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From direct emission factors to inverse and indirect impact factors of road traffic: Influence of emerging and unregulated pollutants

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Abstract. Even though emissions regulations have successfully reduced concentrations of some road traffic-originated air pollutants, road traffic is still an important source of many unregulated emerging pollutants such as semi-volatile ultrafine particles, non-exhaust emissions and volatile organic compounds (VOCs) in urban areas. In addition, road traffic can have season- and location-dependent inverse and indirect effects on the urban aerosol which are not well understood. In our study, we investigate wintertime vehicle fleet emission factors (EF) of road traffic in a street canyon environment in central Helsinki, Finland, by using CO_2 as a tracer for fuel consumption. We report EFs for particle number and size distribution, black carbon, PM_{10}, $PM_{2.5}$, NO_x, NO_2, NO, CO as well as for organic and inorganic chemical components of particulate matter, and VOCs, including aromatic hydrocarbons, alkanes, and polycyclic aromatic hydrocarbons. The obtained fleet EFs of PM and NO_x were considerably higher than required for new vehicles in Europe since 2014, showing the major role of older vehicles and heavy-duty traffic on the urban aerosol. In addition to direct EFs, we show that CO_2-based EF determination enables detection of inverse and indirect effects of road traffic. For example, road traffic had an inverse effect, i.e., negative EF, on O_3 and small ion (< 2 nm) concentrations. Also, we found that road traffic contributes to compounds that are not necessarily considered to be traffic-derived such as terpenoid VOCs. We introduce a term impact factor (IF) to describe the found inverse and indirect effects of road traffic.

1 Introduction

Road traffic is generally known to be a significant contributor to poor air quality in urban areas. Traffic exhaust and traffic-influenced air pollutants such as fine particulate matter ($PM_{2.5}$) and nitrogen oxides (NO_x) have been associated with various adverse health endpoints and premature deaths (Raaschou-Nielsen et al., 2010; Park et al., 2018; Vodonos et al., 2018). Due to the adverse health effects, regulations have been implemented to control vehicle emissions. For example, the first EURO

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emission standard by the European Union (EU) was introduced in 1992, limiting emissions of CO, hydrocarbons, NO_x and particulate matter of vehicles (only diesel). Since 2011, the EURO standards have also regulated the number concentration of emitted nonvolatile particles larger than 23 nm from diesel vehicles (gasoline since 2014). Advancements in vehicular technologies and tightening emission regulations have led to declining trends in many traffic-related pollutants at urban roadside monitoring sites in Europe and globally (Wallington et al., 2022; Garcia-Marlès et al., 2024; Krecl et al., 2024). These pollutants include ultrafine particles (UFPs, < 100 nm), particulate matter (PM_{10} and $PM_{2.5}$), NO, NO_2, SO_2, lead (Pb), and black carbon (BC).

While the emissions of many traffic-related pollutants have reduced due to the regulations, road traffic is still an important source of many emerging pollutants, i.e. species that have potential adverse effects on health and climate but remain unregulated. For example, road traffic is a major source of particles smaller than 30 nm, including nanocluster aerosol (< 3 nm) (Rönkkö et al., 2017). However, only the number concentration of nonvolatile particles larger than 23 nm until 2026 and larger than 10 nm from 2026 onward is regulated in the EURO standards. Diesel particulate filters have proven effective in reducing emissions of BC and accumulation mode particles (> 100 nm) (Saarikoski et al., 2024). Yet, they are still ineffective at controlling nucleation mode particles (< 30 nm) (Damayanti et al., 2023), which are likely associated with (semi-)volatile aerosol emissions. Despite their low contribution to particle mass, UFPs have been linked to adverse health effects (Vallabani et al., 2023; Lloyd et al., 2024) although more data on these health effects are still needed. In addition to UFPs, road traffic is an important source of non-exhaust emissions: There are indications that even majority of PM_{10} emissions of road traffic would be non-exhaust originated (Bukowiecki et al., 2010; Singh et al., 2020). For example, metallic brake-wear particles include compounds that are harmful to health (Grigoratos and Martini, 2015). Non-exhaust emissions will be included in the EURO 7 standard for the first time in 2026.

Alongside the particulate emissions, road traffic contributes to gaseous emissions, including various volatile organic compounds (VOCs) and polycyclic aromatic hydrocarbons (PAHs). VOCs contribute to secondary aerosol formation (Chaturvedi et al., 2023), and organic aerosol is the most abundant compound in PM_1 in Europe (Chen et al., 2022). According to Daellenbach et al. (2020), secondary organic aerosol is the most important contributor to the oxidative potential of PM_{10} in most parts of Europe. In addition, VOCs are participating in ozone formation together with NO_x. Some of the VOCs are also directly harmful, especially, but not limited to, the BTEX group VOCs (benzene, toluene, ethyl benzene, and xylenes) that have been recognized as carcinogenic compounds. Also, effects of VOCs e.g., on central nervous and immune systems have been identified (Li et al., 2021). Ambient VOCs are mainly unregulated, but EU has set a limit for the annual average concentration of benzene in ambient air (Directive (EU) 2024/2881).

In addition to direct effects, traffic emissions can have indirect effects by affecting other airborne compounds. As an example, NO_x and O_3 are known to have inverse dependency: Decreasing NO_x emissions can cause an increase in O_3 concentrations in urban ambient air as NO depletes O_3 (Keuken et al., 2009; Anttila et al., 2011). Also, particle emissions from road traffic increase the condensation and coagulation sink which affect the scavenging of semi-volatile aerosol precursors and UFPs. On the other hand, road traffic increases the concentrations of aerosol precursors in ambient air (Olin

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et al., 2020). Both these attributes can influence new particle formation events as well as growth of UFPs into sizes where they could act as cloud condensation nuclei (Bousiotis et al., 2021).

Emission factors (EFs) derived from roadside measurements are useful when quantifying the impacts of road traffic on ambient air. For example, by measuring CO_2 simultaneously with other pollutants, emissions can be normalized to the amount of fuel burned, as elevated CO_2 levels at roadside measurements likely originate from the nearby road traffic. In contrast to laboratory measurements, roadside EFs include the real-life direct and short-term effects of vehicular emissions, such as the particle formation due to exhaust cooling and dilution. Also, roadside measurements enable the determination of the EFs for the entire vehicle fleet. Yet, roadside EFs are sensitive to site-specific conditions such as meteorology, traffic composition, driving conditions, and built environment (Choi et al., 2016; Pirjola et al., 2016; Wang et al., 2018a, Wang et al., 2021). Hence, they enable estimation of traffic emissions in different conditions and situations. For example, a study by Wang et al. (2018b) found that particle number (PN) EFs were 2.0 times higher in winter compared to summer at an urban near-road site. Conversely, toluene EFs in summer were 2.5 times higher than in winter (Wang et al., 2018b). Thus, varying conditions and seasons can considerably affect the impacts of road traffic on urban air quality. Up-to-date fleet EFs are, therefore, important when estimating how well and quickly technological advances and tightening regulations decrease the impacts of road traffic in varying urban areas with different conditions. Such insights are valuable when considering future regulatory actions.

In earlier studies, EFs from roadside measurements have mainly been determined for particulate matter (mass), black carbon and gases (CO, NO, NO_2). Additionally, some VOC EFs—mostly the BTEX group VOCs—have been studied (e.g., Majumdar et al., 2009; Gaga et al., 2018). For various emerging airborne compounds, fleet EFs have been determined less frequently, including those for several other VOCs, PN down to sizes below 10 nm, ozone (O_3), airborne ions, and chemical components of PM. Furthermore, literature of EFs derived in winter conditions remain limited.

The aim of our study is to extend the CO_2-based fleet EF calculation beyond regulated pollutants to include emerging pollutants and compounds that have not been previously necessarily considered to be directly traffic derived. The road traffic emissions were studied in a street canyon environment in Helsinki city center, Finland, in winter 2022. EFs were calculated for PN (including size distribution), $PM_{2.5}$, PM_{10}, BC, NO, NO_2, NO_x, O_3, alveolar lung deposited surface area ($LDSA^{al}$) as well as for ions and various VOCs and chemical compounds of PM. Furthermore, influence of increased regional and long-range transported pollution events on the determined EFs was investigated. Overall, in addition to up-to-date fleet EFs of wintertime road traffic, the study brings a new and wider perspective on the use and interpretation of EFs determined in ambient measurements to recognize the direct and indirect effects of road traffic on urban air quality.

