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An integrated assessment of the impacts of $PM_{2.5}$ and black carbon particles on the air quality of a large Brazilian city

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Abstract

Data on airborne fine particle ($PM_{2.5}$) emissions and concentrations in cities are valuable for traffic and air quality managers, urban planners, health practitioners, researchers, and ultimately for legislators and decision makers. Emissions and ambient concentrations of $PM_{2.5}$ and black carbon (BC) were assessed in the city of Curitiba, southern Brazil. The methodology combined a month-long monitoring campaign with both fixed and mobile instruments, development of emission inventories, and dispersion model simulations on different scales. The mean urban background $PM_{2.5}$ concentrations during the campaign were 7.3 μg m^{-3} in Curitiba city center, but three- to fourfold higher (25.3 μg m^{-3}) in a residential area on the city's outskirts, indicating the presence of local sources, possibly linked to biomass combustion. BC concentrations seemed to be more uniformly distributed over the city, with mean urban background concentrations around 2 μg m^{-3}, half of which due to local traffic emissions. Higher mean BC concentrations (3–5 μg m^{-3}) were found along busy roads. The dispersion modeling also showed high $PM_{2.5}$ and BC concentrations along the heavily transited ring road. However, the lack of in situ data over these peripheral areas prevented the verification of the model output. The vehicular emission factors for $PM_{2.5}$ and BC from the literature were found not to be suitable for Curitiba's fleet and needed to be adjusted. The integrated approach of this study can be implemented in other cities, as long as an open data policy and a close cooperation among regional, municipal authorities and academia can be achieved.

Keywords Integrated air quality assessment · Emission source apportionment · Mobile measurements · Short-lived climate pollutants

Highlights
- Monitoring campaign for $PM_{2.5}$ and BC in city center, residential area, and along roads.
- Real-world emission factors determined for the road vehicles in Curitiba.
- $PM_{2.5}$ and BC maps obtained through combined use of in situ and model results.
- High $PM_{2.5}$ levels in residential area point to unidentified local sources.
- Mean BC levels fairly homogeneous over the city, except close to busy roads.

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Introduction

It is generally accepted that the fast pace of global urbanization is associated with the expansion of modern industries and non-agricultural sectors, especially in low- and middle-income countries (Chen et al. 2014). The urbanization and industrialization have increased the concentrations of outdoor air pollutants from vehicles and factories, contributing to the global burden of asthma and allergic diseases (Zhang et al. 2015). Active policies toward a sustainable growth, e.g., investing in renewable energy (Mujtaba and Hussain Shahzad 2021), may allow economic growth to occur together with reduction in air pollution. In developing economies, the increase in incomes will first lead to a deterioration in air quality, but in later stages—with better environmental awareness and technologies—this relation can change. An analysis of the BRIC countries (Brazil, Russia, India, and China) suggests that for Brazil, despite having a comparatively high rate of renewable energy, more preventive actions and investments in renewable technology are needed to bend the air pollution curve downward (Klafke et al. 2015).

A recent global model assessment with relatively high spatial resolution (11 km × 11 km) revealed that the human exposure to ambient $PM_{2.5}$ (fine particles with an aerodynamic diameter smaller or equal to 2.5 μm), ranked fifth as mortality risk factor in 2015, with more than four million deaths (Cohen et al. 2017). The urgent need to take better care of cities' environment has recently been manifested in the Sustainable Development Goal (SDG) 11 addressing urban planning and development. One specific SDG indicator, 11.6.2, is the annual mean level of $PM_{2.5}$ weighted by population (United Nations Statistics Division 2018), for which a global database was developed by the World Health Organization (WHO 2016). Improved information on the spatial distribution of $PM_{2.5}$ exposure in cities will be required to monitor future progress of the SDG 11 air pollution target.

Ongoing research is trying to identify which components of inhalable particulate matter contribute the most to the observed health effects. The Review of Evidence on Health Aspects of Air Pollution (REVIHAAP) assessment (WHO 2013) could not pinpoint the constituents within the $PM_{2.5}$ matrix more related to specific health effects. Nevertheless, strong research outcomes have shown that black carbon (BC) particles—a fraction of $PM_{2.5}$ emitted by combustion processes—is a relevant additional air quality indicator to assess the health risks of air pollution dominated by primary combustion emissions (Janssen et al. 2011). Besides the health impact, BC is also a short-lived climate forcer (SLCF), contributing to warm up the atmosphere (Bond et al. 2013). Hence, curbing BC emissions has the double benefit of reducing health effects as well as contributing to the mitigation of climate change.

Global assessments of air pollution have revealed that the highest $PM_{2.5}$ exposure levels are found in Asia, whereas lower exposure occurs in the American continent (WHO 2016). However, there is no lower $PM_{2.5}$ threshold below which the adverse effects on human health are negligible. On the contrary, Burnett et al. (2014) showed that a reduction of 1 μg m^{-3} in $PM_{2.5}$ concentrations provides a more significant health benefit in a city with relatively low levels (i.e., in the range of 10–30 μg m^{-3}), as compared to cities with much higher mean concentrations, close or above 100 μg m^{-3}.

Janssen et al. (2011) reported that another important advantage in reducing BC concentrations is that a 1 μg m^{-3} decrease in BC exposure would lead to an increase in life expectancy between 3.1 and 4.5 months per person.

The national ambient air quality standards (NAAQS) in Brazil were implemented in 1990 and legislate particulate matter as PM_{10} and black smoke (another metric for exhaust emissions). However, in 2014, only 12 of the 27 federal units of Brazil had at least one air quality monitoring station (Instituto de Energia e Meio Ambiente 2014). Black smoke is regulated with very tolerant thresholds (mean daily and annual concentrations of 150 μg m^{-3} and 60 μg m^{-3}, respectively), but monitoring occurs in only 12% of all stations in the country (Instituto de Energia e Meio Ambiente 2014). Until November 2018, São Paulo was the only state in Brazil where a $PM_{2.5}$ standard had been implemented, with daily maximum and annual limits of 60 μg m^{-3} and 20 μg m^{-3}, respectively. In December 2018, the $PM_{2.5}$ standard was extended to the other states of the federation. However, with no previous legislation supporting the monitoring of $PM_{2.5}$ and BC, most Brazilian cities lack information on these pollutants, except for a few short-term campaigns, which have been conducted to address specific aspects of atmospheric processes and aerosol characteristics (Krecl et al. 2020a, b; Krecl et al. 2018; Polezer et al. 2018; Polezer et al. 2019; Targino and Krecl 2016; Miranda et al. 2012).

The significant health and climate benefits of reducing $PM_{2.5}$ and BC emissions should be an incentive for the Brazilian environmental agencies to assess their concentrations and spatial distribution across cities and to identify their source contributions. However, given financial and infrastructure constraints, uneven spatial distribution of stations, lack of instrument maintenance, spare parts and technical expertise, the operational monitoring of $PM_{2.5}$ and BC concentrations in Brazil is still challenging. Until the new NAAQS are implemented and stations become operable across the country, there is an urgent need to fill the knowledge gap in terms of local emissions of $PM_{2.5}$ and BC.

A set of recommendations was proposed by the General Assembly of the World Medical Association (WMA) in 2014, including the following: (a) monitoring and limiting the concentrations of nanosize BC particles in urban areas, (b) building professional and public awareness of the hazard of BC and

the existing methods of eliminating the particles, (c) developing strategies to protect people's exposure to BC in motorized transport, homes and in the general environment (World Medical Association 2014).

The present work is the result of a 2-year bilateral Swedish-Brazilian cooperation entitled ParCur ("Particles in Curitiba"), conducted in Curitiba, the capital city of the State of Paraná. In line with the SDG objectives, the WMA's guidance, as well as the SLCF reduction initiatives, such as the Climate and Clean Air Coalition (CCAC), Sweden supports bilateral cooperation with specific countries in the fields of environmental protection, climate change, and sustainable development. Hence, ParCur gathered experts from Brazil and Sweden, local stakeholders and end users. The objective of the cooperation was the integration of fixed and mobile $PM_{2.5}$ and BC measurements at high spatiotemporal resolution, the development of emission inventories and the implementation of modelling tools at different spatial scales for validating the emissions and for determining the spatial distribution of pollutant concentrations across the city.

Method

Study area

Curitiba is located in southern Brazil, on a plateau at approximately 900 m above sea level, and at a distance of 110 km from the Atlantic Ocean. The Curitiba Municipality has an estimated population of 1.9 million inhabitants. From the urbanistic standpoint, the Serete Plan, created in 1964 and implemented in the 1970s, was the backbone of the development plan that shaped the city's current structure and morphology (Santos 2014). This plan organized the Municipality using the triad: public transportation, land use, and road system, which led to the creation of the structural axes or transport corridors, some dedicated to the Bus Rapid Transit (BRT) system, pioneered in Curitiba in 1974. An important feature of the city´s planning is the separation of the city center from the industrial areas. The latter is located in the southwest, near the city of Araucária, downwind of the dominating northeasterly winds.

The official air quality monitoring network is managed by the Environmental Institute of Paraná[1] (IAP) and consists of four automatic stations within the Curitiba Municipality: Boqueirão (BOQ), Cidade Industrial (CIC), Ouvidor Pardinho (PAR), Santa Cândida (STC), and four in the industrial area.

The air quality in Curitiba is affected by emissions generated inside the city, by external sources located elsewhere in the State of Paraná and in other parts of the continent. Observational and modelling studies have shown that smoke plumes generated during the biomass burning season in northern and central Brazil (August–October, with peak activity in September) reach the southern states, including the Curitiba region (Targino et al. 2019; Rosário et al. 2013).

