed in 2011 (wells 4H and 6H, Figure 1) and two horizontal production wells completed in 2015 (wells 3H and 5H, Figure 1). Production from the newer horizontal wells began in December 2015. Dates and duration for phases of operation are shown in Figure S2. The vertical drilling was conducted using three diesel Caterpillar 3512 engines with 1365 kW generators. Horizontal drilling made use of two dual fuel (40% diesel and 60% natural gas) engines. All activities at the well pad followed industry's best management practices (MSEEL 2019).

Air Quality and Meteorological Data Collection

An ambient air monitoring laboratory (18' trailer with ambient air sampled from inlets on the trailer roof) 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 optimized the occurrences of the laboratory being downwind of the well pad. Instrumentation in the laboratory and measured constituents are listed in Table 1. All instruments were maintained and calibrated according to manufacturer's recommended protocols. Details of the laboratory assembly and operation have been previously described (Pekney et al. 2014).

Data collected at the air monitoring site is classified by activity at the well pad. Horizontal drilling occurred September 8 – October 5, 2015, first at well 5H then at well 3H. Hydraulic fracturing occurred October 10 – November 16. Cleanout activities followed on November 20-26, which involved using a diesel-powered coil tubing rig to drill out plugs and flush out residue left in the wells.

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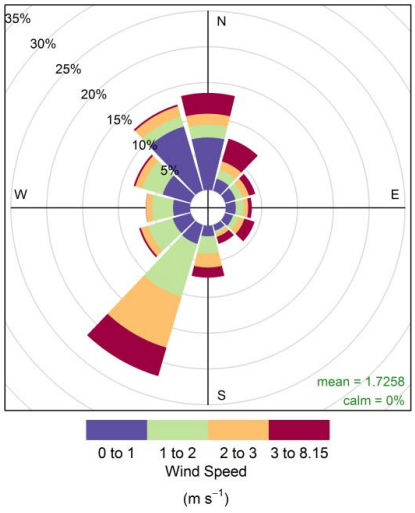

Figure 2. Wind speed and direction during ambient air monitoring campaign at MSEEL (September 2015-February 2016).

157 Flowback, the flowing of gas, formation fluid, and frac fluid up the wells to the
158 surface, took place over December 10-14, after which both wells were in production.
159 A reduced emission completion (REC) was performed; gas produced during this time
160 was captured using portable equipment brought on site that separates the gas from the
161 liquids so that the gas can be retained as a product.

162 Air monitoring began September 18, 2015 and ended February 1, 2016. No data were
163 collected for the vertical drilling phase. Data collection was continuous except for
164 calibration and instrument downtime. The laboratory's meteorological station
165 measured relative humidity, temperature, rainfall, solar radiation, wind direction,
166 wind speed, and barometric pressure at an elevation of 10m.

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171 Table 1. Constituents measured by the MSEEL mobile air monitoring laboratory
172 (Pekney et al. 2018).

Measurement	Unit	Resolution	Sampling Rate	Instrument	Measurement technique
VOCs (52 compounds, see Table S1 for full list)	ppb	0.4 ppb	1 hour	Perkin Elmer Ozone Precursor Analyzer (Waltham, Massachusetts)	Gas Chromatograph with Flame Ionization Detection (GC—FID) with thermal desorption
Ozone, NO_x	ppb	0.4 ppb Ozone, 50 ppb NO_x	1 minute	Teledyne-API Gas Analyzers T400 and T200U (San Diego, California)	UV absorption, Chemiluminescence
Methane, carbon dioxide	ppm	<5 ppb Methane, 1 ppm CO_2	1 second	Picarro G2201-i (Santa Clara, California)	Cavity Ring-Down Spectrometry
Meteorological Parameters: wind speed and direction, temperature, relative humidity, barometric pressure, rainfall, and solar intensity	various	Various; 1 degree for wind direction/ 0.45 m/s for wind speed for Vantage Pro2 Plus; 0.1 degree for wind direction/ 0.01 m/s wind speed for R.M. Young 81000	1 minute	Davis Instruments Vantage Pro2 Plus (Oakland, California) and R.M. Young 81000 ultrasonic anemometer (Traverse City, Michigan)	Various

