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Road-traffic emissions of ultrafine particles and elemental black carbon in six Northern European cities

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A R T I C L E I N F O

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A B S T R A C T

Urban air pollution from vehicular emissions remains a pressing public health concern, particularly in Eastern Europe, where data gaps hinder effective mitigation. This study, conducted in the summer of 2024, presents the first detailed analysis of ultrafine particle (UFP) and equivalent black carbon (eBC) emissions from road traffic across Lithuania's six major cities: Vilnius, Kaunas, Klaipėda, Šiauliai, Panevėžys, and Alytus. We used a custom mobile laboratory to capture real-world emissions, revealing stark spatial disparities. Panevėžys and Vilnius topped eBC levels (10400 ng/m^3 and 10200 ng/m^3, respectively), driven by aging vehicle fleets and a diesel prevalence of 70 % in Panevėžys, which also recorded the highest UFP concentration (97800 particles/cm^3). Emission factors, calculated using an adapted Operational Street Pollution Model (OSPM), identified Vilnius' light-duty vehicles as leading in particle number emissions (8.90×10^{14} particles/(km·veh)), likely due to the prevalence of gasoline direct injection engines. At the same time, Panevėžys dominated eBC emissions (150 mg/(km·veh). Heavy-duty vehicles, including buses and trucks, exhibited emission factors up to five times higher than those of their light-duty counterparts, thereby amplifying their impact in urban areas. These findings illuminate emission dynamics in an understudied region, providing policymakers with precise and actionable insights for targeted interventions, such as fleet upgrades or the establishment of low-emission zones. By addressing a critical knowledge gap, this study empowers the scientific community and public health advocates to devise strategies that combat vehicle-related pollution, reduce exposure to harmful pollutants, and foster healthier urban environments across Eastern Europe and beyond.

1. Introduction

Air pollution worldwide has been recognized as one of the top ten most relevant factors leading to premature deaths (Zhang et al., 2025). As more people live in cities, urban air quality inventories are becoming crucial for identifying the sources, concentrations, and health outcomes of pollutants. One of the primary sources of air pollution in urban environments is the combustion of fossil fuels in motor vehicles (Kumar et al., 2021). The conventional road vehicular fleet, powered by petrol and diesel engines, is associated with the emission of extremely high concentrations of ultrafine particles (UFP; diameter < 100 nm) and soot (black carbon, BC). The size of UFP allows particles to penetrate deeply into the respiratory system, increasing the risks of asthma, cancer, cardiovascular diseases, and even premature death (Kumar et al., 2013; Schraufnagel, 2020). BC pollution is known to have a particularly adverse effect on human health compared to the average composition of

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particles outdoors (Janssen and Joint, 2012). It becomes clear that developing sustainable control strategies for higher air quality and monitoring road vehicular fleet emissions is necessary.

Emission factors of particulate matter (PM_{10} and $PM_{2.5}$, which refer to particulate matter with aerodynamic diameters lower than 10 and 2.5 micrometers, respectively) from on-road vehicular fleets have been widely reported across multiple cities worldwide. For example, a study by Wang et al. (2014) showed that the PM_{10} emission factor in Beijing was 15.5 mg/(km·veh) for light-duty gasoline vehicles and 36 mg/(km·veh) for diesel vehicles. From a tunnel study in Los Angeles, Gillies et al. (2001) reported mixed fleet (97.4 % LD vehicles) $PM_{2.5}$ and PM_{10} emission factors to be 52 and 69 mg/(km·veh), respectively. According to a study in Havana, Cuba, the emission factors of modern and older light-duty vehicles were reported to be 90.6 and 125.4 mg/(km·veh), respectively (Madrazo and Clappier, 2018). Hung et al. (2010) reported a PM_{10} vehicular fleet emission factor of 108 mg/(km·veh) from Hanoi, Vietnam. In the European context, past emission factor studies were primarily conducted in more economically advanced Western and Northern European cities, although studies focusing on particulate pollutants were somewhat limited (Kakosimos et al., 2010). For example, in urban Copenhagen, Denmark, Wang et al. (2010) reported a $PM_{2.5}$ emission factor of 46 mg/km for the total fleet, and 20 mg/(km·veh) for the light-duty vehicles. Ghenu et al. (2008) reported a $PM_{2.5}$ emission factor of 60 mg/(km·veh) from Rouen, France. Ferm and Sjöberg (2015) reported $PM_{2.5}$ and PM_{10} road traffic emission factors to be 21 and 60 mg/(km·veh), respectively, in Gothenburg, Sweden.

Considering emission factors beyond commonly used pollution metrics such as PM_{10} and $PM_{2.5}$, e.g., UFP number and BC mass, were also mainly determined for several economically advanced European countries. For example, a multi-year study by Krecl et al. (2017) reported that BC emission factors for light-duty vehicles in Stockholm, Sweden, decreased significantly between 2006 and 2013: for gasoline-powered vehicles, from 11.0 to 2.6 mg/(km·veh), and for diesel-powered vehicles, from 94.8 to 23.4 mg/(km·veh). In a recent study by Krecl et al. (2024), the authors indicated that during the years 2013–2019, the emission factors of BC and total particle number in Stockholm were reduced due to successful emission mitigation strategies. Wiesner et al. (2020) estimated the emission factors in Leipzig, Germany, to be 48 mg/(km·veh) for BC from a mixed vehicle fleet. When separated by vehicle type, light-duty vehicles (LDVs) emitted 32 mg/(km·veh), and heavy-duty vehicles emitted 522 mg/(km·veh). Furthermore, the authors noticed that despite the increasing share of diesel and direct injection engines, a decreasing trend in BC emission was observed—from 70 mg/(km·veh) at the beginning of the study period (2009) to around 30–40 mg/(km·veh) in later years (2018).

Concerning the UFP number, Straaten et al. (2023) reported LDV emission factors in the range of 1.77×10^{14} particles/(km·veh) for Berlin, Germany. Nickel et al. (2013) reported the LDV emission factor from LDV to be 1.9×10^{14} particles/(km·veh) from Meckenheim, Germany. A comprehensive study in Zurich, Switzerland, was carried out by Imhof et al. (2005), who reported real-world emission factors for size-segregated aerosol particles and BC. The LDV UFP number emission factor ranged from 0.38 to 0.81×10^{14} particles/(km·veh). Other European studies have mainly focused on total particle number (PN) emission factors. It can be seen as a UFP proxy in an urban environment (Kecorius et al., 2024). For example, Birmili et al. (2009) reported a PN emission factor of 0.24×10^{14} particles/(km·veh) in Berlin, Germany. Mårtensson et al. (2006) reported PN emission factors from Stockholm, Sweden, to be 0.3×10^{14} particles/(km·veh). From Copenhagen, Denmark, Wang et al. (2010) reported a PN emission factor of 1.01×10^{14} particles/(km·veh).

The literature review revealed two major research gaps related to real-world emission factor studies. Firstly, although health concerns related to exposure to emerging pollutants, such as UFP and BC, are increasing, the existing studies focusing on this type of pollutant remain outdated, with only a few studies conducted after 2020. And secondly, most road vehicle emission-related studies are conducted in more developed Western and Northern Europe, where the vehicular fleet is more modern. Thus, reported emission values may not represent the rest of Europe. This is especially true for several European Union (EU) countries, such as Romania, Finland, Estonia, Poland, Portugal, Malta, and Lithuania. These countries hold the highest fractions (approx. 30 %) of old passenger cars (20 years or older) as of 2023 (https://www.eea.europa.eu, accessed May 13, 2025). Furthermore, in 13 of the 24 EU countries (for which information is available for 2023), there were more petrol cars than diesel cars, with the highest share of petrol cars (85 %) being in the Netherlands. In the other 11 EU countries, diesel cars outnumbered petrol cars, with their shares ranging from 66 % in Latvia and Lithuania to 49 % in Slovenia. Outdated vehicle engine technologies, combined with a high fraction of diesel-powered vehicles, create favorable conditions for high pollutant emission scenarios and high exposure concentrations, which remain largely underexplored.