Study design

A combination of measurements and dispersion modeling was used to map the $PM_{2.5}$ and BC spatial distributions in Curitiba in July and August 2016. The full assessment included the following components (see map in Fig. 1):

1. Analyzing nitrogen oxides (NO_2 and NO_x) and PM_{10} concentrations collected between 2013 and 2015 at the four IAP monitoring sites in Curitiba (PAR, BOQ, STC, and CIC), with the objective of characterizing the general pollution levels, extreme values, and seasonal variability within the study area, for a period close to the 2016 campaign.
2. Developing an emission inventory for the city of Curitiba, by collecting and complementing available emission data from the State of Paraná.
3. Performing a field campaign at two fixed sites (Fig. 1, left) aimed at: (a) Monitoring of NO_x, $PM_{2.5}$, and BC concentrations within a street canyon (Marechal Deodoro, hereafter MD) in the city center at two levels above ground: street (height of 5 m, hereafter MD street) and rooftop (height of 70 m, hereafter MD roof).(b) Monitoring of $PM_{2.5}$, BC, elemental carbon (EC) and organic carbon (OC) concentrations in a residential area (Sítio Cercado, hereafter SC) located 13 km from the city center, and 750 m from the busy highway BR-376, which is part of Curitiba's ring road ("Contorno" as local designation).
4. Performing a monitoring campaign with instruments onboard bicycles to measure $PM_{2.5}$ and BC concentrations along different types of roads in the city center (see Fig. 1, right).
5. Implementing dispersion models at the regional, urban and street canyon scales to support the interpretation of the monitored data inside the Curitiba Municipality.
6. Consolidating the street canyon data and model output to obtain real-world emission factors (EFs) for $PM_{2.5}$ and BC for road transport in Curitiba.
7. Using the regional and urban modeling, together with the monitored data, to conduct a source apportionment of $PM_{2.5}$ and BC concentrations in the Curitiba Municipality.

The street canyon modeling aimed to assess the impact of vehicles circulating in the city center on the air quality

[1] The name recently changed to Instituto Agua e Terra (IAT)

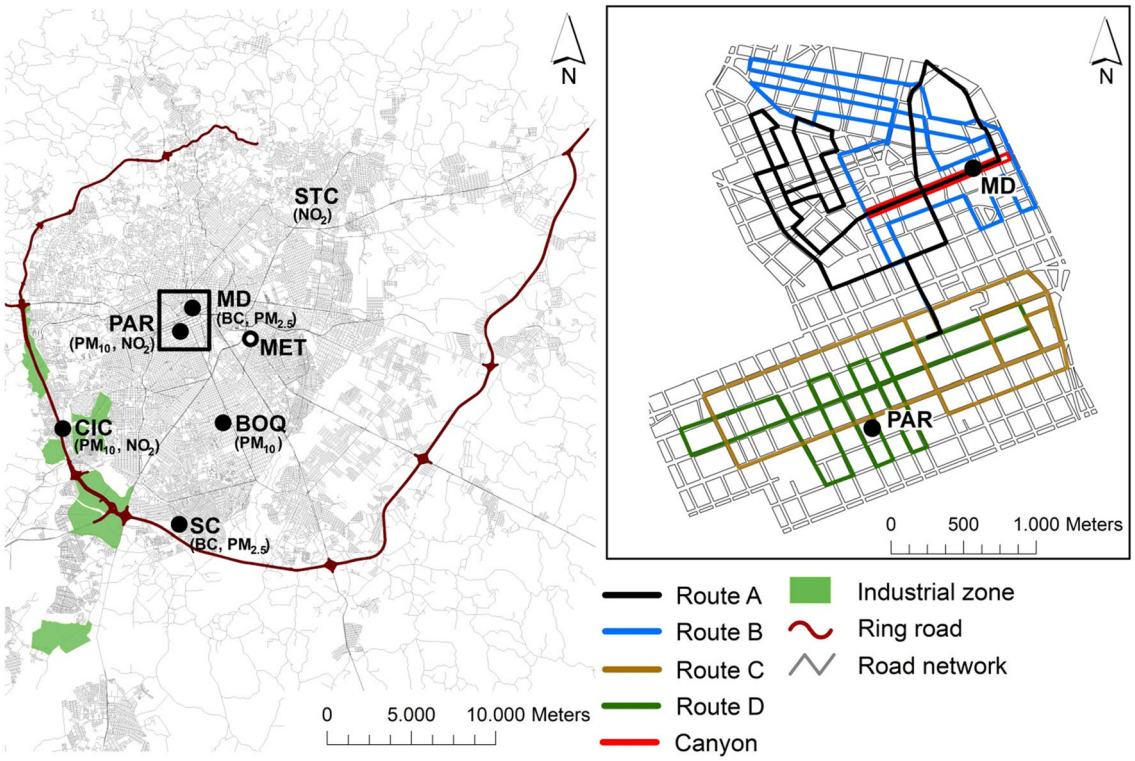

Fig. 1 Maps showing the location of (left) the four official monitoring stations (CIC, PAR, BOQ, STC), the site in the residential area (SC), and the meteorological station (MET), and (right) the street canyon site (MD), and the four biking trajectories used for the mobile measurements

concentrations within a narrow street canyon, and to determine their EFs using in situ data, since vehicular emissions are a major pollution source in such microenvironments. The comparison between measurements and model output was also extended to the urban background of the Curitiba Municipality, by using data collected in the city center and in a residential area. This strategy allowed assessing the validity of the emission inventory covering the Curitiba Municipality, i.e., the area for which high resolution modelling was performed.

Emission inventory

The emission inventory developed for Curitiba considered $PM_{2.5}$, BC, and NO_x for two major economic sectors: industries and on-road transport.

Industrial emissions

Industrial emission values of NO_x, sulfate dioxide (SO_2), and PM_{10} from large industrial sources were compiled from the official regional inventory that covers the State of Paraná (IAP 2013), while the Curitiba Municipality provided data for the inner-city small-scale industries. All industrial emissions were treated as point sources, with stack emission characteristics given by the IAP inventory. This inventory is based on a policy in which the industries are supposed to monitor their own emissions and report to the official authorities. The inventory revealed a cluster of sources at Curitiba's Industrial Site, located in the neighboring city of Araucária, southwest of Curitiba's city center. This industrial site was established in the 1970's following an urban planning which decreed that all industries should be moved from the center to this new area. Araucária is one of the largest industrial sites of Brazil, hosting a state-owned oil refinery together with several steel, cellulose/pulp and chemical industries.

Since the existing industrial inventories only included PM_{10} emissions, there was a need to specify also $PM_{2.5}$ and BC emissions for nearby sources that could have a potential impact on the air quality in Curitiba. A complementary emission inventory was therefore elaborated for the industrial sources in the Araucária area. The inventory was first developed for $PM_{2.5}$ following the methodology proposed by the EMEP/EEA Atmospheric Emission Inventory Guidebook (EEA 2019; Tier 2), considering activity data provided by IAP for the year 2016 and EFs taken from the EMEP guidebook for six fuel types. BC emissions were subsequently calculated using $PM_{2.5}$ emissions and BC/$PM_{2.5}$ emission ratios proposed by EMEP. The calculations included 87 combustion processes and 25 industries.

For the rest of the State of Paraná industrial sources, as well as for the few and small industries in and around Curitiba, we assumed that 70% of the emitted PM_{10} consisted of $PM_{2.5}$ (Ehrlich et al. 2007). We also assumed that these industries

emitted BC in the same amount as the average of the Araucária sources (ratio $BC/PM_{2.5}$ = 7%).

Road traffic emissions in the state of Paraná

Traffic emissions for the State of Paraná, but outside the city of Curitiba, were also extracted from the official regional inventory (IAP 2013). Since only exhaust PM emissions are provided in the IAP inventory, we assumed that PM_{10} and $PM_{2.5}$ contributions from traffic were equivalent to PM, and BC contributions were calculated as fractions of total exhaust PM. From the EMEP/EEA handbook (EEA 2018; Table 3-91) the $BC/PM_{2.5}$ ratios were estimated as 20% for passenger cars and motorcycles (gasoline only, since diesel-fueled passenger cars are not allowed in Brazil), 70% for diesel-fueled light-duty vehicles (LDV) and 65% for diesel-fueled heavy-duty vehicles (HDV), considering the mix of vehicle technology in Curitiba for 2016. According to the Brazilian national vehicular inventory (Ministério do Meio Ambiente 2014), gasoline cars/motorcycles contribute with 10%, LDV 24%, and HDV 66% to the total traffic exhaust PM. Weighting together these estimations, the suggested overall BC/PM ratio for long-range transport (LRT) contributions originating in the State of Paraná was determined as 0.62.

Public transport emissions in Curitiba

Public transport emissions were calculated as line sources along the bus network, as facilitated by the Municipality. Information on bus size, technology (Table 1), average daily distance traveled, and average fuel consumed were obtained for each bus line. Together with the bus timetables, it was possible to describe the number of buses transiting a certain road link on an hourly basis. We used a relation between the Brazilian emission legislation PROCONVE and the European EURO classes (TransportPolicy.net 2018a, b), enabling the use of EFs from the European HBEFA database (INFRAS 2017), as listed in Table 2. We assumed a road speed limit of 50 km h^{-1} and saturated traffic conditions. The bus types listed in Table 1 were aggregated into HBEFA bus classes with weights < 15 t, 15–18 t, and > 18 t. Emission factors for bi-articulated buses were extrapolated using information on fuel consumption provided by the Curitiba Municipality. BC emission factors were taken from the European Environment Agency (EEA 2016) as $BC/PM_{2.5}$ fractions in the range 65–75%. The PM and BC emission factors for buses using 100% biofuel were reduced by 50%, according to the findings published by the U. S. Department of Energy (2018).

