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Multiyear characterization of volatile organic compounds in Zurich: evidence of urban volatile chemical products

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Abstract

Volatile organic compounds (VOCs) are important precursors of ozone and secondary organic aerosols. Here, we present a multiyear analysis of VOC composition and sources at an urban background site in Zurich in Switzerland, based on continuous hourly measurements of non-methane hydrocarbons (NMHCs) and oxygenated VOCs (OVOCs) in 2016, 2017 and 2024.

Between 2016 and 2024 the total concentration of VOCs remained relatively stable, while their composition shifted towards a stronger dominance of OVOCs. Source apportionment using positive matrix factorization (PMF) identified six main factors: road transport, residential heating and wood burning, propane and butanes, volatile chemical products (VCPs), local emissions, and urban background and natural gas. The relative contributions of these sources remained stable between 2016–2017 and 2024. VCPs, mostly explained by OVOCs, contributed 11–20% of total VOCs, showing their importance as a persistent urban source. A comparison with previous studies conducted at the same site in 2005–2006 shows a strong long-term decrease in traditional anthropogenic sources such as traffic and solvent use, consistent with emission inventory trends, and a dominant fraction of ambient VOCs related to VCPs, confirming the need for OVOC measurements in air quality monitoring networks, in line with the new European AAQD (2024/2881).

Introduction

Characterizing sources of volatile organic compounds (VOCs) in urban ambient air is essential for understanding their role in the formation of tropospheric ozone and secondary organic aerosols (SOA). Development of sensitive analytical methods and long-term monitoring have shown that oxygenated VOCs (OVOCs) including alcohols, ketones and aldehydes, are among the most abundant VOCs in urban ambient air, with ethanol often being the dominant compound [1–3]. The growing importance of OVOCs is reflected in the new European Union Air Quality Directive (Directive 2024/2881, 2024) which recommends monitoring an extended list of VOCs including non-methane hydrocarbons (NMHCs), OVOCs and terpenes. Despite their importance, long-term measurements of OVOCs remain limited to only a few European cities such as Zurich, Birmingham, Barcelona and Paris [4] and recently the French air quality network (7 urban and rural sites) due to the analytical challenges associated with their quantification.

Urban VOCs are emitted from a large variety of sources, including car exhaust, residential heating, solvent use, industrial activities and consumer products [5–9]. Recently, in the United States, volatile chemical products (VCPs), including solvents, personal care products and other fugitive emissions, have been identified as dominant contributors to VOC emissions [8, 10]. Nevertheless, many European source apportionment studies still rely primarily on NMHC measurements, which may underestimate the contribution of OVOC sources such as VCPs. In such studies, the contribution of solvent-related emissions was estimated to account for 11–26% of total NMHCs or was not clearly identified [7, 11–13].

Positive Matrix Factorization (PMF) is a receptor-based approach widely used for the source apportionment in studies of ambient VOCs [14–16] and can separate complex mixtures of emissions and quantify the contribution of individual sources. However, PMF studies often focus on NMHCs alone. The

most reactive species, e.g. alkenes and some aromatics, may be underestimated in the chemical fingerprint of a source depending on the photochemical aging of the air masses prior to sampling. OVOCs are both directly emitted and formed secondarily through atmospheric oxidation processes. The inclusion of OVOCs to PMF analysis therefore provides additional information on source contributions and atmospheric processing allowing better VOC source constraints.

The Zürich Kaserne urban background station, part of the Swiss National Air Pollution Monitoring Network (NABEL), offers the possibility to investigate long-term changes in VOC sources. There, VOC measurements have been conducted for several decades, the first OVOC dataset was measured in 2005 and since 2014 continuous OVOC measurements are conducted. Previous studies at this site identified road transport as the dominant VOC source [17] and the importance of local sources, including combustion processes to OVOC emissions [18]. For some OVOCs like acetaldehyde, their origin was mainly attributed to large-scale background although secondary formation could not be excluded [18]. More recent observations based on proton-transfer reaction time of flight mass spectrometry found lower traffic contribution and the importance of OVOC sources indicating a shift of the urban VOC mixture [3]. In parallel, various directives have been introduced in Switzerland aiming to reduce VOC emissions such as the solvent incentive fee introduced in 2000 and the ban of various chemicals, including some VOCs with the Chemical Risk Reduction Ordinance in 2005 [19]. More recently, local strategies, including the city of Zurich's climate and air quality plans (e.g. net-zero targets by 2040), are expected to further influence the composition of urban VOC emissions in the future.

In this study, we present a PMF analysis based on continuous hourly VOC measurements collected at the Zürich Kaserne urban background site in 2016, 2017, and 2024. The dataset combines a broad range of NMHCs and OVOCs measured using thermal desorption gas chromatography with flame ionization detection (TD-GC-FID). The objectives of this work are to (i) characterize the major sources of VOCs in Zurich, (ii) investigate their seasonal and interannual variability, (iii) assess how source contributions have evolved between 2016–2017 and 2024, and (iv) compare the results with previous source-apportionment studies conducted at the same site. This analysis provides new insights into the evolving composition and sources of urban VOCs and highlights the importance of including OVOCs in long-term monitoring.

2 Material and methods

2.1 Measurement site and (O)VOC sampling

Zürich Kaserne station (47.378°N, 8.530°E; 409 m a.s.l.) is located at Kasernenareal, a public backyard in the city center of Zurich. This NABEL station is representative of an urban background site. Sampling campaigns for NMHC measurements, ranging from a few months to several years, have been conducted at this station since 1993, with OVOC measurements measured continuously during a 1-year campaign in 2005. Continuous sampling of VOCs at 1-hour resolution was further conducted between January 2016 and December 2017 and between January 2024 and December 2024, using a preconcentration unit coupled to a TD-GC-FID. A detailed list of the VOCs measured during these sampling campaigns with their corresponding yearly average concentrations is given in Table S1. Ancillary parameters i.e., NO_x, CO,

temperature, wind speed and direction, and total VOC concentrations are available at NABEL [20]. Total VOC concentration (ppb C) refers to the difference between total hydrocarbon and methane concentrations measured simultaneously at the station with dedicated FID instruments (APHA-360 and APHA-370; HORIBA, Ltd.). As the carbon contribution of the OVOCs to total VOC is lower than 1:1 due to the presence of oxygen, the same effective carbon number than the GC-FID's response is utilized to calculate their contribution to total VOC in ppb C.