To improve our understanding of emission factors in under investigated regions of Europe, this work focuses on real-world road traffic emission factors of UFP and BC in Lithuania, an EU country with one of the oldest vehicular fleets and a high fraction of diesel-powered engines. The main objectives of the study are to a) investigate road-traffic emissions of UFP and BC in six largest Lithuanian cities, b) test the hypothesis that due to different diesel-powered engine fraction in six cities the emitted UFP and BC concentrations will show statistical differences, and c) provide estimated emission factors for average vehicular fleet in six Lithuanian cities. This study also extends beyond the two largest Lithuanian cities, providing a more accurate assessment of traffic-related air pollution that accounts for significant differences in traffic patterns and emission sources. The results from this study provide the first comprehensive real-world vehicular emission inventory for Lithuania, addressing the current data gap relative to more advanced European countries. These findings can help to better understand pollution-related health risks in under sampled urban environments and support data-driven revisions of air quality legislation and strategies for sustainable development.

2. Methods

2.1. Measurement domain

An intensive mobile measurement campaign was conducted in Lithuania during the summer of 2024 (June–August) as part of the project “On the Vehicular Fleet Aerosol Particle and Black Carbon Emission Factors in Lithuania” (MOCHA), funded by the Research Council of Lithuania (LMTLT, agreement No S-MIP-22-57). Summer was selected to minimize interference from non-traffic-related sources such as long-range transported biomass burning in spring and residential heating in winter. This also ensured that the measured eBC mass and size-segregated particle number concentrations predominantly reflect vehicular emissions. The absorption Ångström exponent analysis supporting this claim is presented in the supplementary information (SI). The measurement campaign covered six major Lithuanian cities—Vilnius (VLN), Kaunas (KAU), Klaipėda (KLA), Šiauliai (SIA), Panevėžys (PNV), and Alytus (ALY)—with the study domain presented in Fig. 1. The measurements encompassed diverse urban environments and traffic conditions across six Lithuanian cities, covering a cumulative area of 1000 km² over 25 days. Vilnius, the capital, with the largest population and the highest number of registered light-duty vehicles (N ∼325,000), also had the lowest diesel share (50 %). The other listed cities exceed 60 % (Regitra, 2024). The measurements were conducted on non-rainy days between 9 AM and 7-8 PM to ensure consistency. The mean air temperature was in a range from 21°C to 26°C. Except for Vilnius, each city’s routes were covered within a single day. Hence, each planned route covering an entire city was completed four times in four days for the cities PNV, ALY, SIA, and KAU. In the case of KLA, the

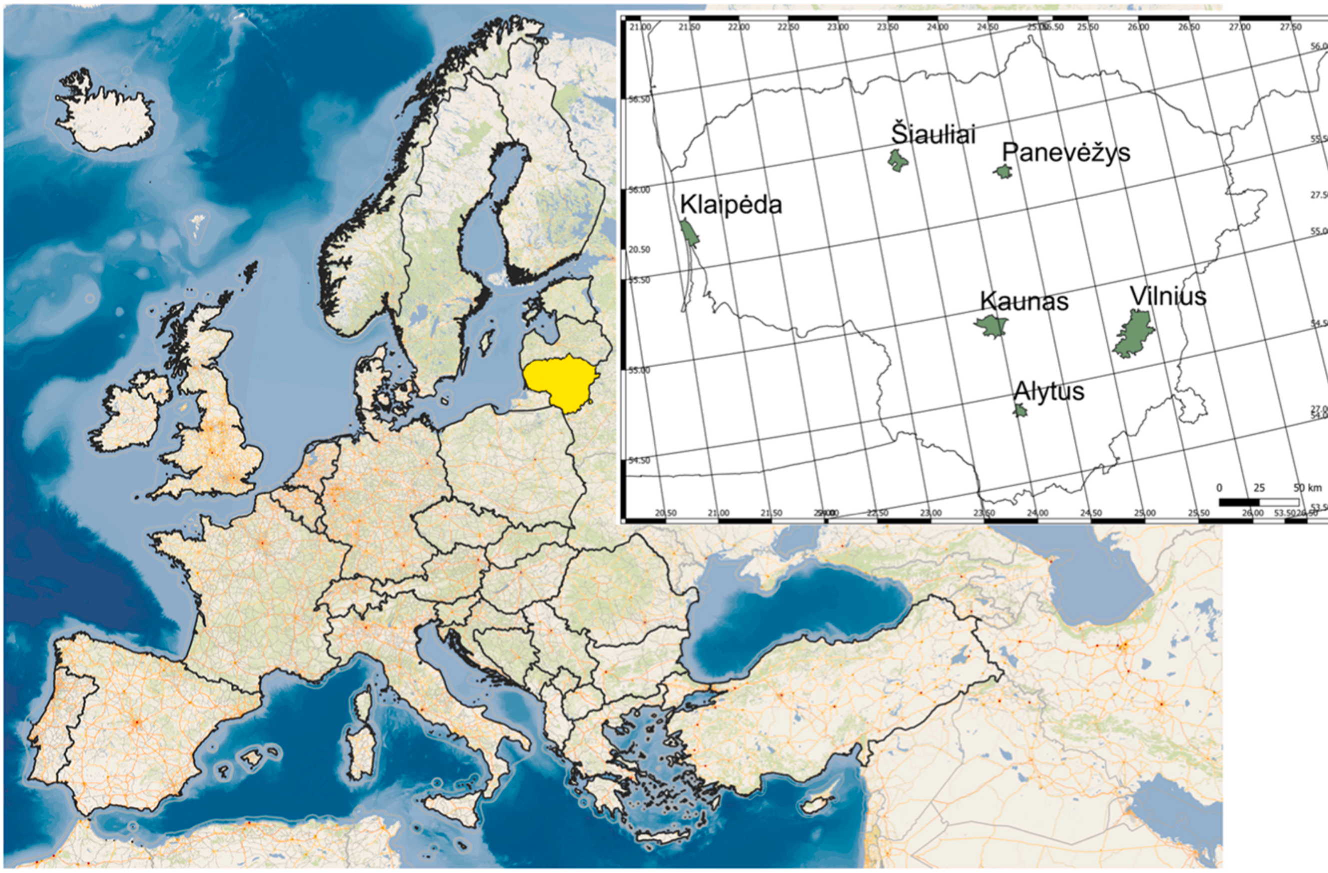

Fig. 1. Geographic coverage of the six Lithuanian cities included in the study, where mobile measurements of particle number size distribution (PNSD) and equivalent black carbon (eBC) concentrations were conducted.

city-wide route was completed five times in five days. In Vilnius, due to its larger area, the city covering route was divided into smaller stretches and completed over four days (each route segment was driven once).

