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Volatile organic compounds and their role in secondary aerosol chemistry in a cold and dark urban environment

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Abstract. The role of volatile organic compound (VOC) emissions in secondary aerosol chemistry in urban subarctic environments remains poorly understood. Previous studies in wintertime Fairbanks, Alaska suggest that aldehydes may serve as precursors of S(IV) species in aerosols, contributing to the high concentrations of wintertime $PM_{2.5}$ pollution that Fairbanks often experiences. It is not known to what extent VOCs participate in secondary aerosol chemistry, or what the sources of largely secondary VOCs (like formaldehyde) are in such a cold and dark environment. Here, we explore the sources of VOCs and examine their role in secondary chemistry. We use measurements from an online proton transfer reaction time of flight mass spectrometer (PTR-ToF-MS), combined with complementary gas and aerosol measurements from the the Alaskan Layered Pollution and Chemical Analysis (ALPACA) wintertime field campaign in 2022,

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to examine VOC sources and their roles in aerosol chemistry in downtown Fairbanks. We find that alcohols, aromatics and carbonyls together account for ~70% of measured VOCs, with methanol, ethanol, formaldehyde, benzene and toluene as dominant species. Positive matrix factorization (PMF) analysis indicates that approximately 56% of VOCs are associated with vehicle emissions, while wood heating and heating oil together contribute about 14%. Formaldehyde is primarily linked to diesel emissions, as well as primary and secondary sources associated with aged air masses. By comparing PMF factors with measured $PM_{2.5}$ S(IV) species, we find that vehicle-related emissions of ammonia and formaldehyde likely play a key role in the formation of hydroxymethanesulfonate (HMS) in Fairbanks.

1 Introduction

Volatile organic compounds (VOCs) are associated with many serious adverse health effects (Sharma et al., 2018; Zhou et al., 2025). While the majority of VOCs are of biogenic origin on a global scale (Guenther et al., 1995), anthropogenic sources can dominate in urban environments. VOCs can undergo secondary processes, typically via photooxidation. This can lead to the formation of secondary products like fine particulate matter and ozone (Grosjean et al., 1983; Seinfeld and Pandis, 2016; Ziemann and Atkinson, 2012), which have been further associated with adverse health effects (Nuvolone et al., 2018; Pope III and Dockery, 2006; Schwartz et al., 2002).

Fairbanks, Alaska is a subarctic town which often experiences episodes of poor air quality. The Fairbanks North Star Borough frequently violates the EPA $PM_{2.5}$ (particles with an aerodynamic diameter of < 2.5 μm) air quality standard of 35 $\mu g/m^3$ in the wintertime, exacerbated by strong inversion layers limiting dispersion (Cesler-Maloney et al., 2022; Simpson et al., 2024). Previous work has shown increased hospitalizations for cerebrovascular disease and respiratory tract infections in Fairbanks during periods of high $PM_{2.5}$ pollution (Kossover, 2010).

Despite a focus on achieving reductions in $PM_{2.5}$ (Campbell et al., 2022; Kotchenruther, 2016; Nattinger, 2016; Ward et al., 2012), VOCs in Fairbanks have received less attention. Wintertime Fairbanks is unique as an urban environment, with temperatures down to -40 °C and only a few hours of daylight. Under these conditions biogenic VOC emissions are insignificant, and secondary VOC production related to UV photooxidation is expected to be small. Dark aging

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of VOCs may be taking place, although to what extent is not clear. Many VOCs have an appreciable decrease in volatility in such cold temperatures, leading to more organic aerosol mass than predicted (Ijaz et al., 2025). Analysis shows that levels of hazardous VOCs like benzene, toluene, and C8 aromatics (ethylbenzene, xylene) can be up to ten times higher in Fairbanks compared to other US wintertime cities (Ketcherside et al., 2025; Simpson et al., 2024).

Certain VOC species can also form organosulfur compounds in the aerosol phase, such as hydroxymethanesulfonate (HMS) (Boyce and Hoffmann, 1984). In downtown Fairbanks, elevated mixing ratios of formaldehyde—comparable to wintertime levels observed in the North China Plain (Campbell et al., 2022; Song et al., 2019)—were found to play a key role in the formation of HMS. Acetaldehyde and other carbonyls can also form similar S(IV) adducts, leading to higher aerosol S(IV) concentrations (Dingilian et al., 2024; Holen et al., 2025). HMS and other S(IV) species account for a significant fraction of $PM_{2.5}$ mass in Fairbanks, particularly when compared to measurements in other urban environments (Campbell et al., 2022, 2024). The sources of formaldehyde and other aldehydes in Fairbanks remain poorly characterized, limiting our capability of developing mitigation strategies.

Positive matrix factorization (PMF) has been used extensively to quantify sources of VOCs in urban settings (Baudic et al., 2016; Gkatzelis et al., 2021; Huang et al., 2024; Ketcherside et al., 2025; McDonald et al., 2018; Rivellini et al., 2024; Sadeghi et al., 2022). Some studies done in the US have reported that the fraction of pollution related to vehicle emissions have decreased, and volatile chemical products (VCPs) from consumer and industrial products have become the dominant source of urban VOCs (Gkatzelis et al., 2021; McDonald et al., 2018). It is not clear whether this trend of decreasing vehicle emissions holds true for Fairbanks; many studies have found that certain VOC emissions can be more than an order of magnitude higher for both gasoline and diesel vehicles when cold-starting at -7 °C compared to 23-24 °C (Cao et al., 2024; Ferrarese et al., 2024; George et al., 2015; Hays et al., 2017; Liu et al., 2023). However, Fairbanks commonly experiences temperatures down to -40 °C and, unlike many places, Alaska does not use ethanol-blended fuels. We are not aware of any work that has examined vehicle emissions at temperatures as low as Fairbanks, nor that uses non-ethanol-blended fuel, so it remains unclear how prominent vehicle VOC emissions are in Fairbanks.

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Several studies have conducted PMF analysis for gas and aerosol measurements during ALPACA field campaign. Ketcherside et al. (2025) combined PTR-ToF-MS VOCs, $PM_{2.5}$, and gaseous measurements of SO_2, CO, ozone, and NO_X to examine sources of gases and aerosols in a residential neighborhood. They find that aromatic VOCs make up 50% of the total VOC mixing ratio, and are mainly attributed to traffic. Formaldehyde was found to come from various sources, and many other VOCs were mainly attributed to residential wood combustion. As the VOC measurements in their work were conducted 3 km away from downtown Fairbanks in a residential area, their analysis is likely more influenced by residential factors such as space heating. Ijaz et al. (2024) used downtown PM_1 measurements from both a PTR-ToF-MS with a CHARON inlet and an AMS to conduct PMF. They found that most PM_1 OA was attributed to residential heating when using the CHARON PTR-ToF-MS measurements. However, the role of VOCs in secondary aerosol chemistry has not been addressed in these studies.

