LA-UR-25-20066
Accepted Manuscript

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Provided by the author(s) and the Los Alamos National Laboratory (2025-01-15).

To be published in: Environmental Research Communications

DOI to publisher's version: 10.1088/2515-7620/ad82b2

Permalink to record:
https://permalink.lanl.gov/object/view?what=info:lanl-repo/lareport/LA-UR-25-20066

ENVIRONMENTAL RESEARCH
COMMUNICATIONS

LETTER • OPEN ACCESS

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To cite this article: Emily Lachenmayer *et al* 2024 *Environ. Res. Commun.* **6** 101013

Environmental Research Communications

CrossMark

OPEN ACCESS

RECEIVED
30 May 2024

REVISED
16 August 2024

ACCEPTED FOR PUBLICATION
2 October 2024

PUBLISHED
26 November 2024

LETTER

Source apportionment of airborne volatile organic compounds near unconventional oil and gas development

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Keywords: PMF, oil and gas, source apportionment, volatile organic compounds

Supplementary material for this article is available online

Abstract

Oil and natural gas (ONG) extraction emits volatile organic compounds (VOCs). Certain VOCs are identified as hazardous air pollutants (HAPS) while others contribute to ozone formation. This study examines the impact of ONG operations on VOC levels during the development of multi-well ONG pads in suburban Broomfield, Colorado. From October 2018 to December 2020, weekly VOC measurements were taken at 18 sites across the area. These included spots near well pads, in adjacent neighborhoods, and at a background site, covering various stages of well pad development including drilling, hydraulic fracturing, flowback, and production. Analysis using Positive Matrix Factorization (PMF) identified six factors, including combustion, background/biogenic sources, light and complex alkanes, drilling activities, and ONG acetylene. Factors linked to local ONG activities exhibited clear temporal and spatial correlations with Broomfield well development. Benzene source analysis revealed distinct contribution gradients, with ONG-related sources notably influencing areas near the well pads, particularly in pre-production. ONG-related weekly benzene contributions varied from 9% to 63% at a community background site and 18% to 89% in a neighborhood close to a well pad.

Introduction

Oil and natural gas (ONG) extraction emits volatile organic compounds (VOCs) that contribute to ozone (O_3) formation (Gilman *et al* 2013, Field *et al* 2014, Cheadle *et al* 2017, Benedict *et al* 2019) and hazardous air pollutants (HAPs) linked with serious health effects (Hazardous Air Pollutants 2021). Advances in hydraulic fracturing and directional drilling techniques have led to substantial ONG production increases, including in heavily populated areas of the Denver-Julesburg (DJ) Basin on the Colorado North Front Range (CNFR).

Prior studies in the CNFR found substantial ONG contributions to regional VOC concentrations using Positive Matrix Factorization (PMF) and simpler regression techniques (Gilman *et al* 2013, Abeleira *et al* 2017, Pollack *et al* 2021). Missing from past studies has been a detailed, community-scale analysis of contributions of ONG operations to concentrations of HAPs and other VOCs during drilling and completion of large, multi-well ONG pads. Hecobian *et al* (2019) reported VOC emissions during specific well development activities in Colorado but did not assess resultant changes in ambient VOC concentrations with distance from well pads. Further, operational modifications, including use of grid-powered electrified drill rigs and closed loop fluid handling systems, have been implemented in some operations in recent years.

Ku *et al* (2024) report temporally and spatially resolved VOC concentrations in Broomfield, Colorado, prior to, during, and following development of several large, multi-well ONG pads. They report increases in ONG-related VOCs during pad development activities. Here we use PMF to quantitatively assess VOC source contributions during development of two Broomfield well-pads. We assess local ONG development impacts on

HAP and VOC concentrations at various stages and distances, using VOC data from up to 18 community locations, starting before well pad development and continuing through drilling, completion, and production. Our findings will (1) help stakeholders assess the physical extent ('footprint') of local ONG emission impacts, (2) quantify the magnitude of air quality impacts from local ONG development among contributions from all VOC sources, and (3) help identify ONG well development periods and practices that have the largest impact on local air quality, presenting targets for future emissions reduction efforts.