2.2. Measurement setup

A specialized mobile laboratory trailer was developed for the campaign and equipped with vibration-dampened instrument racks, a waterproof PM_{10} aerosol inlet mounted 1.5 m above the roof, and an internally routed stainless steel sampling line. The aerosol was passed through a custom-built 0.5 m long diffusion dryer packed with silica gel and then split via a custom isokinetic flow splitter into two lines (10 L/min to instruments, 6.6 L/min bypass). This ensured a total flow of 16.6 L/min, matching the PM_{10} inlet specification. The residence time in the sampling line was approximately 1.5 s. Supporting equipment included high- and low-volume vacuum pumps, an air compressor for dryer regeneration, and an air conditioning system with external venting to maintain internal air quality. The power was supplied via a 2000 W pure sine wave inverter. The particle number size distributions (PNSD) was measured using a high-resolution electrical low-pressure impactor (HR-ELPI, Dekati, Finland), which classifies particles based on aerodynamic diameter via electrical detection across 14 size bins ranging from 6 nm to 10 µm. The instrument operated at 10 L/min with 1 Hz time resolution. Sintered metal impaction plates were used to reduce particle bounce and overloading. Instrument maintenance was rigorously followed, including ultrasonic cleaning of stages after each city campaign, daily zeroing of the electrometer (lasting 30 minutes), and leakage testing. Manufacturer-provided calibration factors, dilution ratios, and operational parameters were applied throughout the study.

Equivalent black carbon (eBC) mass concentrations were determined using a micro-aethalometer (MA200, AethLabs, USA), which measures optical attenuation at five wavelengths (375–880 nm); for this study, the 880 nm channel was used. The instrument operated at a frequency of 1 Hz with a flow rate of 100 mL/min. Before each measurement cycle, the aethalometer was stabilized for 30 min concurrently with the HR-ELPI zeroing.

Additional environmental parameters, including relative humidity and temperature (HYT939, IST AG, Switzerland), as well as onboard video footage, were logged continuously using a combination of a laptop and a Raspberry Pi-based data logging and control system.

2.3. Data evaluation

The raw PNSD data, initially recorded in 14 aerodynamic diameter bins, was interpolated into a high-resolution 500-bin format using the proprietary Data Analysis Tool provided by the HR-ELPI manufacturer (Dekati, Finland). Following the manufacturer's recommendations, an aerosol density of 0.8 g/cm^3 was applied to account for black carbon–dominated particles. Size-dependent particle losses in sampling lines were corrected using the methodology presented by Von der Weiden et al. (2009). The data segments corresponding to calibration, zeroing, and out-of-spec impactor pressures (outside 40 ± 5 mbar) were excluded. Based on the processed PNSD data, the particle number concentrations were classified into ultrafine (UFP, particle diameter, $d_p <$ 100 nm), accumulation (ACCU, 100 nm $\leq d_p <$ 1 µm), coarse (COARSE, 1$\leq d_p <$10 µm), and total (PNC, 6 nm–10 µm) fractions.

The eBC data, recorded at one-second resolution using a micro-aethalometer, were corrected for signal noise using the optimized noise-reduction averaging (ONA) algorithm published by Hagler et al. (2011). A threshold attenuation difference of 0.001 was used, effectively minimizing negative eBC values while preserving short-term variability. Finally, for all measurements, only data corresponding to trailer speeds exceeding 5 km/h were included in the analysis to eliminate potential contamination from the towing vehicle.

The pollutant concentration increments (C_{tr}) attributable to direct tailpipe emissions were estimated by subtracting background levels from the measured values. Background concentrations were determined using a 25th percentile rolling window (20-minute span), following the approach of Kivekäs et al. (2014) and Kecorius et al. (2017).

Traffic density data, distinguishing LDV, buses, and trucks, was calculated from the recorded street videos. A custom video processing and machine learning pipeline was developed in Python to count and categorize moving vehicles using the YOLOv11 (You Only Look Once, version 11) model, implemented with the Ultralytics framework (Jocher et al. (2023), *YOLO by Ultralytics*. GitHub Repository (https://github.com/ultralytics/ultralytics, accessed May 13, 2025). The model processes each video frame to produce bounding boxes, confidence scores, and class predictions for detected objects. The graphics processing unit (GPU) acceleration is utilized for detection, and the pipeline was executed on a high-performance computing (HPC) server (https://www.srce.unizg.hr/en/croatian-centre-hpc). After detection, each object is associated with a unique identification number across frames using simple online and real-time tracking with a deep association metric (DeepSORT, Wojke et al., 2017). Only moving vehicles were counted. This was ensured by comparing the vehicle's current position to the previous position, using a predefined threshold of 30 pixels. If the vehicle's position changes by >30 pixels in any direction, the vehicle is considered to be moving. Vehicle movement was recorded if a vehicle was detected as moving for at least two consecutive frames. Additionally, vehicles are only counted when they pass a predefined count line, ensuring that only vehicles near the measuring car are included in the count, thereby eliminating stationary vehicles from being counted.

As vehicle counts were determined from videos recorded on a driving vehicle, the obtained temporal variation of vehicle flow reflects the dynamics across the entire city, rather than at each location. Such an approach provides a comprehensive view of each city's traffic patterns, encompassing residential, commercial, and transit zones. Higher vehicle flows were observed in larger cities, driven by greater population densities, larger vehicular fleets, and increased economic activity. Vilnius, Kaunas, and Klaipėda, being the three largest cities in Lithuania, averaged approximately. 24, 22, and 19 vehicles per minute (Fig. 2). These elevated flows in larger cities align with their dense historical centers and industrial areas, which sustain greater traffic loads. In contrast, smaller cities exhibit lower vehicle flows, consistent with their smaller population and less extensive infrastructure. The vehicle flows in Alytus, Šiauliai, and Panevėžys were 12, 14, and 18 vehicles/min, respectively, which indicates their reduced congestion and a more dispersed layout.

A custom algorithm was developed to locate pollutant concentration peaks from deconvoluted data, which is presented in the SI. The averaged pollution peak shapes were fitted using a versatile exponentially modified Gaussian (EMG) function, defined as:

$$y(x) = A\frac{\lambda}{2}exp\left[\frac{\lambda}{2}(2\mu + \lambda\sigma^2 - 2x)\right]erfc\left(\frac{\mu + \lambda\sigma^2 - x}{\sqrt{2}\sigma}\right), \quad (1)$$

where A is the amplitude scaling factor, which determines the overall magnitude of the peak, μ is the location parameter, representing the mean of the underlying Gaussian component, σ is the standard deviation of the Gaussian component, controlling the width of the peak, and λ is the exponential decay constant, which governs the rate at which the peak is trailing edge decays. The *erfc(z)* is the complementary error function, which arises from the convolution of the Gaussian and exponential components. The result of the fitting is presented in SI Table T1.

2.4. Emission factor estimation

The emission factors (EF, expressed as particle number or mass per vehicle per kilometer) of measured pollutants in six Lithuanian cities were calculated using an inverse modeling approach (e.g., Madueno et al., 2019):

$$EF = \frac{C_{tr}}{F \cdot N_V}, \quad (2)$$

where C_{tr} is the pollutant concentration increment due to traffic contribution (obtained by subtracting the background concentration from the measured concentration), F – the dilution factor (DF) in s/m^2, and N_V – the number of vehicles per second. The dilution in Eq. (2) needs to be estimated prior to calculating the pollutant emission factor.