Here, we analyze VOC measurements from an online PTR-MS in downtown Fairbanks and use PMF to identify VOC sources during the ALPACA campaign in 2022. We also discuss our PMF solutions with Ketcherside et al. (2025) to highlight the differences and similarities between the downtown site and the House site. To our knowledge this is the first study to have two sets of PTR-MS measurements under the same airshed. We further compare our PMF solutions with complementary gas and aerosol measurements to identify main drivers for formation of S(IV) species in aerosols in Fairbanks.

2 Methods

2.1 Measurements

Measurements were conducted in a trailer in downtown Fairbanks next to the University of Alaska Fairbanks Community and Technical College (64.84064°N, 147.72677°W, elevation 136 m above sea level) from 17 January to 25 February 2022 as part of the 2022 ALPACA campaign, unless stated otherwise. Detailed information about the ALPACA campaign can be found in Simpson et al. (2024).

A proton-transfer-reaction time-of-flight mass spectrometry (PTR-TOF 6000 X2, Ionicon Analytik GmbH, Austria) was used to monitor VOC for 15 minutes at 20-second intervals, each

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hour. The instrument was operated at a low E/N of 65 Td (i.e., drift voltage/pressure; pressure, temperature, and voltage of the drift tube were set at 2.6 mbar, 120°C, and 230 V) and in RF mode for optimal sensitivity. Raw PTR-ToF-MS spectra were processed with the commercial Ionicon Data Analyzer (IDA) software to fit high-resolution mass peaks and extract the corresponding time series. Times series were post-processed (i.e., background subtraction, conversion of raw signal to mixing ratios, 2-minutes temporal averaging, PMF input generation) with an in-house data processing tool, PeTeR Toolkit (version 6.0; Igor 6.37).

Mixing ratios were calculated using Eq. (1):

$$R = \frac{RH^+}{H_3O^+} \times \frac{Tr_{H3O^+}}{Tr_{RH^+}} \times \frac{U_{drift} \times T_{drift}^2}{k \times \tau \times p_{drift}^2} \quad (1)$$

Where RH^+ and H_3O^+ are the signal intensities in counts per second (cps) of the individual protonated ion and the reagent ion corrected by their relative transmission (Tr) respectively, U_{drift}, T_{drift}, p_{drift} and τ are the voltage (V), temperature (K), and pressure (mbar) residence time (s) of the ions in the drift tube respectively, k the reaction rate (cm^3. s^{-1}) between the reagent ion and VOC.

A 12-component mix at ppmV level (Apel Riemer Environmental Inc, Miami, USA) in the range of 32–134 amu diluted in pure nitrogen was used to experimentally determine the relative ion transmission at the beginning and at the end of the measurement campaign.

Compound-specific proton-transfer reaction rate constants (k) were applied for VOCs where available in the literature (Cappellin et al., 2012). For all other species, a rate constant of 2 × 10^{-9} cm^3 s^{-1} was assumed; this is a common value used when a measured rate constant is unavailable (Cappellin et al., 2012; Wang et al., 2020, 2021). An additional correction was applied to ethanol using calibration coefficients obtained from post-campaign laboratory measurements, as significant fragmentation resulted in an underestimation of ethanol mixing ratios by a factor of 9 when default reaction rate constants were assumed.

Alongside the PTR-ToF-MS, two additional formaldehyde measurements were made using a MIRA Ultra Gas Analyzer (Aeris Technologies) and COmpact Formaldehyde FluorescencE Experiment (COFFEE) (St. Clair et al., 2017). Correlations between all three instruments were high (r^2 ranging from 0.69 and 0.81, Fig. S1) but the PTR-ToF-MS measurements were about four times lower than both the MIRA Ultra Gas Analyzer and COFFEE. This is because formaldehyde

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has a proton affinity close to water, so the back reaction of protonated formaldehyde with water vapor becomes relevant and reduces the sensitivity of formaldehyde in the PTR-ToF-MS (Vlasenko et al., 2010). Therefore, the formaldehyde measured by the PTR-ToF-MS was scaled up to match these instruments. Mixing ratios were corrected for background contribution using a cylinder of high purity nitrogen (99.999 purity).

Other non-VOC measurements made at the CTC site were used for comparison with the PMF results. PM_1 OA, sulfate, nitrate, and ammonium were measured by an Aerosol Mass Spectrometer (AMS). Black Carbon (BC) was measured by a Multi-Angle Absorption Photometer (MAAPS). Other CTC measurements include carbon monoxide (CO), SO_2, NO_X, ozone (O_3), temperature, wind speed and wind direction. Total ammonium was measured using a mist chamber coupled with ion chromatography (MC-IC) (Cofer et al., 1985), and total S(IV) was measured using a particle into liquid sampler coupled with ion chromatography (PILS-IC) (Orsini et al., 2003). Both MC-IC and PILS-IC methods have been discussed in other ALPACA papers (Campbell et al., 2024; Dingilian et al., 2024; Simpson et al., 2024).

The PTR-ToF-MS is unable to detect short-chain alkanes with less than C8 such as ethane, propane, and butane due to their low proton affinities. Very few works contain a comprehensive overview of both oxygenated VOCs and short-chain alkane VOCs in the US. The CalNex campaign, which used multiple instruments to measure both categories as described in Heald et al. (2020), found that short-chain alkanes made up on average 36.5% of the total measured $\mu gCsm^{-3}$, excluding organic aerosols. Heald et al. (2008) contains VOC measurements from other North American campaigns, although not all VOCs were measured for all campaigns. Ethane, propane, and butane range from 7.5-37.2% of the measured $\mu gCsm^{-3}$ for measurements done in the US and Canada, excluding organic aerosols. A direct comparison between these works and Fairbanks are difficult due to the unique climate of Fairbanks, but we expect short-chain alkanes and alkenes to be in a similar range.

2.2 Positive Matrix Factorization (PMF)

PMF is a mathematical model commonly used to estimate source contributions of VOCs datasets. Briefly, the PMF model uses a time series dataset matrix and corresponding error matrix as inputs. The user specifies a number of factors—in our case, the number of factors equates to the number of VOC sources. The PMF model attempts to split the main dataset into two matrices: a

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time series of factors, and the contribution of each VOC species to each factor. The user then interprets the resulting matrices to determine what source each factor corresponds to. More detailed information about the model can be found in literature (Paatero, 1999). We used SoFi Pro version 9 which utilizes the ME-2 engine (Canonaco et al., 2013; Paatero, 1999). Over 300 ions were included for PMF analysis. An error matrix was also calculated with the PeTeR tool by propagating uncertainties in ion counts and background signals, assuming Poisson counting statistics (de Gouw and Warneke, 2007). The data was down-weighted based on the recommendations of Paatero and Hopke (2003).