Private vehicle emissions in Curitiba

Private vehicle emissions were calculated based on the number of vehicles per road link (a total of 3061 road links, excluding smaller secondary roads) for the morning and afternoon peak hours, acquired from Curitiba's travel demand model, which was created with the software VISUM (PTV GROUP 2019). VISUM is a macroscopic demand model which, based on socio-economic data and the transport network characteristics, estimates the number of trips per time period between all the defined origins and destinations within the modeled region and assigns those trips to the network, resulting in the traffic volume on each link. The specific model for Curitiba was developed in 2014 and was calibrated and validated based on traffic counts carried out on 80 different links of the road network. Hourly variations of traffic volume over the day were taken from 230 instruments monitoring the speed of individual vehicles and aggregated to different profiles according to the day of the week: Mondays–Thursdays, Fridays, Saturdays, and Sundays (Fig. 2). The same daily profiles were used for all streets.

Table 1 Number of buses in the public transportation system of Curitiba classified by type, technology, and fuel type for the year 2016 (source: Curitiba Municipality)

Bus type and length	Euro II diesel	Euro III diesel	Euro III biodiesel	Euro V diesel	Euro V biodiesel	Total
Micro (8 m)	18	2	0	0	0	20
Special micro (10 m)	0	172	0	0	0	172
Common (12 m)	52	555	0	3	0	610
Semi-standard (13 m)	0	30	0	0	0	30
Standard (13 m)	9	328	0	0	0	337
Standard hybrid (13 m)	0	0	0	28	2	30
Articulated (18.6 m)	38	198	0	0	0	236
Articulated (20 m)	0	35	6	0	0	41
Bi-articulated (25/28 m)	54	92	26	0	0	172
Total	171	1424	32	31	2	1660

Table 2 Emission factors for public transport

Bus classes	Technology	NO_x	PM exhaust		BC		Fuel consumption
		(mg veh^{-1} km^{-1})	Diesel (mg veh^{-1} km^{-1})	Biodiesel	Diesel (mg veh^{-1} km^{-1})	Biodiesel	(ml veh^{-1} km^{-1})
Micro	Euro II	9840	184	92	120	60	326
Standard	Euro II	13080	264	132	172	86	444
Articulated	Euro II	16380	373	187	242	121	568
Bi-articulated	Euro II	19438	443	221	288	144	674
Micro	Euro III	9020	171	86	120	60	344
Standard	Euro III	11740	237	119	166	83	463
Articulated	Euro III	14770	285	143	200	100	588
Bi-articulated	Euro III	17527	338	169	237	118	698
Micro	Euro V	6690	52	26	39	20	298
Standard	Euro V	8370	68	34	51	26	410
Articulated	Euro V	7750	81	41	61	30	535
Bi-articulated	Euro V	9197	96	48	72	36	634

As for the fleet composition, a simplified assumption was made with three different shares depending on the type of road. The following shares were adopted, as suggested by the traffic engineers at the Curitiba Municipality: for inner-city roads (as limited by the ring road): 93% cars, 5% utility vehicles, and 2% trucks; for a few larger thoroughfares open for LDVs: 82% cars, 8% utility and 10% trucks; and for the ring road: 59% cars, 9% utility, and 32% trucks. Table 3 shows the assumptions made on size and technology, together with EFs taken from EEA's emission inventory guidebook (EEA 2016). Utility vehicles and trucks were assumed to be diesel-fueled, while flex-fuel cars were running on gasoline as drivers' primary fuel choice due to favorable price. Stop-and-go emissions due to congestion or traffic lights were not considered. It follows from the EFs displayed in Table 3 that the most critical assumption is the percentages of diesel-fueled LDVs and HDVs.

Other combustion sources in Curitiba

An attempt was made to collect data on the use of wood or coal by restaurants. However, the database gathered was incomplete in space, impeding the inclusion of this source in the emission inventory. Neither was it possible to obtain data on the residential use of wood stoves for cooking or heating. However, municipal authorities informed that residential wood combustion should be minimal, at least in the city center.

Monitoring campaign

Fixed-site measurements were conducted in the winter period from July 25 to August 24, 2016, when pollution levels are expected to peak (see the "PM_{10} and NO_2 concentrations as registered by the official monitoring network" section for

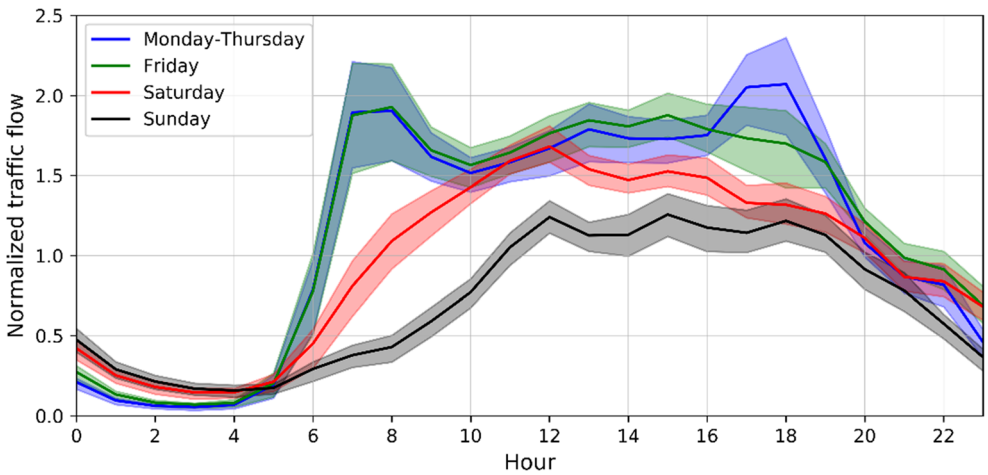

Fig. 2 Traffic volumes normalized by mean flow from the speed radar data collected at 230 locations in Curitiba, used as basis for traffic time variations in the dispersion modelling. Shadowed areas indicate range between 25th and 75th percentiles

Table 3 Emission factors for private vehicles taken from the report by the European Environment Agency (EEA 2016), where BC emission factors are expressed as percentages of $PM_{2.5}$ emissions (see Table 3-91 in EEA 2016). Cars with flex-fuel engines run predominantly on gasoline. Heavy duty vehicles (HDVs) constitute a mix of emissions standards, here simplified to an intermediate technology of Euro III

		NO_x (mg veh^{-1} km^{-1})	PM exhaust (mg veh^{-1} km^{-1})	BC (mg veh^{-1} km^{-1})	Fuel consumption (ml veh^{-1} km^{-1})
Gasoline cars	Euro 4	61	1	0.15	115
Diesel LDV, < 3 t	Euro 4	831	41	36	95
Diesel HDV, 16–32 t	Euro III	6270	130	91	250

further details). $PM_{2.5}$ monitoring was conducted with three types of instruments: a Harvard Impactor (deployed at street and rooftop levels of the canyon site), a MicroVol low volume air sampler (model 1100, Ecotech, Australia) at SC, and a DustTrak unit (model 8520, TSI, USA) deployed at SC.

The Harvard Impactor collected $PM_{2.5}$ samples on 37-mm teflon filters for gravimetric analysis with a 24-hour resolution. Daily integrated samples for gravimetric, EC and OC analyses were collected on 47-mm quartz fiber filters using the Ecotech MicroVol. The gravimetric analyses were performed following the NIOSH Method 5000 (NIOSH 2003), while the EC and OC analyses were conducted at Stockholm University using the NIOSH temperature protocol (Birch 2003). The $PM_{2.5}$ output from the DustTrak instruments was calibrated with the gravimetric data from SC, yielding a correction factor of 1.92 (R^2 = 0.77), which was subsequently applied to all DustTrak outputs. This correction factor is within ranges reported by other studies conducted in urban areas, from 1.70 (McNamara et al. 2011) to 2.78 (Wallace et al. 2011).

Total BC concentrations at street and roof levels were measured with aethalometers (models AE42 and AE33, respectively, Magee Scientific, USA) operating at seven wavelengths (370, 470, 520, 590, 660, 880, and 950 nm), flow rate of 5 L min^{-1} and 5-min resolution. At SC site, BC concentrations were determined with microaethalometers (model AE51, AethLabs, USA) operating at the wavelength of 880 nm, flow rate of 50 mL min^{-1}, and 1-min resolution. These instruments use the wavelength-dependent absorption cross-section values provided by the instrument manufacturers to convert aerosol absorption coefficient into BC mass concentrations. In this study, a site-specific absorption cross section of 18.39 m^2 g^{-1} was determined for SC by correlating daily mean aerosol absorption coefficients with collocated EC concentrations (R^2 = 0.96). The BC data from the AE33 and AE42 instruments were determined using the absorption cross section provided by the manufacturer.

NO_x concentrations were measured at the roof and street levels of the MD site using Ogawa passive samplers (Hagenbjörk-Gustafsson et al. 2010) during two fortnight periods each. The results of the NO_x campaign have been published by Felix et al. (2019). In the present study, the NO_x measurements were used to support the determination of local EFs for vehicle emitted $PM_{2.5}$ and BC.

To supplement the datasets and support the modelling activities, mobile monitoring was performed on 10 selected days and times (morning and evening rush hours) between August 1 and 14, 2016. High spatio-temporal BC and $PM_{2.5}$ concentrations were collected along prescribed routes using two retrofitted bicycles as sampling platforms, equipped with a microathalometer AE51 and a Dusttrak 8520, respectively, following the experimental design by Targino et al. (2016).