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2 Methods

2.1 Measurement site and conditions

The intensive measurement campaign of this study was conducted at an air quality monitoring supersite operated by Helsinki Region Environmental Services Authority (HSY) on Jan 17th – Feb 22nd, 2022 (Teinilä et al., 2025). The station located at Mäkelänkatu street canyon (Mäkelänkatu 50, Helsinki; 60.1964N, 24.9521E), next to a main street leading toward the city center of Helsinki. The street width was 42 m, consisting of two pavements, six lanes, and two tramlines lined with trees in the middle. The speed limit was 40 km/h, and the street had many traffic light junctions. The heights of the surrounding buildings were 16–19 m, limiting the dilution process of pollutants. The station was located on the pavement less than 0.5 meters from the lanes. The measurement instruments utilized in this study were located inside the air quality monitoring supersite as well as right next to it in an additional measurement container. The height of inlets was 4 m above ground level (29 m above sea level). A more detailed description of the station with its air flow patterns, profile and in-depth aerosol characterization are provided by Kuuluvainen et al., (2018) and Barreira et al., (2021).

During the measurement campaign, the traffic rate of the street was about 17 000 vehicles per weekday of which the heavy-duty vehicles account for 10 % (statistics from the City of Helsinki). The shares of gasoline cars, diesel cars, vans, city buses and trucks were 45 %, 34 %, 12 %, 6 %, and 4 %, in 2022, respectively (Table S1). The shares of mileage for different EURO emission classes for gasoline cars, diesel cars, vans, city buses and trucks are collected in Table S2.

The weather conditions during the campaign represented typical wintertime conditions in Southern Finland. The mean temperature during the campaign was -1.4 °C (range -10.0 – +2.9 °C), and the temperature was most of the time near zero centigrade. The mean relative humidity was 89 % (58–100 %). The prevailing wind direction was from south to south-east and the mean wind speed was 4.9 m/s (0.59–11.4 m/s). The average mixing height was 408 m (58–844 m). The measurement campaign conditions are described in more detail by Teinilä et al., (2025).

During the campaign three distinct pollution episodes occurred. These episodes took place January 22 (3 p.m.) – January 23 (10 a.m.), January 31 (7 a.m.) – February 5 (4 p.m.), and February 13 (12 p.m.) – February 17 (11 p.m.). During the episodes the contribution of pollution sources other than road traffic at the site increased compared to the non-episodic data. For example, long-range and regionally transported biomass burning-originated aerosols contributed to increased pollutant concentrations measured at the studied site during the episodes. A cold temperature inversion combined with low wind speeds led to the accumulation of pollutants below the boundary layer, particularly during the second episode. The pollution episodes during the campaign are described in more detail by Teinilä et al. (2025).

2.2 Instrumentation

A detailed description of the instruments used during the campaign is provided by Teinilä et al., (2025). Briefly, PN concentrations and size distributions were measured by utilizing a condensation particle counter (CPC) model A20 (Airmodus), Neutral cluster and Air Ion Spectrometer (NAIS) model 5-27 (Airel Ltd.), and a differential mobility particle

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sizer (DMPS). The particle size range of the CPC was 5.4 nm–2.5 µm, whereas the particle size distributions were measured in the size ranges of ~2–20 nm (NAIS), and 11–800 nm (DMPS). NAIS was additionally used to measure air ion size distributions with both polarities in the mobility diameter range of ~0.8–40 nm, the total duration of the measurement cycle being 1.5 min (30 s ions, 30 s offset, 30 s particles –operation modes) (Mirme and Mirme, 2013; Manninen et al., 2016). For the particle size distributions, the negative particle mode was used with the NAIS data (i.e., negative charger and analyzer polarity). DMPS consisted of a Vienna type differential mobility analyzer followed by A20 model CPC, and a scanning time of 9 min was used. The alveolar LDSA^{al} concentration of aerosol particles from 10 to 400 nm was measured using a Pegasor AQ™ Urban instrument (Pegasor Ltd.) (Kuula et al., 2020).

Particulate matter, PM_{10} and $PM_{2.5}$, concentrations were measured using a Fidas 200 fine dust monitor (Palas). BC in PM_1 was measured with an AE33 dual-spot aethalometer (Magee Scientific) based on the attenuation of light at the wavelength of 880 nm (Hansen et al., 1984; Drinovec et al., 2015). A Soot Particle Aerosol Mass Spectrometer (SP-AMS, Aerodyne Research Inc.) was used to investigate the chemical composition of submicron particles (Onasch et al., 2012). It was operated with a 60–90 s time-resolution of which half was measured in mass spectrum mode and half in particle time-of-flight mode.

Gaseous compounds were measured using following analyzers: CO with APMA 360 (Horiba), CO_2 with LI-7000 (LICOR), NO_x with APNA 370 (Horiba), and O_3 using APOA 370 (Horiba). Gaseous PAHs, VOCs and IVOCs were measured with 30–45 min time resolution using an in-situ thermal desorption unit connected to a gas chromatograph and mass spectrometer (TD-GC-MS, PerkinElmer TurboMatrix 350, Clarus 680, and SQ 8 T). The gaseous compounds were reported at +20 °C and particles in ambient temperatures according to the European Union standards.

2.3 Data processing

The EFs were determined to represent the influence of the entire vehicle fleet passing the measurement station. EF for each pollutant metric was calculated by comparing the measured pollutant concentration with the concurrently measured CO_2 with a time resolution of 1 min. The EF of CO_2 (EF_{CO_2}) was assumed to be 3.141 g/kg_{fuel}, determined for average Finnish road traffic (Yli-Tuomi et al., 2005). The EF calculation method is similar to previous studies, such as Rönkkö et al. (2017) and Lintusaari et al. (2023). The EFs for each studied pollutant (EF_P) were calculated assuming the ideal gas law and standard conditions (STP) by using an equation

$$EF_P = ER_P \frac{EF_{CO_2} R T_{std}}{M_{CO_2} p_{std}} \quad (1)$$

where ER_P is the emission ratio for each pollutant compared to the concurrently measured CO_2, R is the molar gas constant (8.3144621 J/(K mol)), T_{std} is the standard temperature (273.15 K), M_{CO_2} is the molar mass of CO_2, and p_{std} is the standard pressure (10^5 Pa). The ER_P was determined by utilizing a linear fit between the studied pollutant metric and CO_2. Before that, the CO_2 data was divided into 5 ppm bins after which the measured pollutant concentrations were averaged by using geometric mean over the 5 ppm CO_2 bins. The linear fit was applied for the averaged data to determine emission ratios for

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each pollutant in the CO_2 range of 420–465 ppm, representing 99.2 % of the non-episodic data. The lowest CO_2 concentration of the dataset was 420.7 ppm. When CO_2 concentrations exceeded approximately 465 ppm, the average pollutant concentrations for some studied pollutants began to deviate from the linear fit, which was attributed to the limited number of data points. To avoid confusion with the correlations, the R^2 value obtained from linear fit after averaging is called R^2 of the averaged linear fit, as a distinction from the raw data correlation. In addition, 95 % confidence intervals (CI) were calculated for the averaged linear fits. Figure 1 presents an example of how the PN emission factor was calculated.