Methods

Six large, multi-well pads were developed in the city and county of Broomfield (CCOB). As part of an operator agreement, facilities were designed and operated under specific requirements that included using electric drill rigs, operating a closed loop system for fluids handling, and limiting use of storage tanks on well pads (Ku *et al* 2024).

Air quality monitoring program

Air monitoring began prior to pad development to assess impacts of new ONG development and other local/regional air pollution sources, including traffic and approximately 37,000 existing ONG DJ Basin wells (Colorado Oil and Gas Conservation Commission 2022). Here we consider weekly VOC measurements between October 2018 and December 2020 at eighteen strategic sites across CCOB. This period includes pre-development measurements through early ONG production at the Interchange B and Livingston well pads. We analyze key ONG pre-production phases—drilling, coiled and production tubing operations, hydraulic fracturing, and flowback—for any change in concentrations, noting differences in operational timing across pads (see Supplemental information (SI)). Drilling was conducted using electrified, grid-powered drill rigs. Hydraulic fracturing (fracking) pumped high-pressure fluids into isolated well zones to fissure rock formations and increase permeability. Coiled tubing was employed to mill out plugs isolating fracking zones. Production tubing was installed prior to flowback, when water, sand, and hydrocarbons flow up the wellbore. When water production declines, the well goes into production.

Eight monitoring sites were located near a well pad, nine in nearby neighborhoods, and one 5 km from new ONG development selected as a background site. Site locations and sampling schedules are available in SI. Weekly time-averaged whole air samples were collected and analyzed for VOCs. Site sampling frequencies varied with local ONG operations.

Measurements

Each monitoring site included a 6L stainless steel canister with internal Silonite® coating (Entech Instruments). Canisters were cleaned and evacuated prior to deployment (Ku *et al* 2024). Canister sample flow was controlled using a CS1200E Entech Flow Controller. Canister samples were analyzed within one week of collection using a custom-built, 5-channel Gas Chromatograph (GC) (Benedict *et al* 2019, Hecobian *et al* 2019). The system includes three GCs and five detectors (three flame ionization detectors, one electron capture detector, and one mass spectrometer). A targeted analysis measured 48 VOCs in each sample, including light alkanes, alkenes, cyclic and branched alkanes, aromatics, acetylene, and isoprene. The system was calibrated using a certified mixed hydrocarbon standard (HC Mix 56 Airgas, PA, USA) with an accuracy of +/−5%. Multiple working standards were analyzed with each sample batch to monitor system drift. Analytical methods and details are provided elsewhere (Ku *et al* 2024).

Measurement precision was quantified through eight replicate analyses of a large-volume ambient air sample. Relative standard deviations varied between 1 and 15% for measurements of most of the compounds included in the PMF analysis (see SI for details). Limits of detection (LOD) were quantified through analysis of a highly diluted standard. Concentrations below LOD were replaced with half the LOD and the uncertainty associated with the measurement was reported as 5/6 of LOD as suggested by Polissar *et al* (1998). See SI for additional details.

PMF

This study utilized EPA PMF v5.0 (Norris *et al* 2014), previously utilized for apportionment of non-methane VOCs (e.g., Abeleira *et al* 2017, Hecobian and Collett 2019, Pollack *et al* 2021) and explained elsewhere (Hopke 2000, Comero *et al* 2009, Norris *et al* 2014). This section provides a brief overview; more details are in SI.