Dispersion modeling

Dispersion modeling was performed at three spatial scales with three different models, all giving hourly outputs for the monitoring period from July 25 to August 24, 2016. The regional scale modeling was performed with the BRAMS 5.2 modeling system (Freitas et al. 2017), which includes an atmospheric chemistry transport model (CCATT) coupled online with a limited-area atmospheric model. For this specific experiment, the model was configured to simulate aerosol emission, transport, and dispersion during the campaign period. The BRAMS physical parameterizations were configured with Mellor–Yamada level-2.5 turbulence scheme (Mellor and Yamada 1982) and Joint UK Land Environment Simulator (JULES) surface–atmosphere interaction model (Moreira et al. 2013). For shortwave and longwave radiation schemes, the rapid radiative transfer model for general circulation models (RRTMG) was used with 1200s frequency update of the radiation trend (Iacono et al. 2008). Finally, the Grell and Freitas (2014) ensemble version for deep and shallow convection and the single-moment bulk microphysics parameterization from Walko et al. (1995) were used. The model was applied over two domains: one covering large parts of South America with a grid resolution of 50 km × 50 km (G1, Fig. 3, left) and nested down to a 10 km x 10 km grid resolution over the State of Paraná (G2, Fig. 3, left) with anthropogenic emissions taken from a South American inventory (Alonso et al. 2010). Biomass burning sources were taken from the model 3BEM (Longo et al. 2009) and biogenic emissions of gases from MEGAN (Guenther et al. 2012). These emission fields were generated by the preprocessor PREP-CHEM-SRC (Freitas et al. 2011). The preparation of the

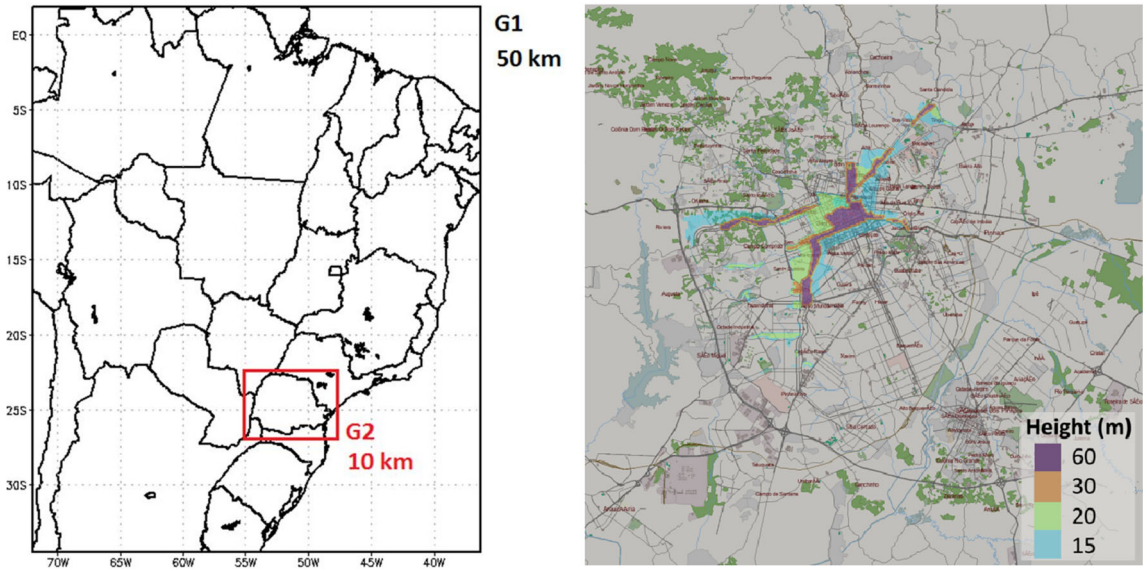

Fig. 3 Domains of the regional model (left) and the urban dispersion model (right). For the latter, building heights above 15 m are marked. The building height was set to 6 m over the remaining built-up urban areas. The simple building height classification was elaborated through inspection of the Google Earth map

$PM_{2.5}$, PM_{10}, and BC emissions for the State of Paraná has been described in the "Emission inventory" section.

For the urban scale impact modeling of emissions inside the Curitiba Municipality, a Gaussian dispersion model was used over a 32 km × 32 km domain (Fig. 3, right) with a grid spacing of 200 m × 200 m. This model is part of the Airviro system (Airviro 2020) and incorporates a diagnostic wind model (Danard 1977) that takes into account surface roughness calculated over built-up areas as a function of building heights, with a maximum roughness of 0.7 m for the city center areas with the highest buildings. The wind model assumes that small-scale winds can be seen as a local adaptation of large-scale winds (free winds) due to local fluxes of heat and momentum at the surface. The free wind is estimated from a vertical profile at the location of a meteorological station, using scaled stability variables. For this application, the input meteorological data were measured at one location (market as MET in Fig. 1, left) and the building heights are illustrated in Fig. 3, right. The regional model output could be directly added to the urban model without double counting, since the sources within the Curitiba Municipality were excluded in the regional model.

The street canyon simulations at the MD monitoring site were performed with the OSPM model (Berkowicz 2000), available in the Airviro system. This street canyon model consists of two components: a plume model that simulates the direct contribution from vehicles, and a box model that calculates the contribution from the recirculation part created by the vortex. Wind input data are taken from the diagnostic wind model, assuming that its output represents the wind at roof level. Neutral stability is assumed within the street canyon. The OSPM model was only used to determine the air pollution contribution from local traffic inside the MD street canyon. Model output was compared to the street canyon increment (defined as the difference between the concentrations measured at street and at rooftop levels). Building heights and the dimensions of the street canyons were determined using 3D Lidar data provided by the Curitiba Municipality. The number of private vehicles passing the MD station was estimated by municipal traffic engineers as 24,075 vehicles per day, as an average over the week, composed by 93% of cars, 5% diesel-fueled LDV and 2% diesel-fueled heavy-duty trucks. The public transport consisted of 469 buses per day.

Results

PM_{10} and NO_2 concentrations as registered by the official monitoring network

The highest PM_{10} concentrations during the period 2013–2015 were recorded at station CIC (Table 4, note the low data capture at this station), located close to the industrial area and the ring road, with both mean and maximum daily values within the NAAQS for Brazil (40 and 120 μg m^{-3}, respectively). The same applies to NO_2, for which the annual mean limit is 60 μg m^{-3} and the maximum measured annual mean concentration was 26.5 μg m^{-3} at station PAR, located at a square in the city center. Averaging over the three years, all stations showed the highest values in August, which incentivized the $PM_{2.5}$ and BC monitoring campaign of this study to be performed between July and August 2016.

The ratios between the mean PM_{10} and NO_2 concentrations during the monitoring campaign and their respective mean

Table 4 PM_{10} and NO_2 concentrations (μg m^{-3}) from Curitiba's official monitoring network for the period 2013–2015, together with present NAAQS (valid from 2018)

Pollutant	Station	Mean	Hourly max	Daily max	Data capture	NAAQS	
						Annual	Daily
PM_{10}	PAR	15.1	180	86	91%	40	120
	CIC	30.3	326	120	43%		
	BOQ*	14.5	197	90	88%		
NO_2	PAR	26.5	201	89	88%	60	-
	CIC	22.5	148	57	59%		
	STC*	13.2	90	43	76%		

* At BOQ only PM_{10} was measured and at STC only NO_2

values in August of 2013, 2014, and 2015 were for PM_{10} 70% at PAR, 50% at BOQ (the monitor at CIC was not in operation during the campaign), and for NO_2 47% at PAR, 76% at CIC, and 86% at STC. Thus, both mean PM_{10} and NO_2 concentrations during the 2016 campaign were lower than in the same period from 2013 to 2015.

While the number of public buses in operation decreased by 6% between 2013 and 2016, the official number of registered private vehicles in Curitiba increased by 4% (http://www.detran.pr.gov.br). Although the registered fleet does not necessarily equal the actual number of vehicles in circulation, it gives the best estimate of the private traffic trend. Regarding the industrial emissions, and despite the lack of data that could sustain an analysis of the trend, to the authors' knowledge, no significant changes occurred over the reported 4-year period. Therefore, local emissions in Curitiba are unlikely to have changed substantially from the period 2013–2015 to 2016, and the identified differences in pollution levels are mostly due to variations in meteorological conditions and the LRT pollution advected to Curitiba. A comparison of meteorological conditions showed considerably more precipitation during August 2016 (163 mm) than expected at this time of the year based on a 30-year climatology (73 mm), while temperature, wind direction and speed were similar to what was registered over the previous 10–20 years (INMET 2019).

$PM_{2.5}$, PM_{10}, BC, and NO_x concentrations measured during the campaign

The mean $PM_{2.5}$ concentration for the whole campaign period at rooftop level at MD site was 7.3 ± 4.3 μg m^{-3} (daily data). Due to equipment malfunctioning, the $PM_{2.5}$ filters exposed at street level could not be used. However, during the mobile monitoring with the bicycles, two 15-min records from weekdays' morning and afternoon rush hours were obtained during stops at the MD street level station. Those data indicated a mean $PM_{2.5}$/BC ratio of 3. Using this scale factor on the BC data obtained with the AE42 at street level (mean concentration of 5.5 μg m^{-3} at 880 nm over the period), a $PM_{2.5}$ concentration of 16.5 μg m^{-3} was estimated for the street level.

At the residential site SC, the gravimetric analysis of 17 filters yielded a mean $PM_{2.5}$ concentration of 36.2 ± 19.0 μg m^{-3} (daily data) of which 11.2 μg m^{-3} was OC and 2.6 μg m^{-3} EC. This means that the total carbon (EC+OC) contributed with 38% to the $PM_{2.5}$ mass and yielded a mean OC/EC ratio of 4.4. The mean $PM_{2.5}$ concentration registered continuously by the DustTrak monitor during 25 days, after gravimetric calibration against the 17 filter samples, was 25.3 ± 32.9 μg m^{-3} (hourly data). The longer time period for the $PM_{2.5}$ mean value obtained with the DustTrak monitor makes it more representative than the filter means.