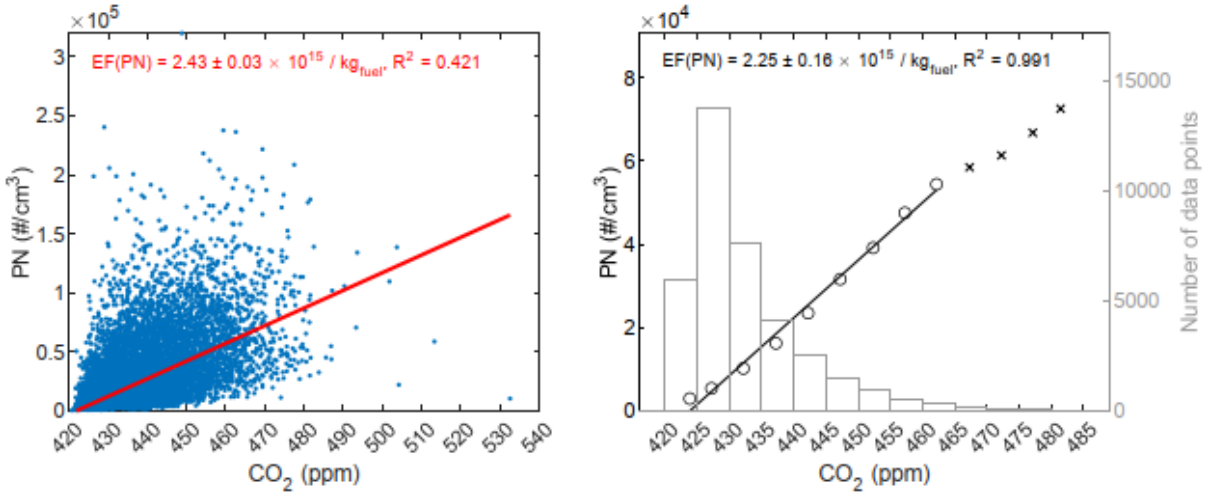

Figure 1: Left: Scatter plot of all individual non-episodic 1-min-average PN and CO_2 concentrations, together with a linear fit. Right: Average (geometric mean) PN and CO_2 concentrations over 5 ppm CO_2 concentration bins, together with a linear fit, which was calculated in the range of 420–465 ppm of CO_2 (circular markers). Also, the number of data points (1 min) in each CO_2 concentration bin shown is shown in the histogram.

In case of the DMPS, NAIS, SP-AMS, and VOC sampling, the measurement intervals were longer than 1 min. Because of the scanning-type of measurement, single datapoint results with the DMPS and NAIS can be uncertain due to the rapidly varying concentrations caused by the road traffic. Hence, with both NAIS and DMPS, 1-hour-resolution was used in the EF calculation. With the SP-AMS, a 10-min-resolution was utilized. The VOC EFs were calculated either with 30 or 45 min resolution, depending on the sampling time. The utilized time resolutions in the EF calculation are collected in Table S3. Due to the longer time resolution with these instruments, some of the bins with the highest CO_2 concentration had limited number of data points. Bins having at least 10 data points were included in the fit; as a result, the upper limits of the fit calculation with these instruments were 450 (for NAIS), 455 (DMPS and VOCs), and 465 (SP-AMS) ppm of CO_2.

In general, the EF calculation method assumes that the measured pollutant and CO_2 dilute at the same rate, and the dilution is faster than other atmospheric processes affecting the measured concentrations (Rönkkö et al., 2023). However, it should be noted that the method is susceptible to non-vehicle influences (Wang et al., 2018a), such as regional wood combustion and long-range transported pollution. On the other hand, road traffic should clearly be the most significant local contributor to the measured concentrations at the supersite (e.g., Saarikoski et al., 2021). To focus on the effects of road traffic, the data

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measured during the mentioned pollution episodes (section 2.1) was not considered in sections 3.1-3.3. The effect of the pollution episodes is investigated in section 3.4.

In addition to EFs, some of the studied pollutant metrics had inverse trends as a function of increasing CO_2 concentration, i.e., the road traffic's contribution to the concentrations of these pollutants was negative (Section 3.2). In these cases, it is not reasonable to interpret the results as EFs as, in principle, EFs describe the amount of emitted pollutants per certain activity (e.g., kg of fuel burnt). Therefore, we use a term impact factor (IF) when reporting the results of pollutant metrics that had an inverse trend as a function of increasing CO_2 concentration, i.e., negative EF. Also, the term IF is used with parameters that are not necessarily considered to be emitted from vehicles itself, but the increased road traffic activity still contributes to their concentrations, like terpenoid VOCs (Section 3.3).

2.4 Bottom-up emission inventory EF calculation

The obtained $PM_{2.5}$ and NO_x EFs were compared to bottom-up emission inventory -based EF calculation. The bottom-up emission inventory was based on the emission factor database of the Handbook of Emission Factors for Road Transport (HBEFA version 4.1). The average EF for NO_x and $PM_{2.5}$ on the studied street were calculated using the estimated shares of mileages of different vehicle types and emission classes (Table S1-2) assuming the mean driving speed of 37 km/h (i.e., close to the speed limit 40 km/h on the street). The estimated mileage shares of different vehicle types and emission classes on the studied street were based on the statistics from the City of Helsinki and VTT's Lipasto Aliisa model 2021. The bottom-up EFs were calculated as a yearly average in 2022.

3 Results and discussion

3.1 Emission factors of regulated and non-regulated emerging pollutants

In this chapter EFs of different variables are presented. The EFs of PN, BC, PM_{10}, $PM_{2.5}$, $LDSA^{al}$, NO_x, NO_2, NO, and CO are presented in Table 1 (Figure S1), whereas EFs for organics, nitrate, sulfate, ammonium, and chloride are presented in Table 2 (Figure 6 and S9) and for VOCs and PAHs in Table 3 (Figure S3-S5).

3.1.1 Particle number and mass

The concentrations of PN, BC, PM_{10}, $PM_{2.5}$ and $LDSA^{al}$ were strongly connected with the CO_2 concentration, and the R^2 values of the averaged linear fits were above 0.93, showing that the pollutants were mainly derived from road traffic at the studied site.

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Table 1: Emission factors (EF) of PN > 5.4 nm, BC, PM_{10}, $PM_{2.5}$, $LDSA^{al}$, NO_x, NO_2, NO and CO, together with 95 % confidence intervals (CI) and R^2 values of the averaged linear fits.

Metric	EF (± 95 % CI)	R^2 of the averaged linear fit
PN > 5.4 nm (#/kg_{fuel})	*$(2.3 \pm 0.2) \cdot 10^{15}$*	*0.991*
BC (g/kg_{fuel})	*0.052 ± 0.002*	*0.998*
PM_{10} (g/kg_{fuel})	*0.31 ± 0.05*	*0.954*
$PM_{2.5}$ (g/kg_{fuel})	*0.11 ± 0.02*	*0.937*
$LDSA^{al}$ (m^2/kg_{fuel})	*1.14 ± 0.04*	*0.998*
NO_x (g/kg_{fuel})	*5.59 ± 0.23*	*0.997*
NO_2 (g/kg_{fuel})	*2.41 ± 0.18*	*0.990*
NO (g/kg_{fuel})	*1.98 ± 0.21*	*0.981*
CO (g/kg_{fuel})	*4.54 ± 0.28*	*0.993*

The obtained PN EF aligns well with earlier studies conducted at the same site. Hietikko et al., (2018) reported PN EFs of $3.3 \cdot 10^{15}$ #/kg_{fuel} (> 3 nm) and $1.5 \cdot 10^{15}$ #/kg_{fuel} (> 7 nm) measured in May 2017 whereas Lintusaari et al. (2023) reported $1.1–1.3 \cdot 10^{15}$ #/kg_{fuel} (> 3 nm) and $0.46–0.56 \cdot 10^{15}$ #/kg_{fuel} (> 10 nm) in May 2018. During the SARS-CoV-2 pandemic in March 2021, PN EFs of $1.8 \cdot 10^{15}$ #/kg_{fuel} (>2.5 nm) and $0.69 \cdot 10^{15}$ #/kg_{fuel} (> 10 nm) were measured by Lepistö et al. (2023). Therefore, it seems that PN EFs have stayed rather constant in central Helsinki during recent years. On the other hand, Enroth et al. (2016) reported fleet PN EFs of $4.9–11.6 \cdot 10^{15}$ #/kg_{fuel} for particles larger than 2.5 nm measured on highways in Helsinki in 2012, suggesting reduction in PN emissions from traffic. If fuel consumption of 11.0 l/100 km was assumed (i.e., 1 kg/10.7 km, with estimated fuel density of 0.85 l/kg), the PN EF would be approx. $2.1 \cdot 10^{14}$ #/km, representing considerably higher EF than the limit for primary emission particles (> 23 nm) utilized by the EU in the EURO standards ($6.0 \cdot 10^{11}$ #/km), likely related to the emissions of (semi-)volatile compounds. The result suggests that the current emission standards are not probably enough in reducing the emissions of the smallest ultrafine particles from road traffic, which has been reported also by Damayanti et al. (2023) and Garcia-Marlès et al. (2024). On the other hand, it should be noted that the winter conditions probably increase the reported EFs in this study which may hide the effect of cleaner vehicle technology. For example, at a comparable ambient traffic site in Toronto downtown, the wintertime PN EF (> 7 nm, fleet mean) was approx. $1.0 \cdot 10^{15}$ #/kg_{fuel} which was the highest among the four seasons (spring: $0.9 \cdot 10^{15}$, summer: $0.5 \cdot 10^{15}$, fall: $0.8 \cdot 10^{15}$ #/kg_{fuel}) (Wang et al., 2018b). Similarly, Saha et al. (2018) found that PN EF was 300 % higher in winter than in summer at a site near a freeway.