There are several different ways to assess it at the street level. For example, a trace experiment can be carried out, where a known concentration of trace gas is released and later measured at a certain distance (e.g., Belalcazar et al., 2009; Mathissen et al., 2011). The observed

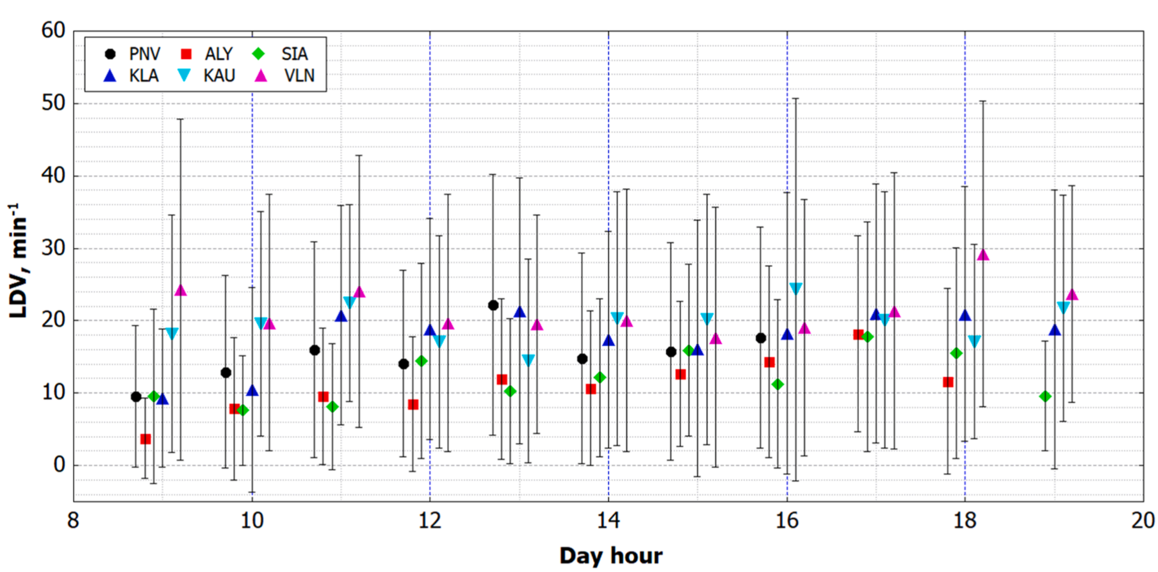

Fig. 2. Average LDV flow per minute across six Lithuanian cities, derived from video footage recorded by the onboard camera during mobile measurement campaigns. Calculations are based on the specific hours of the day when measurements were performed. Error bars indicate one standard deviation. Flow rates for trucks, buses, and the total fleet are available in SI (Figs. S1–S3).

difference in concentrations can be attributed to dilution, which in turn can be used to estimate the emission factor. An alternative approach to estimating pollutant dispersion is to employ high-fidelity computational fluid dynamics simulations. In a study by Plogmann et al. (2023), the authors employed the unsteady Reynolds-averaged Navier–Stokes modeling with the k-ω shear-stress-transport turbulence closure to capture both the large-scale wake structures and the small-scale mixing that drives exhaust dilution. Simulations at 8.33, 13.89, and 22.22 m/s, validated against wind-tunnel drag data, showed that at a moderate speed of 13.89 m/s, the core plume underwent a 100-fold dilution within approximately 0.83 m downstream, 0.81 m laterally, and 0.74 m vertically—underscoring the rapid mixing induced by wake turbulence.

Furthermore, the pollutant dispersion within the street canyon can also be modeled using dedicated software, one of which is the Operational Street Pollution Model (OSPM, Berkowicz et al., 1997), a semi-empirical tool developed specifically for street canyon geometries. The OSPM integrates direct emissions through a plume model, recirculated pollutants via a box model, and the effects of traffic-induced turbulence. It incorporates meteorological parameters, street geometry, and traffic data to compute concentrations at fixed receptor points. To adapt the OSPM, we treated the measurement vehicle as a mobile receptor, capturing plumes at moments of proximity to the target vehicle, as evidenced by sudden concentration increase. This approach enabled us to utilize OSPM's dispersion modeling capabilities for our dynamic setup, simulating the dilution process under conditions which approximate real-world driving.

The assumption was made that the wind direction in the OSPM simulations is parallel to the street and that the wind speed equals the measurement vehicle's driving speed, which ranged from approximately. 2.78 to 9.44 m/s (25th to 75th percentile, median 5.56 m/s). This assumption reflects the relative airflow experienced by the moving vehicle. In the vehicle's reference frame, the ambient air moves in the opposite direction at a speed equal to the vehicle's speed. By configuring the OSPM with this wind speed and direction, we modeled the dispersion conditions relevant to the mobile sensor.

Cross winds were neglected in the simulations, based on the assumption that their influence on plume dispersion was minimal under our measurement conditions. Plogmann et al. (2023) studied the effects of cross winds on vehicle exhaust plumes. They found that although strong cross winds (e.g., 12.50 m/s against a 13.89 m/s vehicle speed, a 90 % ratio) notably enhanced turbulence and dispersion, at light (2.50 m/s, 18 % ratio) and moderate (6.94 m/s, 50 % ratio) cross winds relative to a 13.89 m/s vehicle speed, deflection was not substantial.

To test our crosswind assumption, general wind speed and direction data were obtained from meteorological stations located near each of the investigated cities, representing background conditions. These data were used to identify the maximum wind speeds that could have influenced plume deflection in the urban environment. Throughout the measurement period, the maximum recorded wind speed was approximately half the average vehicle speed, indicating that crosswinds were generally even lower during most of the campaign. This supports our assumption that crosswind effects on plume deflection were minimal and could be reasonably neglected in the modeling framework. That is, in our study, the maximum wind speed recorded at one of the background meteorological stations was 3.33 m/s. The 25th and 75th percentiles of driving speeds ranged respectively from 3.06 to 9.44 m/s (with a median of 6.39 m/s). At the median speed, the cross wind-to-vehicle speed ratio was 51 % similar to the 50 % ratio reported by Plogmann et al. (2023), where deflection was minimal. Only at lower driving speeds and higher winds may the increased turbulence or plume deflection have influenced the results. However, given the typically urban setting with buildings shielding it and the predominance of higher driving speeds, we deemed the impact of the crosswinds to be minor.

The OSPM model was used to simulate a variety of street configurations that reflect the diverse urban environments encountered during measurements (Fig. 3). Street width ranged from 10 to 50 meters, and building heights varied from 0.2 to 80 meters. The configurations included continuous buildings on both sides (no spaces), buildings on one side only, and randomized layouts with spaces between buildings, simulating irregular urban settings. Simulations showed that building height and placement (with and without spaces) did not affect DF. This agrees well with findings by Pan and Ji (2024), who reported that building heights' on pollutant dispersion in parallel wind directions is relatively small compared to oblique and perpendicular winds. These choices account for the uncertainty in actual street geometry, which was not precisely known for each measurement point. By testing extremes, we captured the range of possible dilution factors, ensuring robustness across varied urban conditions.

