Air quality and fossil fuel driven transportation in the Metropolitan Area of São Paulo

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A B S T R A C T

In this paper, we use origin and destination mobility surveys and high-resolution assignment and emission models to study air quality and short-lived climate pollutant impacts related to on-road transportation in the Metropolitan Area of São Paulo (MASP). To begin with, we calculate transport carbon dioxide (CO2) emissions from fossil fuel-driven vehicles (light and heavy-duty) at spatial and temporal resolutions of 500 m and one hour, by means of traffic demand forecasting. These estimates, carry out for 2007 and 2012, are based on passenger and freight trips and the height of the atmospheric boundary layer, among other variables. These proxies depend also on ancillary parameters as particulate matter concentrations and dilution rates. In the second place, we evaluate the changes in CO2 emissions from the MASP (3%/year). Transport emission inventories combine mobility surveys and road network assignments with air quality data. In spite of using different methodologies, bottom-up road link assignments versus top-down vehicle activity-based and fuel consumption approaches, the estimated CO2 emissions are consistent with the Official São Paulo State’s Inventory. This work found that the CO2 emissions in MASP were 10,044 and 11,503 t Ceq./day in 2007 and 2012 (73% light and 27% heavy vehicles), respectively. On-road emission patterns agree with the spatiotemporal variation of transportation journeys and corresponding passenger and freight network assignments. Temporal patterns, diurnal, weekly and monthly, were determined using traffic counts and congestion surrogates. The patterns were also crosschecked with average CO2 measurements, available for 2014 at the road (western area of MASP) and background sites (Jaraguá Peak). Kerbside road measurements showed two prominent peaks associated to the morning (437 ± 45 ppm) and night rush hours (435 ± 49 ppm), coupled to low values of boundary layer height (313 m) and dilution rate (329 m2/h). Background values (414 ± 2 ppm) were subtracted from on-road measurements to estimate excess CO2 (12 ± 8 ppm) directly attributed to vehicles. The inventory reflects the relationships between traffic patterns and emissions, and the developed methodology could be used to evaluate the impacts of forthcoming urban transport and emission control policies. In the future, our estimates will be verified with ground measurements of CO2 concentrations over a bigger monitoring network in the MASP.

1. Introduction

São Paulo State’s compromises to reduce carbon dioxide (CO2) emissions from vehicles will have to be coupled with accurate emission inventories, especially in urban areas such as the Metropolitan Area of São Paulo (MASP). These inventories will be defined at temporal and spatial scales adequate to the emission reduction rates fixed by the greenhouse gas (GHG) international agreements (McKain et al., 2012). Therefore, these inventories will monitor different time periods to report trends and test the results against measured observations following the recommendations suggested by the State Act of Climate Change and the International Panel of Climate Change (IPCC, 2014). Future CO2 surface measurements will help to calibrate the inventory estimates in accordance to the reduction constraints, updating the degree of the objectives fulfilled (Newman et al., 2013; Turnbull et al., 2015).

On-road transportation is the main origin of CO2 emissions in the metropolitan regions and several strategies are defined to decrease vehicular emissions (Huo et al., 2009; Uherek et al., 2010). The report of the IPCC (2014) spotlights the convenience of using combined observations with modeling to accurately estimate vehicular emissions from the metropolitan regions with strong transportation activities and related increments of CO2 atmospheric concentrations. The MASP is one of the largest megacities in the world and the most important conurbation in the southern hemisphere including >20 million inhabitants and >8 million vehicles. The region has

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an important air quality network mainly used to identify environmental problems related to anthropogenic sources as vehicle emissions, especially during the winter season when the concentrations of pollutants are higher (Miranda et al., 2019). Motorization rates have increased drastically and the region is responsible for half of the state’s CO2 emissions (CETESB, 2014a). Transport CO2 emissions are a major contributor to climate change and local strategies are designed to constrain transport demand and emission rates (Pérez-Martínez et al., 2015). These strategies aim to develop methodologies capable to reduce uncertainties on CO2 estimates at a resolution consistent with the transport policy and measures fixed by local governments and international compromises (Gurney et al., 2009).