The size-resolved PN EF, based on the data of DMPS and NAIS, is shown in Figure 2. The shapes of the EF size distributions determined from the DMPS and NAIS data seemed to agree well with each other, but, with particles smaller than 30 nm, the EFs from NAIS data were higher compared to the DMPS which is important to acknowledge. This difference is likely related to the varying measurement methods, especially the different scanning times of the measurements.

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Based on the NAIS data, the highest PN EFs were determined for particles around the size of 7.5 nm. Still, the peak of the EF size distribution could be located even below the obtained 7.5 nm: For example, Lintusaari et al. (2025) found that the PN EFs were highest in the smallest size fraction that they analyzed (2.5–4.0 nm) next to a busy arterial road in Prague, Czech Republic. Also, Rönkkö et al. (2017) have reported traffic to be a significant source of nanocluster aerosol. Hence, it may be possible that the rapid changes in nanocluster aerosol concentration cannot be completely observed from NAIS block data with the utilized measurement cycle. As EURO7 (11/2026) (Regulation (EU) 2024/1257) and the new EU air quality directive (Directive (EU) 2024/2881) have introduced 10 nm as the lower size limit of particle number measurements, the observations highlight that road traffic produces also a high number of particles that are not currently (or planned to be) regulated.

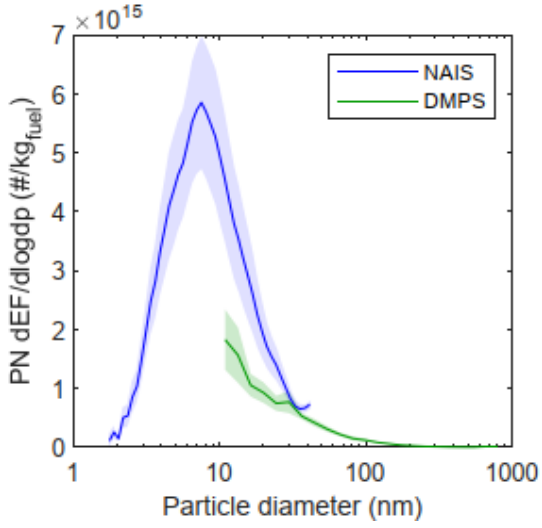

Figure 2. PN EF size distributions measured by NAIS and DMPS with 95 % confidence intervals (shaded areas).

The review by Rönkkö et al. (2023) shows a large variation of BC EFs measured at road sites in different studies: 0.015–2.4 g/kg_{fuel}, average being 0.17 g/kg_{fuel} and median and 0.054 g/kg_{fuel}. Our value, 0.052 g/kg_{fuel}, is close to the median and the value reported by Lepistö et al. (2023) at the same site during winter 2021 (0.042 g/kg_{fuel}). Our BC EF was 47 % of the determined $PM_{2.5}$ EF (0.11 g/kg_{fuel}). This result is reasonable as organic compounds contribute considerably to PM emissions of vehicles (see Section 3.1.3). With the fuel consumption of 11.0 l/100 km, the reported BC EF (0.0049 g/km) would be close to the current EURO PM EF limit (0.0045 g/km), but the obtained $PM_{2.5}$ EF (0.0103 g/km) would clearly exceed this limit. This result could partly be related to heavy-duty traffic as well as to use of older vehicles on the studied street. Also, non-exhaust emissions could increase the $PM_{2.5}$ EF. In this case, it is, however, not likely that the non-exhaust emissions would significantly affect the $PM_{2.5}$ EF because of the clear contributions by organics and BC (see Section 3.1.3). Still, the measured PM_{10} concentrations correlated strongly with the CO_2, suggesting that EF for non-exhaust emissions, such as road dust, tire-wear, and braking, could be determined indirectly even though they do not originate from combustion processes like CO_2 does. The obtained PM_{10} EF was 2.81 times higher than the $PM_{2.5}$ EF, suggesting contribution by non-exhaust

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emissions even though winter conditions typically limit the effects of non-exhaust emissions compared e.g., to spring (Kupiainen et al., 2016; Stojiljkovic et al., 2019). In addition, the diurnal variation shows that PM_{10} EF increased during daytime traffic and at other times was close to zero, supporting the idea of potential non-exhaust contribution on the metric (Figure S2). With the fuel consumption of 11.0 l/100 km, the PM_{10} EF would be 0.0290 g/km, where the coarse fraction (PM_{10}-$PM_{2.5}$) would be 0.0187 g/km.

The obtained $PM_{2.5}$ and NO_x (Section 3.1.2) EFs were compared to the bottom-up emission inventory EF calculation. The emission inventory based $PM_{2.5}$ EF was 0.0123 g/km (Table S4), being slightly higher than the obtained CO_2 based $PM_{2.5}$ EF of 0.0103 g/km (with estimated fuel consumption of 11.0 l/100km). Interestingly, according to the bottom-up calculation, the contribution of gasoline vehicles to the total road traffic originated $PM_{2.5}$ emissions on the street were only 6 %, even though they had 45 % share of the total mileage. With diesel vehicles, the share of $PM_{2.5}$ emissions was 38 % with 34 % share in total mileage. This result is related to the relatively high share of diesel vehicles without a diesel particulate filter (25 %, EURO4 or older). Vans, city diesel buses, and trucks had 22 % share in total mileage, but they contributed 56 % of total $PM_{2.5}$ emissions. These results support the idea that the fleet $PM_{2.5}$ EFs on the street still clearly exceed the current EURO standard limit especially because of heavy duty traffic and the use of vans and older diesel vehicles.

To highlight the potential health effects contributed by particle emissions from road traffic, an EF for $LDSA^{al}$ was also determined. $LDSA^{al}$ is especially dependent on the ultrafine particles, which deposit efficiently in the lung alveoli, as well as on BC. However, it should be noted that the concept of $LDSA^{al}$ EF is tricky: the unit (m^2/kg_{fuel}) describes the surface area of particles that reach the lung alveoli of a single individual per kg of burnt fuel. Hence, the metric assumes that the individual breathes all the exhaust that is emitted as a consequence of burning one kg of fuel, and this breathing occurs at the same exact location where the measurement is conducted. Despite this challenge, strong connection between $LDSA^{al}$ and CO_2 shows the clear contribution of road traffic on the metric. Also, by comparing $LDSA^{al}$ EFs in different locations, effects related to changes in the size distribution of emitted particles could be assessed because the relative change in $LDSA^{al}$ is likely not the same as the relative change in PN or in BC (e.g., Lepistö et al., 2025).

3.1.2 Nitrogen oxides and carbon monoxide

According to the NO and NO_2 EFs in Table 1, the (mass-based) NO_2 EF was higher than the NO EF. However, when considering the volumetric emissions, the shares of NO and NO_2 emissions were 56 % and 44 %. Fresh traffic exhaust typically has higher fraction of NO even though NO_2/NO_x ratio has been suggested to be increasing from EURO0 to EURO6 with passenger cars (Carslaw and Rhys-Tyler, 2013; Carslaw et al., 2019a). Still, the volumetric fraction of NO_2 has typically been below 30 % with passenger vehicles (Carslaw et al., 2019b). Therefore, it is likely that at the studied street canyon, atmospheric oxidation increased the fraction of NO_2. For example, in a study by Xiang et al. (2025), NO_2/NO_x ratios measured from a truck plume were 3-4 times higher than in the tailpipe measurements, suggesting that substantial NO_2 formation occurs in exhaust plumes after the emission. NO_2 formation does not, however, change the total NO_x emission, but it is important to note that ratios of NO and NO_2 can be considerably different in ambient air compared to tailpipe

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measurements, even close to road traffic. Regarding the CO EF, it is generally known that road traffic is not anymore a significant source of CO, a finding that is also supported by our study. For example, the EURO6 limit for CO emissions of passenger vehicles is 1.0 g/km, and the obtained EF was clearly below if 11 l/100 km fuel consumption is assumed (0.42 g/km).