3 Results and Discussion

3.1 Overview of VOC Measurements in Downtown Wintertime Fairbanks

We first examine the campaign average for VOC measurements. The mean VOC mixing ratio during the campaign was 35.6 ppb. Here, for simplicity, we describe VOC composition for species which had a campaign average of 0.05 ppb or higher. This included 46 species which account for 33.6 ppb (94.3% of the total VOC mixing ratio). It should be noted that there were two periods of enhanced pollution during the ALPACA campaign, as described in Simpson et al. (2024). Figure S2 shows PM_1 (measured by aerosol mass spectrometer, described below) and temperature during the campaign, with the pollution events highlighted.

Figure 1 shows the percent contributions and campaign-mean VOC mixing ratios by functional group (see Table S1 for details). Alcohols, aromatics, carbonyls, acids, non-aromatic hydrocarbons, and nitrogen-containing species are the dominant VOC classes. Alcohols were by far the most prominent category due to the high mixing ratio of methanol (6.8 ppb) and ethanol (5 ppb) during the campaign. The species with the third largest mean mixing ratio was formaldehyde at 2.3 ppb. Common species from other functional group categories are (in decreasing order): toluene, C8 aromatics, benzene, and C9 aromatics for aromatics; formaldehyde, acetaldehyde and acetone for carbonyls; $C_2H_4O_2$ (acetic acid) and formic acid for acids. The speciation is similar with median values (Fig. S3).

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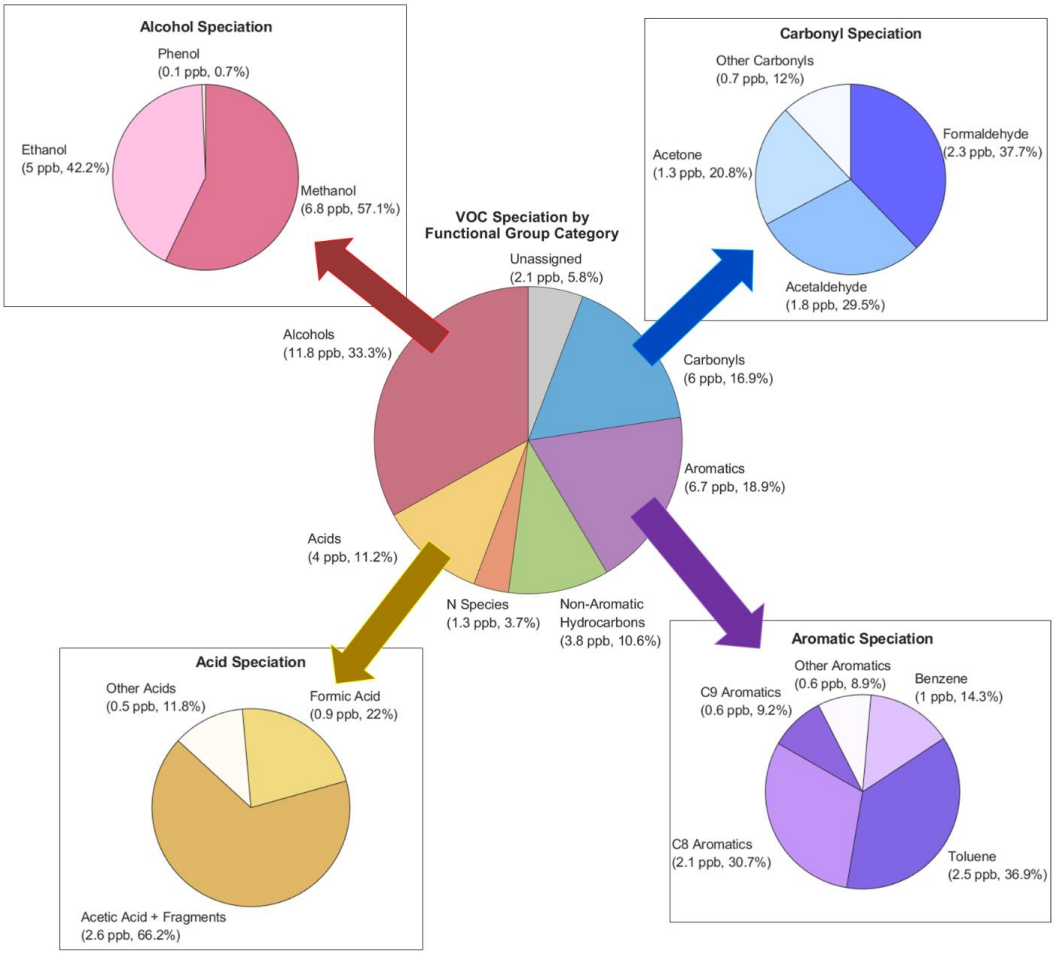

Figure 1. Mean composition of VOCs categorized by their functional groups during ALPACA campaign. The "Unassigned" group is made of up of about 290 species whose individual campaign means are all less than 0.05 ppb.

Methanol and ethanol are highest from Jan 21st to Jan 25th before the first pollution event, with methanol spikes up to 166 ppb and ethanol spikes up to 180 ppb. Interestingly, as shown in Table S2 and Fig. S4a, there was not much enhancement of methanol and ethanol during the cold polluted event compared to periods outside the pollution events, but they were significantly

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increased during the warm pollution event, with methanol spikes up to 133 ppb. Both are similar mixing ratios to measurements at the house site in Ketcherside et al. (2025).

The methanol in wintertime Fairbanks is on average 1.2-2 times higher than other urban wintertime measurements in the US and China (Li et al., 2019; Millet et al., 2005), and 2-6 times higher than measurements in Europe (Languille et al., 2020; Legreid et al., 2007; Patokoski et al., 2014). The large discrepancy of methanol between Fairbanks and Europe may be because the US and Canada use methanol-based windshield wiper fluid and Europe uses ethanol-based. Legreid et al. (2007) attributes the high ethanol mixing ratios in Zürich, Switzerland to windshield wiper fluid. Slowik et al. (2010) attributes high levels of methanol due to windshield wiper fluid in Toronto, Canada, but did not report a mixing ratio. We further discuss the influence of windshield wiper fluid in Fairbanks in Section 3.2. Ethanol in Fairbanks is between 1.2-5 times higher than measurements in the US and Europe (Gkatzelis et al., 2021; Legreid et al., 2007; Millet et al., 2005).

The aromatics were mainly made up of toluene (2.5 ppb) and C8 aromatics (xylenes and ethylbenzene, 2.1 ppb), benzene (1.00 ppb) and C9 aromatics (0.62 ppb). All species show enhancements during the polluted episodes, with similar mixing ratios during the cold and warm pollution events (Fig. S4b, Table S2). The mean aromatic mixing ratio in Ketcherside et al. (2025) is 11.9 ppb, almost double what is reported in this work. All aromatics are higher at the house site, with C8 aromatics having the largest discrepancy—nearly 3 times higher at the house site than downtown. This likely reflects a greater influence of heating oil combustion in residential areas. The mixing ratios of aromatics in this work are 2-12 times higher than those reported during other North American wintertime campaigns in urban locations like NYC, NY (Gkatzelis et al., 2021); Boulder, CO (Gkatzelis et al., 2021); Pittsburgh, PA (Millet et al., 2005); Houston, TX (Sadeghi et al., 2022); and Toronto, Canada (Rivellini et al., 2024). Benzene, toluene and C8 aromatic levels are comparable to urban and suburban wintertime measurements in China (Li et al., 2015, 2019).