173 **Source-Receptor Modeling**

174 Positive Matrix Factorization (PMF), a factor analysis method (Figure S3), was
175 applied to hourly averaged ambient concentrations of measured species to identify
176 possible sources and patterns for the stages of development. PMF decomposes the
177 sample data into two matrices: factor profiles (representative of *sources*) and factor
178 contributions (Brown et al. 2015; Norris et al. 2014). The fundamental objective of
179 PMF is to solve the chemical mass balance (Equation 1) between measured species
180 concentrations and source profiles while optimizing goodness of fit (Equation 2):

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Mass balance (Evans and Jeong 2007):

$$x_{i,j} = \sum_{k=1}^{p} g_{ik} f_{kj} + e_{ij} \quad [1]$$

where $x_{i,j}$ is the data matrix with dimensions of i (observations) by j (chemical species), p is the optimum number of factors, g_{ik} is the factor contribution to the observation, f_{kj} is the species profile of the factor, k is the factor, and $e_{i,j}$ is the residual concentration for each observation.

Goodness of fit:

$$Q = \sum_{i=1}^{n} \sum_{j=1}^{m} \left(\frac{x_{ij} - \sum_{k=1}^{p} g_{ik} f_{ij}}{s_{ij}} \right)^2 \quad [2]$$

where Q is the goodness of fit, n is the total number of observations, m is the total number of chemical species, and s_{ij} is the uncertainty for each observation. Summary of methods for uncertainty calculations are provided in Supplemental Information. Missing values within the data set are replaced with the median value of that species; also, uncertainty for missing values is set at four times the species-specific median by the program. Multiple runs with different numbers of factors are executed for each data set. The output of the PMF analysis needs to be interpreted by the user to identify the number of factors that may be contributing to the samples and the possible sources they represent. One of the main strengths of PMF analysis is that each sample is weighted individually, which allows the user to adjust the influence of each sample based on the measurement confidence.

Signal-to-noise ratio (S/N), an indicator of the accuracy of the variability in the measurements, can be used to identify a species as “Strong”, “Weak”, or “Bad”.

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Generally, if this ratio is greater than 0.5 but less than 1 that species has a "Weak" signal. "Strong" is the default value for all species with an assumption of S/N greater than 1. "Bad" category excludes the species from the rest of the analysis. We considered the number of samples that are missing or below the detection limit when choosing the category for each species. (Norris et al. 2014). The expected goodness of fit ($Q_{expected}$) is calculated for each scenario (Norris *et al.*, 2014):

Expected goodness of fit:

$$Q_{expected} = (i \times j\ ) - \{(p \times i) + (p \times j)\} \quad [3]$$

where (i x j) is the number of non-weak data values in Xij and (p x i) and (p x j) are the number of elements in G and F, respectively. Q_{robust} is the calculated goodness-of-fit parameter that excludes points that are not fit by the model. The lowest $Q_{robust}/Q_{expected}$ is calculated to compare different factor scenarios; when changes in Q become small with increasing factors, it can indicate that there may be too many factors in the solution (Brown et al. 2015).

In addition to these calculated parameters, factor profiles and error estimation diagnostics are used to compare the output of different simulations. Marker species (chemical species that are unique to a particular source) and temporal or seasonal variations can be used to aid in identifying the possible emission sources (Figure 3). Associations between factors can also provide useful information for profile characterization. Moreover, meteorological data can provide useful information about the geographic location of the sources.