The bottom-up emission inventory based NO_x EF was 0.545 g/km at the studied street canyon. This result is close to the obtained CO_2 based EF, which was 0.522 g/km with the fuel consumption of 11 l/100 km. Together with the emission inventory based $PM_{2.5}$ EF, the results show that the CO_2 based fleet EF determination agreed well with the emission inventory based calculations. Also, the results suggest that the estimated average fuel consumption of 11 l/100 km is reasonable at the studied street canyon. In comparison with the bottom-up $PM_{2.5}$ EFs, the share of NO_x contributed by gasoline vehicles was still low (5 %), but the contribution of diesel vehicles in total NO_x emissions was clearly higher (52 %) than in case of $PM_{2.5}$ (38 %). The contribution of vans, diesel city buses and trucks was 43 % of total NO_x emissions.

Overall, both the CO_2 and emission inventory based NO_x EFs indicate that the fleet NO_x EF is significantly higher than the EURO6 emissions standard limits of 0.06–0.08 g/km. Similar to $PM_{2.5}$, this result is most likely related to the heavy duty traffic and the use of vans and older diesel vehicles. In general, it is good to note that NO_x emissions have been shown to be temperature and season dependent (Saha et al., 2018; Wang et al., 2018b; Grange et al., 2019; Li et al., 2020). For example, Saha et al. (2018) measured a 20 % higher NO_x EF in winter (4.7 g/kg_{fuel}) than in summer (3.8 g/kg_{fuel}). Also, colder temperatures may cause cooler engine temperature and influence the operation of exhaust after treatment systems (Suarez-Bertoa and Astorga, 2018) used in vehicles. Hence, the winter-time conditions could have affected the obtained CO_2 based NO_x EF. On the other hand, the CO_2 based EF agreed well with the emission inventory based EF, suggesting that the winter-time condition is not likely the most important reason behind the result. Overall, it is important to note that the EURO6 emissions standard has been applied since 2014 and, yet the fleet NO_x and $PM_{2.5}$ emissions still clearly exceed the limit. Exceedances were also observed with particle number, but, in case of PN, it is easy to understand that the regulations do not include the volatile particle fraction. However, in case of nitrogen oxides and $PM_{2.5}$, it is an important question to ask, why the fleet EFs are that much higher compared to the regulations. As mentioned, heavy-duty traffic and older passenger vehicles (i.e. so called super emitters) could partly explain this result which has been shown e.g., by Ježek Brecelj et al. (2025). Thus, policies to limit the emissions of individual super emitters would be beneficial when reducing the $PM_{2.5}$ and NO_x of traffic, in addition to emission regulations of new vehicles.

3.1.3 Chemical composition of submicron PM

Organics had clearly the highest EF (74.2 mg/kg_{fuel}) of the studied chemical compounds. Organics had also a strong R^2 of the averaged linear fit (0.977), whereas other compounds, i.e., sulfate, ammonium, nitrate and chloride, had larger confidence intervals. Therefore, road traffic may not necessarily have been a direct source of these compounds, especially in case of chloride and sulfate. The results show that PM mass emissions from road traffic are mainly dominated by organic compounds (74.2 mg/kg_{fuel}) and BC (52.5 mg/kg_{fuel}), organics to BC -ratio being approx. 1.4. The sum of the EFs of BC and

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organics (127 mg/kg$_{fuel}$) slightly exceeds the determined EF of $PM_{2.5}$ (110 mg/kg$_{fuel}$), indicating that there are uncertainties when comparing the results obtained with the different measurement techniques and time-resolutions. For example, the lowest detected particle size with the Fidas 200 measurement is 0.178 µm, which could indicate underestimation of fresh traffic particles (e.g., BC) in $PM_{2.5}$.

Table 2: EFs, including confidence intervals (CI), and R^2 values of the averaged linear fits for the chemical components measured by the SP-AMS.

Component	EF and 95 % CI (mg/kg$_{fuel}$)	R^2 of the averaged linear fit
Organics	74.2 ± 8.6	0.977
Nitrate (NO_3^-)	7.6 ± 3.1	0.775
Sulfate (SO_4^{2-})	0.3 ± 1.9	0.011
Ammonium (NH_4^+)	3.7 ± 2.0	0.667
Chloride (Cl^-)	0.77 ± 0.36	0.728

The results of the significant contributions of BC and organics agree well with laboratory studies performed with modern technologies, where the relative fraction of organics has been especially high with cars including gasoline/diesel particulate filters (Karjalainen et al., 2019; Saarikoski et al., 2024). However, according to authors knowledge, fleet EFs of different chemical components of PM have rarely been reported. In a highway study in Helsinki, fleet EFs for organics have been between 240 and 330 mg/kg$_{fuel}$ (Enroth et al., 2016), and for heavy-duty diesel trucks in a highway tunnel in San Francisco Bay, 240 mg/kg$_{fuel}$ (Dallmann et al., 2014). These are significantly higher than our result, 74.2 mg/kg$_{fuel}$, which is reasonable when considering the general reduction of PM mass emissions from road traffic during recent years.

Overall, the results in Table 2 show that the CO_2-based EF fleet determination is a suitable method to investigate the emissions of different chemical compounds of particles. Typically, the contribution of road traffic to urban air quality and aerosol chemical composition is estimated by using e.g., matrix-factorization methods. Our results show that CO_2-based EF calculation could provide an alternative and supporting approach to estimate the effects of road traffic on certain aerosol compounds. One benefit of the CO_2-based method is that it provides a clear link between the measured concentrations of ambient pollutants and the activity producing those pollutants.

3.1.4 Volatile organic compounds

All 18 studied aromatic hydrocarbon compounds as well as gaseous PAHs (Table 3) had a strong correlation with CO_2 (R^2 of the averaged linear fit > 0.76). In addition, most alkane hydrocarbons correlated well with CO_2. Clearly, the highest fraction of the studied VOCs were aromatic hydrocarbons, especially toluene and p/m-xylene. Sums of the aromatic hydrocarbon, alkane and gaseous PAH EFs in Table 3 were 149.2, 28.4, and 1.8 mg/kg$_{fuel}$, respectively. The high fraction of aromatic

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hydrocarbons was expected but, interestingly, in comparison e.g., with studies conducted in China, the relative fraction of alkanes was lower (Qi et al., 2021; Liu et al., 2024). In general, the relative contribution of alkanes has decreased with stricter emission regulations (Liu et al., 2024). Also, total VOC emissions from vehicle exhaust have strongly decreased during the last decades (Harrison et al., 2021). As the emission regulations, fuels and vehicle fleets in general vary in different countries, these findings highlight the importance of understanding VOC emissions at the local level. In addition to these factors, also weather conditions can play a role in the VOC emissions of vehicles. For example, Wang et al. (2018b) studied toluene EFs in Toronto downtown across four seasons from 2013 to 2015, and found that the EF was the lowest in winter, about 90 mg/kg$_{fuel}$, and highest in spring, 220 mg/kg$_{fuel}$, potentially explained by the seasonal variation with changes in seasonal fuel composition. The local level understanding is also important since the direct effects as well as the O_3 (Carter, 1994) and secondary aerosol (Zhao et al., 2014) formation potentials vary considerably with different VOCs. In general, the information on fleet EFs of VOCs in different locations is currently rather scarce. The utilized CO_2 based fleet EF determination enables a potential and relatively simple method to estimate vehicle fleet VOC EFs in different locations and conditions.

Table 3. EFs of studied aromatic hydrocarbon, alkane and gaseous PAH VOCs with confidence intervals (CI) and R^2 values of the averaged linear fits.