There are several studies in the international literature, which have researched the relationships between the emission patterns and atmospheric dynamics and performed high-resolution GHG emission fluxes at urban environments using different methodologies (Huo et al., 2009; McKain et al., 2012). Consequently, urban regional CO2 emissions have been estimated with mass-balance downscaling methodologies using spatial distributed top-down parameters and atmospheric surface and space observations (Mays et al., 2009; Zimnoch et al., 2010; Brondfield et al., 2012; Jacobson et al., 2015). Other works employed fuel and vehicle counts to estimate emission products following an inventory-based and a bottom-up approach (Pérez-Martínez, 2012; Dallmann et al., 2013; McDonald et al., 2014). Combining both approaches, other studies used CO2 fluxes and carbon monoxide (CO) to CO2 emission ratios to define vehicular contributions (Newman et al., 2013; Turnbull et al., 2015; Kort et al., 2014).

However, most of them fail to accurately estimate vehicle emission trends at high urban scales to monitor GHG reduction strategies and seeking the fulfilment of sovereign commitments. In this context, Djuricin et al. (2010, 2012) reported trace CO2 emission trends in Californian urban environments for three decades (1980–2008), using CO2 mixing ratios from fossil fuel consumption based on radiocarbon (14C) contents of tree rings. Another study presented by Levin et al. (2011) used long-term CO2 concentrations combined with 14CO2 measurements to estimate trends since 1996 at a regional scale in Heidelberg (Germany). Long-term regional studies are unusual in the literature due to monitoring constraints (Turnbull et al., 2015; Locosselli et al., 2018).

High-resolution emission inventories and models are missing especially in Brazil as in other megacities of the southern hemisphere (Duren and Miller, 2012). High-resolution transportation models have been used in previous works to estimate on-road CO2 emissions (Brondfield et al., 2012; McDonald et al., 2014). These bottom-up inventories combine transport activity, meteorology and emission factor data. These inventories are well supported by travel demand models, mobility surveys and laboratory vehicle emission measurements. However, these studies are scarce in Brazil due to the lack of transport activity and emission data (Alonso et al., 2010). This study uses vehicle travel demand modeling combined with station measures across an air quality network to extrapolate and estimate CO2 across the MASp. The fossil fuel-driven CO2 estimates represent real-world traffic operating conditions for an average weekday over the year.

2. Methods and experimental setting

Fig. 1 shows transport area zones (TAZs), road network and air quality network systems in the Metro São Paulo from the “Centro de Estudos da Metrópole” (CEM, 2010) and “Centro Tecnológico de Saneamento Básico do Estado de São Paulo” (CETESB, 2014b), respectively. In the figure, the road network represents urban freeways (1550 km), arterials (9713 km) and residential roads (29,328 km). The urban freeways include the 212 km urban ring roads, named “marginals” (86 km central lines and 126 km express lines), and 1281 km of radial roads (named highways) intersecting the marginal in several directions and connecting the city centre to suburban areas. MASp is divided into two major zones: São Paulo and the rest of the residential municipalities. The region inside the Pinheiros and Tietê marginals corresponds to the commercial expanded city centre and has traffic restrictions during peak hours (Jacobi et al., 1999). The region outside the urban marginals corresponds to the São Paulo and metropolitan residential municipalities which have been increasing vehicle activity. Finally, the metropolitan residential region outside the outer ring road of the Metro São Paulo, named “Rodoanel”, represents the new areas of the city resulting of recent urban sprawl. The road total length is 40,590 km (Table 1) in a total surface of 7966 km² (5.1 km/km²). In the figure, black and pink polygons represent the limits of the 460 transport area zones in the Metro São Paulo. TAZs are homogeneous common units used in standard transportation planning models constructed by census block data according to socio-demographical parameters. In the MASp, the spatial extent varies between few city blocks in the central business district (CBD) to very large areas in the periphery. This study analyses the transport demand on internal combustion engine light and heavy-duty vehicles (LDVs/HDVs) and CO2 emissions in the areas of Metro São Paulo, especially in the city centre and urban freeways where most of the transport activities take place (Fig. 1).

