https://doi.org/10.5194/egusphere-2025-3241
Preprint. Discussion started: 25 August 2025

Measurement report: 30 years of monitoring aromatic hydrocarbons (BTEX) at a suburban site in Europe

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Abstract. Since 1994, benzene, toluene, ethylbenzene and xylene isomers (BTEX) are monitored in the ambient air at Dübendorf (DUE) in the suburban area of Zurich city in Switzerland. Overall, BTEX concentrations decreased up to 89 % in ambient air in DUE notably due to the introduction of regulations concerning the air quality such as limiting benzene concentrations in car fuel or the introduction of the incentive fee on VOCs in 2000 in Switzerland. While BTEX was one of the major VOCs compound classes in 1994 (33 % of the total non-methane hydrocarbons (NMHC)), BTEX contribution to total NMHC significantly decreased to 8.4 % in 2022. Before 2000, traffic exhaust emissions were the dominant source of BTEX to the ambient air with a toluene-to-benzene (T:B) ratio of 2.4 ± 0.1. Since 2000, the contribution of vehicle emissions to toluene concentrations in ambient air in DUE during summer has decreased from 82 % to 65 %, with the remaining proportion emitted from solvent emissions. In addition, BTEX are important ozone and secondary organic aerosol (SOA) precursors. While the BTEX contribution to ozone formation potential (OFP) has decreased from 25 % to 8 % between 2005 and 2024, their relative contribution to SOA formation potential remains high, contributing to 80 % of the SOA formation potential of the total VOCs measured in Zurich in 2024.

1 Introduction

Benzene, toluene, ethylbenzene and xylene isomers, referred to as BTEX compounds, are volatile aromatic hydrocarbons found in ambient air which are predominately of anthropogenic origin and are precursors of tropospheric ozone and secondary organic aerosols (SOA) (Atkinson, 2000). Benzene, one of the most abundant BTEX in urban areas, is classified as known carcinogen to humans (Group 1) (Althouse et al., 1980). Long-term or high-level exposure to BTEX showed adverse effects on human health (Demirel et al., 2014). Since inhalation is the primary route of BTEX entry into human body, monitoring these volatile organic compounds (VOC) in ambient air remains crucial for assessing associated health risks. These aromatic hydrocarbons can represent an important fraction (up to 40 %) of non-methane hydrocarbons (NMHC) emitted in (sub)urban areas (Derwent et al., 2000; Borbon et al., 2002). Car exhaust and industrial activities represent the main emissions of BTEX in ambient urban air (Caselli et al., 2010). Combustion of benzene-containing petrol and coal, as well as the evaporation from gasoline service stations represent the main sources of benzene (Schnatter, 2000; Ballesta et al., 2006). Toluene, commonly used as a solvent, can also be emitted from fuel combustion and biomass burning (Kavouras and Zielinska, 2012). Ethylbenzene and xylene isomers have similar sources, mostly originating from petrol industries and solvents (Monod et al., 2001). Once released into

https://doi.org/10.5194/egusphere-2025-3241
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the atmosphere, these compounds undergo oxidation by hydroxyl radicals with lifetimes between 2 weeks (benzene), two days (toluene, ethylbenzene) and some hours (xylenes) (Monod et al., 2001). They can form highly oxygenated compounds that contribute to higher levels of tropospheric ozone and to the formation of secondary organic aerosols (SOA) (Molteni et al., 2018; Calvert et al., 2002). Higher tropospheric ozone events, primarily occurring during heatwaves, can have severe damages on human health and vegetation (Lippmann, 1991). Consequently, reducing BTEX emissions and subsequently the production of tropospheric ozone are critical for effective air pollution management and to reduce human risks in (sub)urban areas.