For the analysis, ambient air is drawn from a glass line with a flow of 2000 L h^{-1}, at 15 mL min^{-1} for 20 minutes via a sampling line (PFA, 1/4" diameter, 4 m long) equipped with a particle filter (5 µm). The sampling line is continuously flushed at 300 mL min^{-1} to ensure a representative sampling. Then, the sampled air passes through a glass trap maintained at -43°C to remove residual water. The VOCs are then trapped onto a multi-sorbent trap (Ozone Precursors, Markes International) maintained at -30°C. Subsequently, the trap is heated to 250°C for 5 min and the desorbed VOCs are transferred to the GC. Two GC capillary columns are installed with distinct FID detectors for the simultaneous detection of the non-methane hydrocarbons (NMHC), i.e., alkane, alkene and alkyne classes, and the OVOCs utilizing a Deans Switch with a DB-1 restrictor column (2.48 m x 0.25 mm). OVOCs are separated utilizing an OxyPLOT column (30 m × 0.53 mm), while the NMHCs are separated on a PLOT column (Al_2O_3, Na_2SO_4; 50 m × 0.53 mm). First, all the analytes are transferred to the OxyPLOT before NMHCs reach the PLOT column. Calibrations are performed monthly, utilizing a 4 ppb 30-compound NMHC standard in nitrogen certified from the National Physical Laboratory (NPL). If no standard is available for the compound of interest, e.g., terpenes and OVOCs, its concentration is calculated according to the response factor of benzene in the calibration tank and to its effective carbon number (ECN) given in Sternberg et al. [21]. Since 2024, the recovery of the OVOC measured by the GC-FID system has been investigated monthly by diluting a newly acquired NPL standard containing OVOCs (1 ppm) with nitrogen 6.0 at a dilution ratio of 1:100 and saturated with ultra-pure water. Monthly quality assurance and quality control (QA/QC) process following the GAW and ACTRIS guidelines [22] is then conducted on the datasets to remove outliers due to potential contamination (e.g., maintenance of the backyard utilizing motorized lawnmowers or instrumental issues). The uncertainty estimation was also assessed following the GAW and ACTRIS guidelines [22]. The validated datasets can be accessed through the EBAS database at https://ebas.nilu.no/data-access/.

2.2 Source apportionment by positive matrix factorization (PMF)

2.2.1 PMF model description

For the purposes of this study, the Environmental Protection Agency's PMF 5.0 model was employed [23]. The dataset has been separated by seasons for each year of the dataset for a total of 10 seasons. Spring 2017 and autumn 2024 were excluded from the analysis due to insufficient data to produce robust results. Details about the method are available in Dufresne et al. [7] where the method used was similar.

2.2.2 Dataset missing values and limit of detection

The presence of missing data in a dataset can be attributed to various factors, including calibration and blank measurements, instrumental issues or data invalidated during the QA/QC process. To conduct the PMF analysis, the input matrix cannot contain missing values. Therefore, missing values were replaced by the hourly median of the month and their associated uncertainties with the hourly median of the month multiplied by 4. For values of a given compound below the limit of detection (LoD), they were replaced by the LoD of the corresponding compound, divided by two. In addition, the associated uncertainties were equivalent to $\frac{5}{6}\ \mathrm{of\ the\ LoD}$.

2.2.3 Determination of the optimal results and associated uncertainties

For each season in 2016 and 2017, a total of 20 base runs were applied, with the number of factors ranging from 3 to 12, similar to Dusfrene et al. [7]. The factors for the 2024 dataset were determined according to the optimized runs of 2016 and 2017 with source factors varying from 5 to 7 factors per season for the base runs. VOCs that are poorly resolved during a season are subsequently dismissed: i.e., methanol and acetone in winter 2017; m,p-xylenes and o-xylene in summer 2017. The VOCs were then classified as strong, weak or bad, based on the value of their signal-to-noise ratio (S/N) [23], which have an impact on how strongly VOCs will influence the PMF solution. Species classified as bad are not considered in the PMF analysis such as α-pinene for all seasons of 2016 and of 2017, methanol, acetone and acetonitrile for winter 2017 and xylenes in summer 2017. A total of six factors were identified for all the 2016 and 2017 seasons, except for the winter 2017 season, for which only five factors were identified. Seven factors were identified in winter, spring and summer 2024. This additional, well-isolated factor explains the majority of the measured propanols and is potentially linked to a pollution due to the use of a butanol-based Condensation Particle Counter (CPC) at the measurement station. Therefore, this factor is not considered further in this study.

Subsequently, the potential origins of each identified factor were addressed following their chemical fingerprint, their diurnal and seasonal variability and their correlation with other pollutants. Finally, the FPeak, explained in Paatero et al. [24] was evaluated to determine if there was any rotational ambiguity on the base run solution and if the base run solution can be optimized. In conclusion, the bootstrapping (BS) method was employed to assess the final solution's stability. These methods are employed to evaluate rotational ambiguity and the effect of random errors on the PMF solution. Further details on these methods can be found in Brown et al. [25] and in Paatero et al. [24, 26].

Given the substantial background levels and occasional high peaks in ethanol concentration, i.e., hundreds of ppb, caused by nearby activities like summer parties, where large quantities of alcoholic drinks were consumed, ethanol was not incorporated into the final PMF input data. Consequently, this compound has been utilized as ancillary data. A comprehensive overview of all quality indicators can be found in Table S2 including $Q_{robust}/Q_{expected}$, the r^2 model vs measurement, the number of species with a $r^2 > 0.75$ for model vs measurement and the FPeak %dQ which is the difference between the Q value of the base run and the Q value of the FPeak run (in %)

3 Results and discussion

3.1 General overview of the Zurich station, meteorological and VOC variability between 2016–2017 and 2024

3.1.1 Mobility and energy use in Zurich city

Overall, the primary energy balance in Zurich City is dominated by electricity (42%), followed by residential heating (37%) and the use of combustible fossil fuel (21%) [27]. The means of transport in Zurich is dominated by public transport (41%) with electrified trams and trolleybuses representing 80% of the public transport volume. Car passengers remain an important component of the urban mobility (25%) with gasoline cars representing a major component of the fleet (89% in 2005; 67% in 2016 and 56% in 2024) with a progressive electrification of the fleet with the use of hybrid (11%) and electric cars (5.5%) in 2024 [28]. Additionally, emissions of ethanol and its oxidation products, e.g., acetaldehyde and formaldehyde [29–32], are expected from fuel emissions as gasoline is mixed with up to 5%v/v of ethanol and coupled with increasing production of bioethanol since 2005 in Switzerland [33]. Residential energy use is strongly influenced by natural gas (47%), followed by heating oil (21%), district heating (19%) and heat pump (12%) while wood represents only 1% in 2024. Heat pumps are becoming increasingly common in newer buildings (53%) [34]. Road transport and residential heating are thus expected to contribute to urban VOC concentrations while the progressive change in mobility and residential heating systems may influence the chemical fingerprints over time.

3.1.2 Meteorological conditions, air quality parameters and VOC variability between 2016–2017 and 2024

Meteorological conditions were comparable between 2016, 2017 and 2024 with only slightly warmer winter temperatures in 2024 (Fig. S1). NO_x concentration decreased by 50% between 2016 and 2024 with an annual concentration of 23 ppb in 2016 and 12 ppb in 2024. O_3 did not significantly change over the sampling years with seasonal average concentration varying between 13 and 36 ppb. Yearly CO concentrations significantly decreased by 32% between 2016 and 2024 with season average concentrations ranging between 0.18 and 0.32 ppm in 2016 and between 0.12 and 0.21 ppm in 2024. These observations suggest that change in VOC composition and source contribution are unlikely driven by meteorological differences between years.