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Class	VOC	EF and 95 % CI (mg / kg_{fuel})	R^2 of the averaged linear fits
Aromatic hydrocarbons	Benzene	*18.4 ± 2.8*	*0.972*
	Toluene	*47.2 ± 2.8*	*0.996*
	Ethylbenzene	*10.1 ± 1.2*	*0.982*
	p/m-xylene	*33.1 ± 3.1*	*0.989*
	Styrene	*1.6 ± 0.4*	*0.920*
	o-xylene	*11.3 ± 1.1*	*0.987*
	Propylbenzene	*1.8 ± 0.2*	*0.986*
	3-ethyltoluene	*5.2 ± 0.7*	*0.975*
	4-ethyltoluene	*2.2 ± 0.3*	*0.975*
	1,3,5-trimethylbenzene	*2.4 ± 0.3*	*0.982*
	2-ethyltoluene	*1.8 ± 0.2*	*0.985*
	1,2,4-trimethylbenzene	*8.3 ± 0.8*	*0.988*
	1,2,3-trimethylbenzene	*1.8 ± 0.3*	*0.975*
	1,3-diethylbenzene	*0.4 ± 0.1*	*0.761*
	1,4-diethylbenzene	*2.1 ± 0.4*	*0.946*
	2-propyltoluene	*0.4 ± 0.1*	*0.955*
	2-ethyl-p-xylene	*0.6 ± 0.1*	*0.967*
	1,2,4,5-tetramethylbenzene	*0.5 ± 0.1*	*0.935*
Alkanes	Heptane	*1.5 ± 2.8*	*0.192*
	Octane	*2.0 ± 2.2*	*0.405*
	Nonane	*3.0 ± 0.5*	*0.963*
	Decane	*6.0 ± 2.0*	*0.879*
	Undecane	*5.6 ± 1.3*	*0.940*
	Dodecane	*3.9 ± 1.0*	*0.920*
	Tridecane	*3.3 ± 1.0*	*0.893*
	Tetradecane	*2.0 ± 0.8*	*0.836*
	Pentadecane	*1.1 ± 0.5*	*0.827*
Polycyclic aromatic hydrocarbons (PAHs)	Naphtalene	*1.7 ± 0.2*	*0.986*
	Acenaphtylene	*0.08 ± 0.03*	*0.801*

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3.2 Traffic-related inverse impact factors

3.2.1 Ozone

In addition to EFs, where concentration of a certain pollutant increases as a function of CO_2, road traffic can have negative contributions to some pollutant metrics. For example, O_3 was inversely proportional to CO_2, i.e., its IF was negative (-2.17 g/kg_{fuel}) (Figure 3). The O_3 level was the highest when CO_2 concentration was at its lowest level (420-425 ppm) and decreased steadily as CO_2 increased. O_3 was almost depleted at around 465 ppm of CO_2 and was near zero at higher CO_2 concentrations. Especially, when considering the data measured during the pollution episodes (Figure 3), it can be seen that at the same point the increase of NO_2 as a function of CO_2 almost stopped, whereas the concentration of NO increased at a steeper pace (see also the diurnal variation of NO_x, NO_2, NO, and O_3 EFs in Figure S6). This behavior can be explained by the chemical reactions where O_3 oxidizes NO into NO_2. During the pollution episodes, atmospheric mixing and dispersion were reduced (see increase in CO_2), explaining why the behavior was more clearly observed during the pollution episodes. Significant differences in the O_3 concentrations during inversion and non-inversion events have been reported before (see e.g., Janhäll et al., 2006).

In the context of long-term air quality trends, O_3 levels have gradually decreased in urban environments since the 1960s (Wallington et al., 2022) but, unlike many other pollutants, the concentrations have increased near traffic sites in Europe (Garcia-Marlès et al., 2024). At the measurement site of the present study, an increasing trend in O_3 concentrations has been identified during 2015-19, with an annual growth rate of 3.52 % (subject to large uncertainties), while NO and NO_2 levels decreased by 12.53 % and 8.58 % per year, respectively (Garcia-Marles et al., 2024). This pattern is in line with observations from many European air quality monitoring stations. Hence, as NO_x emissions from road traffic are expected to continue decreasing, it is possible that exposure to O_3 in urban areas will increase. Therefore, controlling tropospheric O_3 levels likely remains a significant challenge in terms of urban air quality in the future. The varying decreasing trends of NO and NO_2 observed by Garcia-Marlès et al., 2024 are also interesting in relation to the findings shown in Figure 3. As NOx emissions decrease, it is more likely that O_3 is not depleted at traffic sites which would enhance the oxidation of NO to NO2, thereby increasing the NO_2/NO_x -ratio. Increasing NO_2/NO_x -ratios have also been found in emissions of newer cars (Carslaw and Rhys-Tyler, 2013; Carslaw et al., 2019a). In current air quality recommendations by the WHO and the EU's new air quality directive (Directive (EU) 2024/2881 WHO, 2021), only the concentration of NO_2 is considered. Thus, it is worth discussing whether the NO_2 concentration is the most appropriate metric to estimate the air quality impacts of NO_x emissions in highly trafficked locations.

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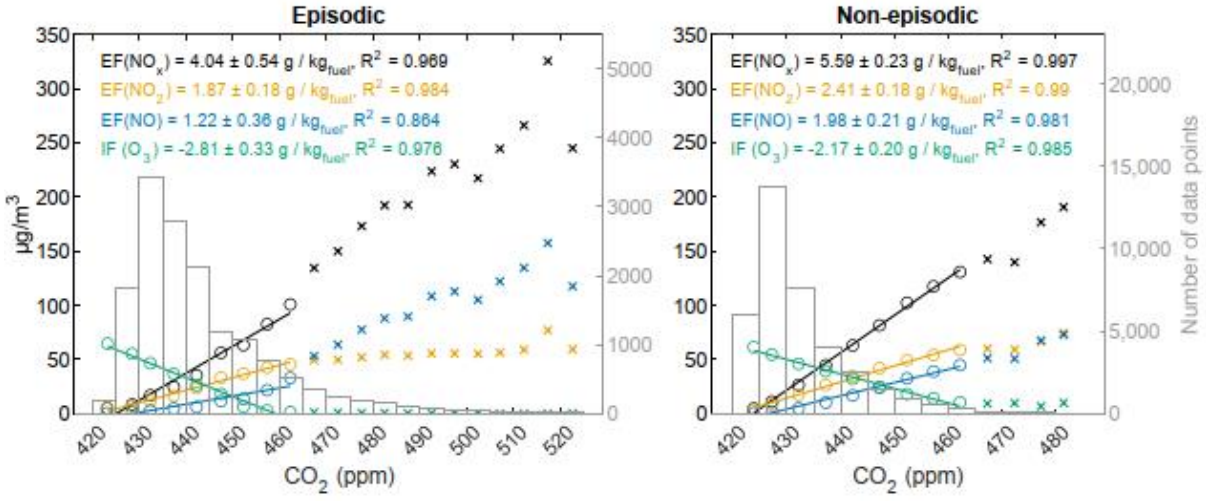

Figure 3: NO_x, NO, NO_2 and O_3 concentrations as a function of CO_2 concentration and the determined EFs for NO_x, NO, NO_2 and the inverse IF for O_3, including 95 % confidence intervals and R^2 of the averaged linear fits. The episodic data on left and non-episodic data on right. Notice the different x-axis. Also, the number of data points in each CO_2 concentration bin is shown.

3.2.2 Small ions

Ions, including electrically charged small particles, are constantly present in the atmosphere. Ions are formed, for example, due to ionization of air molecules by cosmic rays, natural gamma radiation and Radon decay. Also, traffic is known to produce significant amounts of ions via vehicle exhaust (Yu et al., 2004; Jung and Kittelson, 2005; Maricq, 2006; Hirsikko et al., 2007; Ling et al., 2010). Figure 4 presents the number concentrations of positive ions in the size ranges of 0.8–1.5 nm, 1.5–5.0 nm, and 5.0–38.5 nm as a function of CO_2, respectively. Also, the size-resolved IFs of each negative and positive ion size bin are shown.