Fleet-specific emission factors for LDV, buses, and heavy-duty trucks were derived by isolating individual exhaust plumes under on-road sampling conditions where only vehicles from one category occupied the measurement zone. Transient concentration peaks, corresponding to the instantaneous passage of each vehicle, were combined with dilution-factor estimates (at the corresponding speed) to yield emission factors for each fleet segment. This methodology eliminates inter-fleet interference by constraining each sampling event to a single vehicle class and applying consistent driving-speed criteria. It enables direct, quantitative comparison of pollutant emissions across LDVs, buses, and trucks under comparable real-world conditions. The EFs for different vehicular fleets were derived from a robust dataset of 90–297 plume events per pollutant

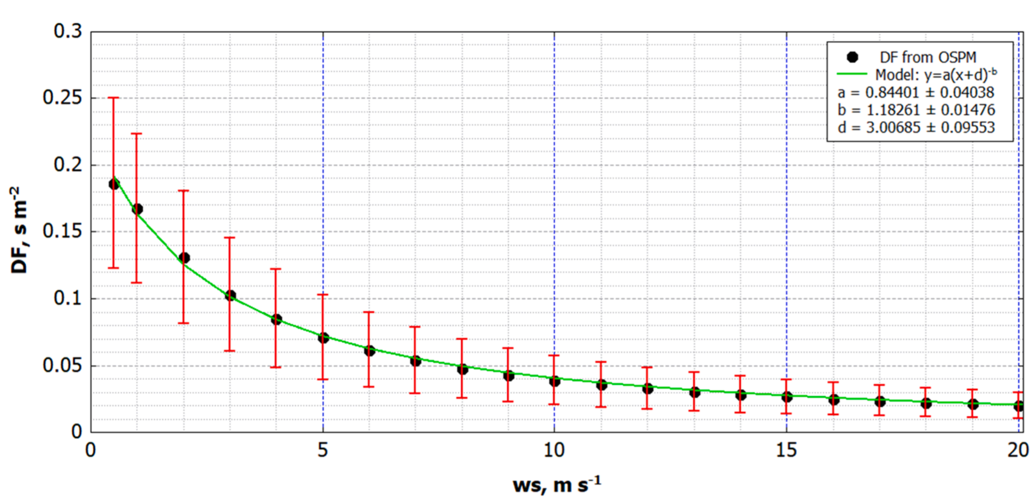

Fig. 3. Dilution factors (DF) estimated using the OSPM model under varying wind speeds parallel to the road and different street configurations, reflecting a range of typical urban driving conditions.

across six cities. LDV plume counts spanned 129–268 (PNC), 90–247 (UFP), 160–297 (ACCU), 105–203 (COARSE), and 108–243 (eBC). Bus emission factors relied on a smaller but representative dataset (0–6 plumes per pollutant), while truck factors utilized 4–25 plumes per pollutant.

2.5. Software

The measurements were evaluated and analyzed using the open-source programming language and software environment R (R Core Team, 2013; version 4.2.2). BC and PNSD data were analyzed using open-source tools developed for air quality data analysis (Carslaw and Ropkins, 2012) and custom functions. Maps were created using QGIS (QGIS Development Team, 2022). Figures were generated using QtiPlot (QtiPlot, 2008; version 0.9.8.3) and finalized with the free graphical design tool Inkscape (version 1.2, 2020). The dilution factors for pollutants were obtained from the OSPM (Berkowicz et al., 1997). The programming language Python was used for the deep learning models using video data (v3.12, www.python.org).

3. Results and discussion

3.1. Overview of pollutant plume peak concentration statistics

A key indicator of combustion-related pollution, primarily from diesel exhaust, the eBC exhibits distinct spatial trends across the studied cities. Median eBC concentrations ranged from 7700 ng/m^3 in Alytus to 10400 ng/m^3 in Panevėžys, with Vilnius close behind at 10200 ng/m^3 (Fig. 4). Kaunas, Klaipėda, and Šiauliai record intermediate values of 8700, 8600, and 8300 ng/m^3, respectively. The 25th and 75th percentiles show the highest eBC variability in Panevėžys (5600 to 20300 ng/m^3) and the lowest in Alytus (4200 to 18500 ng/m^3), indicating that while median values differ, peak pollution events can be comparably high across cities. Statistical analysis using the Kruskal-Wallis test (Metzler, 2021) with Bonferroni correction (Weisstein, 2004) identified significant pairwise differences ($p < 0.05$) in eBC concentrations between Alytus and Panevėžys, Klaipėda and Panevėžys, and Alytus and Vilnius. These results confirm that Panevėžys and Vilnius experience significantly higher eBC pollution than Alytus and Klaipėda. The higher eBC levels in Panevėžys can be attributed to its vehicular fleet, where 70 % of light-duty vehicles are diesel-powered, combined with an aging fleet averaging 16.5 years, one of the oldest in Europe. Older diesel vehicles emit substantially more black carbon due to less stringent emission controls, amplifying pollution despite the city's smaller size and lower traffic density.

In contrast, Vilnius, with a lower diesel fraction of 50 %, still records high eBC, most likely due to its dense urban structure and higher traffic volume (part of which may include vehicles with no/outdated exhaust after-treatment technologies). It is worth mentioning that, unlike many Western European cities, Lithuania had not adopted a comprehensive low-emission zone (LEZ) framework at the time of our field measurements. On 1 August 2024, the city of Kaunas introduced a limited form of a LEZ in its historic center; however, rather than restricting access for older, higher-emitting vehicles, the regulation imposes a flat €2 entrance fee (Kaunas Tourism, 2024). This approach allows continued access by the most polluting vehicles upon payment and is unlikely to reduce emissions from the legacy fleet significantly. Fee-based LEZ schemes, while politically easier to implement, often lack the strength to drive meaningful behavioral change or fleet modernization. In contrast, more robust LEZ policies—such as tiered restrictions based on vehicle emission standards or outright bans on the highest-emitting vehicles—would be more effective in improving urban air quality by directly targeting the most polluting segments of the vehicle fleet (Rasch et al., 2013).

Median UFP concentrations were highest in Panevėžys at 97800 particles/cm^3, followed by Klaipėda at 90500 particles/cm^3, meanwhile, Alytus records the lowest at 73100 particles/cm^3. Kaunas, Vilnius, and Šiauliai show intermediate values of 86900, 83700, and 82000 particles/cm^3, respectively. Significant differences were observed between Klaipėda and Panevėžys, Kaunas and Vilnius ($p < 0.05$), and Klaipėda and Vilnius. Accumulation mode particle number concentrations were relatively consistent between cities. Significant differences occur between Alytus and Vilnius, as well as between Šiauliai and Vilnius. Similarly, coarse mode particles showed somewhat limited variation. Primarily from non-exhaust emissions like tire and brake wear, as well as resuspended dust, coarse mode particle concentrations ranged from 45 in Kaunas to 52 particles/cm^3 in Klaipėda. No significant differences were found indicating consistent coarse particle emissions across cities. The ultrafine particle number fraction was highest in Panevėžys and