Although total VOC concentrations measured by NABEL decreased by 17% from 77 to 64 ppb C between 2016 and 2024 (Fig. 1, a), the summed concentration of VOCs quantified by GC-FID did not significantly change with 25 ppb in 2016 and 2024 (Fig. 1, b). OVOC class was the most abundant class measured in Zurich, contributing with a yearly average of 58% in 2016 and 66% in 2024 to the total quantified VOCs, followed by the alkane class with 27% in 2016 and 24% in 2024 (Fig. 1, b). Minimum OVOC concentrations were observed in winter and maximum values occurred in spring or summer, with the timing of peak concentrations varying across sampling years (Fig. S2).

3.2 PMF factor identification and contribution: seasonal variability in 2016, 2017, and 2024

3.2.1 Road transport

The road transport factor accounts for 40–60% of the heavier measured alkanes (C5 to C7) and for 30–70% of the aromatics across all seasons (Fig. 2). Methyl-tert-butyl-ether (MTBE), a gasoline additive used since the replacement of leaded fuels in 1970's [35, 36], contributes between 30 to 70% to this factor. These compounds are typically retrieved from vehicle exhaust and fuel evaporation in source apportionment studies [7, 37]. Ethyl acetate contributes up to 20% to this factor in 2024 but not in 2016–2017 (Fig. 2). Ethyl acetate has nevertheless been reported in traffic-influenced environments, such as in a Swiss highway tunnel in a previous study [38]. Therefore, it is assumed that this compound is a secondary product of the burning process. This factor exhibits a clear morning and evening peaks corresponding to rush-hour traffic [7, 39] (Fig. 3) and correlates positively with combustion-related markers such as NO_x (R = 0.45) and CO (R = 0.37) (Table 1). Stronger contributions to this factor are measured for the low wind speeds, indicating more local emissions without any specific wind direction as the monitoring station is surrounded by roads (Fig. S5).

Table 1
R Pearson correlation coefficient of identified factors with ancillary data between 2016 and 2024 with a p-value below 5%.

Road transport	**NO_x**	**CO**	**CH_4**	**$PM_{2.5}$**	**PM_{10}**	**O_3**	**Temperature**	**Ethanol**
	0.45	**0.37**	**0.37**	**0.14**	**0.21**	**-0.33**	**0.05**	**0.40**
Residential heating and wood burning	0.36	0.40	0.34	0.43	0.41	-0.22	-0.10	0.30
Propane and butanes source	0.57	0.48	0.40	0.25	0.29	-0.39	-0.20	0.15
Volatile chemical products	0.38	0.34	0.23	0.20	0.24	-0.21	-	0.14
Local emissions	0.08	0.09	-0.08	0.11	0.20	0.20	0.41	0.22
Urban background and natural gas	0.21	0.31	0.36	0.31	0.21	-0.37	-0.53	-0.02

In addition, the factor shows correlations with methane (R = 0.37; Table 1). Methane emissions can be related to natural gas-based fuels[40, 41] but due to the low proportion of vehicles in Zurich using natural gas-based fuels (Sect. 3.1.1), methane is likely associated with emissions from catalytic converters [42, 43]. A strong correlation is observed with ethanol (R = 0.40) as expected due to its presence in gasoline formation and in window-wiper fluids [18] (Fig. 2).

The factor accounts for approximately 17–23% of total VOC concentrations and showed relatively stable contributions across years. Contribution to this factor is higher in summer than in winter by 13–15%. As the seasonal diurnal profile of the vehicle count per year is comparable over the year (Fig. S4), the seasonal variations observed for this factor reflect the temperature-dependent evaporative emissions as observed in previous studies [7, 37]. In summer, a small contribution of methanol was associated with this factor (Fig.

S6) due to enhanced biogenic emissions and photochemistry [44]. A part of the background level of methanol cannot be totally excluded from this source which may influence its estimated contribution and its variability.

3.2.2 Residential heating and wood burning

The residential heating and wood burning factor explains 40–70% of ethene and propene, 20–60% of ethyne, 25–45% of benzene and 30–80% of 1,3-butadiene (Fig. 2). The elevated contributions of light alkenes and ethyne are characteristic of combustion processes [7, 37, 45, 46]. Positive correlation with NO_x (R = 0.36), CO (R = 0.40), $PM_{2.5}$ (R = 0.43) and PM_{10} (R = 0.41) further supporting its attribution to combustion and wood burning (Table 1) [47]. The diurnal profile is characterized by a morning peak and a stronger evening-to-nighttime maximum (Fig. 3), consistent with residential heating activities such as emissions from boilers or localized combustion sources during evening and nighttime hours [39, 48]. Contributions of this factor remain relatively stable throughout the years with 9–10%. This factor likely represents a mix of two combustion processes: residential heating and local barbecuing activities in the park located close to the station. Residential heating is expected to dominate during colder periods, whereas barbecue emissions are more likely to contribute during warm and sunny periods.

At this site, repeated barbecuing activities in the nearby park have been identified as local source of VOCs. Maximum contributions to this factor occur under low wind speed (< 2 m s^{-1}) from both north-west and south-east and exclusively from south-east at moderate wind speeds (2–4 m s^{-1}) (Fig. S5) supporting the influence of local, spatially located sources. During summer 2016 and 2024 (but not 2017), approximately 70% of the highest contributions were associated with air masses originating from the south (135–225°), corresponding to the location of the park with barbecuing activities and were predominantly observed between 15:00 and 21:00 UTC.

3.2.3 Propane and butanes

The propane and butanes factor explains 15–40% of propane, 35–60% of 2-methylpropane and 40–60% of n-butane (Fig. 2). The predominance of light alkanes (C_2-C_4) suggests that this fingerprint is attributed to unburned fuel and evaporative processes linked to road transport and represent emissions linked to cold-start conditions, diffuse fuel evaporation and combustion-related local activities. Furthermore, minor contributions from additional compounds, e.g., butenes, pentenes and limonene indicate that some of the urban background emissions are included to this factor. This factor exhibits correlations with combustion markers such as NO_x (R = 0.57), CO (R = 0.48). In addition, this factor shows the strongest correlation with CH_4 (R = 0.40). Together with the low contribution of ethane, this suggests that the observed CH_4 is associated with local urban emissions rather than natural gas leakage. The highest contributions observed under low wind-speed conditions further support a local origin (Fig. S5).

The temporal variation of this factor is characterized by a generally low contribution to VOC concentrations, punctuated by frequent high-intensity peaks. The diurnal profile exhibits twin peaks with a more pronounced morning peak than in the evening (Fig. 3) with higher contribution in winter (Table S3) due to a combination of shallow winter planetary boundary height increasing near-surface VOC concentrations [49,

50] and increased emissions from combustion-related activities during colder periods. Furthermore, on 28 November 2016 in the morning, all factor contributions increased, although especially for the propane and butanes factor, coincided with the presence of many stationary delivery trucks close to the monitoring station. This observation is consistent with contributions from cold-start engines and unburned fuel emissions.

3.2.4 Volatile Chemical Products

VCPs factor is characterized by ethyl acetate (20–100%), acetone (10–80%), butanone (30–70%) and smaller proportion of aromatics such as toluene (14%; Fig. 2). These compounds are associated with solvent use, coating, cleaning and personal care products or industrial sources such as for ethyl acetate [51]. Compared to other factors, VCP fingerprint shows higher variability, reflecting irregular and sporadic emissions coupled with the heterogeneous and diffuse nature of its sources.