The high aromatic mixing ratio in wintertime Fairbanks compared to other US cities is also likely due, in part, to differences in gasoline composition. Yano et al. (2016) measured the aromatic content of Anchorage, AK gasoline in 2013, after new gasoline regulations took place nationwide. They found benzene content was 1.3% v/v, and the total aromatic content (benzene, toluene, ethylbenzene, and xylene) was 25% v/v. This is more than double the average benzene content in

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gasoline in 2013 (0.6% v/v), and about 1.3 times higher than the total aromatic content (19% v/v) provided by the United States Environmental Protection Agency (https://nepis.epa.gov/Exe/ZyPDF.cgi?Dockey=P100T5J6.pdf). The relatively high aromatic content in Alaskan gasoline, combined with increased vehicle emissions at cold temperatures, leads to high gas-phase aromatic mixing ratios.

The greatest carbonyl species was formaldehyde (2.3 ppb), followed by acetaldehyde (1.8 ppb) and acetone (1.2 ppb). Nine other carbonyls had mean values between 0.05-0.17 ppb (Table S1). Formaldehyde, acetaldehyde and acetone are all enhanced during the pollution periods (Fig. S4c, Table S2). Formaldehyde is noticeably much higher during the cold event, whereas acetaldehyde is highest during the warm event. In contrast, measurements of formaldehyde (1.4 ppb) and acetaldehyde (1.3 ppb) at the house site are on average 40-60% lower compared to downtown measurements. Acetone has a similar mixing ratio range to other wintertime measurements in the US, European, and China, although acetaldehyde is 2-3x higher (Gkatzelis et al., 2021; Legreid et al., 2007; Li et al., 2019; Millet et al., 2005). In another study, formaldehyde, acetaldehyde, and acetone are in a similar range to wintertime non-haze periods in Beijing, and about half as much when compared to haze periods (Rao et al., 2016).

Acetic acid (1.7 ppb) and formic acid (0.9 ppb) are the two most prominent acid species. Although these compounds often spike at the same time, in many periods formic acid increases while acetic acid decreases (Fig. S4d, see Jan 28-29, Feb 3-4, Feb 21-23), and they are poorly correlated (Fig. S5), suggesting that they are likely arising from different sources, instead of a shared source like wood heating. Acetic acid at the house site is lower (1.3 ppb) compared to the downtown site, but formic acid is similar. Formic acid is unique in that it slightly lower during both pollution events compared to the clean periods (Table S2). This indicates a lack of primary sources of formic acid in downtown Fairbanks, and may be evidence of dark aging, which is discussed more in Section 3.2. Both formic acid and acetic acid are moderately lower than urban and suburban wintertime measurements in China (Li et al., 2019).

Mixing ratios of nonaromatic hydrocarbons and nitrogen species are generally low. The two most prominent nonaromatic hydrocarbon species are C_3H_6 (1.7 ppb) and C_5H_8 (0.4 ppb) (Fig. S6a), but these ions may also include fragmentation of larger VOCs (Coggon et al., 2024; Gueneron et al., 2015). Similarly, the most commonly measured nitrogen-containing ion was NO_2^+

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fragments (Fig. S6b), a common fragment for many organic nitrate species (Aoki et al., 2007; Hansel and Wisthaler, 2000).

3.2 Nine Factor PMF Solution

Nine factors were resolved from the PMF analysis. These factors are: gasoline, diesel, windshield wiper fluid, acetic acid, wood heating, heating oil, volatile chemical products (VCPs), vertical mixing, and aged air. Section S1 describes how VOC profiles, diurnal cycles, and correlation with external measurements were used to assign the PMF factors. A summary of the factors is shown in Fig. 2 and Table S3. A log-scale version of Fig. 2 is shown in Fig. S7, all species are shown in Fig. S8-S10, and time series of all factors is shown in Fig. S11. In the following, we define a VOC as negligible to a factor's composition when a VOC contributes less than 3% of its average campaign mixing ratio to a factor. The likely assignment of the VOC "$C_2H_4O_2$" is acetic acid, however we refer to it by its chemical formula to avoid confusion with the acetic acid factor.

3.2.1 Traffic Factors: Gasoline and Diesel

There are two traffic factors related to direct vehicle emissions—gasoline (6.4 ppb, 18.2%) and diesel (3.7 ppb, 10.6%). The gasoline factor is by far the largest source of aromatic species like benzene (65.8%), toluene (82.6%), C8 aromatics (74.1%), and C9 aromatics (63.4%). A little more than 5% each of formaldehyde, acetaldehyde, and acetone are explained by the gasoline factor. 6.9% of total benzene and 3.5% of total C9 aromatics are associated with the diesel factor, but toluene and C8 aromatics are negligible. The diesel factor is the largest primary source of formaldehyde, accounting for 30.8%; acetaldehyde and acetone are lower at 17.0% and 11.0%, respectively.

Both traffic factors are lower on weekends than during the weekdays, with diesel decreasing the most by nearly 50% (Fig. 2). This may indicate that heavy duty diesel vehicles that do not operate on weekends (school busses, public transportation, garbage trucks, etc.) account for much of the diesel emissions.