In the MASp, new CO2 monitoring systems (surface and flux measurements) are being purchased recently to develop and assess techniques for estimating GHG emissions at the megacity level (Duren and Miller, 2012). The methodology described in this paper starts using air quality measurements registered by the State’s Company CETESB (2014b) and LAPAT (“Laboratório de Análise dos Processos Atmosféricos”) joint with atmospheric Planetary Boundary Layer (PBL), meteorological data and transportation models (Ribeiro et al., 2016). This methodology can be extrapolated in the future to other Brazilian cities and compared to historical transport energy consumption figures and emission inventories. This work defines an innovative bottom-up methodology in the region based on modeling of transport activities as well as on atmospheric measurements to estimate CO2 temporal and spatial trends. Therefore, we present a high scale emission model, at a megacity level, where measured atmospheric data and the height of the boundary layer are linked to the transportation demand model. Metropolitan transportation flows are estimated using a stochastic link-based procedure. Transportation activity data consist of repetitive surveys that describe the urban mobility related to atmospheric observations, which represent spatial and temporal pollutant concentration enhancements, and GHG emission inventory that reproduces the temporal and spatial distributions of vehicle flows. We test the methodology to the Metro São Paulo at the resolution of 500 m and 1 h during the period 2007–2012, using available country’s unique data of air quality and mobility surveys. We perform a sensitivity analysis of the methodology’s parameters for estimating trends in vehicle emissions from this metropolitan region and statistically assess the contribution of the parameters on GHG estimates. The emission estimates, using the Metro São Paulo as a case study, permit to evaluate transport-related policy measures and can aid air quality and chemistry forecasting models. Finally, we give some recommendations to improve the methodology through future GHG observations and model definitions.

Fig. 2 provides a descriptive basic flow chart of the different parts integrating the urban transportation and CO2 emission model. The diagram includes the different steps for inputting the model: urban transport and emission inputs and meteorological and chemical parameters. The model starts characterizing the transport activity of the on-road transportation in the region by means of transport demand modeling modulated by mobile source parameters such as vehicle type, fuel consumption, traffic flows and road network characteristics (capacity, travel times and level of service). The estimated transport activity can be compared and cross-check with available emission inventories based on regional vehicle fleet and fossil fuel historical sales. The transport activity is the basis to extrapolate and estimate high-resolution air quality and CO2 emissions across the Metro São Paulo. In the emission model it is important the appropriate determination of vehicle-based emission factors. To this aim, we are using travel demand modeling combined with collected air quality measurements across a network of 28 stations. Air quality measurements reflect the atmospheric chemistry and physics of the region and the emissions depend on meteorological and chemical parameters as the height of the planetary boundary layer and the pollutant dilution rates.
2.1. CO2 sources from the metro São Paulo

The São Paulo State Air Quality Management Program has focused on urban source assignments and was initially launched to control the pollutant emissions of road transport (CETESB, 2014a). The Program estimated vehicular emission factors based on dynamometer measurements of new vehicles and emission deterioration factors, cross-checked with energy consumption figures (Pérez-Martínez et al., 2015). Complementing these top-down dynamometer studies and emission inventories, Sánchez-Ccoyllo et al. (2009), Martins et al. (2006) and Pérez-Martínez et al. (2014) used CO2 measurements, together with carbon monoxide (CO) as a tracer of atmospheric concentrations, to determine vehicular pollutant emission factors in urban tunnels. Salvo and Geiger (2014) and Miranda et al. (2018) established a good relationship between air quality measurements and traffic emissions collected from upwind kerbside sites in the MASP. The importance of meteorology and anthropogenic sources on observed atmospheric concentrations was demonstrated in the ambient studies of the Metro São Paulo (Silva et al., 2012; Andrade et al., 2017).

We developed a stochastic model to assign vehicle kilometers travelled (VKT) in the MASP’s network based on the mobility travel survey data from the São Paulo Company METRÔ-SP (2012) and using the transportation-engineering model TRANSCAD (2014). Fig. 1 shows the limits of 460 transport area zones (TAZs represented by black and pink lines), urban freeways (represented by bold thick lines) and arterials (represented by lighter fine lines) in the MASP. In the figure, blue crosses and squares represent the locations of the 28 stations of the São Paulo State Environmental Company CETESB within the São Paulo and the Metropolitan Municipalities. The red star represents the location of the Jaraguá peak (background station). The assigned VKT, broken down by different transport modes (cars, buses, vans and trucks), were used to estimate the CO2 emissions. The VKT will be combined with air quality measurements. Particulate matter 10 μm or less in diameter (PM10), carbon monoxide (CO), nitrogen oxide (NOx)
and ozone (O₃) measurements were obtained from the network of observation sites operated by CETESB at several locations since 2000 (CETESB, 2014a). These measurements were complemented recently with CO₂ mixing ratios measured at the supersite located at the Institute of Astronomy Geophysics and Atmospheric Sciences-IAG (inside the University of São Paulo’s Campus, western side of the Metro São Paulo). The IAG’s supersite has a secondary spot outside the Metro São Paulo influenced by similar atmospheric conditions and air masses. This secondary site represents a mountain-peak top (Pico de Jaraguá, 312 m above the mean level of the Metro São Paulo) and is mainly used as a background to estimate CO₂ anthropogenic exceedances (CO₂xs).