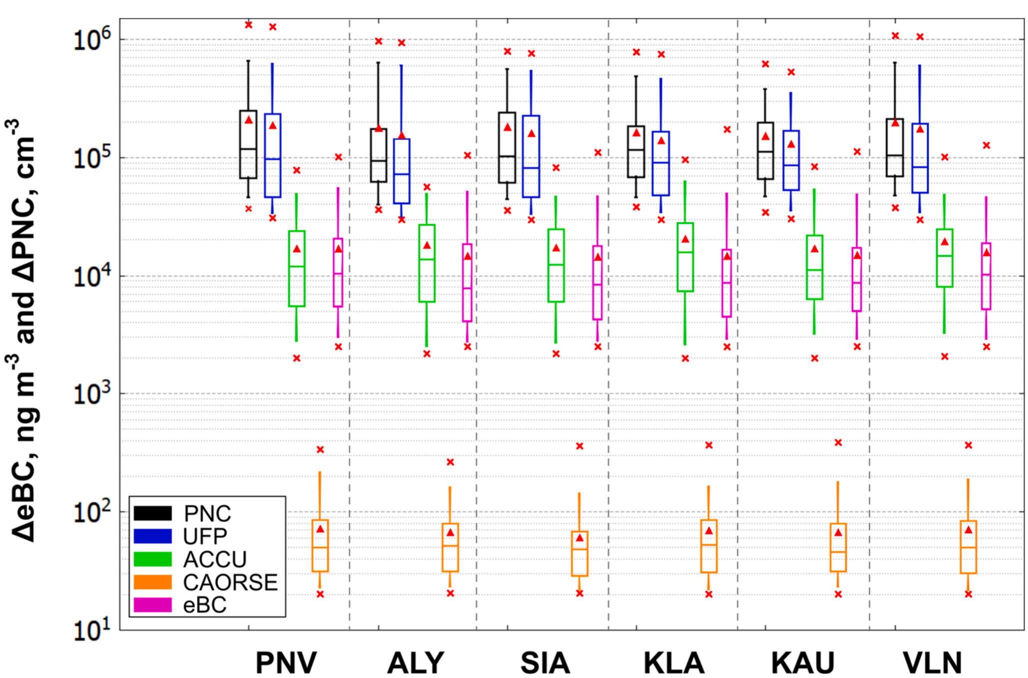

Fig. 4. Pollutant concentration increase measured on the streets during the mobile measurement campaign in six Lithuanian cities.

Kaunas, at 90 %. The accumulation mode particle number fraction was highest in Klaipėda at 20 %, followed by Vilnius at 16 %. Coarse particle number fractions were comparably low and stable across the cities, ranging from 0.04 % to 0.06 %.

Median eBC concentrations measured in Lithuania (approx. 8000 to 10000 ng/m^3) are slightly lower than the 10000 – 25000 ng/m^3 plume values reported by Ježek et al. (2015) in vehicle-chasing experiments with Euro-3 and Euro-5 compliant fleets. Zheng et al. (2022) recorded roadside eBC levels of 2000 – 10000 ng/m^3 at 5 m from the curb in Shanghai—somewhat lower than our on-road, tailpipe-proximate measurements—likely reflecting plume dilution with increasing distance from the emission source. Several studies (e.g., Vogt et al., 2003; Wang et al., 2011; Ježek et al., 2015) reported tailpipe PNC ranging from 3×10^4 to 1.2×10^5 particles/cm^3, closely matching the PNC levels observed in this study.

3.2. Analysis of pollution plume behavior using EMG function

The observed sharp rise in concentration, followed by an exponential decay driven by atmospheric dispersion was analysed using EMG function (Fig. 5). The plume dynamics can be inferred from EMG parameters σ and lambda λ. Across the six cities, the (σ, λ) pairs for PNC reveal noticeable variability tied to urban characteristics. In Vilnius, the capital and most densely populated city, PNC exhibits a (σ, λ) pair of (2.02, 0.35), indicating a broad plume with slow decay. This suggests a vast, persistent pollution plume, likely driven by high traffic volumes and limited dispersion due to the dense street network in its compact urban layout. Similarly, Kaunas shows a comparable pair of (2.03, 0.36), reflecting its dense vehicle flow and built environment, which hinders pollutant dispersal.

In contrast, a more open layout and smaller Alytus with lower traffic presents a (1.79, 0.63) pair for PNC. The smaller σ indicates a narrower plume, while the larger λ points to faster decay, consistent with enhanced dispersion in less congested settings. Intermediate cities, such as Klaipėda (1.94, 0.45), Šiauliai (1.94, 0.53), and Panevėžys (1.83, 0.49), fall between these extremes, with moderate plume widths and decay rates reflecting their balanced urban density and traffic influences. Klaipėda's slightly larger (compared to Vilnius and Kaunas) λ may be influenced by its coastal location, which could enhance air movement and dispersion. The UFP, a significant fraction of PNC, exhibits closely aligned (σ, λ) pairs, suggesting similar dispersion behavior.

The ACCU display subtly different plume characteristics. In Vilnius, the ACCU (2.05, 0.41) pair indicates a slightly broader plume than the PNC (2.02, 0.35), with a faster decay rate. This wider spread could stem from ACCU's intermediate size, allowing greater initial dispersion, while the larger λ suggests moderate deposition or removal processes. Kaunas shows a reversal, with ACCU at (1.94, 0.35) compared to PNC's (2.03, 0.36), indicating a narrower plume but similar persistence, possibly reflecting differences in emission sources. Alytus maintains a narrower ACCU plume at (1.88, 0.46), consistent with its open structure, though its decay is slower than PNC's (0.63).

The COARSE exhibit a starkly different profile, characterized by narrow plumes and rapid decay. In Kaunas, COARSE has a (1.55, 22.55) pair, with a small σ indicating a highly confined plume and an exceptionally large λ reflecting rapid dissipation, likely due to gravitational settling of these heavier particles. Vilnius follows suit with (1.61, 20.92), reinforcing this trend of narrow, quickly dispersing plumes across dense cities. Overall, COARSE plumes contrast sharply with finer particles, their rapid decay aligning with the physical expectation of faster settling for larger particles.

The eBC, primarily originating from diesel exhaust, exhibits the most extreme plume behavior, characterized by exceptionally wide and persistent plumes across all cities. In Vilnius, eBC's (3.56, 0.09) pair indicates a plume far broader than PNC or UFP, with a minimal λ

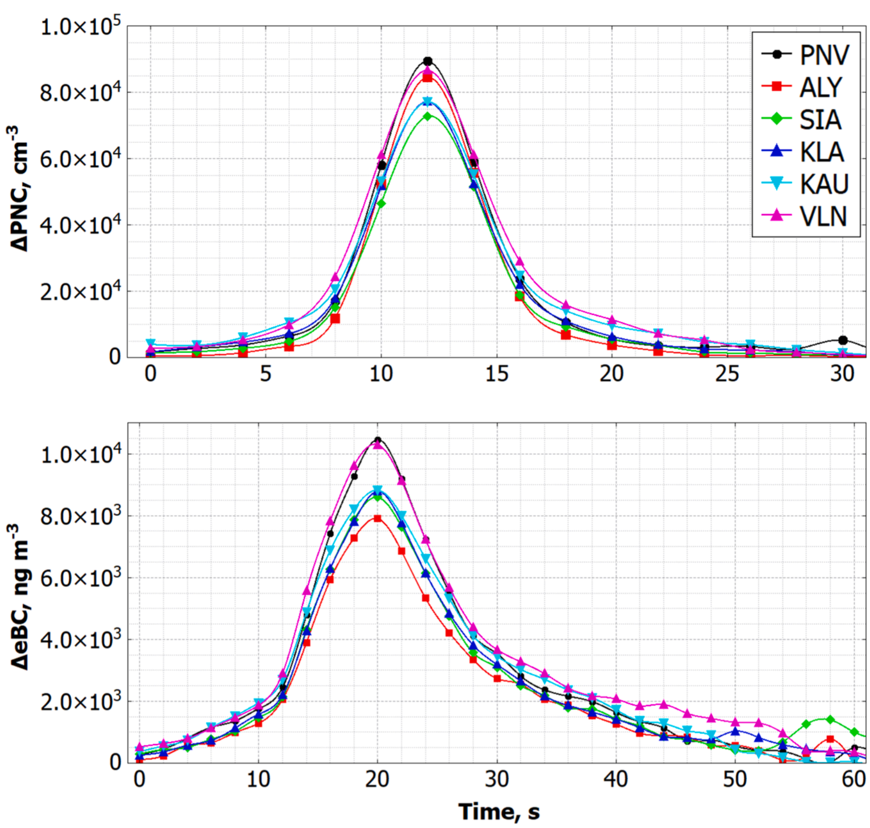

Fig. 5. Median pollutant plume profiles showing increases in total particle number concentration and equivalent black carbon derived from the difference between total and background pollutant concentrations across six Lithuanian cities. Plume profiles for ultrafine, accumulation, and coarse mode particles are provided in the SI (Figs. S4–S6).

denoting slow decay, reflecting its resistance to removal processes like deposition. Kaunas amplifies this pattern with (4.22, 0.1), the widest plume observed, which is potentially linked to its high proportion of diesel vehicles (62.1 %). Even in Alytus, eBC maintains a (3.52, 0.1) pair, underscoring its pervasive nature regardless of urban scale. The consistently small λ (~0.1) across cities suggests that eBC's decay is minimally influenced by urban structure, likely due to its strong association with traffic emissions and fine particulate properties, which resist rapid dispersion.