Various regulations on the emissions of VOCs have been implemented in Switzerland and Europe to mitigate their potential harmful effects to human health. Since 1987, gasoline engines are equipped with 3-way catalysts reducing the emissions of aromatic hydrocarbons and other VOCs from car exhausts in Europe (Heeb et al., 2000). In 2000, the ordinance on the incentive tax on VOCs has been introduced in Switzerland, regulating the use of solvents, such as toluene and xylene (Swiss Confederation, 1997). In addition, from 2000 onwards, the benzene content in gasoline engines in Switzerland has been restricted stepwise from 5 % vol. to less than 1 % vol. of benzene according to the Ordinance on Air Pollution Control (Swiss Confederation, 1986). In 2005, toluene and benzene were added to the list of substances with restricted use under the Swiss Chemical Risk Reduction Ordinance (ORRChem; Swiss Confederation (2005)). For example, preparations containing of benzene levels of 0.1 wt% or more is prohibited on the market and adhesives and spray paints destined for the general public should contain less than 0.1 wt% of toluene.

Thus, long-term monitoring of BTEX in the ambient atmosphere is essential for identifying trends in their concentrations and establishing their background concentrations, providing valuable data on the impact of ongoing and future air quality policy management. The longest continuous BTEX monitoring time series in Europe began at the suburban London Eltham station in late 1993 within the UK Automatic Hydrocarbon Monitoring Network, followed by Dübendorf in Switzerland in early 1994. Here, we present the BTEX data collected over 30 years in Dübendorf at a ground-based monitoring station in the suburban area of Zurich, one of Europe's longest continuous BTEX time series. This time series is complemented by BTEX measurements from traffic, urban, and rural sites across Switzerland.

2 Material and methods

2.1 Sampling areas

Dübendorf station, later referred as DUE, is located in the suburban area of Zurich city (47.405°N; 8.609°E; 435 m a.s.l.) in Switzerland and is considered as a suburban monitoring station. Benzene and toluene are monitored continuously since 1994 in the ambient air at DUE while the monitoring of xylenes and ethylbenzene was only performed from 1994 until 2010, due to their decreasing concentrations in ambient air making it very laborious to measure these compounds with the required precision. As a consequence, BTEX are monitored since 2011 at the road side station in Bern (BER; 46.570°N, 7.263°E; 536 m a.s.l.), which exhibits higher traffic influence and higher BTEX concentrations. At both stations, BTEX are measured with a gas chromatograph equipped with a photoionization detector (GC-PID, GC955 BTEX Analyzer, Synspec, the Netherlands).

In addition, non-continuous sampling of BTEX has been conducted in Zurich city, considered as an urban background site

(ZUE, 47.378°N, 8.530°E; 409 m a.s.l.) from 1993 to 1994, from 2001 to 2017 and since 2022 with a GC-flame ionization detector (GC-FID; details are given in Lanz et al. (2008)) with 1-hour resolution. Online measurements of BTEX using GC-FID are conducted since 2018 at the rural Beromünster station (BRM; 47.18961391°N, 8.175433684°E; 797 m a.s.l.). All monitoring sites are part of the Swiss National Air Pollution Monitoring Network (NABEL).

2.2 BTEX measurement and ancillary data

Air samples for BTEX analysis are taken from the main inlet of the monitoring station in DUE and BER every 20 minutes and every hour at ZUE and BRM from a sampling line continuously flushed with air and analyzed by GC-PID or GC-FIDs respectively. BTEX concentrations are aggregated to 1-hour resolution. Calibration and QA/QC assessments are monthly conducted. GC-PID instruments are calibrated with real-air standards filled with urban ambient air and are calibrated against a 4 ppbv 30-compound standard in nitrogen from the National Physical Laboratory (NPL) while GC-FIDs instrument are monthly calibrated using the 4 ppbv 30-compound NPL standard. Detection limits for all BTEX are 10 ppt. BTEX data are available at the EBAS database (https://ebas-data.nilu.no/, accessed on April, 2024).

Ancillary data, i.e., temperature, wind speed and direction, total NMHC concentration, NO_x, CO, ozone and global irradiation, are aggregated to 1-hour resolution and available from the NABEL database at luftreinhaltung@bafu.admin.ch.