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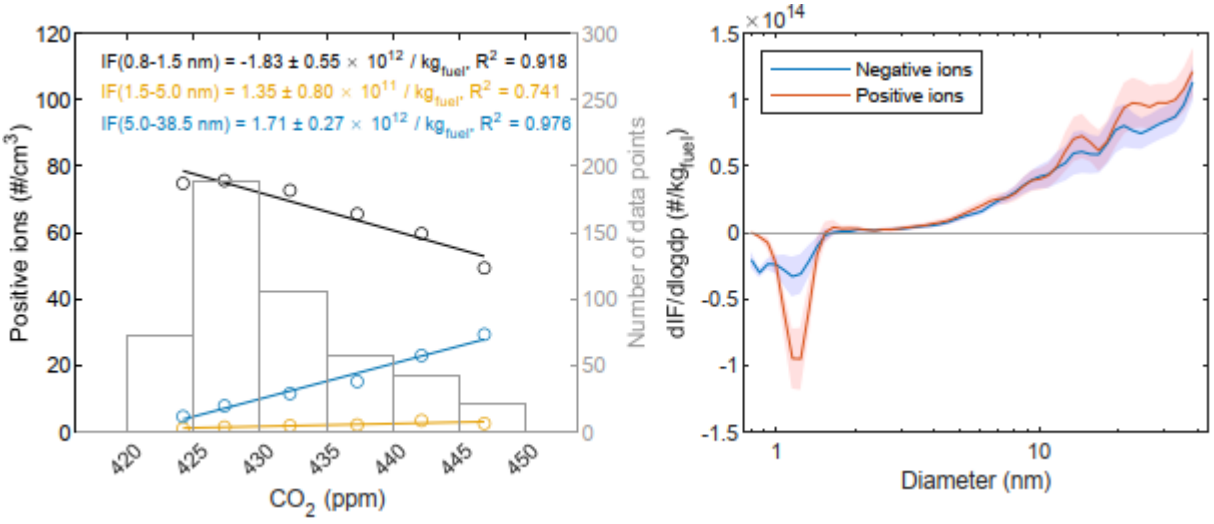

Figure 4: Left: Concentrations of selected positive ions with mean diameters of sizes 1.15 nm, 2.37 nm and 38.5 nm as a function of CO_2 concentration and the determined IFs with 95 % confidence intervals and R^2 of the averaged linear fits. Also, the number of data points in each CO_2 concentration bin is shown. Right: Size-resolved IFs of negative and positive ions with 95 % confidence intervals (shaded areas).

The results show that the small ions (< ~2 nm, Tammet, 1995) have inverse IFs, i.e., their concentration decreases as the contribution of traffic increases, whereas the concentrations of intermediate ions (3-7 nm, Tammet, 1995) and larger ions increase as a function of increased traffic. The negative IFs for the smallest ions may be a result of increased coagulation sink: As shown in Section 3.1, increasing CO_2 concentration increased particle concentrations, which, in principle, should increase the rate at which ions collide with other particles. On the other hand, also recombination could affect the inverse IFs of the small ions which could partially explain why the impact of CO_2 is more significant with the positive ions in Figure 4. This result could be related also to the general higher concentrations of positive ions (Figure S8). The road traffic's inverse contribution to small ions can also be seen when examining the diurnal ion concentration variation (Figure S8), where the concentration of small ions is reduced during rush hours compared to less trafficked hours. The negative effect of traffic on small ion concentrations has been observed before e.g. by Hirsikko et al., (2007). Interestingly, direct engine exhaust measurements have indicated that road traffic is a source of these smallest ions (Lähde et al., 2009). Thus, the results show a paradox: Road traffic emits small sub-2 nm ions but, at the same time, increased road traffic decreases the concentration of these smallest ions in ambient air.

Road traffic is a source of secondary aerosol precursors, but it is also a sink for small ions, which both have impacts on new particle formation. In general, ion-induced new particle formation is not considered as a dominant mechanism of new particle formation in urban areas even though the process can be important in clean and pristine environments (Lehtipalo et al., 2025). Overall, the analysis of the small ion concentrations as a function of CO_2 enables in-detail detection of the effects of road traffic as a sink for small ion concentration. By combining this analysis with size-resolved EF of particle number, the method could be potential for estimating the production and removal processes of ions in urban environments.

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3.3 Traffic-related positive impact factors of terpenoids

Many terpenoid VOCs, which are typically considered to originate mainly from biogenic sources, had a clear correlation with CO_2 at the Supersite (Figure 5, Figure S7 and Table 4). Out of the studied 8 terpenoids, only β-pinene and 1,8-cineol had poor correlation (Table S5) with CO_2 in the averaged linear fits. The connection with CO_2 seemed to be especially strong with α-pinene and limonene (IFs > 2 mg/kg$_{fuel}$). In winter, natural sources should emit negligible amounts of terpenoids (Hellén et al., 2021; Hakola et al., 2023). Elevated terpenoid concentrations have been observed in urban areas, and the elevated concentrations have been linked to population density, indicating human influence. For example, wood combustion, detergents and cosmetics have been suggested as potential sources of terpenoids in urban areas (Gilman et al., 2015; McDonald et al., 2018; Coggon et al., 2021). However, the results suggest that terpenoids could be more directly linked to road traffic than previously considered. In previous source apportionment studies, some fractions of terpenoids, especially isoprene and α-pinene, have been included in factors linked to traffic and combustion (Borbon et al., 2001; Hellén et al., 2012; Xiong et al., 2020; Zhang et al., 2020) but, overall, the mechanism linking road traffic to terpenoid emissions remains poorly understood. As limonene is found in many cleanser products, the correlation with the CO_2 and road traffic could potentially be related to the number of recently washed vehicles on the street and the use of windshield washer fluid. As the contribution of natural sources in this study should be minimal, it is likely that road traffic contributes directly also to the other terpenoids. Hence, the results indicate that road traffic could have previously unconsidered direct effects on terpenoid concentrations in urban environments.

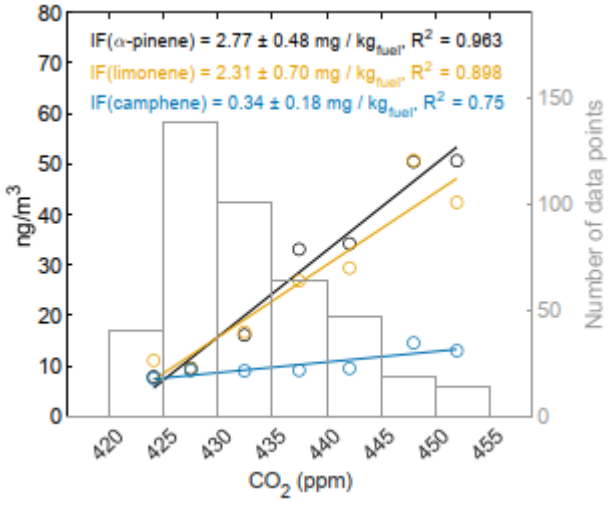

Figure 5: Concentrations of selected terpenoids (α-pinene, limonene and camphene) as a function of CO_2 concentration and the determined IFs with 95 % confidence intervals and R^2 of the averaged linear fits. Also, the number of data points in each CO_2 concentration bin is shown.

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Table 4. Terpenoid IFs with confidence intervals (CI) and R^2 values of the averaged linear fits.

Compound	IF and 95 % CI (mg/kg_{fuel})	R^2 of the averaged linear fits
α-pinene	*2.77 ± 0.48*	*0.963*
limonene	*2.31 ± 0.70*	*0.898*
camphene	*0.34 ± 0.18*	*0.750*
myrcene	*0.49 ± 0.14*	*0.902*
β-pinene	*0.10 ± 0.21*	*0.149*
3Δ-carene	*0.98 ± 0.17*	*0.966*
p-cymene	*0.60 ± 0.21*	*0.866*
1,8-cineol	*0.07 ± 0.14*	*0.180*

3.4 Comparison of episodic and non-episodic data

The three regional and long-range transported pollution episodes that occurred during the campaign had effects on the relationship between the studied metrics and CO_2 concentration. For example, the EFs of BC (+31 %), CO (+74 %), PM_{10} (+65 %), benzene (+137 %), α-pinene (171 %) and especially $PM_{2.5}$ (+219 %) increased during the episodes compared to the non-episodic periods (Table S5 and S6). On the contrary, the EFs of PN (-54 %) and nitrogen oxides (-38–(-)22 %) were considerably lower during the episodic periods. This result could be affected by many potential factors. First, during the episodes, temperature inversions likely decreased the dilution and dispersion of emissions at the studied sites. Hence, the measured aerosol could have been relatively more aged, increasing the particle mass and decreasing the number concentration. Second, other meteorological conditions themselves could affect the road traffic emissions: colder temperatures may cause cooler engine temperature and influence the operation of exhaust after-treatment systems (Suarez-Bertoa and Astorga, 2018) used in vehicles. Third, the increased PM, CO, benzene and BC EFs may be related to residential wood combustion, which has been shown to be a significant contributor to absolute pollutant concentrations during the episodes (Teinilä et al., 2025). Thus, even though CO_2 is generally a good tracer for road traffic, the CO_2-based EF-calculation could be challenging if there are other major sources of the studied pollutants and CO_2 nearby. The decreased EFs of PN and nitrogen oxides support this theory as the contribution of residential wood combustion to these metrics is typically lower compared to traffic.