CETESB (2014a) has developed CO₂ emission inventories of anthropogenic sources and fuel combustion from road transportation. CETESB provides annual CO₂ emissions for the whole Metro São Paulo, top-down vehicular-based estimates, to be used as a reference to the high-scale modeling of this study. Similarly to the CETESB’s inventory, the EDGAR database could be also integrated into the model framework as an external reference. The EDGAR database has temporal and spatial resolutions of 1 month and 10 km, respectively, and was combined with annual on-road CO₂ emission values to perform the comparison (v4.3, http://edgar.jrc.ec.europa.eu).

2.2. Transportation model

A stochastic user equilibrium model was used as the traffic assignment model to estimate the traffic flows expressed in VKT on the Metro São Paulo’s network from the “Centro de Estudos da Metrópole” (CEM, 2016). The model uses, as input, the matrix cells of transportation volumes of the Metropolitan Company METRÔ-SP (2012). Split by vehicle mode, the cells represent road trips corresponding to 460 transport area zones of the MASP (Fig. 1). The model was determined with network topological fields such as link characteristics and performance functions from the CEM’s network (“logadouros da região metropolitana de São Paulo”, CEM, 2016). The volumes between each origin and destination pairs were loaded into the network based on the travel impedances and times of the different paths that can carry the transport flows (De la Barra, 2005; Ortízár and Willumsen, 2011). The assignment procedure for modeling the regional urban traffic followed an interactive algorithm to achieve a convergent solution at the equilibrium point. This algorithm uses the method of successive averages and the passengers cannot ameliorate their travel impedances and times by shifting routes (Sheffi, 1985). The travel time for each origin and destination pair on all used links is less or equal to the travel time on any alternative path at the equilibrium point (Aashtiani, 2004). The stochastic model assumes that passengers do not know the network attributes perfectly and also have divergent perceiving costs (Sheffi and Powell, 1982; Chen et al., 2011).

The model computes network link traffic flows by different vehicle types (light and heavy-duty) that incorporate link constraints (road capacity) and travel times (impedances). Traffic flows were extrapolated at a 500 m gridded resolution using a searching radius of 5 m. Traffic flows represent hourly values of normalized VKT for a mean working day and retain the signs of the mobility survey (times and weekdays). Flows were scaled to annual values to allow comparison with CETESB’s and EDGAR’s estimates. The number of VKT can be considered as the product of vehicle flows (determined by the transportation model) on-road segment at hour t for vehicle type k (N(t,k)j) and length of road segment (l):

\[ N(t,k,j) = \eta_{j,t,k} l_j \] (1)

We assume that the vehicle flows remain constant during the hour t at working days. The traffic flow estimates along with some road corridors were compared with data from earlier campaigns of the “Companhia de Engenharia de Tráfego” to adjust the transport demand model’s parameters (CET, 2013). CET provides annually with hourly average flows derived from the results of the driving campaigns at certain significant urban routes. In this manuscript, we considered the segments of freeways (2964 links and 1550 km), arterials (58,131 links and 9713 km) and residential roads (320,087 links and 29,328 km), as line sources derived from the road network of CEM’s geographical information system (Fig. 1 and Table 1). The MASP is classified into 460 TAZs, according to the locations and mobility features of the origin and destination survey (METRÔ-SP, 2012). Finally, traffic flows are aggregated to the TAZs according to the total lengths Lij of freeway, arterial and residential roads of each TAZ i.

2.3. Vehicle emission model

Using the concentration measurements, the vehicle flows (stochastic user equilibrium modeling) and the planetary boundary layer height data, CO₂ emissions can be estimated on an hourly basis for a mean working day for each transport area zone (TAZ) in the Metro São Paulo. Consequently, the vehicle emission factor of CO₂ (EFCO₂) at the TAZ i can be estimated using the following equation:

\[ EFCO₂,i \ [g/km] = \frac{CO₂xs [\text{mg/m}^3] \times S_i [\text{ha}] \times PBL [\text{m}] \times t_i [\text{h}]}{k_i [3\times10^{-11}\times10^3] \times \eta_{total}} \times \left[ \frac{10^6}{\text{VKT}} \right] \] (2)