The seasonal variability of the VCPs factor does not show a consistent pattern across the years. Contributions were generally higher during winter than summer in 2016 and 2017, whereas the opposite is observed in 2024 (Table S3). Although evaporation emissions from VCPs are expected to increase with temperature, no significant correlation with temperature was observed (Table 1), indicating that the use and/or production of VCPs is influenced by episodic events rather than seasonal variability.

The VCPs factor shows correlations with combustion markers with NO_x (R = 0.38) and CO (R = 0.34; Table 1). A clear relationship between the VCPs factor and NO_x/CO is observed in 2016 and to a lesser extent, in 2017 but not in 2024 (Tables S4-S6). This relationship may partly reflect the use of solvent-containing products associated with road transport activities, such as windshield washer fluids, that can exceed the emissions from exhaust and evaporative fuel losses [1]. The factor exhibits little diurnal variability and no clear dependence on wind direction or wind speed (Fig. 3; Fig. S5), consistent with spatially distributed urban sources.

In this study, VCPs contributed between 11% and 20% which is consistent with the findings of Zhang et al. [51] but remain lower than the estimations from emission inventories from North America [10] and from EMEP inventory [52] where the VCPs emissions represents around 33 and 35% of total VOCs emission between 2016 and 2022. This difference may result from the exclusion of ethanol from the PMF input dataset and from the limited number of VCP-related compounds measured by the GC-FID system.

3.2.5 Local emissions

The local emissions factor is characterized by high contribution of methanol (40–90%; except in autumn 2017 with 20%), isoprene (20–70%) together with smaller contributions from alkanes, mainly butenes (10–40%; Fig. 2). This fingerprint suggests a mixture of biogenic and local anthropogenic emissions. Although isoprene and methanol are commonly associated with biogenic emissions [53, 54],[55], the presence of additional alkenes and other OVOCs, including MTBE, indicates that combustion-related emissions are also partly represented within this factor. Isoprene can also originate from anthropogenic sources such as rod transport [56–58].

The local emissions factor showed a clear seasonal variability with a maximum contribution during spring and summer and minimum contribution during winter (Fig. 3, Table S3). In wintertime, the diurnal profile includes an evening peak (Fig. 3, Fig. S3) likely due to unidentified combustion-related emissions. In summertime, the diurnal profile is characterized by a daytime maximum around midday (Fig. 3) and the factor shows a positive correlation with temperature (R = 0.41; Table 1). Enhanced contributions to this factor are associated with air masses coming from the east and south-east at wind speeds above 6 m s^{-1} (Fig. S5), corresponding to the forests located approximately 2–3 km east from the site. Additionally, the highest contributions are measured south-east for at low wind speed corresponding to the location of the Kasernenareal, a vast green area. These features are consistent with enhanced biogenic emissions and photochemical production during warmer periods. Elevated contributions are also observed for a large area covering from west to north-east corresponding to the urbanized areas of Zurich and likely reflecting anthropogenic emissions contribution to this factor.

3.2.6 Urban background and natural gas

The urban background and natural gas factor is characterized by high contribution of long-lived VOCs, such as ethane (50–75%), propane (15–45%), ethyne (10–45%), benzene (5–50%), acetaldehyde (10–55%), and acetonitrile (15–60%) (Fig. 2). This factor is commonly identified in PMF analyses and represents the background concentration of VOCs in the troposphere reflecting both local and long-range transported emissions [7, 59]. This factor likely represents a mixture of background and local sources rather than pure natural gas leakage. Nature gas emissions from pipelines in Switzerland are expected to have an ethane-to-propane ratio between 6.5 and 7.5 [60] while a ratio ranging between 1.4 and 3.9 is observed at the station suggesting that the natural gas emissions alone cannot contribute to this factor. For acetonitrile, this factor explains the largest fraction of concentrations in summer each year (Fig. S11), potentially due to distant wildfires.

The factor shows no dominant wind direction (Fig. S5), consistent with a regional background contribution. The contribution to this factor is higher during winter than summer (Fig. 3, Table S3) and anti-correlated with temperature (R=-0.53; Table S3). In summer, contributions decrease during daytime as the boundary layer develops, whereas this pattern is less pronounced in winter (Fig. 3). These observations suggest that seasonal and diurnal variability is largely controlled by atmospheric mixing and dilution processes.

3.3 PMF factor contributions: comparative analysis across 1993–1994, 2005–2006, 2016–2017, and 2024

3.3.1 PMF analysis over three decades

The relative contribution of each identified PMF factor showed seasonal and interannual variability between 2016–2017 and 2024 (Fig. 4). Road transport remained relatively stable across all years, contributing 17–23% of total VOCs. In winter, the relative contribution of the PMF factors was stable for most of the factors between the two periods with urban background and natural gas (21%) followed by local emissions (21%) contributing the most in 2016–2017 and urban background and natural gas (25%) and road transport (22%) in 2024. VCPs factor showed the highest relative contribution difference in winter

between both periods with 18% in 2016–2017 and 11% in 2024 emphasizing the irregular variety of emissions covered by this factor. More pronounced differences in factor contributions were observed in summer with the contribution of local emissions decreasing from 36% in 2016–2017 to 23% in 2024, while VCPs increased from 12% to 20%. Despite this seasonal variability, the annual relative contribution of the identified PMF factors was not significantly different between the two periods indicating that the VOC sources in Zurich have not significantly changed between 2016 and 2024.

A PMF analysis based on VOCs measurement at the same site was conducted in 2005–2006 and compared to results from 1993–1994 in Lanz et al. [17] without including OVOC measurements and using only 13 NMHCs: ethane, propane, 2-methylpropane, n-butane, 2-methylbutane, n-pentane, isohexanes, n-hexane, ethene, propene, ethyne, benzene and toluene. Eight PMF factors were identified in 1993–1994 and seven in 2005–2006. To compare both PMF results, four general PMF factors were determined: road traffic, solvent use, wood burning and gas leaking. Thus, the same methodology has been applied for 2016–2017 and 2024 included these 13 NMHCs to estimate the contribution of these four general PMF factors. The road traffic, solvent use, wood burning, and gas leaking factors correspond respectively to the road transport, VCPs, residential heating and wood burning and the combined urban background, natural gas, propane and butane sources identified in this study. The contribution of the four factors for 1993–1994, 2005–2006, 2016–2017 and 2024 is indicated in Table 2 and Fig. 5. As the comparison between 1993–1994 and 2005–2006 was already performed [12], the comparison is conducted with the most recent datasets, 2005–2006, 2016–2017 and 2024.

In general, considering the four PMF factors together, their contributions decreased by 57% between 2005–2006 and 2024, from 13 ppb to 5.5 ppb. A strong decrease is observed for the four factors between 2005–2006 and 2016–2017 with decreasing factors of 2.5, 3.4, 1.5 and 1.9 ppb for road traffic, solvent use, wood burning and gas leaking respectively. Between 2016–2017 and 2024, only slight decreases (0.2–0.3 ppb) are observed for solvent use, wood burning, gas leaking while road traffic shows a small increase from 1.4 to 1.6 ppb. The strongest decrease is observed for the road traffic factor with the most reduction occurring before 2016–2017. Gas leaking factor has the smallest decrease and remains relatively stable compared to the other factors after 2005–2006. This limited decrease likely reflects the relatively constant composition of the gas used for residential heating over the past decades, as well as its widespread use in residential areas surrounding the measurement site.