PMF aims to solve a chemical mass balance (CMB) equation, producing solutions only containing positive values. CMB decomposes a speciated data matrix with i samples and j chemical species along with uncertainties, u, into a summation of p factors containing three matrices: g (mass contribution), f (species profile), and e (residuals) (equation (1)). It aims to minimize Q, a squared difference between the fit of the summed predicted

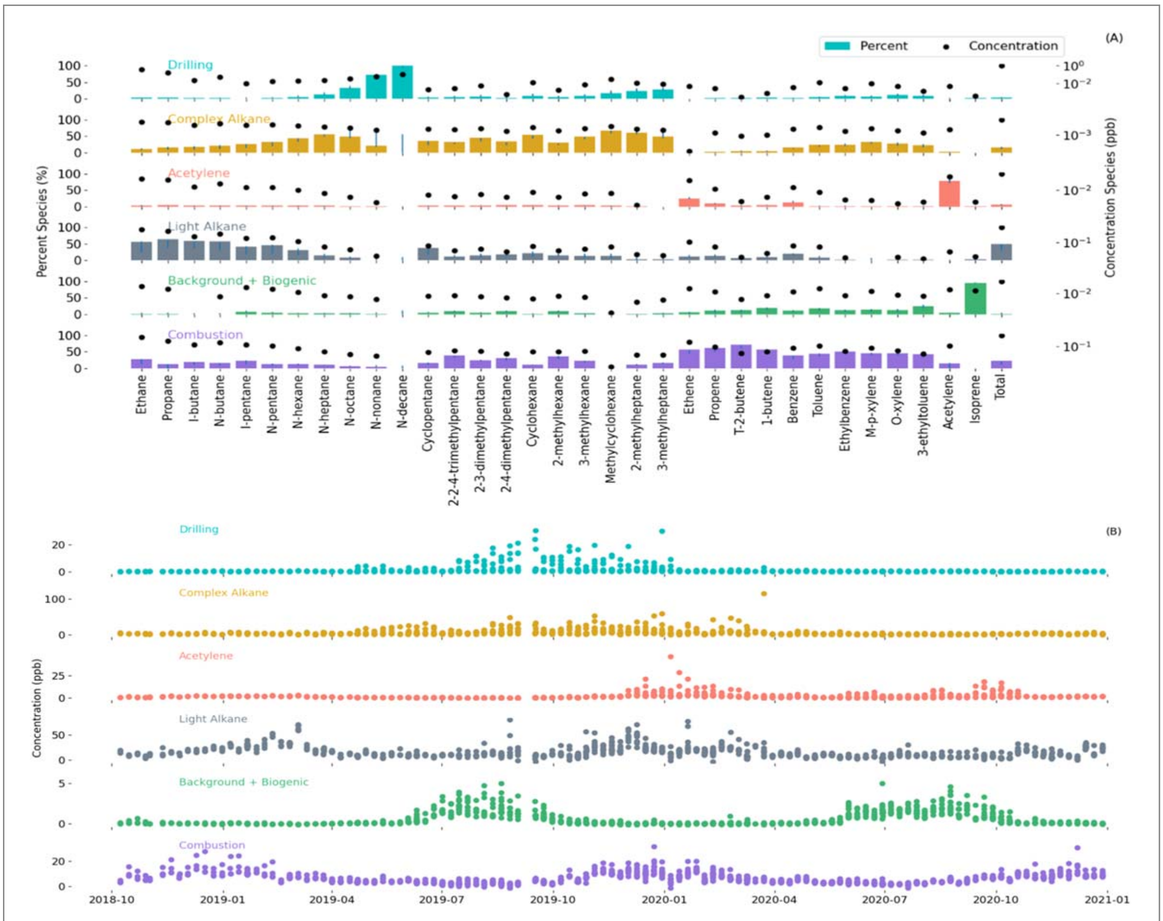

Figure 1. (A) Species concentrations and percentages associated with individual PMF factors and (B) Factor time series and concentrations at CCOB monitoring sites (mutiple points during each week represent different monitoring sites).