where CO₂xs are the concentration exceedances for CO₂ considering a background concentration of 380 ppm (695 mg/m³) in the urban environments of the Metro São Paulo (McKain et al., 2012), S_i is the surface of transport area zone TAZ i, PBL is the height of the planetary boundary layer corresponding to TAZ i, t_i is the 3600 s time period equivalent to 1 h and k_i is a dimensional constant equal to 100–300. Comparing modeled to monitored CO₂ emissions is a brand-new technique that is innovative to most transportation and air quality planners. The approach matches modeled and monitored data to link near-road monitoring studies in Brazil. This approach is more difficult to be applied especially with secondary reactive pollutants. Since CO₂ is not a local air pollutant, the approach appears most useful for pinpointing spatial and temporal patterns of emissions and trends in those emissions, rather than looking at air quality and health impacts as might be done with hazardous air pollutants. Finally, \( \eta_{total} \) is the estimated traffic counts of the total vehicles at the sampling links of each 460 TAZs and corresponding to the 1 km segment length (Fig. 1), expressed in vehicle kilometers travelled per hour (VKT/h, \( \eta_{total} = \eta_{LDV} + \eta_{HDV} \)). To split up the shares of light and heavy-duty vehicles (LDVs and HDVs), we can provide the emission factors, \( f_{1,i} (EFCO₂,LDV,i) \) and \( f_{2,i} (EFCO₂,HDV,i) \), fit individually by the linear regression model:

\[ EFCO₂,i = f_{1,i} \eta_{LDV,i} + f_{2,i} \eta_{HDV,i} \] (3)
Emission factors in Eq. (2) are directly proportional to CO₂ atmospheric dilution rates ($D_{CO2}$) and inversely proportional to the fraction of vehicles $F(1/h)$:

$$EFCO2, i \, [g/km] = \frac{k_2 \times 10^{-3} \times CO2_{xs}[ug/m^3] \times D_{CO2}[m^2/h]}{F[1/h]}$$  \hspace{1cm} (4)

where $k_2$ is dimensionless constant equal to $10^{-3}$. $D_{CO2}$ expresses the ratio between emissions and concentrations and are related to the height of the planetary boundary layer (PBL) and the traffic counts ($\eta$) per surface unit ($S$):

$$D[m^2/h] = \frac{PBL[m]}{k_1 \times 3 \times 10^{-8} \times \eta_{total} \times [VKT]/[S/ha]}$$  \hspace{1cm} (6)

During the daytime, the boundary layer is high and partially counteracts higher traffic volumes. Oppositely, during nighttime lower boundary layer drives also to lower dilution rates despite low traffic volumes. CO₂ exceedances (CO₂xs) and traffic-related dilution rates ($D_{CO2}$) work as key parameters on vehicle emissions and are proxies of the vehicle traffic conditions (characterized by congestion, engine load, speed and acceleration rates) and distribution of vehicle types (characterized by age, mileage, fuel used, weight and technology), especially during the morning rush-hours where weather conditions are more stable (atmospheric stratification) and the impacts of increased emissions from traffic are more evident.

Different surveys have been conducted to gather vehicle activity data in the MASP in 2012 (Table 2). Because of the availability in transport activity data, light vehicles in the Metro São Paulo were classified into three types: interurban cars and motorcycles, urban cars and motorcycles (private and public) and urban vans (light-duty freight vehicles). Similarly, heavy vehicles were classified into two types: interurban trucks (heavy-duty freight vehicles) and urban buses (public and private buses). In the MASP they were about 5.8 million private and business cars and motorcycles (gasoline, ethanol and flex-fuel), characterizing 84.4% of the total vehicle fleet. 54 thousand buses (public and private), 174 thousand trucks (light, medium and large diesel fuel trucks) and 850 thousand vans (gasoline, ethanol, flex and diesel fuel), represented 0.8, 2.5 and 12.3% of the total fleet, respectively. The average ages of the cars and motorcycles, buses, trucks and vans were 8.4, 10.0, 11.5 and 7.6 years. The average mileages (and average CO₂ emission factors) of the cars and motorcycles, buses, trucks and vans were 14,035 (165), 39,921 (729), 36,062 (698) and 16,422 km (265 g CO₂/km) at the year 2012 when the survey was conducted by the CETESB (2014a). During the period 2007–2012, the growth in the number of passenger cars and motorcycles (23%), and buses (18%) was lower than the growth in the number of freight trucks (61%) and vans (57%).