Table 2
Mean contribution of general PMF identified factors in Lanz et al. (2008) and applied to this study using 13 NMHCs in common (in ppb). The gas leaking factor corresponds to the combined urban background, natural gas, propane and butane sources factors identified in this study.

Road traffic	**1993–1994 (Lanz et al., 2008)**	**2005–2006 (Lanz et al., 2008)**	**2016–2017 (this study)**	**2024 (this study)**
	9.9	**3.5**	**1.4**	**1.6**
Solvent use	4.4	2.7	0.8	0.6
Wood burning	4.2	2.1	1.4	1.1
Gas leaking	6.0	4.6	2.4	2.2

13 NMHCs were used to estimate the contribution of the four general PMF factors. However, in 2016 these compounds accounted for 39% (6.0 ppb) of the total VOCs quantified by the TD-GC-FID and 30% (5.5 ppb) in 2024. As shown in 3.1.3, the OVOCs contribute more than half of the total VOCs at this station after 2005–2006, indicating that analysis focusing only on NMHCs are becoming less representative of the urban environments. Depending on the VOC species included in the PMF analysis, the identified factors can be strongly impacted as observed with two studies conducted in Zurich in 2005–2006 using different VOCs [12, 18]. In Lanz et al. [17], only NMHCs were monitored and the following PMF factors were identified: gasoline evaporation, solvent, propane, ethane, wood burning and fuel combustion. In contrast, Legreid et al. [18] which included OVOCs and a few of NMHCs, such as butane, 1,3-butadiene, and monoaromatics, identified broader factors including regional background, large-scale background, combustion and other sources. The combined use of NMHCs and OVOCs allows better resolution of the source apportionment, as illustrated in this study by the identification of the VCPs and local emissions factors, which are largely characterized by OVOCs (Fig. 2; Fig. 5b).

3.3.2 Comparison with EMEP emission inventories

The road traffic and solvent use factors determined by the PMF analysis conducted in Zurich are compared with the European Monitoring and Evaluation Programme (EMEP) inventory since 1993 in Switzerland (Table 3). Overall, the PMF results and the EMEP inventory show comparable long-term decreasing trends for road traffic emissions and solvent use. The road traffic emissions in Switzerland from the EMEP inventory decreased from 23.62 and 21.22 kT in 2005 and 2006 respectively, to 9.08 and 8.73 in 2016 and 2017, corresponding to an overall reduction by a factor of 2.5. A similar reduction factor between the PMF analysis and the EMEP inventory is observed for the periods between 1993–1994 and 2016–2017. Between 2016–2017 and 2024, the PMF analysis indicates a slight increase in the road traffic factor (factor of -1.1), whereas the EMEP inventory still suggests a moderate decrease (factor of 1.3).

For the VCP and solvent use emissions, a similar reduction is observed between 1993–1994 and 2005–2006 for the PMF analysis and the EMEP inventory with decreasing factors of 1.6 from the PMF analysis and 1.8 from the EMEP inventory. However, between 2005–2006 and 2016–2017, the PMF-derived solvent use factor decreases much more strongly, with a reduction factor of 3.4 compared with only 1.4 in the

EMEP inventory. In contrast, between 2016–2017 and 2024, a similar reduction factor was observed with 1.3 and 1.0 for the PMF analysis and EMEP inventory, respectively. A possible explanation for the discrepancy observed between 2005–2006 and 2016–2017 is the limited numbers of compounds available for the comparison between the two periods. Only thirteen NMHCs were included for 2005–2006, which likely limits the representativeness of the solvent use factor, which is primarily characterized by aromatic compounds, whereas in the present study the VCPs factor is more strongly associated with OVOCs such as ethyl acetate, acetone and butanone.

Table 3
Decreasing factor of road traffic and solvent use factors in Zurich according to measurements and in Switzerland according to the EMEP emission inventory. A negative value corresponds to an increasing factor.

	1993–1994 vs 2005–2006		**2005–2006 vs 2016–2017**		**2016–2017 vs 2024**	
	Observation in Zurich	**EMEP inventory for Switzerland***	**Observation in Zurich**	**EMEP inventory for Switzerland**	**Observation in Zurich**	**EMEP inventory for Switzerland****
Road traffic	2.8	2.3	2.5	2.5	-1.1	1.3
Solvent use	1.6	1.8	3.4	1.4	1.3	1.0
*: Before 2000 only the years 1990 and 1995 are available for Switzerland in the EMEP inventory. Hence, the year considered is 1995 and not 1993 nor 1994						
**: As the 2024 data are still not available in the EMEP inventory the year considered is 2023						

4 Conclusion

A long-term VOC dataset, including both NMHCs and OVOCs, based on hourly measurements conducted at Zürich Kaserne in 2016, 2017 and 2024, was analyzed to assess the sources contributing to urban VOC concentrations. Between 2016 and 2024, significant decreases were observed for several pollutants, including NOx (50%), CO (32%) and total VOCs (17%). In contrast, the total VOC concentrations of all VOC species quantified by the TD-GC-FID did not significantly change since 2016 with the OVOCs becoming more dominant, by contributing to 58% to the total quantified VOCs in 2016 and to 66% in 2024.

In this study, the source-apportionment analysis identified 6 factors according to the PMF results: (i) road transport containing both traffic exhaust and fuel evaporation; (ii) residential heating and wood burning impacted in the summertime by barbecuing activities; (iii) propane and butanes factor, possibly linked to unburned fuel; (iv) VCPs corresponding to the use and production of solvents and their diffusive emissions; (v) local emissions corresponding to a mix between unidentified combustion processes and biogenic emissions and (vi) urban background and natural gas, dominated by long-lived compounds such as ethane and propane. The relative contribution of these sources remained stable between 2016–2017 and 2024 with seasonal variations. In the summertime, road transport and local emissions contribute the most with

23% and 23–36% respectively while propane and butanes, and the urban background and natural gas factors contribute the most during the wintertime with 16–19% and 21–25% respectively. The VCPs factor contributed between 11 and 20% of the VOC concentration and showed no clear seasonal pattern, consistent with its diffuse and heterogeneous emission sources.

The comparison with earlier measurements from Lanz et al.[17] based on 13 NMHCs highlights a strong long-term decrease of the four sources identified such as road traffic, solvent use, wood burning and gas leaking since the 2000s. This decrease is consistent with EMEP inventory trends for road traffic, but appears more pronounced for solvent-related emissions, likely due to the limited VOC speciation in earlier datasets. The inclusion of a broader range of VOCs, particularly OVOCs, demonstrates that analyses relying solely on NMHCs are no longer sufficient to represent urban VOC composition or to accurately constrain emission sources. In this study, VCPs accounted for 11–20% of the measured VOC concentrations, confirming their role as a significant urban emission source, although lower than reported in recent U.S. studies. Further investigations across European cities are required to confirm the contribution of the VCPs and other OVOC-related sources to VOC concentration. Overall, this study shows the importance of long-term VOC monitoring, including OVOCs, to accurately assess the evolution of source contributions throughout the year and support effective air quality management strategies.