In order to perform the PMF analysis, we utilized a user-friendly graphical user interface (GUI) developed by the U.S. Environmental Protection Agency (EPA), EPA PMF 5.0 (Norris et al., 2014). Hourly average data was used for each pollutant to

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unify the measurement intervals. All pollutants included in the matrix were identified as "strong" (signal to noise: S/N > 2). Fifty base runs were performed, and the run with the minimum Q value was selected as the base run solution. In each case, the model was run in the robust mode with a number of repeat runs to ensure the model least-squares solution represents a global rather than a local minimum. First, the rotational (linear transformation) Fpeak variable was held at the default value of 0.0. However, there can be almost infinite possibilities of F and G matrices that produces the same minimum Q value, but the goal is producing a unique solution. As a result, rotational freedom is one of the main sources of uncertainty in PMF solutions (Paatero et al. 2014). Therefore, Fpeak values were adjusted (-1.0, -0.5, 0.5, and 1.0) to explore how much rotational ambiguity exists in PMF solutions. In other words, the model adds and/or subtracts rows and columns of F and G matrices based on the Fpeak value, which is typically between -5 and +5 (Norris et al. 2014). Positive Fpeak values cause a sharpened F-matrix and smeared G-matrix; negative Fpeak values result in subtractions in the G-matrix. The factor contributions were analyzed to find the optimum Fpeak value.

The PMF analysis was completed with error estimation. We used three methods of error estimation: Bootstrap (BS), Displacement (DISP), and BS-DISP, which guide understanding the stability of the PMF solution (Norris et al. 2014). BS analysis is used to determine whether a set of observations affect the solution disproportionately. The main idea of BS analysis is resampling different versions of the original data set and perform PMF analysis. Random errors and rotational ambiguity affect BS error intervals. The main reason of rotational ambiguity is the existence of infinite solutions similar to the solution generated by PMF solution. DISP analysis helps to analyze the PMF solution in detail. Only rotational ambiguity affects DISP error intervals.

BS-DISP is a hybrid method that gives more robust results than DISP results.

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Results and Discussion

Overview of Results for Measured Compounds

Figure 3 shows a box-and-whisker graph of the measured NO_x, NO, NO_2, Ozone, and ethane during the whole monitoring campaign at the study site. Similarly, Figure 4 shows a statistical summary of methane and carbon dioxide. The y-axis represents concentrations and the x-axis represents the phases of the well development. The black line on each of the boxes represents the median for that particular data set. The small circles represent outliers. The blue circles represent the mean. Since most of the VOCs concentrations measured were consistently below 10 ppb, only ethane is included. There was an increase for NO_x (25^{th} percentile (q1)=12.5 ppb) and NO (q 1= 2.7 ppb) during the *fracturing* phase compared to other phases. The whiskers show the high variability for this phase, which can be a result of small sample size for the *fracturing* phase. NO/NO_2 ratio for 25^{th} and 75^{th} percentiles was 1.2, indicating fresher, less oxidized emissions. The skewness of the data for this phase indicates that the data may not be normally distributed. NO_2 graph shows a similar trend for the *fracturing* phase. We did not observe significant differences for different development phases for ozone, which is not surprising as it is a secondary pollutant and it can be related to winter season of the data collection period. (Peter M. Edwards et al. 2014). There was a dramatic increase for the flowback phase for ethane concentration. This 25^{th} percentile was 24 ppb, while this concentration ranged between 0 and 11 ppb for other phases. The 75^{th} percentile was 89 ppb, which is a significantly higher value compared to other phases. We observed a similar trend for methane concentration. The 25^{th} percentile (2.5 ppm) and the 75^{th} percentile (4.3 ppm) were significantly higher than other phases. Differences for development phases for CO_2 were not statistically significantly different. CO_2 has many emissions sources and variable

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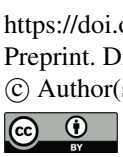

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280 background concentrations so distinguishing emissions from the well pad activities is
281 difficult.