speciated factors and the original matrix normalized by the measurement uncertainties (equation (2)). A user-determined number of factors (k) are analyzed for their mathematical robustness using displacement and bootstrapping techniques. If solutions with minimized error similar to the initial set of factors have similar timelines and compound profiles, the solution is considered rotationally robust (Norris *et al* 2014).

$$x_{ij} = \sum_{k=1}^{p} g_{ik} f_{kj} + e_{ij} \quad (1)$$

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

Results and discussion

PMF was used to analyze weekly VOC concentration data from October 2018—December 2020, covering pre-development to early production at the Interchange and Livingston pads. The analysis included 33 compounds which were quantifiable in more than 40% of the samples and had a signal to noise ratio that exceeded the compound's uncertainty—as suggested by the PMF 5.0 User Guide (Norris *et al* 2014). Compounds included: ethane, propane, i-butane, n-butane, i-pentane, n-pentane, n-hexane, n-heptane, n-octane, n-nonane, n-decane, cyclopentane, 2,2,4-trimethylpentane, 2,3-dimethylpentane, 2,4-dimethylpentane, cyclohexane, 2-methylhexane, 3-methylhexane, methylcyclohexane, 2-methylheptane, 3-methylheptane, ethene, propene, t-2-butene, 1-butene, benzene, toluene, ethylbenzene, m + p-xylene, o-xylene, 3-ethyltoluene, acetylene, and isoprene. Six identified factors (see figure 1) were labeled as follows: 1) Combustion, 2) Background + Biogenic, 3) Light Alkanes, 4) Complex Alkanes, 5) Drilling, and 6) Acetylene. The Light Alkane, Complex Alkane, Drilling, and Acetylene factor composition and/or spatiotemporal nature aligned well with development activities. The Combustion and Background + Biogenic factors exhibited compositions and timelines unrelated to ONG emissions.

Previous VOC apportionment studies in the CNFR and Piceance Basin have reported factors similar to the Light and Complex Alkane profiles (Abeleira *et al* 2017, Hecobian and Collett 2019, Pollack *et al* 2021). While the split of these factors has previously been linked to differing atmospheric lifetimes (Pollack *et al* 2021), current findings, near active ONG sources suggest a factor split reflecting significant local complex alkane emissions during specific well development activities.

The Light Alkane factor, comprised of short-chain alkanes, represented a significant proportion of observed ethane (63%), propane (73%), i-butane (66%), n-butane (66%), and n-pentane (52%). Ethane and propane are commonly utilized as ONG tracers (Gilman *et al* 2013, Collett *et al* 2016, Oltmans *et al* 2021, Rossabi *et al* 2021). This factor exhibits a seasonal cycle (figure 1) with elevated winter concentrations when surface emissions are often trapped in a shallow boundary layer. Spatiotemporal factor analysis suggests possible influence from local well-pad tubing and flowback operations. Given the large regional influence of light alkane emissions from many producing ONG wells in the DJ Basin, however, influence by local ONG development is less obvious than with other factors.

The Complex Alkane factor, rich in longer chain, branched, and cyclic alkanes, represented 50% or more of concentrations for n-heptane, cyclohexane, methylcyclohexane, 2-methylheptane, and 3-methylheptane. This factor increased in prominence from spring 2019 through spring 2020—aligning with extensive drilling and well completion activities at Interchange B and Livingston pads. Elevated concentrations coincided with drilling operations at all active pads, tubing operations at Livingston and Interchange pads, and flowback operations at the Interchange pad.

The ratio of i-pentane to n-pentane serves to distinguish VOC emissions from combustion or ONG sources (Gilman *et al* 2013, Abeleira *et al* 2017, Hecobian *et al* 2019, Pollack *et al* 2021). ONG emissions typically have i/n pentane ratios <1.0, while traffic and combustion emission ratios usually exceed 2.0 (Swarthout *et al* 2013). Gilman *et al* (2013) reported a ratio of 0.86 ± 0.02 for the DJ Basin Wattenberg field. Here, the Light and Complex Alkane PMF factors showed similar ratios at 0.88 and 0.81, respectively.