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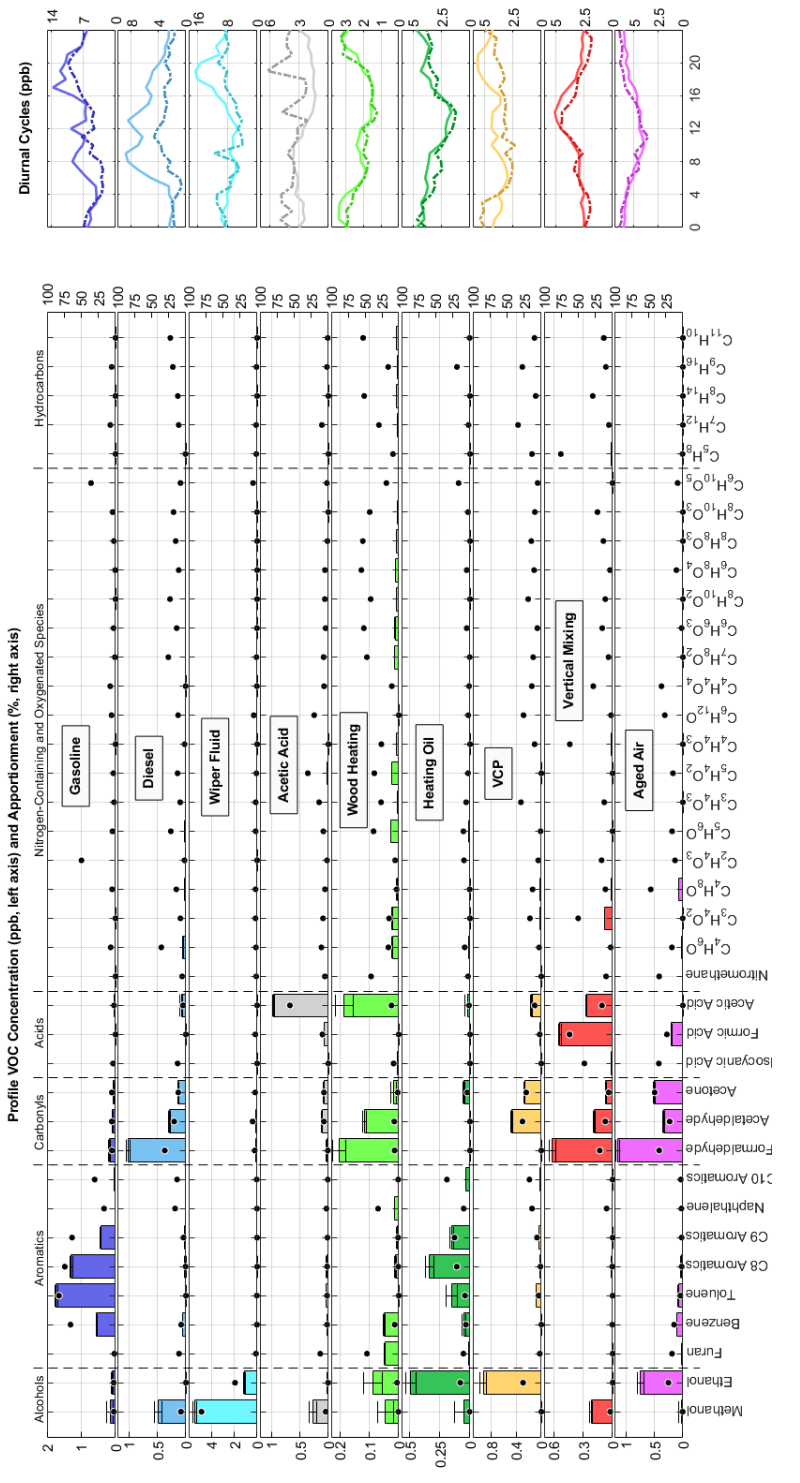

Figure 2. The left figures show the mixing ratio (left y-axis, ppb) and percent apportionment (right y-axis) of VOCs used to assign each factor. The x-label shows either the chemical name for species which are likely to be dominated by one isomer, or the chemical formula for species which are likely to have multiple isomers. The mixing ratio and percent for each species in each factor can be found in Tables S7 and S8, respectively. The right figures show the weekday (solid line) and weekend (dashed line) mean diurnal cycles of the factors.

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The gasoline and diesel factors do not have the same diurnal cycles, although this could be due to differences in cold start emissions between gasoline and diesel vehicles. The weekday and weekend diurnal cycles for each factor are shown in Fig. 2. The gasoline and diesel factors both increase early in the morning. Gasoline continues to increase until it peaks around 7 pm, whereas diesel starts to decrease around 2 pm. Since the site is downtown (as opposed to the residential areas outside of downtown), measurements in the morning will mainly be characterized by vehicles that have been started elsewhere and are driven into town, so the full impact of cold start emissions are not expected to be observed. After the typical workday is over, vehicles will be cold-started downtown, which would lead to an increase in gasoline vehicle emissions (Cao et al., 2024; George et al., 2015). However, diesel vehicle VOC emissions are less affected by cold starts overall compared to gasoline vehicles (Ferrarese et al., 2024; Marques et al., 2022; Wang et al., 2022), which leads to a much lower afternoon peak for diesel compared to gasoline. Some diesel vehicle emissions, especially carbonyls, can increase at low temperatures (Ferrarese et al., 2024; Hays et al., 2017; Liu et al., 2023; Suarez-Bertoa et al., 2022; Wærsted et al., 2022; Zhang et al., 2026), but to our knowledge emission measurements for VOCs diesel vehicles have not been carried out at the extremely low temperatures Fairbanks experiences.

3.2.2 Other Vehicle-Related Factors: Windshield Wiper Fluid and Acetic Acid

The windshield wiper fluid factor (7.2 ppb, 20.7%) is made up of 81.2% of the total measured methanol and 32.3% of the total ethanol. Contributions from other species are very small. The wiper fluid factor is the largest source of VOCs due to high mixing ratios of methanol measured during the campaign.

The acetic acid factor (2.4 ppb, 6.8%) may be related to vehicle emissions, as described in the SI. The acetic acid factor mainly consists of $C_2H_4O_2$ (acetic acid), and 56.1% of the total $C_2H_4O_2$ measured is attributed to this factor. It does not have a well-defined diurnal cycle, as it is affected by large spikes of $C_2H_4O_2$ which do not occur at regular intervals.

If the windshield wiper fluid and acetic acid factors are included with the gasoline and diesel factors, the total VOCs related to vehicles increases from 28.8% to 56.3%.

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3.2.3 Space Heating Factors: Wood Heating and Heating Oil

Two factors were related to residential heating: wood heating (2.0 ppb, 5.6%) and heating oil (3.1 ppb, 8.8%). Although wood heating and heating oil are the largest and second largest contributors to $PM_{2.5}$ mass, respectively (Nattinger, 2016), their contribution to total VOC mixing ratios are relatively small at 14.2% total.

Residential wood heating is a moderate source of small carbonyl species, associated with 6.2% of total formaldehyde and 6.7% of acetaldehyde. It is also associated with 5.9% of benzene, although other aromatics are negligible. Heating oil, which can be residential or commercial in downtown Fairbanks, is the second largest contributor to total aromatics, including 19.3% of C8 aromatics and 22.6% of C9 aromatics. The wood heating factor is 6.1 times higher in the cold polluted period than the warm polluted period, a large increase in wood burning at these extremely cold temperatures.

Heating oil is associated with no formaldehyde or acetaldehyde, and 4.1% of the total acetone. The lack of formaldehyde assigned to the heating oil factor may be because heating oil boilers operate without after-treatment systems. In contrast, diesel vehicles can produce higher formaldehyde emissions due to oxidation of unburned fuel in their exhaust after-treatment systems, especially under cold conditions (Suarez-Bertoa et al., 2022).