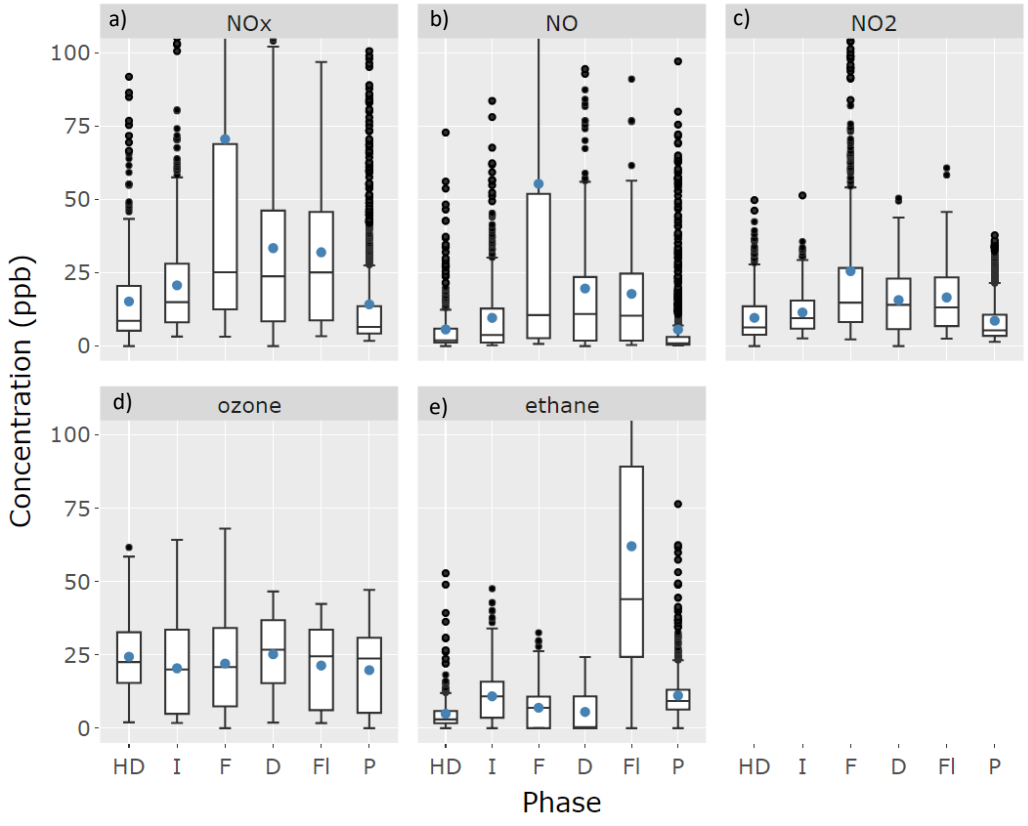

Figure 3. Summary statistics of input parameters for (a) NO_x, (b) NO, (c) NO_2, (d) Ozone, € ethane (HD: Horizontal Drilling, I: Idle, F: Fracturing, D: Drillout, Fl: Flowback, P: Production. The idle phase consists of gaps of time between other operational phases, when there was little to no emissions-generating activity on the well pad.

282 The average concentrations of methane and ethane for the entire monitoring campaign
283 are shown in Figure S4. The highest ethane concentrations occurred during the
284 *flowblack* stage (565.7 ppb). A mean that is significantly higher than the median
285 comes from a distribution that is skewed due to peak events ($mean_{ethane}$= 11.4 ppb,
286 $median_{ethane}$= 8.5 ppb). Propane and isobutane had the second and third highest
287 average concentrations, respectively, for each phase of development. Similarly, the

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hourly concentration graphs of NO_x, O_3, and CH_4, and CO_2 were used to analyze the factor solutions (Figure S5).