Even though the results of this study show that CO_2 EF-calculation is a good method to observe the direct, and indirect, effects of road traffic, the mentioned uncertainty related to other contributing pollution sources should generally be better

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acknowledged. For example, an EF of 88 mg/kg_{fuel} for sulfates was determined during the episodes, whereas during the non-episodic periods, the connection between the sulfates and CO_2 was poor (Figure 6). Therefore, according to the non-episodic data, road traffic is not a direct source of sulfates near traffic, but based on the episodic data, one could misinterpret the data to conclude road traffic as a clear source of sulfate. The correlation between CO_2 and nitrate was also higher during the episodes, which is reasonable, as it was speculated that during the pollution episodes aerosols were relatively more aged, and NO_x emitted from vehicles could have oxidized to nitrate. In addition, changes with the determined VOC EFs (Table S5) were observed during the episodic periods, especially for benzene and terpenoids, which are known to have contributions higher than those from traffic exhausts in wood combustion emissions (Gilman et al., 2015). On the other hand, the correlations were generally weaker during the episodic periods, supporting the idea that the non-episodic results better describe the influence of road traffic. Overall, the results show that without identification of interfering external pollutant sources, the obtained EFs could be misleading. This uncertainty needs to be considered when determining EFs in ambient measurements as well as when comparing EF results between different studies, sites, and environments. In this study, the non-episodic EFs and IFs identified can be considered to accurately represent the effects of wintertime road traffic in Helsinki central area, as the regional pollution levels during the non-episodic periods were low and the linkages of studied pollutants were strong with CO_2.

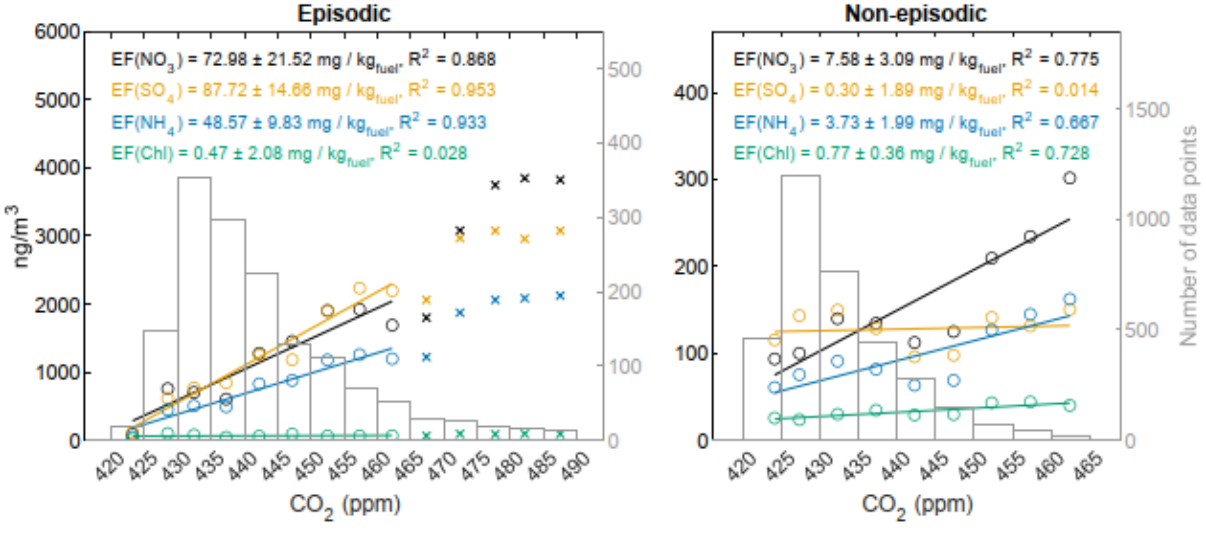

Figure 6: Concentrations of nitrates (NO_3), sulphates (SO_4), ammonium (NH_4) and chloride (Chl) as a function of CO_2 concentration and the determined EFs, including 95 % confidence intervals and R^2 of the averaged linear fits. The episodic data on left and non-episodic data on right. Notice the different x- and y-axes. Also, the number of data points in each CO_2 concentration bin is shown. See Figure S9 for organics.

4 Conclusions

Wintertime road traffic emission factors for various air pollutants were determined in a street canyon environment in Helsinki city center, Finland. In case of regulated pollutants such as $PM_{2.5}$, BC, PN and nitrogen oxides, the determined fleet

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EFs were considerably higher than required for new vehicles in EURO6 standard since 2014, showing that despite tightening regulations, effects of road traffic on urban air quality can remain high. This result is likely related to the slow renewal of vehicle fleet, as well as the presence of super-emitters such as old heavy-duty vehicles, vans and vehicles with malfunctioning exhaust aftertreatment systems. In case of PN, it is worth considering whether the current or proposed emission regulations are indeed effective in reducing the concentrations of the UFPs that are formed in exhaust during exhaust cooling and dilution after the tailpipe. The EFs of particle chemical components showed that the particle mass emissions from road traffic exhaust are mainly composed of organics and black carbon. The clearly higher PM_{10} EF compared to $PM_{2.5}$ EF also suggests an important contribution of non-exhaust emissions to particulate emissions. The VOC emissions of road traffic were the most attributable to aromatic hydrocarbons, especially to toluene and p/m-xylene.

In addition to direct EFs, we showed that the CO_2-based EF-calculation is suitable when investigating inversely proportional impact factors (IF) related to road traffic. Concentrations of O_3 and small ions decreased linearly as a function of increasing CO_2. Even though the negative contributions of road traffic to these metrics have been reported before, the approach utilized can be fruitful in terms of air quality modelling and when estimating the formation and removal processes of atmospheric gases and ions. For example, it was found that in cases of high CO_2 concentration, i.e. high traffic volume, NO_2 concentration remained constant regardless of increasing CO_2 and the increase of NO_x was almost completely contributed by NO. This phenomenon was linked to near-zero O_3 concentration which caused that NO was no further oxidized to NO_2. The approach could, hence, be useful when estimating NO/NO_2 concentrations in urban environments.

Furthermore, the contribution of road traffic to compounds that generally have not been considered to be traffic-influenced was found. Positive IFs of road traffic were found for terpenoid VOCs, especially α-pinene and limonene. Even though terpenoid VOC concentrations have been earlier connected with human activity, the results indicate that road traffic could also have direct effects on these compounds, which is not generally well understood.

Overall, the determined up-to-date fleet EFs of vehicles in winter conditions provide valuable insights into the performance of current emission regulations as well as potential future regulatory actions. Also, the findings related to the terpenoid VOCs, together with the observed inverse IFs on O_3 and small ions, show that the CO_2-based EF calculation is an effective approach to investigate not only the direct emissions but also the indirect and unconventional effect of road traffic on urban air. Due to these effects, the influence of road traffic should not be considered only as a linear multiplication of direct emissions and traffic rate, as with some pollutants, the influence of traffic may follow a step-function-like behavior. On the other hand, the data measured during the pollution episodes showed that the method is also vulnerable to misinterpretations if background concentrations or effects of other sources cannot be recognized. In all, our results show the benefits of determining and reporting CO_2-based EFs in traffic-influenced ambient measurements to observe the impacts of road traffic on urban air with a broad perspective, highlighting the potential to gain insights into the spatial variability of these effects if utilized in further studies in different locations and regions.

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Data availability

The raw data is from the measurement report by Teinilä et al. (2025): https://doi.org/10.5194/acp-25-4907-2025. Data shown in the figures and tables will be available open access.

Author contributions

LS: Conceptualization, Methodology, Validation, Formal analysis, Investigation, Data Curation, Visualization, Writing – Original Draft, Writing – Review & Editing

TL: Conceptualization, Methodology, Supervision, Investigation, Writing – Original Draft, Writing – Review & Editing

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MDM: Writing – Review & Editing

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Competing interests

At least one of the (co-)authors is a member of the editorial board of Atmospheric Chemistry and Physics.

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