On average, BC concentrations were highest at street level (5.5 μg m^{-3}), followed by rooftop level (2.3 μg m^{-3}, representing urban background conditions), and 2.2 μg m^{-3} at the residential SC site. The mean BC street increment (calculated as the difference between the concentrations at MD street and MD roof) equaled 3.2 μg m^{-3} and can be attributed to local traffic within the street canyon. Figure 4 shows the daily BC cycle in the city center with peak values occurring in the morning (ca. 10 μg m^{-3}) and evening (ca. 7 μg m^{-3}) rush hours.

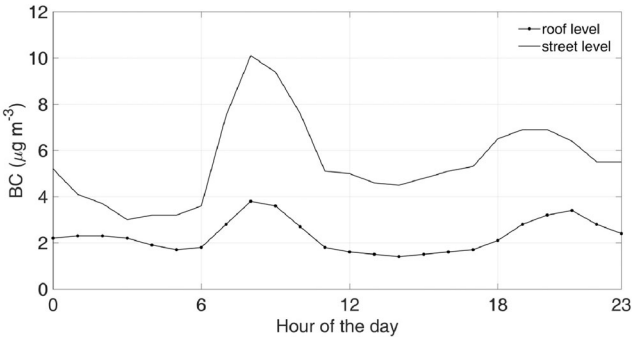

Fig. 4 Mean diurnal cycle of BC concentrations (at 880 nm wavelength) measured at roof and street levels at MD site. Period: July 25–August 24, 2016 (all days of the week)

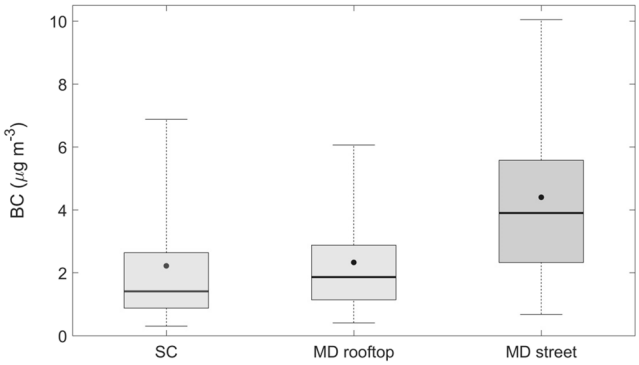

Fig. 5 Boxplots of hourly BC concentrations measured at MD and SC sites in the period July 25–August 24, 2016. The midline is the median, the upper and lower limits of the boxes are the 75th and 25th percentiles, the whiskers are the 5th and 95th percentiles, and the black dot is the mean

Figure 5 summarizes the descriptive statistics of BC concentrations measured in the city center (MD street and roof) and at the SC site. The BC concentrations at MD roof were fairly similar to those recorded at the residential site (SC), and substantially large inside the street canyon.

Figure 6 displays boxplots of BC data gathered simultaneously in the city center with mobile monitoring and at MD street. The median BC within the canyon was larger than the one obtained in the mobile samplings (5.5 and 4.0 μg m^{-3}, respectively). However, the mobile data showed a larger variability with lower 5th percentile and larger 95th percentile, illustrating the heterogeneity of BC concentrations, i.e., the large variations in traffic intensity and street layout (ventilation) characterizing the biking routes across the city center.

The mean NO_x concentrations at MD site were 43 ± 2.8 and 55 ± 1.8 μg m^{-3} at roof level, and 105 ± 1.1 and 122 ± 6.8 μg m^{-3} at street level, for the two 14-day periods of the passive samplers. From this, the street canyon increment was determined as 62 and 67 μg m^{-3}, respectively, for the two time periods.

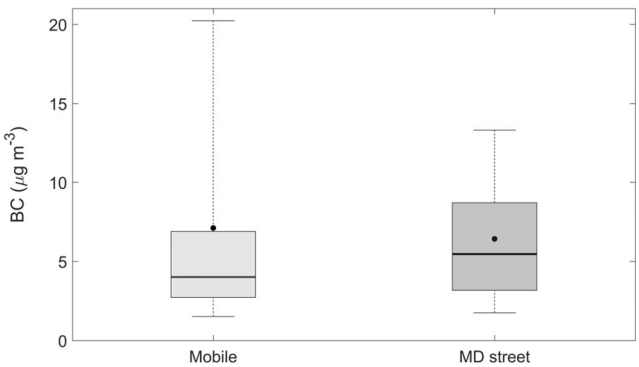

Fig. 6 Boxplot of BC data collected with mobile monitoring (10-s resolution) along the four biking routes and from 5-min concentrations simultaneously measured at MD street site. The midline is the median, the upper and lower limits of the boxes are the 75th and 25th percentiles, the whiskers are the 5th and 95th percentiles, and the black dot is the mean

Figure 7 illustrates the temporal variability of the $PM_{2.5}$ concentrations at the residential site SC. For comparison purposes, PM_{10} concentrations at the station BOQ (in a residential part of the city halfway between station SC and the city center) and at the station PAR (in the city center) are also shown. While the time series partly follow each other, there are a number of events—most pronounced around August 7 and 13—when the $PM_{2.5}$ concentrations at SC are much higher than the PM_{10} at BOQ and PAR.

The strong co-variation of BC concentrations (r = 0.68 for hourly data) between sites MD roof and SC—spaced 13 km from each other—suggests the influence of a common air pollution source across the study area (Fig. 8). A similar temporal variation as for BC can be seen in NO_x, yielding a correlation coefficient of 0.75 between BC at station SC and NO_x at station CIC.

LRT as simulated by the regional model

The impact of sources outside the Curitiba Municipality, including both nearby industrial sources in the Araucária Municipality southwest of Curitiba, and remote contributions from the State of Paraná and the rest of the continent, was simulated by the regional model BRAMS 5.2 (see the "Dispersion modeling" section). The simulated contributions of LRT to Curitiba air quality are presented in Table 5 (PM_{10} is also included for comparison purposes). The contributions to $PM_{2.5}$ from sources outside the Curitiba Municipality were predominantly industrial, with small contributions originating from mobile sources and biomass burning. The transport of BC into Curitiba from sources outside the city comes in similar amounts from industries and mobile sources, while the biomass burning contribution—at least during this month-long campaign—was small.

Simulations of local traffic impact inside the street canyon

We ran the street canyon dispersion model OSPM separately for public transport and private vehicles for the period July 25–August 24, 2016, with output at the MD site. Table 6 shows the comparison of measured and simulated NO_x, $PM_{2.5}$, and BC concentrations due to local traffic inside the street canyon, using EFs from the literature (Tables 2 and 3). The agreement was relatively good for NO_x, whereas the $PM_{2.5}$ and BC simulated concentrations were much lower than the measurements. Since uncertainties related to the meteorological data and model assumptions (including vehicle fleet composition) are equal for all three species, it is reasonable to suggest that the larger discrepancies found for BC and $PM_{2.5}$ were caused by the assumptions in the EFs. For the comparison of simulated and measured $PM_{2.5}$ concentrations, it should be noted that the non-exhaust fraction of $PM_{2.5}$ due

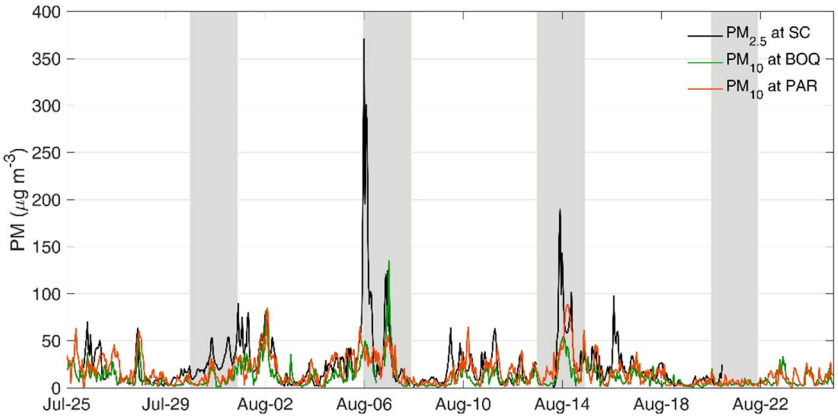

Fig. 7 Monitored $PM_{2.5}$ concentrations at the residential site SC, together with monitored PM_{10} concentrations at site BOQ (southern residential part of the city) and at site PAR (city center). The shadowed areas represent weekends

to road dust suspension, tire and break wears, was not considered in the model simulation.

Since BC measurements were available as hourly data, it was possible to perform a multiple regression analysis using the measured BC increment as dependent variable and the simulated BC concentrations from buses and private traffic as two independent variables. The regression—with a forced zero intercept—was based on 696 hourly data points from the monitored increment and from model output for the two simulations. The regression analysis suggests that the public transport signal should be multiplied by a factor of 1.2 and the private traffic by a factor of 5, yielding an adjusted R^2 of 0.78. Given the linearity between EFs and concentration contributions of inert pollutants in the street canyon model, Fig. 9 and the mean values of Table 7 show that with a corresponding correction of the EFs, the model output would match the measured street increment in the street canyon. With the assumed fleet composition, the real-world BC emission factor for the mixed private vehicle fleet in Curitiba was corrected from 4 to 19 mg veh^{-1} km^{-1}.