3 Results and discussion

3.1 Decrease in BTEX concentrations from 1994

The longest time series of BTEX monitoring in Switzerland, conducted at the suburban station in Dübendorf, shows an overall decline in BTEX concentrations from 1994 to 2024 with decrease of up to 89 % depending on the aromatic compound considered (Fig. 1). The trends for the other Swiss stations are shown in Fig. S1. Due to the low overall concentrations of ethylbenzene and xylenes after 2010 in Dübendorf, their concentrations from 2011 onwards are estimated according to their relative contribution to BTEX between 1994 and 2000 (Fig. S2). The estimated and measured concentrations of both compounds showed good agreement (R^2=0.94 and R^2=0.97 for ethylbenzene and xylenes, respectively; Fig. S3).

In 1994, BTEX compounds constituted 33 % of the total NMHC concentrations in DUE and 2.3 % in 2024 corresponding to a decrease by 93 %. Toluene is the most abundant BTEX compound in DUE with its yearly average concentration decreasing by 89 % between 1994 and 2024 (from 3.3 pbb to 0.36 pbb). For the same time period considered, benzene concentrations decreased by 87 % (from 1.1 pbb to 0.14 pbb). Ethylbenzene and xylenes decreased by 75 % (from 0.42 to 0.10 ppb and from 1.7 to 0.46 ppb, respectively) between 1994 and 2010. The strongest decrease in BTEX concentrations (Fig. 2) occurred between the period 1994-2000 and 2001-2010, notably due to the restriction of the benzene-content in gasoline engines implemented in 2000 and the introduction of the incentive tax on VOCs in Switzerland (Swiss Confederation, 1997).

Emissions from the Swiss emission inventory (Bundesamt für Umwelt (BAFU), 2023), calculated according to the territorial principle, i.e., considering all emissions emitted within Switzerland showed an overall decrease in benzene emissions of

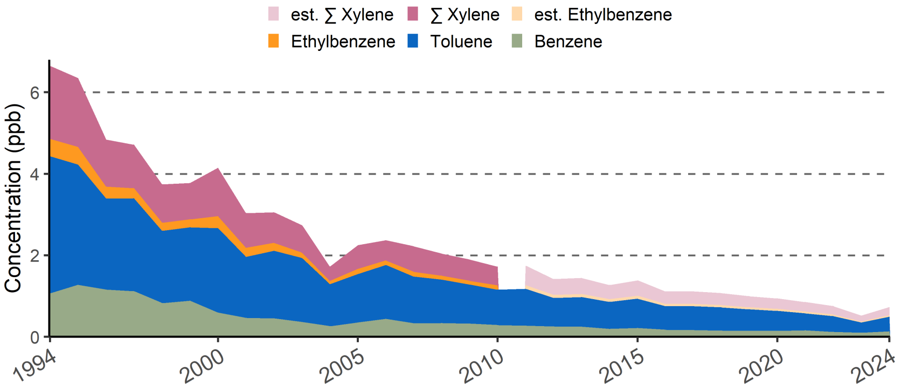

Figure 1. Monthly averages of benzene, toluene, ethylbenzene, xylenes (sum of m-, o- and p-xylene) concentrations from 1994 to 2024 at the suburban station DUE. After 2011, xylenes and ethylbenzene concentrations were estimated based on their relative contribution to BTEX before 2011, further details given in Fig. S2, S3.