Factor Profiles

The three-factor model was chosen for the PMF analysis based on the interpretation of the factor profiles, $Q_{robust}/Q_{expected}$ ratios, factor contributions, error estimation results, and hourly peak concentrations of pollutants (Figure S6). The three-factor solution was resolved to the following factors: *natural gas* for the natural gas-related emissions sources; *regional transport/photochemistry* for the atmospheric regional molecular transport and oxidized background air; and *engine emissions* for emissions from vehicles, drill rigs, generators, and pumps used at the site (Figure 5). The summary of PMF models with various Fpeak values for well development activities are shown in Table S4. The DISP, BS, and BS-DISP results for 2, 3, and 4 factor PMF solutions are summarized in Table S2. For the 3-factor analysis, the DISP results indicate that there are no swaps and the PMF solution is stable, which means there are no exchange factor identities and it is a well-defined solution for the case. According to BS results, there is a small uncertainty; this can be an impact of high variability in concentration. BS-DISP captures both random errors and rotational ambiguity; these results also indicate that the solution is reliable because there are no swaps between factors for the PMF model. Error estimation summary plots (Figure S6) show range of concentration by species in each factor: Base Value, BS 5th, BS Median, BS 95th, BS-DISP 5th, BS-DISP Average, BS-DISP 95th, DISP Min, DISP Average, and DISP Max.

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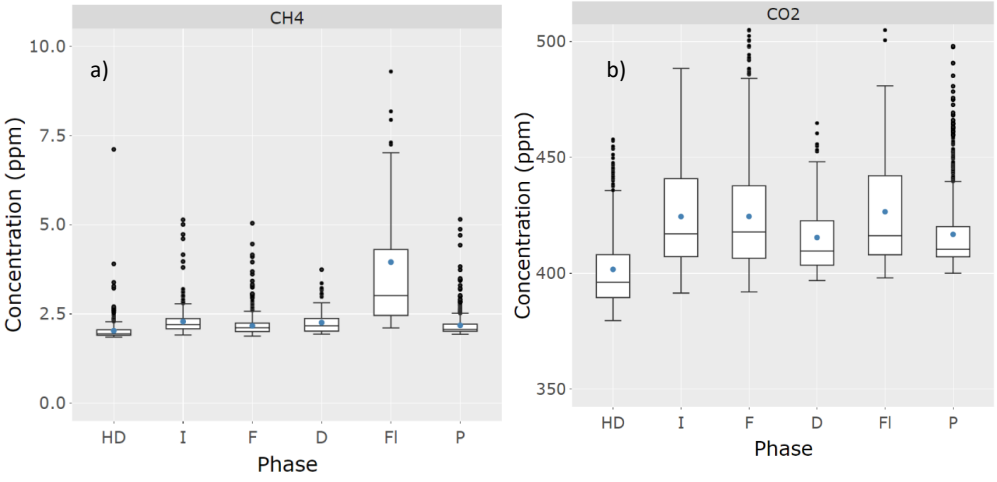

312 Figure 4. Summary statistics of input parameters for methane (a) and carbon dioxide
313 for (b) (HD: Horizontal Drilling, I: Idle, F: Fracturing, D: Drillout, Fl: Flowback, P:
314 Production.

315 **Source Profiles**

316 **The natural gas factor** was named as such due to its composition of species that are
317 present in natural gas: 1% methane, 3% ethane, 1.5% propane, 0.5% isobutane, 1% n-
318 butane, 0.1% pentane, and 0.2% isopentane. Ethane is a particularly good marker for
319 natural gas emissions sources due because its atmospheric sources are almost
320 exclusively from natural gas extraction, production, processing and use (Liao et al.
321 2017). Ninety-two percent of ethane mass is explained by the natural gas factor. The
322 highest contribution for this factor occurred during the flowback phase.

323 **The regional transport/photochemistry factor** was characterized by high
324 contributions from ozone (12%), CH_4 (1%), and CO_2 (86%). Ninety-nine percent of
325 the ozone mass was explained by this factor. Ozone is a product of photochemistry
326 and not directly emitted by any of the sources on the well pad. Although CH_4 and
327 CO_2 would be emitted by well pad sources, they are also present in background
328 ambient air and could be transported to the monitoring location from other sources in

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329 the region. Contributions of this factor were relatively steady for all phases of
330 operation during the entire monitoring campaign.