We could not perform a similar regression analysis for $PM_{2.5}$ because there were no hourly data available. However, scaling up the summed model output of the bus and private vehicle contributions to $PM_{2.5}$ by a factor of 5, yielded a local contribution similar to the averaged monitored street canyon increment.

Urban and regional simulations of $PM_{2.5}$ and BC

The Gaussian urban model was applied with the corrected EFs for the on-road vehicles, and the results were subsequently added to the regional LRT contributions. Table 8 (excluding the values within parentheses) shows the contributions at the two sites, and Fig. 10 displays the mean spatial distribution of the simulated $PM_{2.5}$ and BC concentrations. The emission inventory shows local $PM_{2.5}$ emissions of 643 t $year^{-1}$ from the transport sector (public and private together) within the urban model domain over Curitiba, while the industrial emissions from industries located inside the city and from the industrial Araucária area just outside the city were 342 tons $year^{-1}$ and 2050 t $year^{-1}$, respectively. For BC, we estimated 18 t $year^{-1}$ from public transport and 375 t $year^{-1}$ from private traffic. The industries inside Curitiba contributed 23 t $year^{-1}$, while the industrial BC emissions in the nearby Araucária area were 135 t $year^{-1}$.

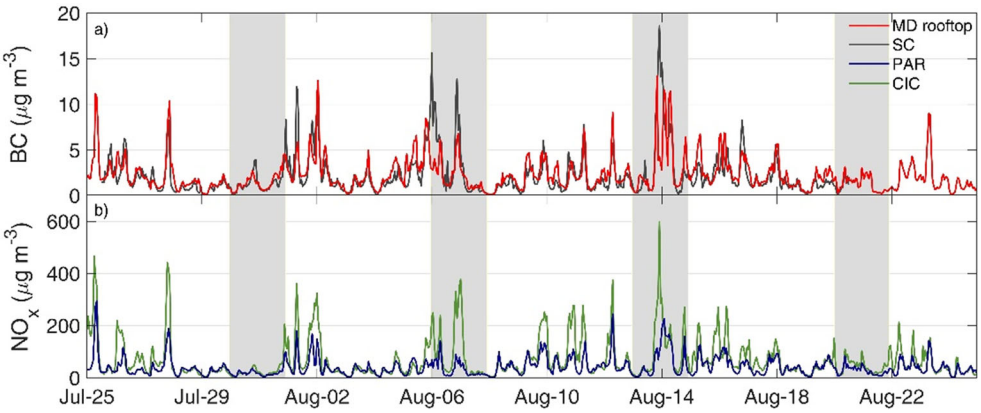

Fig. 8 **a** Monitored BC concentrations at the urban center MD roof and at the residential area SC (top) and **b** monitored NO_x concentrations at site PAR (city center) and at site CIC (close to the ring road) (bottom). The shadowed areas represent weekends

Table 5 Contribution of long-range transported pollution (from sources outside the Curitiba Municipality, evaluated at MD in the city center and at the residential site SC. Values within parenthesis show the contribution of industrial/mobile sources. Period: July 25 to August 24, 2016

Emission sources	Pollutant	MD rooftop (μg m^{-3})	SC (μg m^{-3})
Anthropogenic including IAP inventory	PM_{10}	2.2 (2.0/0.3)	3.6 (3.3/0.3)
Biomass burning		0.5	0.5
Anthropogenic including IAP inventory	$PM_{2.5}$	1.7 (1.4/0.3)	2.6 (2.3/0.3)
Biomass burning		0.2	0.2
Anthropogenic including IAP inventory	BC	0.27 (0.09/0.18)	0.32 (0.15/0.17)
Biomass burning		0.03	0.03

* The IAP inventory provided the PM_{10} industrial emissions and mobile emissions. Industrial $PM_{2.5}$ and BC emissions were detailed for the nearby Araucária area. For the rest of the state of Paraná, $PM_{2.5}$ and BC emissions were estimated as fractions of the IAP PM_{10} emissions for industrial and mobile sources (see Section 2.3 for further details)

Discussion

Pollution concentrations in Curitiba in general

The information from the official monitoring network in Curitiba for 2013–2015 (Table 4) revealed mean annual PM_{10} concentrations between 15 and 30 μg m^{-3}, which are within the range of values presented in the WHO global assessment for high-income countries and for low- to middle-income countries in the Americas (see Fig. 3 in WHO 2016). Comparatively, cities of the size of Curitiba in the eastern Mediterranean and southeast Asia have much higher annual mean concentrations, ranging from 100 to 200 μg m^{-3} (WHO 2016).

The monitoring campaign was only a month long and planned to be representative of wintertime conditions in the Southern Hemisphere, when the highest pollution levels are usually observed. However, the meteorological conditions, and likely also a smaller contribution from LRT than usual, caused the PM_{10} and NO_2 concentrations to be considerably lower during the 2016 field campaign, as compared to average concentrations in previous years. This indicates that the monitored $PM_{2.5}$ and BC concentrations reported for the 2016 winter month campaign may also be lower than normally observed for the months of July and August. The urban background $PM_{2.5}$ mean concentrations (± standard deviations of the daily means) registered at the roof top in the city center (7.3 ± 4.3 μg m^{-3}) agreed reasonably well with the reported from a year-long measurement in 2014–2015 at the Federal University campus (10.3 ± 6.3 μg m^{-3}) (Polezer et al. 2018), even when considering the fact that the latter was located approximately 100 m from an interstate highway. On the other hand, measurements conducted in 2007 and 2008 at the same campus location showed a slightly higher mean $PM_{2.5}$ concentration of 14.4 μg m^{-3} (Miranda et al. 2012; Andrade et al. 2012) possibly reflecting higher vehicular emissions at that time.

$PM_{2.5}$ concentrations in the residential area (SC site)

The mean $PM_{2.5}$ concentration at the residential SC site was 25.3 μg m^{-3}, substantially higher than the concentrations at MD roof (7.3 μg m^{-3}). On the other hand, the model output (Fig. 10, left) does not show any $PM_{2.5}$ concentrations in the range of 20–30 μg m^{-3} around SC. This suggests the existence of additional local $PM_{2.5}$ sources not included in the emission inventory. The mean OC/EC ratio of 4.4 (range 3.3–9.0) found in $PM_{2.5}$ sampled at SC indicates that the dominant local emissions in this area were unlikely to originate from

Table 6 Comparisons between simulated and measured air pollutants from local traffic inside MD street canyon. Note that the measurements are reported here as street increments (given as the difference between street and roof level concentrations) to be comparable with the model output, which only gives the impact of the road traffic inside the street canyon

Period		Measurements (μg m^{-3})	Model (μg m^{-3})		
			Buses	Private vehicles	Total
July 29–August 12, 2016*	NO_x	62	43	32	75
July 15–August 29, 2016*	NO_x	66	40	29	69
July 25–August 24, 2016	$PM_{2.5}$	9.3*	0.9	0.7	1.6
July 25–August 24, 2016	BC	3.2	0.6	0.5	1.1

* $PM_{2.5}$ increment estimated by using a $PM_{2.5}$-to-BC ratio of 3 at street level, based on mobile measurements (see previous comment of $PM_{2.5}$ measurements at MD street site, "$PM_{2.5}$, PM_{10}, BC, and NO_x concentrations measured during the campaign" section)

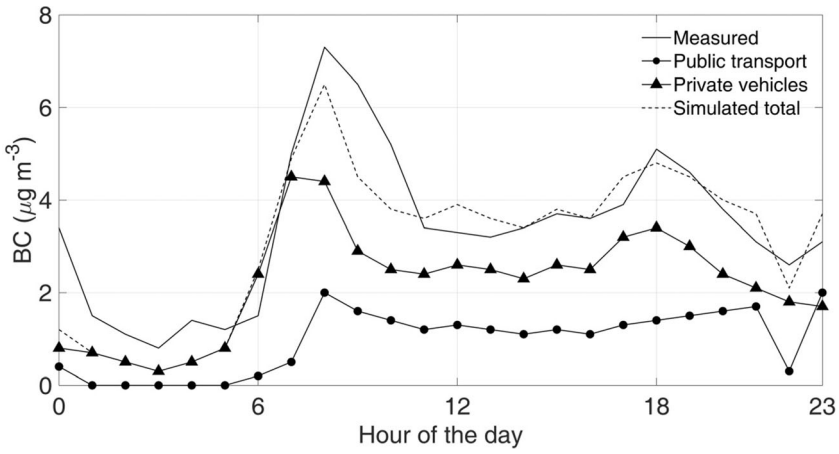

Fig. 9 Daily mean variation of measured and simulated BC contributions from local traffic at MD street site. The simulated impact was corrected with a factor of 1.2 for public transport and a factor of 5.0 for private vehicles

diesel traffic emissions along the nearby ring road, since the EC fraction in environments dominated by diesel emissions is larger than the OC (Harrison and Yin 2008). For example, in a Brazilian study, Santos et al. (2016) reported OC/EC ratios at four urban sites in São Paulo ranging between 0.56 and 1.89. The smallest OC/EC ratio was found at a street canyon site and attributed to the large contribution from diesel-fueled vehicles. Thus, the high OC/EC ratio found at the Curitiba residential station makes it unlikely that traffic emissions and the closeness to the ring road were the cause of the high $PM_{2.5}$ concentrations. This conclusion is also supported by Fig. 7 that shows the larger variability and a number of high $PM_{2.5}$ peaks not visible at the PM_{10} monitoring sites PAR and BOQ. Although a large part of the temporal variability seen in Figs. 7 and 8 can be explained by meteorological conditions that, at this scale, affect all pollutants in a similar way, we hypothesize that the intermittent high $PM_{2.5}$ peaks at SC were due to wood and/or waste burning around this site, which also contributed to an elevated OC/EC ratio. Curitiba Municipality lacks information on such residential wood furnaces, considering them to be of minor use and without need for control. However, the staff operating the monitoring site occasionally reported the smell of wood smoke and noted the presence of several houses with small chimneys in the neighborhood.