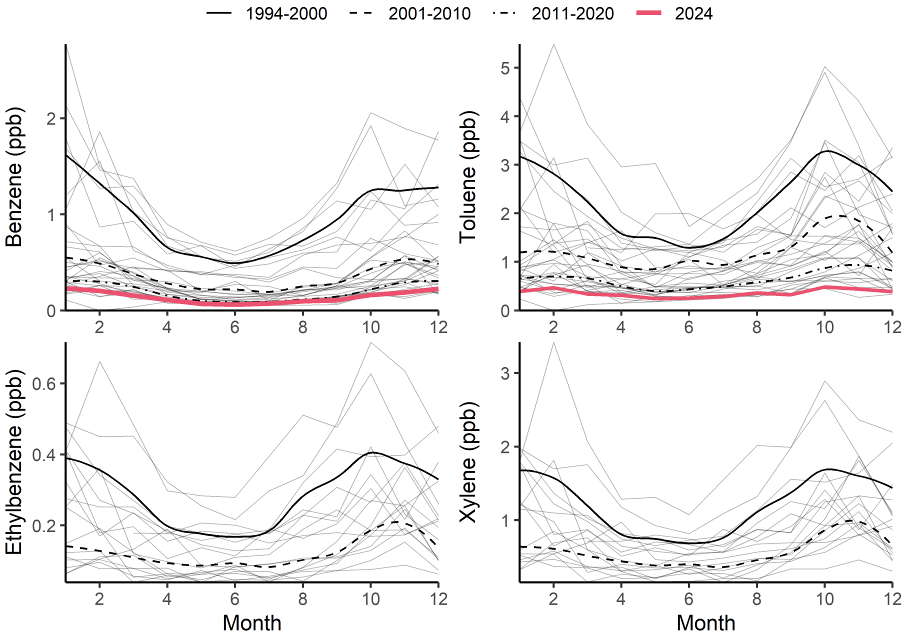

Figure 2. Monthly average of benzene, toluene, ethylbenzene, xylenes (sum of m-, o- and p-xylene) concentrations from 1994 to 2024 at the suburban station DUE.

85 % between 1994 and 2022 (Fig. 3; A). Benzene emissions are dominated by the transport sector (70 % in 1994; 65 % in 2022) and an increasing contribution of the household sector, including the use of wood combustion, over the years from 11 % in 1994 to 20 % in 2022. This agrees well with the measurement data at DUE, showing a strong positive linear relationship

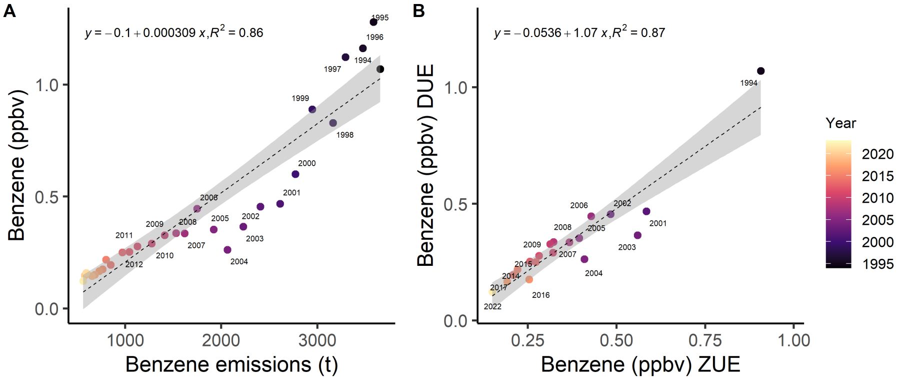

Figure 3. Comparison between modeled benzene emissions calculated according to the territorial principle, i.e., emissions emitted within Switzerland (Bundesamt für Umwelt (BAFU), 2023) and the benzene concentrations measured in ambient air in Dübendorf (DUE) (A) and benzene concentration in ambient air in DUE and Zurich (ZUE) stations (B).

(R^2=0.86; Fig. 3, A) between benzene concentrations emitted at the suburban DUE station and the modeled benzene emissions. Discrepancies are observed between the modeled benzene emissions and measured benzene in Dübendorf between 2000 and 2005. Based on the long-term correlation between Zurich and Dübendorf since 1994 (Fig. 3, B), higher benzene concentrations would have been expected in 2001, 2003 and 2004 in Dübendorf. However, this correlation alone does not account for the discrepancies observed between modeled and measured benzene data during this period, which remain unclear.