The CCOB Drilling and Acetylene factors, to our knowledge, have not been previously reported. These factors are directly associated with local ONG emissions, being clearly correlated in time and space with well pad activities.

The Drilling factor, characterized by abundant levels of octane, nonane, and decane, representing 33%, 73%, and 100% of their overall concentrations, exhibited an i/n pentane ratio of 0.55, within the range observed during CCOB drilling operations (Ku *et al* 2024). This factor was named for (1) its similarity in composition to a headspace VOC analysis (see Ku *et al* 2024 for details) of the synthetic (Neoflo) drilling mud used during Livingston pad drilling and (2) spatiotemporal association with known CCOB drilling periods and locations. Drilling mud lubricates the drill bit and helps convey drill cuttings up the bore hole. While grid-powered drill rigs eliminated engine emissions, unenclosed shakers used for separating drill cuttings and drilling mud remain an important source of VOC emissions. Smaller increases in the PMF Drilling factor were observed during earlier Interchange B pad drilling operations that used a different petroleum-based drilling mud (Gibson) which emits fewer C_8-C_{10} alkanes (see Ku *et al* 2024). The switch from Gibson to Neoflo-based mud addressed local odor complaints.

The Acetylene factor accounts for 77% of total acetylene, 24% of ethene, and 13% of benzene concentrations. This factor exhibited elevated concentration periods in winter 2019/2020 and summer/fall 2020, primarily around the Livingston pad. Other studies of ONG emissions (Schade and Roest 2016, 2018) have linked elevated acetylene, alkene and benzene emissions to flaring; however, reported benzene:toluene ratios are much lower than the 4.7 ratio seen here. The prevalence of the acetylene factor during flowback operations is especially surprising since combustion sources are typically limited on-pad during flowback. Flares were not operating at the Livingston pad during this period and infrared camera inspections by CCOB inspectors did not find improperly operating enclosed combustion devices. Separate mobile and high time resolution measurements in fall 2020 further confirmed the Livingston pad as the source of plumes rich in acetylene, ethene, and benzene without significant toluene (Ku *et al* 2024), lending confidence that this PMF factor represents a real, if unusual, source. Such a PMF factor has not previously been reported for ONG producing regions. The dominant contributions of this source to CCOB acetylene concentrations during extended periods suggests that caution is needed in using acetylene as a tracer for non-ONG combustion sources for VOC source apportionment.

The Combustion PMF factor showcased elevated alkenes and toluene and an i/n-pentane ratio of 1.74, expected for combustion emissions. A clear factor seasonal cycle, peaking during colder months, is consistent with year-round traffic emissions concentrated within a shallower boundary layer during winter. The combustion factor was most prevalent at the Commons background site, that is near busy traffic routes and closer to the Denver metro area.

Isoprene was the dominant species in the Background + Biogenic PMF factor that peaked in summer, when vegetation growth and elevated temperatures increase biogenic emissions. Concentrations were elevated in more highly vegetated neighborhoods and lower near well pads and the adjacent highway with their limited vegetation.

Spatial analysis of pad emissions

The long-term, spatially resolved CCOB VOC measurements offer valuable insights into the localized impact of ONG emissions. The Livingston pad was analyzed due to its proximity to residential neighborhoods and clear drilling factor signature. Site-specific ONG-related factor observations from six monitoring sites, ranging from near the well pad (Livingson02, 110 m distance) to the Commons background site (5 km distance), were analyzed. Spatial gradients of ONG-related factors were examined during periods of Livingston pad activity and compared to a background period without ONG activity. As seen in SI, no gradient is apparent in ONG-related PMF factors with distance from the pad during the background period.