Wood burning has been identified as a major source of local $PM_{2.5}$ in the Nordic countries, most notably in the Norwegian capital Oslo (Kukkonen et al. 2020), and dominating over the emissions from on-road traffic. Studies on biomass burning in Brazilian cities are scarce, but Krecl et al. (2020b) reported that the burning of waste in residential areas may create local $PM_{2.5}$ hotspots with concentrations higher than those found in the city center and along busy roads.

BC concentrations in Curitiba

The simulated mean BC concentrations in the urban background (i.e., excluding street canyons and the immediate vicinity of the major roads) were fairly homogenous over the city. In addition, the few existing measurement data points indicated a spatial homogeneity, with mean values just above 2 μg m^{-3} at the sites MD roof and SC reported in this paper. Similar concentrations were reported at the UFPR university campus for 2014–2015 (Polezer et al. 2018), revealing that the BC is a significant contribution of $PM_{2.5}$ associated to the vehicular emission. Within street canyons and close to highways where diesel traffic dominates, the BC concentrations are usually high (Johansson et al. 2017; Krecl et al. 2016; Andrade et al. 2012).

Table 7 Measured street increments and simulations of local traffic impact inside the canyon where the monitor station MD (street level) was located, after correcting for the emissions factors

Period		Measurements	Model		
		(μg m^{-3})	(μg m^{-3})		
			Buses	Private	Total
July 25–August 24, 2016	$PM_{2.5}$	9.3			8.0*
July 25–August 24, 2016	BC	3.2	0.8**	2.2**	3.0**

* Total simulated $PM_{2.5}$ output of Table 6 corrected with a factor of 5

** Bus impact of BC corrected with a factor of 1.2 and private traffic impact of BC corrected by a factor of 5, with values given by a regression analysis of hourly data

Table 8 Simulated contributions of LRT from sources outside the Curitiba Municipality (regional model output) and local sources inside Curitiba (urban model output) to ambient concentrations at the MD rooftop and SC sites. Values within parentheses indicate concentrations after applying a correction factor of 5 on traffic emissions taking place outside Curitiba (see the "Spatial distribution of $PM_{2.5}$ and BC over Curitiba (comparison between simulated and measured concentrations)" section for more details). Period: July 25–August 24, 2016. Unit: μg m^{-3}

Contribution	Model	MD roof ($PM_{2.5}$)	SC ($PM_{2.5}$)	MD roof (BC)	SC (BC)
Industrial	regional	1.4	2.3	0.09	0.15
Mobile sources	regional	0.3 (1.4)	0.3 (1.4)	0.18 (0.90)	0.17 (0.85)
Biomass burning	regional	0.2	0.2	0.03	0.03
Industry inside Curitiba	urban	0.5	0.4	0.03	0.03
Public transport	urban	2.3*	1.8*	0.13	0.07
Private vehicles	urban			1.07	0.98
Summed model		4.6 (5.8)	5.1 (6.1)	1.53 (2.25)	1.43 (2.11)
Monitored		7.3	25.3	2.29	2.22
Not explained		2.6 (1.5)	20.2 (19.2)	0.76 (0.04)	0.79 (0.11)

* The contribution could only be simulated as the sum of public and private traffic since the same correction factor was applied

Even with spatially homogenous mean BC concentrations in the urban background, we observed large temporal variations mainly due to changes in meteorological conditions (wind and boundary layer stability). The impact of the boundary layer stability on the air quality can be deduced from the highly similar variations in both BC and NO_x concentrations (Fig. 8). Clearly, there is a large BC contribution from traffic, which is intense along the ring road during all days of the week. However, it is likely that there can be intermittent BC contributions also from industrial plumes and potentially from the wood and/or waste burning, as indicated to take place in peripheric residential areas. Note that the latter source may produce a relatively stronger impact on $PM_{2.5}$, as compared to BC.

The mean BC concentration observed at MD street level (5.5 μg m^{-3}) was similar to values reported for a street canyon in Stockholm on weekdays in 2006 (5.1 μg m^{-3}). A later assessment for the same site in Stockholm in 2013 showed reduced levels (2.2 μg m^{-3}), which has been explained by technologically improved vehicles circulating on this street (Krecl et al. 2017). This gives an indication of what is possible to achieve with progressively stricter limits on vehicle emission standards.

Long-range contribution of air pollutants as assessed by the regional model

The regional model was used to estimate the contributions of LRT to PM_{10}, $PM_{2.5}$, and BC concentrations in the city.

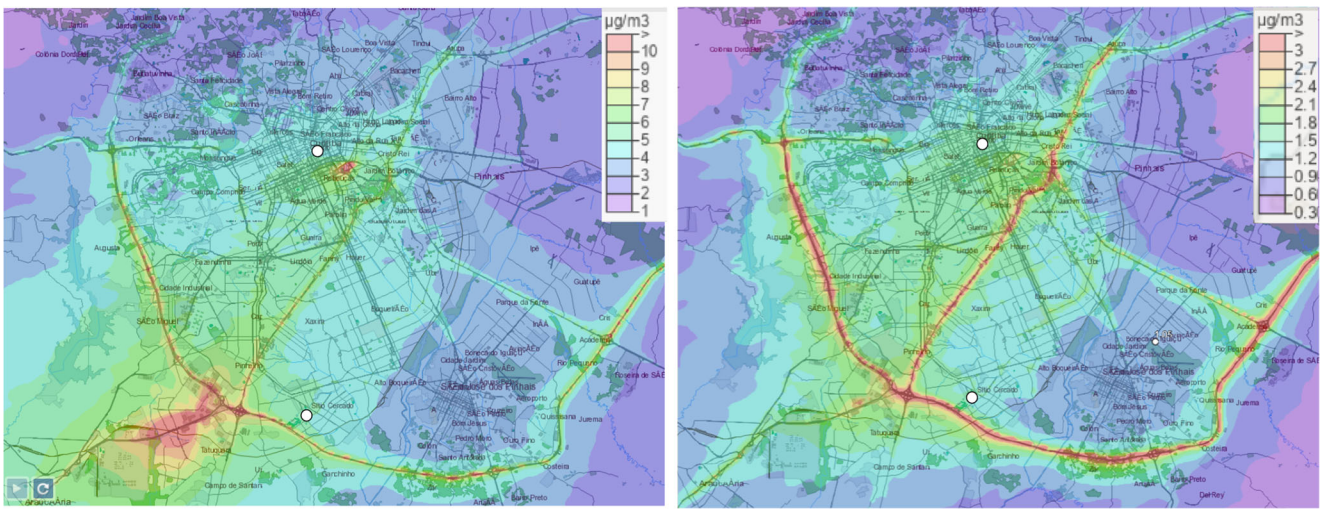

Fig. 10 Modeled mean $PM_{2.5}$ (left) and BC (right) concentrations in the period July 25–August 24, 2016. Regional (anthropogenic and biomass) and local (industry inside Curitiba, public transport and private vehicle impact) contributions were included. The city center (MD) and the residential (SC) sites are marked as white circles

Table 5 shows that PM_{10} and $PM_{2.5}$ contributions were dominated by industrial and mobile source emissions, with smaller impact of biomass burning. Since the regional model did not include secondary aerosol formation, it is likely that the results, based solely on primary emissions, give a somewhat underestimated magnitude of the long-range transported aerosol mass. An upper bound of the magnitude of the lacking secondary aerosol can be taken from the difference between the mean simulated (4.6 or 5.8 μg m^{-3}) and monitored (7.3 μg m^{-3}) $PM_{2.5}$ concentrations at the central MD roof station, yielding 2.6 or 1.5 μg m^{-3} (Table 8, further discussed in the "Spatial distribution of $PM_{2.5}$ and BC over Curitiba (comparison between simulated and measured concentrations)" section). The difference of almost 20 μg m^{-3} registered at the residential site SC can obviously not be explained by the lacking LRT contribution of secondary aerosols, but instead there should be a large unaccounted local source (as discussed in the "Spatial distribution of $PM_{2.5}$ and BC over Curitiba (comparison between simulated and measured concentrations)" section).

The BC arriving at Curitiba Municipality due to LRT consists of contributions from industrial and mobile sources in the State of Paraná. BC contributions from the surrounding states and other South American countries simulated over the large domain (G1, Fig. 3, left) include biomass burning, but no anthropogenic sources. However, plumes from mobile sources in states situated far away should likely have little impact in comparison to traffic and industries inside Paraná.

Contribution from local traffic to BC concentrations

An important aim with the ParCur assessment in Curitiba was to determine the emissions of $PM_{2.5}$ and BC from sources inside the city. To this end, the street canyon measurements were used for an in situ evaluation of the emission characteristics of Curitiba's vehicular fleet. As shown previously, the OSPM model output agreed well with the NO_x observations when using EFs taken from the literature, but the BC increment within the street canyon was largely underestimated (Table 6). The OSPM has been used and evaluated in many urban environments and an extensive comparison showed good results for NO_x (Ketzel et al. 2012). Felix et al. (2019) used the same NO_x emission factors as input to the Gaussian model (the same as used in the present study, see the "Emission inventory" section), yielding urban background NO_x concentrations that compared well with those measured by 10 passive samplers and at three IAP monitoring stations located across the city, for the same period of our campaign. Hence, considering that both the OSPM and the Gaussian model give robust results, as shown by the good agreement between simulated and monitored NO_x concentrations, it is reasonable to consider that the NO_x emission factors are fairly accurate. Hence, we suggest that the model output could be improved by adjusting the $PM_{2.5}$ and BC vehicle exhaust EFs according to the results of the "Simulations of local traffic impact inside the street canyon" section.