3.2 T:B ratio changes over the years

The toluene-to-benzene (T:B) ratios can be used as an indicator to assess the contribution of various sources of atmospheric pollutants, coming from traffic, solvent use or biomass burning. Areas impacted preliminary by car exhaust emissions have been estimated to have a T:B ratio ranged between 1.3 and 3.4 based on the European fleet composition (Ait-Helal et al. (2015); Liu et al. (2025) and references therein). For wood combustion, a T:B ratio below 1.3 has been determined from measurements conducted in an Swiss Alpine valley using wood as main heating source (Gaeggeler et al., 2008). Higher T:B ratio than 3.4 are linked to industrial activities, solvent and fuel evaporations (Liu et al., 2025).
During the period 1994 - 2000, the T:B ratio showed little variation throughout the day and the year, remaining at 2.4 ± 0.1, indicating that vehicle emissions were the dominant source of toluene and benzene in ambient air. After 2000, hourly T:B ratios varied throughout the day, with the highest T:B ratio measured at night time (3.6 ± 0.1), due to nocturnal stagnation of benzene and toluene in the atmosphere and during the rush hour time between 6 and 9am (3.7 ± 0.01) followed by a decrease during the day due to their degradation (2.9 ± 0.06). After 2000, the pronounced summer peak in the T:B ratio suggests a shift in source dominance, highlighting the increasing contribution of solvent and fuel evaporation to toluene concentrations (Fig. 4).

Assuming that for benzene emissions (i) traffic exhaust remain constant throughout the year, (ii) biomass burning is negligible in summer and (iii) biomass burning are negligible during the winter between 8 and 9 am, the theoretical summer

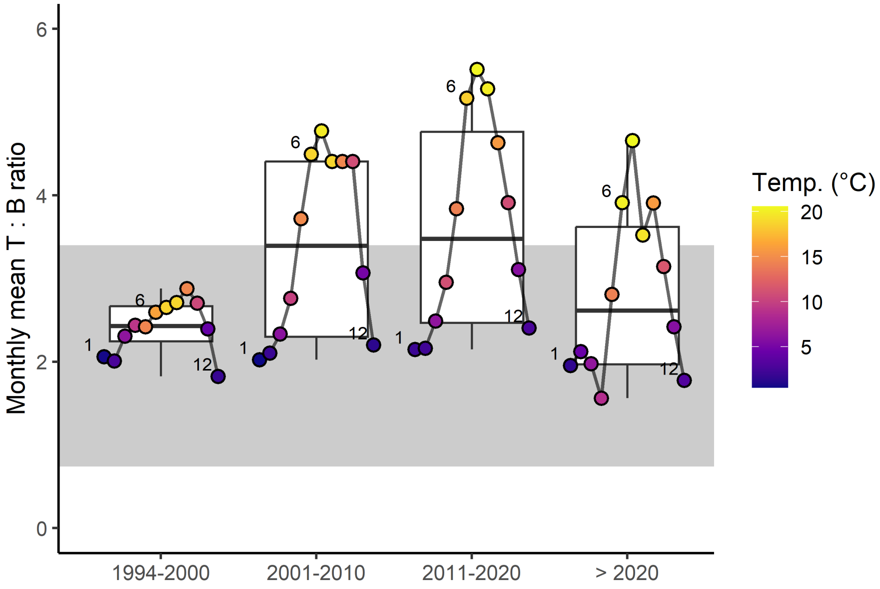

Figure 4. Monthly averages for T:B ratio from 1994 to > 2020 grouped by time periods (1994–2000, 2001–2010, 2011–2020, and > 2020). Each dot represents the average T:B ratio for a given month, color-coded by the corresponding mean temperature (°C). The gray area marks the T:B range for typical vehicle emissions (1.3 - 3.4). Labels for January (1), June (6), and December (12) are indicated.

concentration of toluene (toluene$_{th}$), i.e., assuming traffic emissions as the only source of toluene, can be estimated as follows in Eq. 1, with toluene$_{winter}$: benzene$_{winter}$ calculated for each year during winter between 8 and 9am, corresponding only to traffic emissions during rush hour. The solvent contribution to toluene is then calculated according to Eq. 2.

$$toluene_{th} = \frac{toluene_{winter}}{benzene_{winter}} \cdot benzene_{summer} \tag{1}$$

$$solvent\ contribution\ (\%) = \left(1 - \frac{toluene_{th}}{toluene_{summer}}\right) \cdot 100 \tag{2}$$

For the period 1994-2000, the solvent contribution to toluene concentration was calculated to be 18 % and approximately doubled afterwards to 44 % for the period 2011-2020 as shown in Fig. 5. Since 2021, less emissions from solvent are observed (35 %) for toluene, suggesting either a more controlled release of toluene to ambient air or a change in the source dominance of toluene during the summer.