Declarations

Competing Interests

One of the co-authors was involved as a Guest Editor for the Short-lived climate pollutants Collection.

Competing Interests Statement

One of the co-authors was involved as a Guest Editor for the *Short-lived climate pollutants* Collection.

Author Contribution

MD conducted the PMF analyses. SR was responsible for and supervised the monitoring of VOCs in Zurich, with the assistance of ZLB in data processing. MD and ZLB wrote the manuscript with support and feedback from TS and SR. TS conceived, designed, and supervised the study. All authors contributed to reviewing and revising the manuscript and approved the final version.

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Data Availability

The validated datasets for Zürich Kaserne can be accessed through the EBAS database at https://ebas.nilu.no/data-access/.

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Figures

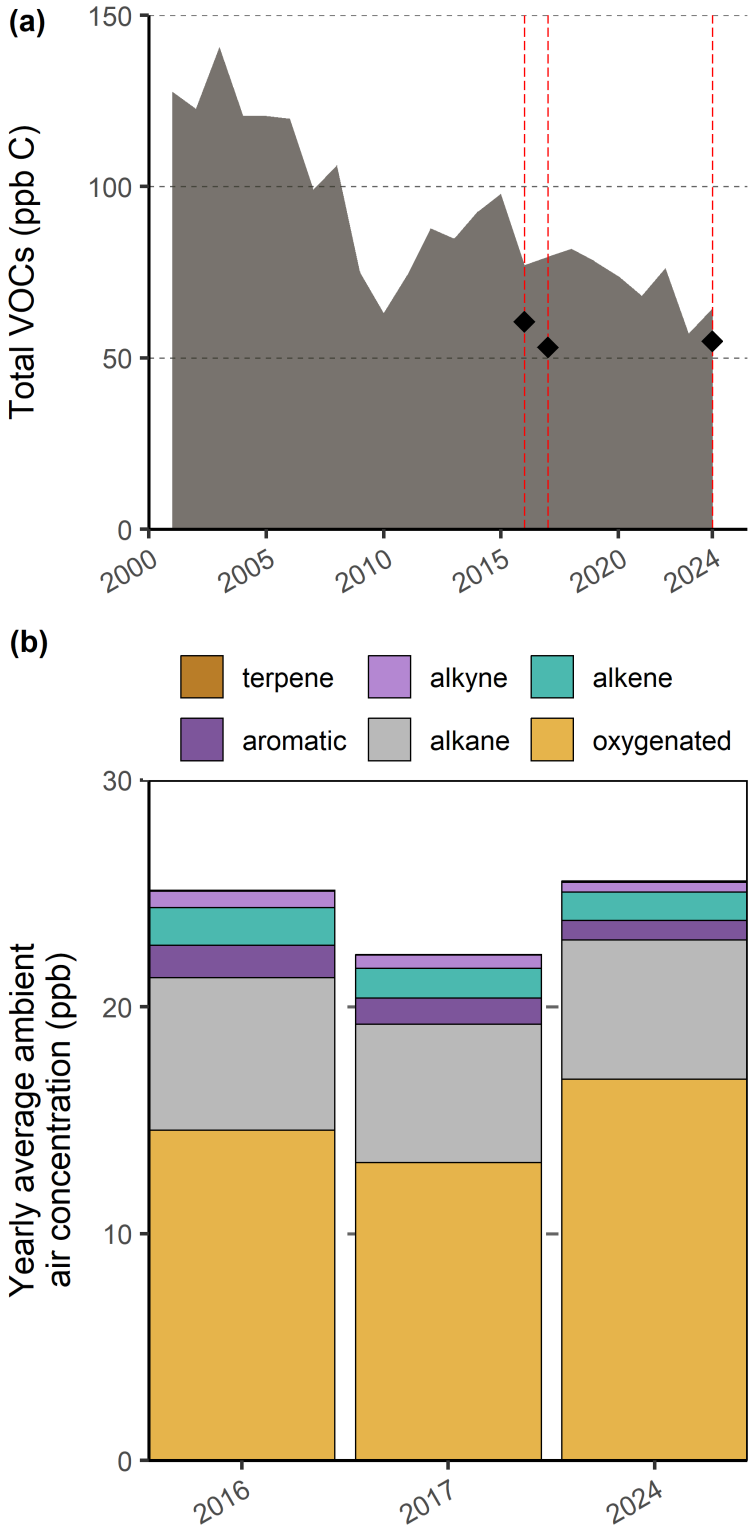


Figure 1

(a) Total yearly average VOC concentration (ppb C) measured by the NABEL and the corresponding average concentration of the VOC (ppb C) calculated according to their carbon response factor by the GC-FID (black diamond) and (b) the yearly average VOC class concentrations measured in 2016, 2017 and 2024 at the Zurich station. The monthly variations in the VOC classes are given in Fig. S2.