Observed spatial gradients in concentrations of the four ONG-associated PMF factors (see SI) reveal that local ONG pre-production activities impact regions past the pad fence line and beyond required setback distances for new ONG wells (610 m in Colorado). The Drilling factor shows a clear, decreasing concentration away from the pad during drilling operations. The Complex Alkane factor also showed clear enhancement at sites closer to the pad with somewhat smaller near-pad enhancement factors than for drilling, reflecting a greater background concentration of this factor.

A local increase in the abundance of the Light Alkane PMF factor was apparent during the coiled tubing (mill-out) period. A shallower gradient moving away from the pad for this factor reflects elevated levels of light alkane concentrations across the region due to extensive DJ Basin ONG operations and the long atmospheric lifetimes of these compounds. Substantial increases in concentrations of the Acetylene factor were also observed closer to the Livingston pad during periods when emissions associated with this factor were active.

Benzene source apportionment

ONG emissions of Hazardous Air Pollutants (HAPs) are of interest given potential impacts on health of nearby residents (Weisner *et al* 2023). The PMF analysis allows us to apportion source contributions to observed HAP concentrations during ONG development phases. Results are presented here for benzene, which is emitted by a variety of sources including ONG and traffic. Benzene is carcinogenic and has been identified as the HAP most likely to present exposure concerns near ONG facilities (McMullin *et al* 2018, Holder *et al* 2019). Findings for other HAPS (n-hexane and n-nonane) are in SI.

Measured benzene concentrations were reconstructed well by the PMF solution (Pearson's correlation 0.79). Over the study period across all CCOB sites, the Combustion factor accounted for an average 40% of benzene concentrations, followed by the Light Alkane factor at 22%, the Complex Alkane factor at 16%, and the Acetylene factor at 12%. These contributions varied in time and space (see figure 2). Sites closer to an ONG pad (e.g., Livingston) show larger benzene concentrations associated with ONG factors during pre-production activities. January 2–9, 2020, during Livingston hydraulic fracturing operations, for example, ONG-related factor contributions to benzene at Livingston 02 totaled 95% versus 26% at the Commons background site. Benzene contributions from ONG-related factors ranged from 26% to 100% at the Livingston near-pad sites, 18% to 89% at the nearby Anthem neighborhood site, and 9% to 63% at the Commons background site.

Across the full study period the Livingston and Interchange near-pad sites averaged 61% and 56% of benzene concentrations from ONG factors. These monitoring sites were closer to well pads than were any residences; however, further away we still see an average 55% ONG factor benzene contribution at the Anthem neighborhood site. At the Commons background site an average of just 30% of benzene was associated with ONG sources. Other PMF studies within the CNFR attributed the largest benzene contributions to a mixture of sources (Pollack *et al* 2021) with traffic contributing an overall minor amount (Abeleira *et al* 2017). Elevated benzene periods were associated with variations in combustion and light alkane factors (Pollack *et al* 2021). Gilman *et al* (2013) utilized a multivariate regression method to apportion source contributions finding an average 32% of benzene at the Boulder Atmospheric Observatory north of CCOB attributed to ONG activities, similar to our 30% average for Broomfield Commons.

While study findings reveal significant regional and even larger local ONG contributions to benzene concentrations, levels in CCOB weekly observations (maximum of 0.80 ppbv with site averages 0.14–0.22 ppbv) fall consistently below the 3 ppbV Agency for Toxic Substances and Disease Registry (ATSDR) non-cancer, chronic health guideline value (HGV). HGVs do not consider cumulative and additive impact exposure to multiple HAPs. Other CCOB research concluded that cumulative impact exposure may explain an increase in upper respiratory and acute symptom reporting in residents living within 1.6 km of ONG pads (Weisner *et al* 2023). Increases in benzene exposure can increase cancer risk, although the largest increases were during pre-production activities lasting days to months compared to lifetime exposures considered for cancer risk. Higher