3.3 Role of BTEX in tropospheric ozone and secondary aerosol formation potential

The role of BTEX in ozone formation can be estimated using their contribution in ozone formation potential (OFP) based on the maximum incremental reactivity (MIR) coefficient calculated by Carter (1990) for various individual VOCs. The MIR coefficient corresponds to the amount of ozone formed per amount of VOC added to an initial VOC-NO$_x$ mixture under

https://doi.org/10.5194/egusphere-2025-3241
Preprint. Discussion started: 25 August 2025

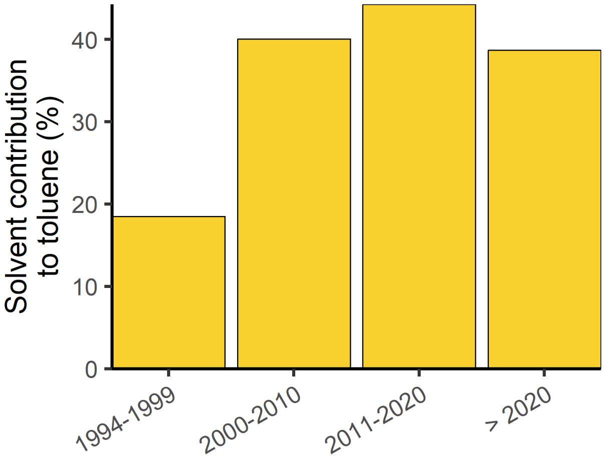

Figure 5. Solvent contribution to toluene concentration in the summer calculated according to Eq. 1 and Eq. 2 between 1994 and > 2020 grouped by time periods (1994–2000, 2001–2010, 2011–2020, and > 2020). Only June, July and August were considered for the summer months.

relatively high NO_x conditions, which are characteristic of (sub)urban environments. The OFP_i for a given VOC compound i is calculated according to Eq. 3 with MIR_i coefficient values updated from Carter (2009).

$$OFP_i = [VOC]_i \cdot MIR_i \quad (3)$$

At the monitoring station in Zurich, a wider range of NMHC - from C_2 to C_8 - are quantified by GC-FID including those VOC classes: alkane, alkene, alkyne, terpene, BTEX and oxygenated VOC (OVOC). Given the proximity of the DUE and ZUE stations, along with the similar BTEX trends observed at both sites, observations from ZUE can be extrapolated to DUE in terms of their potential to form tropospheric ozone and secondary aerosols. The yearly NMHC average ambient air concentration measured by the GC-FIDs in Zurich increased by 53 % from 20.1 ppb in 2005 to 30.7 ppb in 2024 (Fig. 6). In 2005, alkane was the dominant NMHC VOC class (45 %) followed by OVOC (20 %) and BTEX (15 %). OVOC became the most dominant class in 2015 with 54 % (70 % in 2024) while BTEX became less abundant (6 % in 2015; 3 % in 2024).

Considering the non-methane VOC classes in Zurich and the MIR values of each single VOC used to estimate OFP, the highest MIR values are observed for the alkene, terpene and BTEX classes. Due to the overall low concentrations of terpenes, i.e., isoprene, limonene and α-pinene measured at the Zurich station, their contributions to OFP contributed to less than 2 % in 2024 to total OFP and thus remain negligible. Alkene contribution to total OFP decreased from 48 % in 2005 to 25 % in 2024. Although OVOC compounds have low MIR values in comparison to the other VOCs measured in ZUE, their contribution to total OFP increased from 8 % in 2005 to 53 % in 2024 due to their increasing concentration and the decreasing abundance of other VOCs. In contrast, BTEX contribution to total OFP significantly decreased from 25 % in 2005 to less than 8 % in