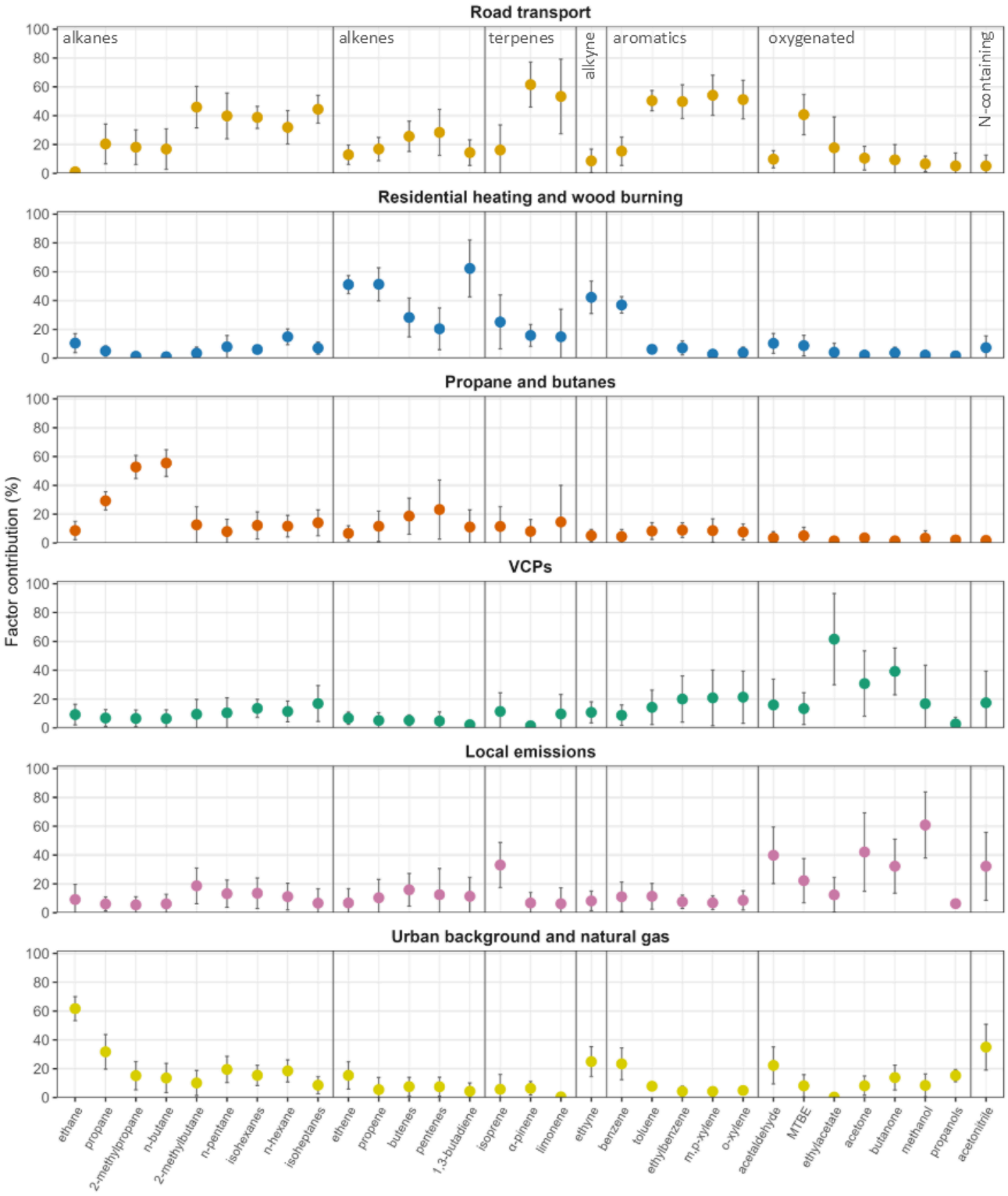


Figure 2

Fingerprints of road transport, residential heating and wood burning, propane and butanes, VCPs, local emissions, urban background and natural gas. The black bars are the standard deviation of the fingerprints between all the seasons.

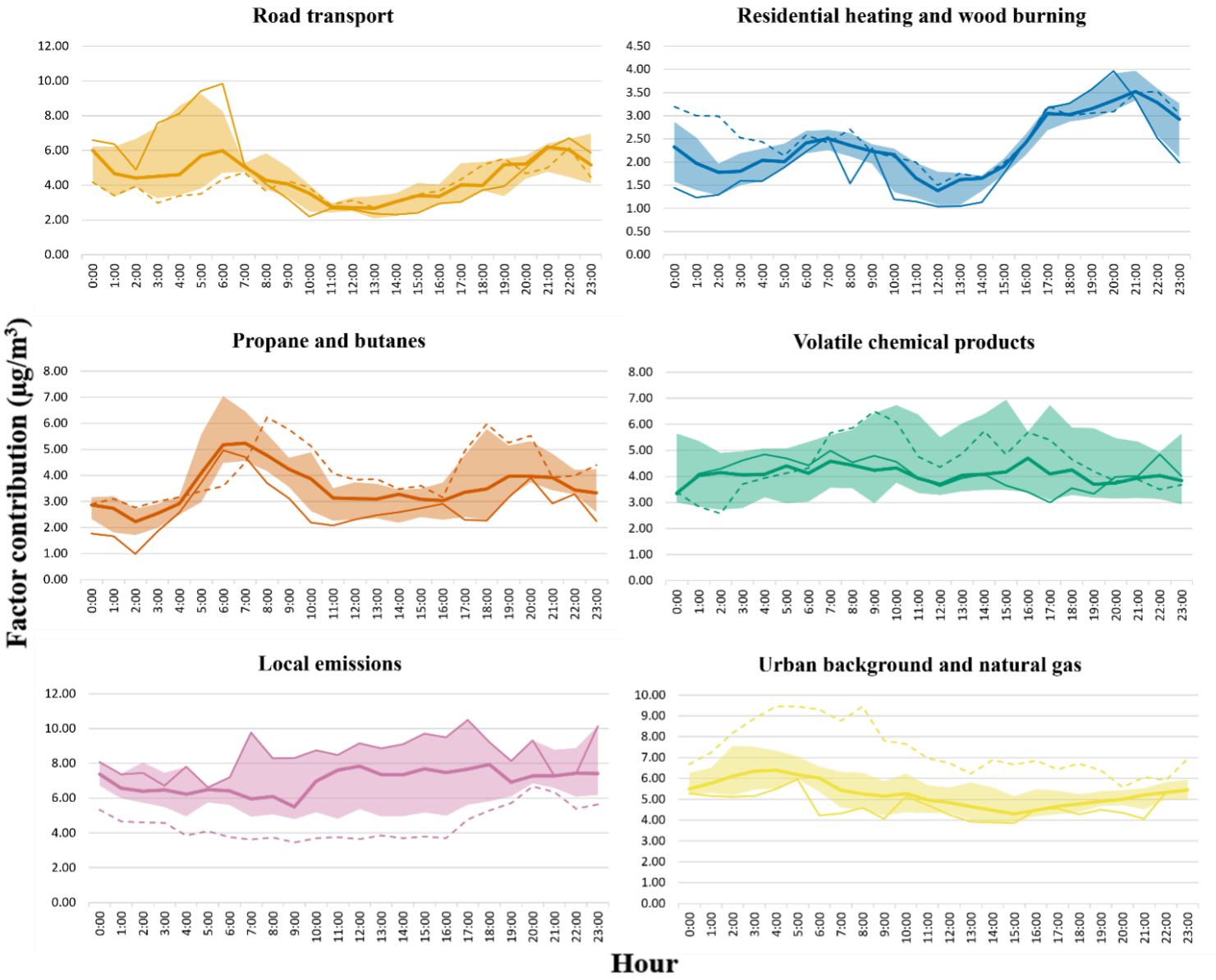


Figure 3

Median of the diurnal profiles of all factors during the whole campaign. The colored area corresponds to the interquartile range. The thin solid and dashed lines correspond to summer and winter diurnal profiles respectively. The diurnal profiles for each individual season are shown in Fig. S3.