3.3. Emission factors

City segregated emission factors of total particle number (PN), UFP, ACCU, COARSE particles, and eBC mass are presented in Table 1. Although the Kruskal-Wallis test, followed by Dunn's test and Bonferroni correction, revealed no statistically significant differences in emission factors across cities, notable variations were observed, which can be attributed to differences in diesel vehicle fractions and engine technologies. In the following, we discuss the observed variations in EF and contextualize the findings by comparing them with emission factors reported in previous studies.

For LDVs, primarily passenger cars (M1 vehicle category), the median PN emission factor ranged from 6.53×10^{14} in Šiauliai to 8.90×10^{14} particles/(km·veh) in Vilnius. Despite Vilnius having the lowest diesel-powered vehicle fraction (50 % for M1 vehicles; Regitra, 2024), it exhibited the highest median PN EF. The reason for this may be a higher prevalence of gasoline direct injection (GDI) vehicles, which emit high pollutant concentrations. Suarez-Bertoa et al. (2019) reported PN emissions for GDI vehicles ranging from 8.9×10^{11} to 2.1×10^{12} particles/(km·veh), significantly higher than vehicles with port fuel injection technology. However, the reported values are than those observed in Lithuanian, suggesting that older GDI technologies or real-world driving conditions may elevate emissions (Suarez-Bertoa et al., 2019).

The 25th percentile PN EF for LDVs, representing fewer polluting vehicles, ranged from 3.54×10^{14} in Alytus to 4.47×10^{14} particles/(km·veh) in Klaipėda. These values are closer to those reported in European cities, such as Berlin (1.77×10^{14} particles/(km·veh)) by Straaten et al. (2023) and Meckenheim, Germany (1.9×10^{14} particles/(km·veh)) by Nickel et al. (2013). This indicates that the cleaner segment of Lithuania's LDV fleet emits particles at levels similar to those in regions with more modern fleets. In contrast, the 75th percentile, representing more polluting vehicles, reached up to 2.07×10^{15} particles/(km·veh) in Vilnius, significantly exceeding European benchmarks. For instance, Beddows and Harrison (2008) reported field-measured PNC for LDVs up to 8×10^{13} particles/(km·veh), approximately an order of magnitude lower than Lithuania's upper quartile, highlighting the impact of older, high-emitting vehicles.

The UFP number EFs followed a similar pattern, with Vilnius showing the highest median at 9.51×10^{14} particles/(km·veh) and Šiauliai the lowest at 5.92×10^{14} particles/(km·veh). The 25th percentile UFP number EF ranged from 4.13×10^{14} in Šiauliai to 4.59×10^{14} particles/(km·veh) in Vilnius, somewhat like values reported by Imhof et al. (2005) for Zurich (0.38–0.81 $\times 10^{14}$ particles/(km·veh)), suggesting that less polluting LDVs in Lithuania perform comparably to those in some European cities two decades ago. However, the 75th percentile UFP number EF reached 2.07×10^{15} particles/(km·veh) in Vilnius, far exceeding European norms, reflecting the presence of high-emitting vehicles, possibly older diesel or GDI models lacking advanced emission controls.

Accumulation mode and coarse mode PN EF showed less variability. Median accumulation mode PN EF ranged from 6.11×10^{13} in Vilnius to 8.23×10^{13} particles/(km·veh) in Panevėžys. Explanation for Panevėžys showing the highest emission factor for accumulation mode particles can be drawn from the fact that it also has the highest emission factor for BC particles. This correspondence is driven by the city's notably high diesel vehicle fraction (70 %), the largest among the surveyed locations.

Table 1
Estimated emission factors for various pollutant metrics, including particle number concentrations across different size ranges and equivalent black carbon, stratified by vehicle type (light-duty vehicles, buses, and trucks) in six Lithuanian cities. The values represent fleet-average emissions derived from mobile measurements and are intended to reflect real-world urban traffic conditions.

Category	City	PNC ($\times 10^{14}$ #/(km veh))	UFP ($\times 10^{14}$ #/(km veh))	ACCU ($\times 10^{14}$ #/(km veh))	COARSE ($\times 10^{11}$ #/(km veh))	eBC (mg /(km veh))
LDV	PNV	7.63 (4.38 – 18.53)	8.44 (4.39 – 17.82)	0.82 (0.43 – 1.56)	4.50 (2.13 – 7.87)	148.65 (55.90 – 307.62)
	ALY	8.08 (3.54 – 12.29)	8.15 (4.27 – 13.44)	0.74 (0.37 – 1.54)	4.83 (3.31 – 8.25)	102.19 (50.41 – 289.33)
	SIA	6.53 (4.40 – 16.73)	5.92 (4.13 – 18.35)	0.71 (0.34 – 1.31)	4.29 (2.25 – 7.69)	111.40 (55.03 – 226.97)
	KLA	7.54 (4.47 – 15.62)	8.21 (4.25 – 15.20)	0.74 (0.30 – 1.74)	4.22 (2.20 – 8.47)	91.18 (38.92 – 215.63)
	KAU	7.55 (4.04 – 13.41)	7.44 (4.21 – 14.22)	0.73 (0.39 – 1.53)	4.54 (2.18 – 8.58)	99.62 (52.77 – 242.55)
	VLN	8.90 (4.43 – 20.73)	9.51 (4.59 – 20.67)	0.61 (0.36 – 1.56)	4.33 (2.23 – 8.30)	110.79 (50.40 – 227.22)
Bus	PNV	38.05 (36.35 – 39.74)	41.15 (35.37 – 67.36)	–	7.62 (6.32 – 20.81)	–
	ALY	11.03 (9.75 – 12.31)	10.17 (8.55 – 11.81)	–	–	–
	SIA	–	–	0.47 (0.44 – 0.55)	14.10 (11.38 – 19.77)	–
	KLA	7.49 (5.78 – 10.09)	6.92 (5.79 – 9.95)	0.77 (0.67 – 1.08)	5.68 (3.32 – 8.04)	–
	KAU	7.78 (6.07 – 59.59)	6.29 (4.95 – 58.76)	1.12 (0.76 – 2.48)	–	239.89 (153.50 – 326.27)
	VLN	12.28 (5.66 – 15.12)	12.53 (5.99 – 15.13)	0.56 (0.43 – 0.85)	4.64 (3.19 – 7.23)	134.12 (107.61 – 163.93)
Truck	PNV	13.82 (8.75 – 31.44)	9.94 (8.20 – 20.98)	0.78 (0.34 – 1.23)	7.51 (5.00 – 10.76)	131.70 (105.50 – 155.65)
	ALY	23.35 (14.98 – 26.77)	16.59 (9.24 – 23.29)	1.08 (0.63 – 2.24)	5.89 (4.15 – 12.33)	172.71 (87.90 – 487.37)
	SIA	7.52 (4.76 – 9.85)	5.92 (3.87 – 10.22)	0.77 (0.31 – 1.78)	4.26 (3.28 – 11.68)	192.15 (80.77 – 341.61)
	KLA	9.24 (5.80 – 29.84)	12.13 (5.75 – 18.15)	1.38 (0.55 – 2.50)	6.82 (3.54 – 14.46)	110.15 (70.56 – 374.00)
	KAU	10.92 (7.35 – 29.53)	11.02 (6.86 – 15.56)	0.87 (0.47 – 2.20)	6.26 (3.03 – 8.72)	104.46 (76.34 – 262.42)
	VLN	9.02 (5.71 – 26.24)	9.93 (7.00 – 29.42)	1.07 (0.55 – 2.07)	8.82 (4.25 – 13.54)	245.38 (108.03 – 451.10)