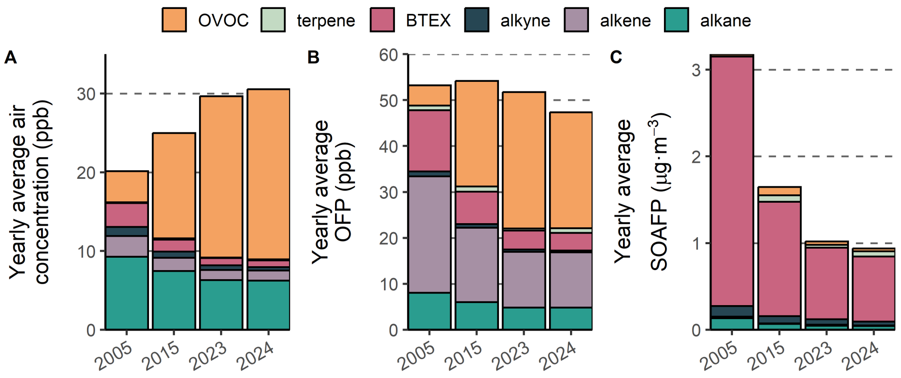

Figure 6. Yearly average ambient air concentrations (A), ozone formation potential (OFP; B) and secondary organic aerosol potential formation (SOAPF; C) of the NMHC classes measured at the Zurich station between 2005 and 2024.

2024. These results show a significant shift in the VOC-induced ozone formation in Zurich over the past two decades towards a marked transition from the predominance of alkenes and BTEX in OFP to the more abundant OVOCs since 2015.

Secondary organic aerosol formation potential (SOAFP) can be calculated in a similar way as OFP with the SOAP yield, instead of the MIR value, determined for each single VOC in Derwent et al. (2000); Gu et al. (2021). Li et al. (2023) showed that aromatic compounds and alkanes contribute most to the formation of low-condensable organic vapors necessary for the growth of new particles, with aromatic compounds accounting for 59 ± 20% of low-volatility organic compounds. In Zurich (Fig. 6, C), although BTEX remain the dominant contributors to SOA formation potential (90 % in 2005; 80 % in 2024), their concentration significantly decreased by 74 % - from 2.9 μg m^{-3} in 2005 to 0.75 μg m^{-3} in 2024. These results show the dominant role of BTEX compounds within the different measured VOC classes in SOA formation.

4 Conclusions

This study focuses on BTEX concentrations in ambient air in Switzerland since 1994. Using continuous online BTEX measurements over three decades, the effect of the implementations of various regulations showed a significant net decrease in BTEX concentrations. As a consequence, the monitoring of ethylbenzene and xylenes at Dübendorf stopped in 2011 due to their low concentrations. The yearly average concentration of toluene and benzene was 4.4 ppb in 1994 and decreased to 0.5 ppb in 2024 in Dübendorf. In addition, changes in the T:B ratio since the introduction of benzene reduction in car fuel in 2000 indicate a shift in dominant sources, with a significant increase in the contribution of solvents to the toluene concentrations during the summer. Although BTEX compounds contributed less to overall ozone formation potential, they accounted for up to 80 % of the SOAFP at the Zurich station in 2024. Given their role as key precursors for both ozone and secondary organic aerosol formation, continuous monitoring of BTEX concentrations in (sub)urban environments remains essential for the development and implementation of effective air quality management strategies.

165 *Data availability.* Data related to VOCs are available at EBAS Database at https://ebas-data.nilu.no/.

Author contributions. Z.L.B. wrote the manuscript with the input from all authors. S.R. was in charged of and supervised the continuous monitoring of BTEX in Switzerland at DUE, ZUE, BER and BRM stations. P.R. helped in processing the collected BTEX data at the monitoring stations. C. H. supervised the data collected within the NABEL framework.

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

170 *Acknowledgements.* We acknowledge the technical staff involved in the BTEX data collection since 1994 at the different Swiss monitoring stations of the NABEL network.

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