3.2.4 Volatile Chemical Products (VCP) Factor

The VCP factor (3.1 ppb, 8.9%) is a minor contributor to VOCs in Fairbanks, in contrast to other US cities like LA, NYC, and Boulder, where it is becoming a main anthropogenic source of VOCs; McDonald et al. (2018) and Gkatzelis et al. (2021) find that the ratio of vehicle emissions to VCP emissions is between 0.3-1.4. In Fairbanks, we find that vehicle emissions are 3.2 times higher than VCPs when looking at only the combined gasoline and diesel factors, and 6.3 times higher when including the windshield wiper fluid and acetic acid factor. The relatively high contribution of VOCs related to vehicles is likely due to the increase in both gasoline and diesel vehicle VOC emissions at low temperatures (Cao et al., 2024; Ferrarese et al., 2024; George et al., 2015; Hays et al., 2017; Liu et al., 2023), and decreased volatility of VCPs at cold temperatures leading to less off-gassing. Overall, vehicle emissions remain the dominant source of VOCs in wintertime Fairbanks.

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3.2.5 Vertical Mixing Factor

The vertical mixing factor (2.8 ppb, 7.9%) describes air masses that have been transported from outside downtown Fairbanks and have undergone secondary chemistry. 18.7% of total formaldehyde, 10.7% of acetaldehyde, and 5.5% of acetone are associated with the vertical mixing factor. Benzene, toluene, C8 aromatics and C9 aromatics are all negligible. This factor is the largest source of formic acid, accounting for 62.8%. The large fraction of formic acid, formaldehyde (18.7%), and maleic anhydride (64.3%), combined with the factor's correlation to ozone, may be indicative of secondary formation via ozonolysis and/or NO_3 radicals (Eliason et al., 2003; Li et al., 2023; Paulot et al., 2011). The vertical mixing factor explains much of the observed $C_3H_4O_2$ (50.3%), $C_2H_4O_3$ (16.1%), $C_3H_4O_3$ (75.6%), $C_4H_4O_3$ (62.5%), $C_5H_6O_3$ (28.9%), $C_4H_4O_4$ (28.2%), and $C_5H_6O_4$ (29.7%). These species are associated with aged residential biomass burning plumes (Coggon et al., 2019; Li et al., 2023) and decomposition of aromatic VOCs (Lannuque et al., 2023; Mehra et al., 2020). And, as mentioned earlier, formic acid mixing ratios are lower during both polluted events with strong surface inversions, indicating few primary sources of formic acid in downtown Fairbanks. As discussed in Brett et al. (Brett et al., 2025), residential space heating emissions are emitted above the surface. Thus, wood heating plumes may be transported aloft from areas with prominent wood heating emissions, during which longer chain VOC species are oxidized by ozone and/or NO_3 radicals, and break down into smaller chain VOCs, producing formaldehyde and especially formic acid. A similar $OVOC_{Ox}$ factor was observed in Wang et al. (2024). It had similarities to the time series of O_3+NO_2, contributions from low mass oxidized compounds like formic acid, and high contribution from $C_XH_YO_3$ ions, which they relate to daytime photochemistry.

We further evaluate the vertical mixing factor by comparing it and ozone to wind speed and direction. Ozone, as shown in Fig. S12, increases significantly when wind speed is greater than 3 mph. These higher wind speeds coincide with weakly stable periods and lower NO_X, and more vertical mixing, as discussed in Brett et al. (2025). In contrast, strongly stable stagnant conditions correspond to low wind speeds, and are associated with high NOx which titrates ozone. At the surface, during periods of higher wind speeds, air masses mainly come from 0-100° (northeast to east direction) or 210-280° (southwest to west). However, as shown in Fig. S12, the vertical mixing factor (at the surface) only increases when wind directions are from the east (80-

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100°). This shows that not only wind speed, but also the wind direction matters for the vertical mixing factor. Figure S13 show the high wind speed vectors overlaid on a map of Fairbanks, with points colored by the mixing ratio of the vertical mixing factor. We can see that periods associated with a higher mixing ratio come from Fort Wainwright and the northern part of the town Badger. It is also possible that plumes from the nearby town of North Pole, which has greater use of wood heating compared to Fairbanks (Nattinger, 2016), are included in these air masses. Higher wind speeds from less populated areas like Steele Creek and Ester have lower mixing ratios.

3.2.6 Aged Air Factor

The aged air factor (4.4 ppb, 12.6%) contains relatively stable products of atmospheric reactions involving a wide range of VOCs, and accounts for 21.2%, 34.7%, 19.4%, and 41.2% of the variability of ethanol, formaldehyde, acetaldehyde, and acetone, respectively. Notably, it is also the factor with the second largest amount of formic acid (23.5%), which we associate with secondary chemistry. It has a low contribution of markers of primary emissions, does not correlate well with any usual tracers or external measurements, and has small diurnal variability. This indicates that this factor is secondary in nature, representing the regional background. It has a low contribution from primary emission markers (12.7% of benzene and 3.7% of toluene is related to this factor, but other aromatics are negligible).

3.2.7 Factor Summary

According the PMF result, vehicle-related factors are the largest source of VOCs in downtown Fairbanks. Gasoline vehicles are by far the largest source of all major aromatic species like benzene, toluene, C8 aromatics, and C9 aromatics. A smaller but still significant fraction is attributed to heating oil, especially C8 (19.3%) and C9 aromatics (22.6%). Sources of carbonyl species are more variable. Diesel is the largest primary source of formaldehyde (30.8%). The vertical mixing and aged air factors account for about 53% of the formaldehyde, which may be a mixture of primary and secondary sources. The three largest sources of acetaldehyde are (in decreasing order) VCPs, aged air, and diesel. The three largest sources of acetone are (in decreasing order) aged air, VCPs, and diesel.

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3.3 Comparison with Other PMF Work in Fairbanks

Ketcherside et al. (2025) also used PTR-ToF-MS data in Fairbanks to conduct PMF of VOC measurements in Fairbanks, but there are a few main differences. First, their instrumentation was located in a residential area 2.6 km away from the CTC site. Therefore, their analysis is likely more influenced by residential factors, like space heating. Second, in their PMF analysis they included species that were measured at the house site by instruments besides PTR-ToF-MS, like the gas-phase species CO and NO_X, as well as aerosol-phase PM_1 species like BC, sulfate, nitrate, and ammonium. They also include SO_2 measured at the CTC site. In our analysis, we only use PTR-ToF-MS data as inputs for PMF analysis, but we do compare the PMF outputs to the corresponding CTC measurements of these species. Third, their analysis is based on hourly measurements, while ours is based on 2-minute data. They also use the EPA PMF 5.0 software while we use SoFi Pro V9; however, both are based on the ME-2 engine (Canonaco et al., 2013; Norris et al., 2014) so the PMF procedures should be largely similar. More information on the differences between these two sites and the instrumentation at the house site can be found in Simpson et al. (2024) and Ketcherside et al. (2025).