Diesel engines are known to emit soot particles, which predominantly fall within the accumulation mode size range and are a primary contributor to black carbon, thus explaining the elevated levels of both pollutants in Panevėžys (Kecorius et al., 2017).

The eBC emission factors for LDVs were highest in Panevėžys (median: 150 mg/(km·veh)), correlating with its high diesel fraction. The 25th percentile eBC ranged from 40 mg/(km·veh) in Klaipėda to 56 mg/(km·veh) in Panevėžys. Krecl et al. (2024) documented a reduction in BC emissions for diesel LDVs from 95 mg/(km·veh) in 2006 to 23 mg/(km·veh) in 2013, indicating that Lithuania's less polluting vehicles emit more eBC than modern diesel vehicles elsewhere. Meanwhile, more polluting vehicles have emissions comparable to those of older, uncontrolled diesel engines. Compared to global studies, Lithuania's eBC values (90–150 mg/(km·veh) median) are higher than Zurich (10 mg/(km·veh)) and São Paulo (16 mg/(km·veh)) but similar to Manila (180 mg/(km·veh)), reflecting the impact of an aging fleet (Madueno et al., 2019).

The buses and trucks in Lithuania emit UFP numbers at rates far above modern heavy-duty standards, reflecting an aging fleet (Grigoratos et al. 2019; Giechaskiel 2018). Median PN EF for buses range from 7.5 × 10^{14} particles/(km·veh) (Klaipėda) to 3.8 × 10^{15} particles/(km·veh) (Panevėžys), and for trucks from 7.5 × 10^{14} particles/(km·veh) (Šiauliai) to 2.3 × 10^{15} particles/(km·veh) (Alytus) - orders of magnitude above Euro-6 limits and older vehicles with diesel particulate filters. The UFP, ACCU, and COARSE PN EFs follow similar trends. The eBC median EFs (buses: ~240 mg/(km·veh) in Kaunas; trucks: 104–245 mg/(km·veh) across Lithuanian cities) lie between Manila's extreme (~1260–1620 mg/(km·veh)) and Zurich's cleaner fleets (427 mg/(km·veh); (Madueño et al., 2019) and references therein), underscoring regional technology and traffic-pattern impacts.

4. Summary and conclusions

Addressing a critical knowledge gap in regions characterized by aging vehicular fleets and elevated diesel engine usage, this study provides an in-depth examination of road-traffic emissions of UFP and eBC across the six largest cities in Lithuania: Vilnius, Kaunas, Klaipėda, Šiauliai, Panevėžys, and Alytus, conducted during the summer of 2024.

Mobile, on-road measurements revealed pronounced spatial disparities in pollution levels among the cities. Panevėžys and Vilnius exhibited the highest median eBC concentrations (traffic increment), registering 10400 ng/m^3 and 10200 ng/m^3, respectively, while Alytus recorded the lowest - 7700 ng/m^3. Statistical analyses substantiated significant differences in eBC concentrations between specific city pairs, notably Alytus and Panevėžys, and Klaipėda and Panevėžys, which were linked to variations in diesel vehicle prevalence and fleet age. Panevėžys, with a diesel-powered vehicle fraction of 70 % and an older fleet, emerged as a hotspot for elevated eBC emissions. A parallel trend was observed for UFP, with Panevėžys recording the highest median concentration at 97800 particles/cm^3, reinforcing the influence of diesel engines on fine particle emissions.

The EMG-based analysis of pollution plumes revealed that high σ and low λ values, typical in larger cities like Vilnius and Kaunas, correspond to broad, persistent plumes caused by dense traffic and limited dispersion. In contrast, smaller towns like Alytus showed narrower plumes and faster decay (low σ, high λ), reflecting better ventilation. The results highlighted differences in urban structure and traffic intensity, and how it shapes the spread and persistence of traffic-related air pollution.

To estimate emission factors, the study adapted the OSPM framework for mobile measurements, treating the measurement vehicle as a dynamic receptor to account for pollutant dispersion in urban settings. This innovative approach yielded detailed emission factors for each vehicle category. For LDVs, Vilnius exhibited the highest median PN EF at 8.90 × 10^{14} particles/(km·veh), potentially reflecting a greater presence of gasoline direct injection vehicles, emitting higher particle numbers. Panevėžys, however, led in eBC emission factors for LDVs at 150 mg/(km·veh), aligning with its diesel-dominated fleet. Buses and trucks demonstrated significantly higher emission factors, with buses in Panevėžys reaching a median PN EF of 38.05 × 10^{14} particles/(km·veh) and trucks in Alytus peaking at 23.35 × 10^{14} particles/(km·veh). This underscores the outsized contribution of heavy-duty vehicles to urban pollution, particularly in cities with outdated fleets. While the hypothesis of statistically significant differences in emission factors due to diesel fractions was not supported, the observed variations offer critical insights into the interplay of fleet composition and air quality.

This study provides a comprehensive assessment of real-world vehicular emission factors for UFP number and eBC mass in Lithuania, shedding light on a region where older vehicles and high diesel usage prevail. The findings presented here can pave the way for data-informed revisions to air quality regulations and the advancement of sustainable urban mobility strategies, promising improved health outcomes and environmental quality for Lithuania and comparable regions worldwide.

CRediT authorship contribution statement

Simonas Kecorius: Writing – review & editing, Writing – original draft, Visualization, Validation, Supervision, Software, Resources, Project administration, Methodology, Investigation, Funding acquisition, Formal analysis, Data curation, Conceptualization. **Leizel Madueño:** Writing – review & editing. **Kristina Plauškaitė:** Writing – review & editing, Supervision, Project administration, Funding acquisition. **Steigvilė Byčenkienė:** Writing – review & editing, Supervision, Project administration, Funding acquisition. **Mario Lovrić:** Writing – review & editing, Software. **Valentino Petrić:** Writing – review & editing, Software. **Manuel Carranza-García:** Writing – review & editing, Software. **Manuel J. Jiménez-Navarro:** Writing – review & editing, Software. **María del Mar Martínez-Ballesteros:** Writing – review & editing, Software. **Gaudentas Kecorius:** Writing – review & editing, Data curation.

Declaration of competing interest

The authors declare that they have no known competing financial interests or personal relationships that could have appeared to influence the work reported in this paper.

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Supplementary materials

Supplementary material associated with this article can be found, in the online version, at doi:10.1016/j.envadv.2025.100661.

Data availability

Data will be made available on request.

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