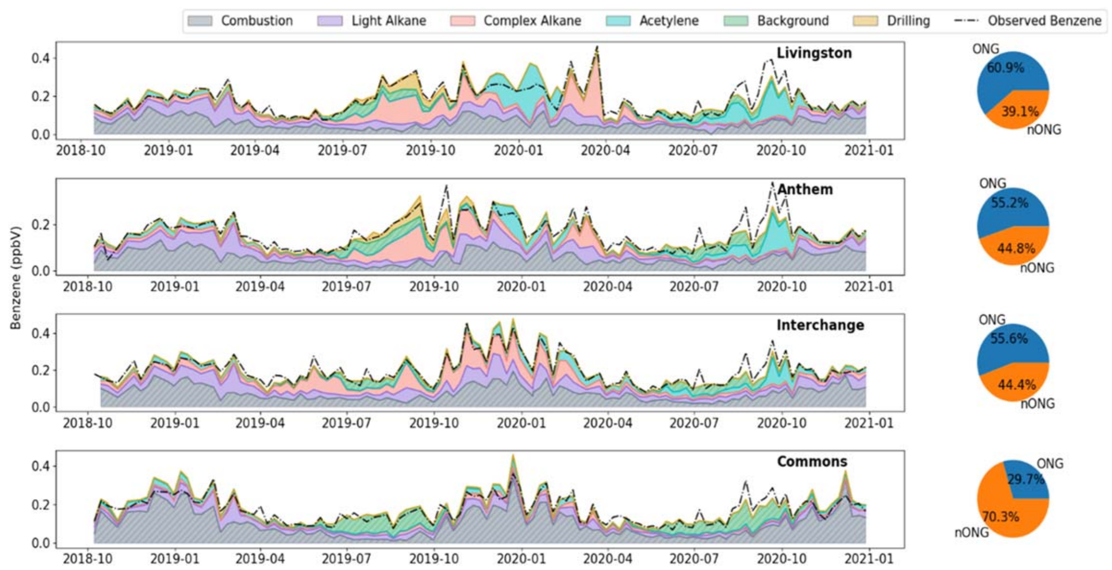

Figure 2. Reconstructed PMF benzene concentrations and their allocation to factor profiles at (A) Livingston, (B) Anthem 01, (C) Interchange, and (D) Commons 01. Results are averaged for the near-pad sites at Livingston and Interchange. Non-ONG related factors are denoted with hash marks. Pie charts depict the average contributions of ONG related factors (blue) and non-ONG factors (orange) over the study.

time resolution CCOB HAPs measurements relevant for considering acute exposure are summarized by Ku *et al* (2024).

Conclusions

This study demonstrates that PMF analysis of spatially resolved, weekly speciated VOC data can provide good insight into the time-varying impacts of ONG operations and other sources on local air quality. Four of six identified PMF source factors were associated with local and/or regional ONG operations. Two (Light Alkanes and Complex Alkanes) have been previously reported in the CNFR. New factors were associated with (1) drilling mud volatilization and (2) well pad emissions of acetylene, typically considered a non-ONG combustion tracer. Temporally and spatially resolved speciated VOC observations and information about ONG well drilling and completion timelines were critical in identifying these factors.

Analysis of the spatial footprint of air quality impacts and PMF factors associated with local ONG development activities demonstrated that emissions exerted the largest influence close to the well pad but continued to influence local air quality in surrounding neighborhoods beyond Colorado's required 610 m well setback distance. While neighborhoods close to ONG operations experienced elevated concentrations of benzene and other HAPs during well drilling and completions activities, weekly benzene concentrations across the community remained well below a 3 ppbv health guideline value for chronic exposure.

Acknowledgments

Funding for this work was provided by the City and County of Broomfield (CCOB). We are grateful to CCOB staff for their assistance with monitor siting and for providing information about well pad operations.

Data availability statement

The data that support the findings of this study are openly available at the following URL/DOI: https://doi.org/10.5061/dryad.fqz612jzs.

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