Despite the different methodology, there are many similarities between our work and the results in Ketcherside et al. (2025). Both studies found factors related to vertical mixing (which they call ozone), aged air, heating oil, wood heating (i.e., biomass burning), and traffic. Their study identifies two biomass burning factors (one for short-lived VOCs and one for long-lived), while we only have one. We find two traffic factors, while in their six factor solution they only find one. Their traffic diurnal cycle is most similar to our gasoline factor. We find the wiper fluid, VCP, and acetic acid factor, which they did not. They found that methanol was largely associated with traffic emissions, which is similar to our assignment of methanol to windshield wiper fluid. They also did not observe the large spikes of $C_2H_4O_2$ that we find, which helps to confirm that our analysis of $C_2H_4O_2$ are local to the CTC site. They mainly attribute $C_2H_4O_2$ and formic acid to biomass burning, while find that much of the $C_2H_4O_2$ is in its own factor, and formic acid is mainly attributed to the vertical mixing factor. Figure S14 presents pie charts of the PMF factor contributions from our work and Ketcherside et al. (2025) It is notable that wood heating and heating oil are a significantly smaller fraction in our work compared to theirs. Figure S15 can be used for further comparison of PMF factors with their Fig. S5.

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Both our study and Ketcherside et al. (2025) find traffic to be the largest source and heating oil to be the second largest source of aromatic VOCs. However, in our results the gasoline factor dominates, making up 75% of the total benzene, toluene, C8 aromatics, and C9 aromatics, while heating oil only makes up 13%. In Ketcherside et al. (2025), gasoline makes up 48% and heating oil makes up 39%. This is large difference likely due to the increased influence of space heating at the residential site compared to the CTC site.

Ketcherside et al. (2025) attributes 18% of formaldehyde to heating oil, whereas we do not see any formaldehyde associated with heating oil. Here we describe some possible reasons for this discrepancy. This difference could be due to the different time resolution used in our works. The measurements in Ketcherside et al. (2025) were averaged to one hour, while our measurements are every two minutes. Heating oil and diesel fuel are compositionally similar but the combustion process is different, so the emissions are comparable but not identical. Our two-minute data may be able to resolve the differences between heating oil and diesel emissions more effectively. Another reason is that since their work was conducted at a residential site with less traffic than our CTC site, diesel emissions are expected to be much smaller and thus may be more difficult to distinguish from heating oil. Since the CTC site is much more dominated by traffic emissions, the variation of formaldehyde related to heating oil may be difficult to distinguish from other factors. Qualitatively, their heating oil factor contains characteristics of both our diesel and heating oil factors—for example, a 5 am increase similar to our diesel factor, and an afternoon increase present in our heating oil factor.

Our wood heating factor appears to be most comparable to the short-lived biomass burning factor in Ketcherside et al. (2025), including similarities between the furan, methylfuran, furfural, and methylfurfural assignments, and their diurnal cycles (Fig. 2, Table S4). Meanwhile, our vertical mixing factor has similarities to the chemical composition of their long-lived biomass burning factor (notably formic acid and formaldehyde, as well as furans and other aldehydes, Table S4) but the diurnal cycle of the vertical mixing factor is closer to their ozone factor (diurnal cycles). The discrepancy between chemical composition and diurnal cycles may be because 98% of ozone gas was applied to their ozone factor. Although our vertical mixing factor is correlated with ozone gas, there are also periods where ozone gas and the vertical mixing factor are not in agreement

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(Fig. S12). Their ozone factor describes the transport of background levels of ozone from all wind directions, but our vertical mixing factor is highest when wind direction is from the east.

We cannot compare the source apportionment of species like CO, NO_X, BC, and PM_1 sulfate because we used these species as external validation to compare with the PMF results. However, we do find that CO, NO_X, and BC are well-correlated with traffic (Fig. S16 and S17), which is consistent with these species being associated with their traffic factors.

Ijaz et al. (2024) conducted PMF in Fairbanks using PM_1 measurements with both a PTR-ToF-MS with CHARON inlet and an AMS. We focus on their PTR-ToF-MS results here since that is the same instrument used in our analysis. They found four residential heating-related factors, including three biomass burning factors and one heating oil factor, which contribute on average 2.6 $\mu g/m^3$ (62%) of total measured OA. Although this is a much larger fraction of OA mass compared the 14% of total VOCs we attribute to residential heating, the absolute mass is much lower than our wood heating (8.6 $\mu g/m^3$) and heating oil (12.1 $\mu g/m^3$) factors. Ijaz et al. (2024) contribute about 1.6% (0.06 $\mu g/m^3$) of measured OA to traffic, significantly less than the 29% (36.6 $\mu g/m^3$) of VOCs that we contribute to the gasoline and diesel factor. They also identify a cooking factor, which we did not see resolved in our PMF analysis. This comparison shows major differences between the sources of PM_1 OA and gaseous VOCs. Comparison of VOC and aerosol partitioning is also explored in Ijaz et al. (2025).

3.4 PMF Comparison with Mist Chamber Total Ammonium and PILS S(IV)

High concentrations of HMS and related aldehyde-S(IV) species are a unique feature of Fairbanks wintertime $PM_{2.5}$ (Campbell et al., 2022, 2024; Dingilian et al., 2024; Holen et al., 2025; Moon et al., 2023) and so are investigated more here. The MC-IC measures gas-phase and $PM_{2.5}$ aerosol-phase species. This is important for ammonium, which could have a significant portion in the gas-phase. In extremely cold environments like Fairbanks, high concentrations of total ammonium can increase aerosol pH to 4-5, greatly facilitating secondary aqueous chemistry of species like HMS and sulfate (Campbell et al., 2024). We compare MC-IC total ammonium (TA) measurements to the PMF factors in order to help determine the sources of TA.

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Gasoline vehicles may be a large source of total ammonium in wintertime Fairbanks. We find that TA correlates best with the gasoline factor (R^2 = 0.43), followed by the VCP factor (R^2 = 0.38) as shown in Fig. 3. TA also has a similar diurnal cycle to gasoline (Fig. S18), and is correlated with external factors CO (R^2 = 0.47), NO_X, (R^2 = 0.42), and somewhat with BC (R^2 = 0.36), which are commonly associated with vehicle emissions (Fig. S19). Vehicle emissions have also been shown to be a large source of ammonia (Ji et al., 2025). Therefore, a large fraction of TA may be coming from gasoline vehicle emissions. VCPs may also be a large source of TA, but since VCPs include a broad category of industrial and personal care products, it is more difficult to pinpoint exact sources.