331 **The engine emissions factor** was composed of 39% NOx, 33% NO, and 11% NO_2 as
332 well as 0.02% toluene and 0.04% benzene. The portions of the mass of these species
333 explained by this factor are 74%, 87%, 60%, 20%, and 54%, respectively. Toluene is
334 released mainly from motor vehicle emissions and chemical spills (Gierczak et al.
335 2017). Contribution of this factor was significantly highest during hydraulic
336 fracturing, when there were emissions from many diesel engines operating
337 continuously on the well pad. Contribution during flowback was also elevated.
338 Several peaks of contribution were observed during production, which could be due to
339 maintenance vehicles and other short-lived vehicle-based activities on the well pad.

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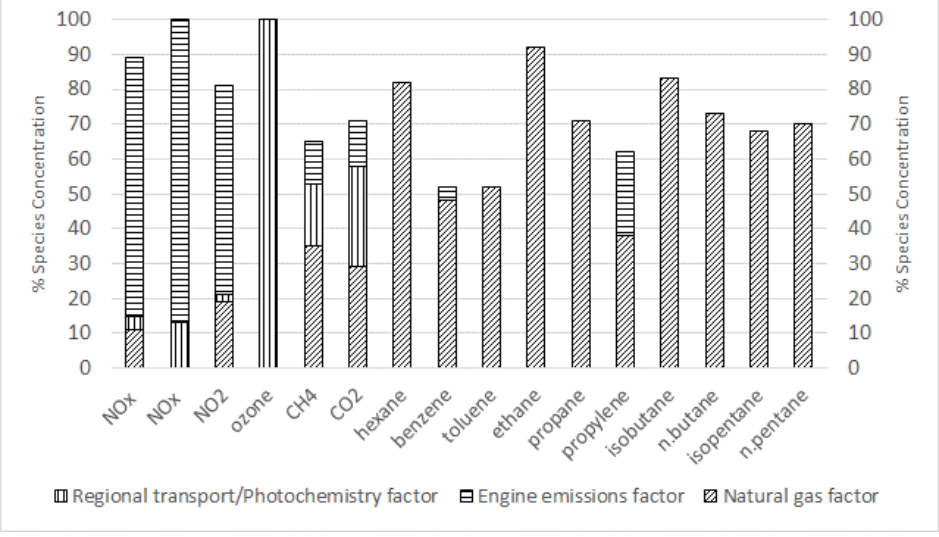

341
342 Figure 5. The three-factor solution fingerprints for Drilling through Production
343 Monitoring Period, F_{peak}=1.

344 The main limitation of the study is having uneven number of data points for each
345 operational phase. This limitation affects the analyses; however, we do not have
346 control of the durations of the operational phases. As a future work, integrating more
347 data from different fields can decrease the inherent uncertainty.

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Conclusion

We investigated the effect of unconventional natural gas development activities on local air quality by using ambient air monitoring laboratory near Marcellus Shale well pad in Morgantown, Western Virginia. The results of PMF solutions for well pad development phases show that there were three potential factor profiles as outlined in Figure 5: *natural gas*, *regional transport/photochemistry*, and *engine emissions*. Horizontal drilling stage had an important contribution to the *natural gas* factor. In addition, there was a significant concentration contribution at the end of the horizontal drilling phase. An increasing contribution to *engine emission* factor was observed over different well pad drilling through production phases. The peak concentration was observed during the drillout stage. Even though it is difficult to compare the *regional transport/photochemistry* contributions due to high variability, highest contributions occurred during horizontal drilling and drillout.

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

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important since it has potential to help inform future decision-making and constrain cumulative impact assessments.

Conflicts of interest

There are no conflicts to declare.

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Author Contribution

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Nur H Orak: Conceptualization, Methodology, Software. Visualization, Writing **Natalie J. Pekney:** Supervision, Methodology, Writing. **Matthew Reeder:** Methodology, Validation.

Code/Data availability

Model simulations presented in this paper are available upon request.

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