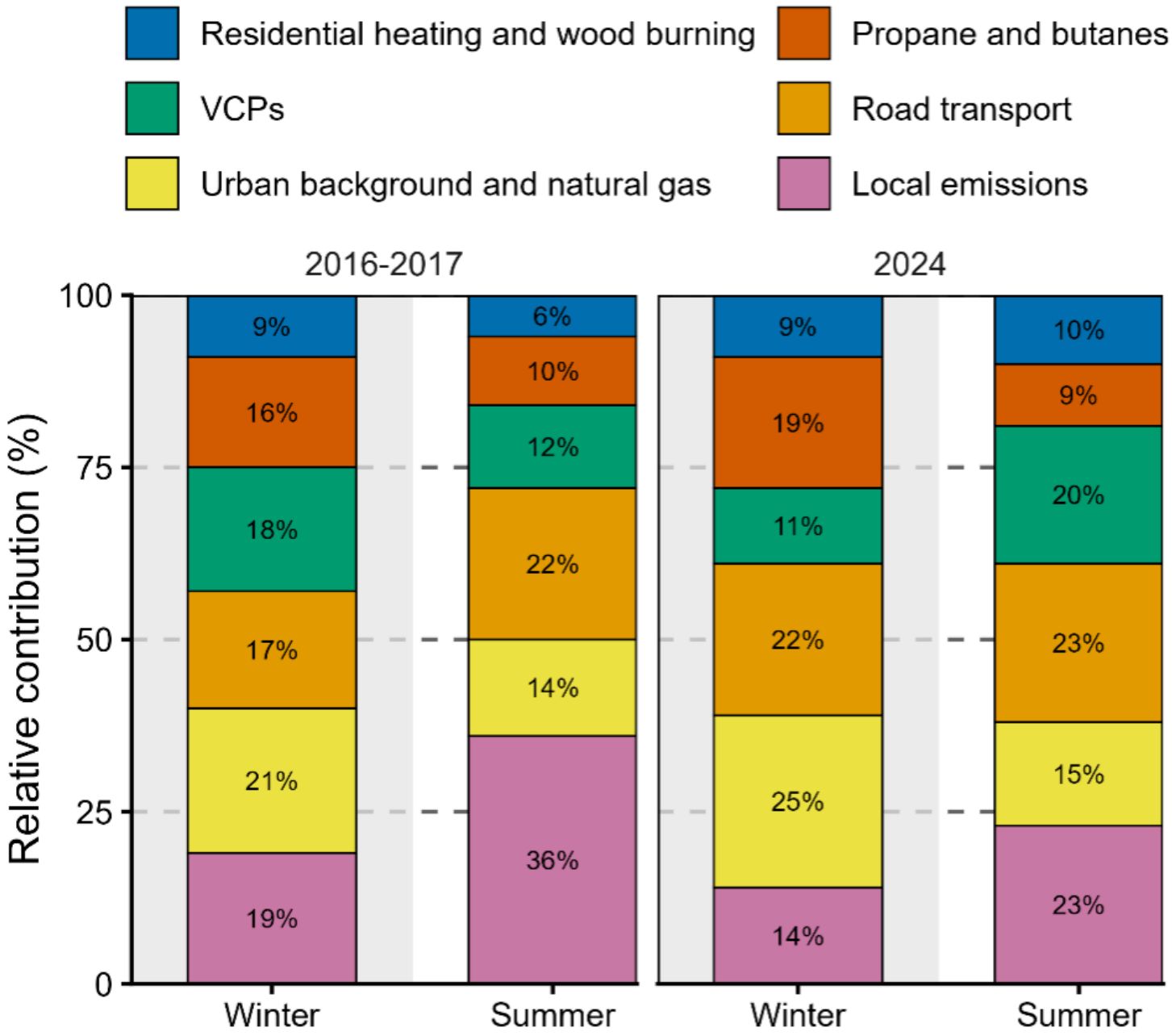


Figure 4

Relative contribution of each identified factor in winters 2016 – 2017, summers 2016 – 2017, winter 2024 and summer 2024.

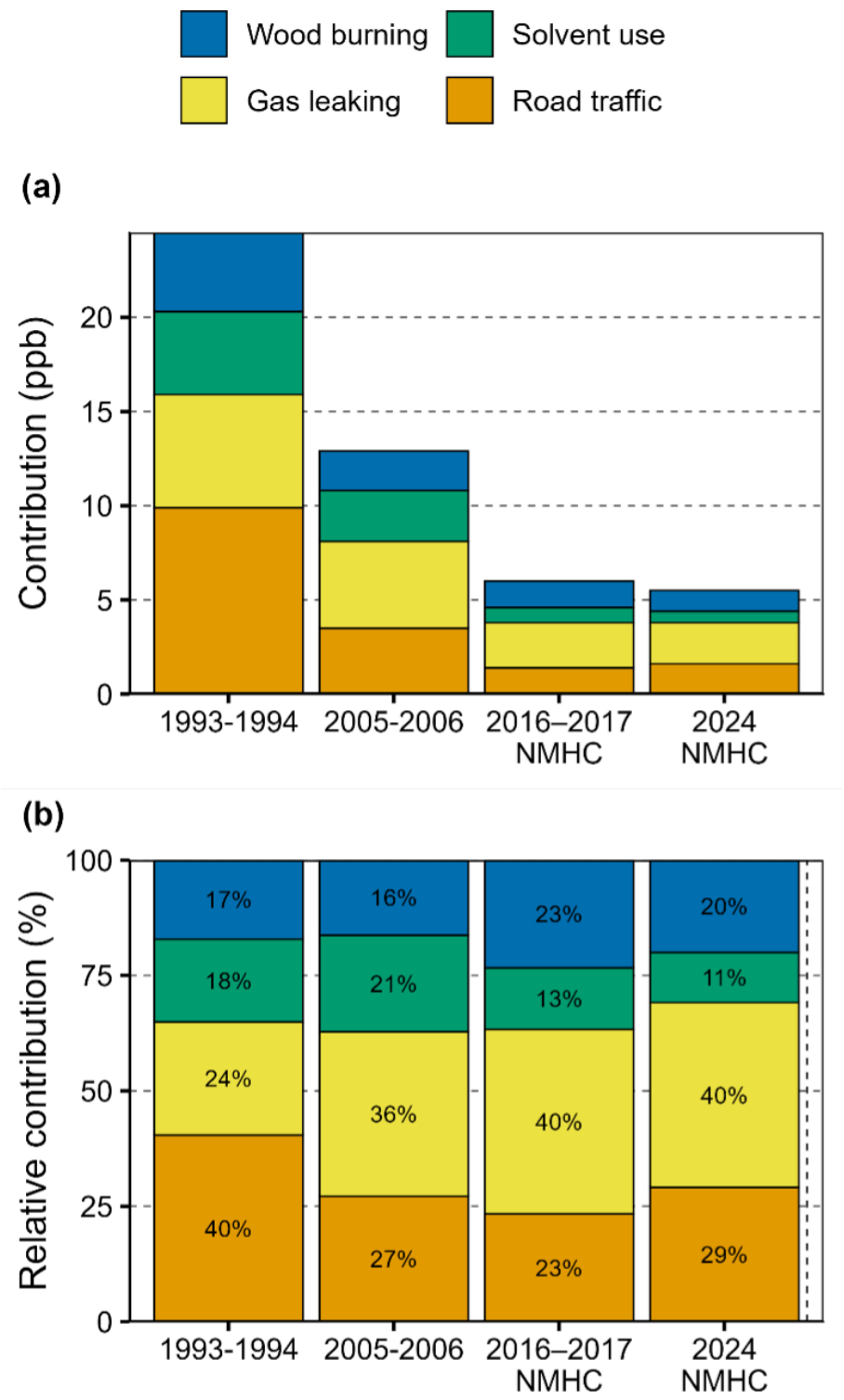


Figure 5

Contribution (a) and relative contribution (b) of the general PMF factors determined for the different datasets in 1993-1994, 2005-2006, 2016-2017 and 2024 at the Zurich station utilizing the same thirteen NMHCs determined by GC-FID (NMHC).

Supplementary Files

This is a list of supplementary files associated with this preprint. Click to download.

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