An interesting fact is that the regression analysis confirmed the BC emission factor for buses, whose fleet composition, age and equivalent Euro class we know in detail, while a large underestimation (a factor of five) was found for the mixed private vehicle fleet emissions, for which the composition and technical status was much less known. A difference by a factor of five may be seen as a too large discrepancy. However, BC emission factors reported in the literature show a large range, but both the original and the corrected EFs (4 and 19 mg veh^{-1} km^{-1}, respectively) for the mixed private vehicle fleet circulating through the MD street canyon fit into the span of real-world EFs. For example, through chasing individual vehicles, BC emission factors ranged from 10 to 32 mg veh^{-1} km^{-1} for gasoline cars (Ježek et al. 2015), while for diesel-fueled HDV they went from 53 mg veh^{-1} km^{-1} (Park et al. 2011) to values tenfold higher (Wang et al. 2012). Real-world BC emission factor estimated within a street canyon in Stockholm (Krecl et al. 2017) for the year 2006—when BC concentrations within the street canyon were similar to those reported for M. Deodoro in Curitiba—was 11 mg veh^{-1} km^{-1} for gasoline cars, which is substantially higher than the 0.15 mg veh^{-1} km^{-1} used originally in the present study (Table 3), justifying the higher EFs coming out of the regression. A lesson learnt for other Brazilian cities is thus the necessity to either assess local BC and $PM_{2.5}$ emission factors for the private vehicle fleet, using similar experiments as in Curitiba, or to search for comparable real-world EFs in the literature.

Contribution from local traffic to $PM_{2.5}$ concentrations

Because $PM_{2.5}$ at the MD station was only measured as daily averages, it was only possible to compare the simulated total impact of all vehicles in the street canyon with the measured increment, i.e., without separation between private traffic and public transport vehicles. Also, the simulated $PM_{2.5}$ contribution of the local traffic was largely underestimated when compared to the measured increment. For $PM_{2.5}$, contributions from non-exhaust particles generated by the vehicles are also expected, namely those consisting of wear particles from brakes and tires, together with resuspension of dust from the pavement itself (none of these were included in the EFs of Tables 2 and 3). However, since the aim was to estimate the total $PM_{2.5}$ emissions, no separation was made between exhaust and non-exhaust $PM_{2.5}$ originating from traffic in Curitiba. Based on the comparison between the street canyon model output and the measured increment ("Simulations of local traffic impact inside the street canyon" section), a general correction with a factor of five was applied to EFs of both

buses (Table 2) and the mixed fleet of private vehicles (Table 3).

Spatial distribution of $PM_{2.5}$ and BC over Curitiba (comparison between simulated and measured concentrations)

One of the goals of the ParCur project was to map the annual concentrations of $PM_{2.5}$ and BC over the city, identifying the contribution of local sources and the magnitude of LRT. Table 8 summarizes the different contributions in the city center (MD roof) and in the residential area SC. The concentrations outside parentheses reflect the impacts given by the regional model (Table 5) and the local Gaussian model after adjusting the EFs, as discussed in the "Contribution from local traffic to BC concentrations" and "Contribution from local traffic to $PM_{2.5}$ concentrations" sections. An alternative model simulation of the impact of the LRT contribution from anthropogenic sources outside Curitiba is given within parentheses along with the modified total concentration. These alternative results illustrate the regional impact by correcting the contributions from mobile sources in the State of Paraná in a similar way as for the private vehicles inside Curitiba (an EF five times higher for both $PM_{2.5}$ and BC). Considering that the mobile emissions for the whole State reported in the IAP inventory were based on EFs recommended by an environmental state agency in Brazil, one could think that they were underestimated, as were the original mobile emissions assessed inside Curitiba.

For $PM_{2.5}$, the summed model output attained 4.6, alternatively 5.8 μg m^{-3}, compared to 7.3 μg m^{-3} measured in the city center. The missing 36 or 21% contribution could possibly be attributed to sources not included in the emission inventory (e.g., restaurants and residential sources), and also to underestimated particle mass in the regional model output which did not include secondary aerosol formation (as already suggested in the "Long-range contribution of air pollutants as assessed by the regional model" section). At SC site, the simulated $PM_{2.5}$ concentrations only accounted for 20-24% of the measured concentrations, indicating a strong local source not being included. As discussed earlier, there are evidences of wood combustion taking place in this residential area on the city's outskirts, but not in the city center.

For BC, the simulated concentrations constituted 67%, alternatively 98%, of the measured concentrations in the city center and 65%, alternatively 95%, at the residential location. It is interesting that for BC the adjusted and increased long-range transported vehicle exhausts from other parts of the State of Paraná (the alternative within parenthesis in Table 8) practically takes the simulated concentrations at both locations to the monitored levels. With this adjustment of the regional contribution, it accounts for about half of the BC in the Curitiba urban background, the remaining half being emitted by the traffic inside Curitiba and, to a very minor degree, from industries within the city.

The higher $PM_{2.5}$ and BC concentrations in the model output (Fig. 10) along the ring road "Contorno" and around the industrial area southwest of Curitiba, could not be verified by local monitoring. The model also included various methodological simplifications, such as the vehicle fleet composition, which makes the concentrations for this area more uncertain. In connection to this problem, great uncertainty subsists in knowing the actual characteristics of the gross polluters, notably old trucks (Fig. 11) in transit through Curitiba's ring road. Many of them are not registered in the State of Paraná and, consequently, not taken into account in our inventory.

Lessons learnt from the Curitiba case study and its applicability to other Brazilian cities

This study highlights the need to develop emission inventories for $PM_{2.5}$ and BC at local, regional, and national levels that are currently missing in Brazil. Other local activities, such as small combustion from backyard burning and wood burning appliances, and emissions from pizzerias, bakeries, and steakhouses should be also considered in the inventories.

Data on airborne fine particle emissions and concentrations in cities are valuable for traffic and air quality managers, urban planners and landscape architects, health practitioners, researchers, and ultimately for legislators and decision makers. Most Brazilian cities lack this kind of information, which hinders an accurate assessment of their ambient concentrations, the source apportionment and the potential health outcomes. Air quality monitoring networks with sufficient spatial coverage are unlikely to be available in Brazil in the near future. Hence, methods, instruments and models to identify air pollution hotspots and their sources are an urgent need to safeguard the population exposure to toxic airborne species. This paper, as a result of the Brazil-Sweden cooperation materialized in the ParCur project, addressed this gap taking Curitiba as case study. The integrated approach used here can be implemented in other Brazilian cities, as long as an open data policy and a close cooperation between municipal authorities and academia can be achieved.

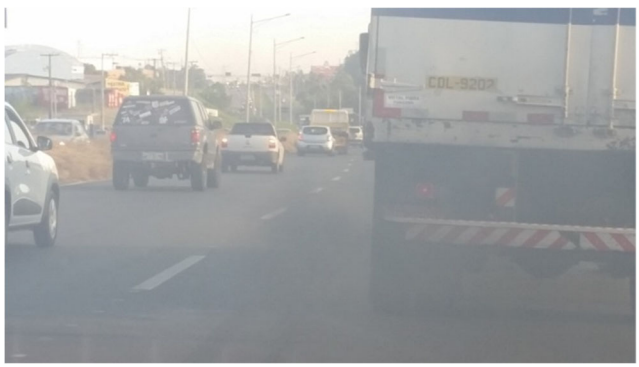

Fig. 11 Photo of a truck circulating in Curitiba's ring road, illustrating the intense exhaust emissions.

Conclusions

Through the use of an integrated approach, combining monitored data, emission inventories and dispersion model outcomes, the emissions and impacts of $PM_{2.5}$ and BC on the air quality in Curitiba have been determined. The main results from this case study are summarized as:

- Vehicular EFs from the literature were found not to be suitable for Curitiba's fleet. Local real-world EFs for $PM_{2.5}$ and BC were determined for on-road vehicles.
- The mean $PM_{2.5}$ concentration in central Curitiba was 7–8 μg m^{-3} in the urban background, doubling inside street canyons. $PM_{2.5}$ concentrations in a residential area were three- to fourfold higher than in the center, probably due to local biomass or waste combustion.
- Local traffic emissions contributed to about half of the urban background BC concentrations of around 2 μg m^{-3} in Curitiba.
- The mean BC concentrations seemed to be fairly homogenously distributed over the city, except for areas close to busy roads and inside street canyons where they reached 3–5 μg m^{-3}.

Limitations

Some important limitations of this $PM_{2.5}$ and BC assessment in Curitiba have been identified:

- The measurement campaign was limited in time (one month) and space (two fixed stations and mobile measurements along four routes covering an area of 4.8 km^2): An attempt was made to relate the one month-long campaign data to the wintertime conditions observed during the three preceding years, this in order to assess the representability of our collected $PM_{2.5}$ and BC data. The two fixed sites were well selected; however, there is a need for future monitoring campaigns at additional locations (e.g., in the more polluted areas along the ring road and toward the industrial zones just southwest of the Curitiba Municipality).
- Technical failures during the campaign contributed to increased uncertainties in the $PM_{2.5}$ assessment in the street canyon. Moreover, a future campaign could give more reliable information on EFs for the Curitiba's vehicle fleet.
- While the model input information for simulating the impact of public transport was very detailed and of high quality, there was a lack of information on fleet composition for the private vehicles circulating the road network.
- Lack of information on emissions from some local sources of potential importance (e.g., restaurants, residential wood combustion).

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Data availability The datasets generated during and/or analyzed during the current study are available from the corresponding author on reasonable request.

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