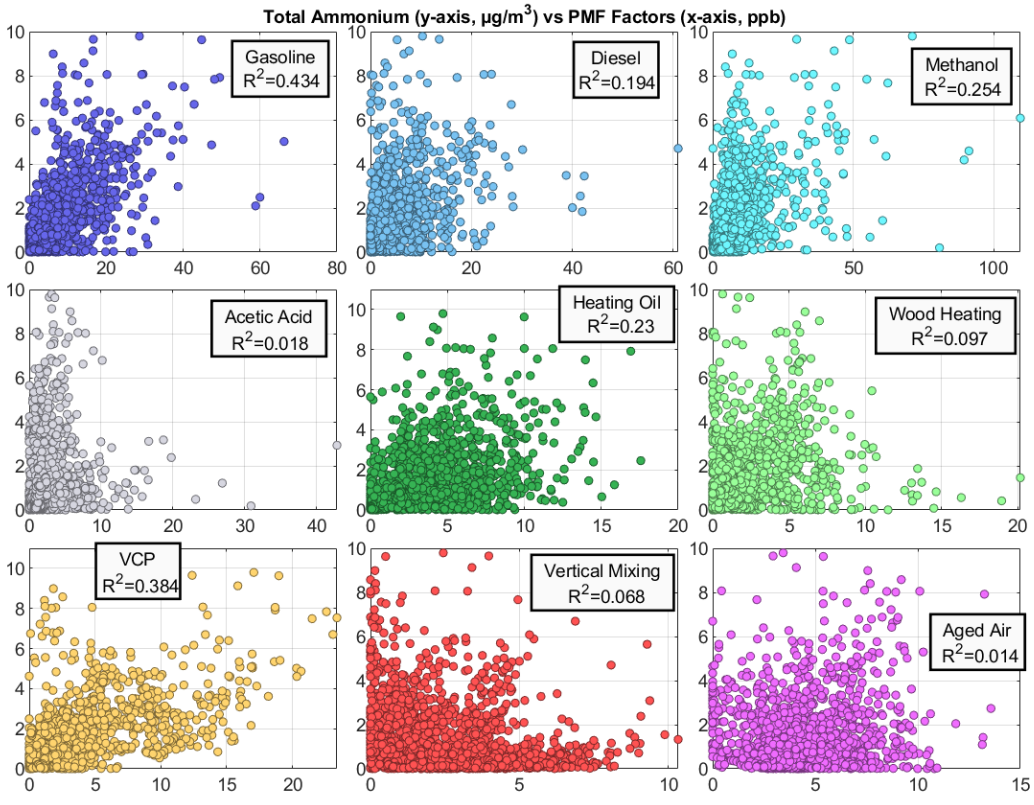

Figure 3. Correlation of total ammonium (y-axes) with the nine PMF factors (x-axes).

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HMS formation may be highly dependent on ammonia emissions (because of its effect on aerosol acidity) from gasoline vehicles and formaldehyde emissions (an HMS precursor) from diesel vehicles. PILS-IC total S(IV) correlates best with diesel (R^2 = 0.40), followed by wood heating (R^2 = 0.25) and gasoline (R^2 = 0.24), as shown in Fig. S20. The total S(IV) during the ALPACA campaign was found to be mostly HMS in Dingilian et al. (2024) and Holen et al. (2025). Total S(IV) has a diurnal cycle most similar to diesel during weekdays and most similar to gasoline on the weekends (Fig. S21). Furthermore, total S(IV) has a similar diurnal cycle to formaldehyde on weekdays and NH_X on the weekends (Fig. S22). Although SO_2 gas is a precursor to HMS, there are few similarities to the S(IV) and SO_2 diurnal cycles (Fig. S22), which may be because SO_2 gas (campaign mean 9.6 ppb) is often much higher than formaldehyde gas (campaign mean 2.3 ppb), making formaldehyde mixing ratios more often the limiting factor in HMS formation (Dingilian et al., 2024). The diesel factor is not the largest source of formaldehyde, but formation of HMS related to the gasoline and diesel factors is likely helped by both factors being emitted in the same place, and the mixing of air that occurs from moving vehicles.

Since the formation of HMS and other S(IV) species are secondary reactions, many other factors need to be considered, and this is not a complete picture of what drives aqueous S(IV) chemistry in Fairbanks. However, this first look seems to indicate that vehicle emissions are involved in S(IV) formation.

4. Conclusions

In this work, we use online PTR-ToF-MS measurements at one minute time resolution to characterize VOCs in downtown Fairbanks during the winter 2022 ALPACA campaign. We combine these measurements with PMF analysis to examine the sources of VOCs. We further compare these sources with other complementary measurements, like total ammonium and S(IV), to help understand how VOCs can contribute to secondary aerosol formation in extremely dark and cold environments like Fairbanks.

High mixing ratios of many hazardous VOCs like formaldehyde, benzene, and toluene were measured in wintertime Fairbanks, with mixing ratios comparable to and often exceeding

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measurements in other US cities. We attribute 29% of total VOC emissions to vehicle emissions and 56% to vehicle-related factors overall, with the largest factor being windshield wiper fluid. Unlike many other US cities, VCPs are a relatively minor source of VOCs. High mixing ratios of aromatics were measured and a majority of all prominent aromatic species (benzene, toluene, C8 aromatics, C9 aromatics) were attributed to gasoline vehicles, followed by heating oil as the second largest aromatic factor. Sources of various carbonyls are more complex. Large portions of formaldehyde can be attributed to diesel vehicles, wood heating, vertical mixing, and possible secondary chemistry; other species like acetaldehyde and acetone can be attributed to diesel, vertical mixing, and volatile chemical products (VCPs).

In both our work and Ketcherside et al. (2025), a significant amount of aromatic VOCs were assigned to heating oil factors. This is in contrast to the Alaska Department of Environmental Conservation's emission inventory for ALPACA, which contributes little to no aromatic species to heating oil, as described in Zhang et al. (Zhang et al., 2026). The results of these works show a need to update the emission inventories of heating oil emission to include contributions from aromatic species.

We hypothesize that particulate S(IV) formation may be largely linked to vehicle emissions. Total ammonium, which allows for moderate aerosol pH in wintertime Fairbanks, is best correlated with the gasoline factor. S(IV) (largely made up of HMS) is best correlated with the diesel factor, which is the largest primary source of formaldehyde (an HMS precursor), and S(IV) has a similar diurnal cycle to both gasoline and diesel vehicles. Reductions in vehicle emissions could lead to a reduction in both harmful VOC species and secondary $PM_{2.5}$ formation, in Fairbanks as well as other cold and dark urban environments. This is somewhat limited by lack of ammonia measurements; simultaneous VOC, ammonia, and S(IV) measurements combined with pH modeling would help confirm these results.

Data Availability Meteorological data used in this study are available at https://doi.org/10.18739/A27D2Q87W. MC-IC, PILS-IC, and COFFEE HCHO data is available at https://doi.org/10.18739/A2VD6P65Q. PTR-ToF-MS data will be made available at https://arcticdata.io/catalog/portals/ALPACA/Data before publication.

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Author Contributions SoFi PMF analysis was performed by JRC and BT-R. JRC, BT-R, B'DA, KL, RJW, KSL, MB, KKD, JMSC, and JM contributed to data collection and data analysis. JRC prepared the manuscript with key contributions from BT-R, BD'A, KL, JM, and contributions from all coauthors.

Competing Interests The authors declare that they have no conflict of interest.

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