2.4. Spatial and temporal distributions

The CETESB's network of air quality measurement sites have been operated at 28 locations in the Metro São Paulo since 2000 (Fig. 1). We used air quality data from 2007 and 2012 to compare with the corresponding transport data from origin and destination surveys. For this manuscript, we extrapolated the observations from the 16 stations at the São Paulo Municipality and from the 12 remaining stations of the Metro São Paulo. Therefore, we obtained boundary layer height data for 2012 from the Weather Research Forecast model coupled with chemistry (WRF-Chem). Fig. 3a shows 24-day averaged boundary layer heights estimated at the 28 locations of the air quality network. We also extrapolated the estimates to the whole Metro São Paulo to represent the mean dispersion conditions and daily profiles during the working days (Fig. 3b). We evaluated boundary layer heights at a resolution of 0.5 km for the Metro São Paulo. The road network and transport activity data were modeled to the same gridded resolution. We modeled road transport CO₂ emissions at this spatial resolution to interact with the air quality and boundary layer height data according to Eqs. (2) and (4). This scale is among the finest resolutions used in air quality and near-road air pollution modeling (Karner et al., 2010; Kort et al., 2013; Kort et al., 2012). At this high scale, transport emission gradients are easier to be identified (McDonald et al., 2014). The stochastic transport model, which integrates data sources from origin and destination surveys and cross-checked with new traffic sensors (CET, 2013), was used to characterize the traffic patterns keeping time and weekday signatures providing transport demand data not only on highways but also on arterials and residential roads.

Table 2
Vehicle trip surveys in the Metropolitan Area of São Paulo during average working days by types (2012).

| Vehicle type | Daily vehicles (1000 trips/day) | Hourly vehicles (1000 trips/h) | Sampling year | Source |
|--------------|---------------------------------|-------------------------------|---------------|--------|
| Urban cars   | 9063                            | 378 ± 199                     | 2012          | Metro (2012) |
| Int. cars    | 878                             | 37 ± 19                       | 2012          | CETESB (2014a) |
| Urban bus    | 416                             | 17 ± 11                       | 2012          | Metro (2012) |
| Int. trucks  | 182                             | 8 ± 4                         | 2012          | CETESB (2014a) |
| Urban vans   | 732                             | 30 ± 23                       | 2012          | This study |

Fig. 3. Planetary boundary layer height (PBL) for 2012 as derived from modeling estimates. (a) Time profiles of the boundary layer height during mean working day. (b) Spatial distribution of the boundary layer height (18 h UTC). Notes: Each marker in panel a represents estimates at an individual air quality location shown in panel b. Error bars represent standard deviations (SD) for the means across all the Metro São Paulo sites.
The CO2 emissions developed by the bottom-up inventory of this paper can be compared with aggregated CO2 flux fields from other top-down approaches and/or spatial resolutions as the EDGAR’s and CETESB’s anthropogenic emission databases. EDGAR provides estimates of fossil fuel-driven CO2 emissions every month from the transport sector for the São Paulo State at spatial resolution of 10 km; analogously, CETESB provides yearly estimates of vehicle emissions for the different regions of the São Paulo State. These two databases can be compared to deduce emission trends and changes over time.

3. Results and discussion

We calculated fossil fuel-driven CO2 emissions from gasoline light vehicles (cars, motorcycles, and vans) and diesel heavy vehicles (buses and trucks) on every road link of the Metro São Paulo by the hour for an average weekday over the year. The total CO2 emissions and emission factors of light and heavy vehicles, using the methodology of Eqs. (1) to (6) and the data collected, are summarized in Table 3. 16.8 Mt. CO2 eq./year are emitted annually in the Metro São Paulo: 12.2 light and 4.6 heavy vehicles. Light vehicles accounted for about 92% of the total daily vehicle kilometers travelled (91.4 106 VKT/weekday) but contributed to 73% of the total emissions due to lower emission factors per VKT (366 g CO2 eq./km). Heavy vehicles accounted for 8% of the vehicles travelled (8.4 106 VKT/weekday) but 27% of the CO2 emissions due to higher rates (1500 g CO2 eq./km), which are about 4 times those of light vehicles. The emissions of highways and arterials correspond to >75% of total emissions. Table 3 shows also the carbon emissions estimated by the CETESB using local fuel sales, vehicle activity data and dynamometer-based emission factors (i.e. model year and vehicle fleet distribution, technology engine levels and driving conditions). Our estimates are close to the CETESB estimates (∼16% higher), due to the differences in the emission factors, the distribution of the activity by vehicle types and the methodologies used. In terms of total daily traffic, although we used similar aggregated activity data than CETESB, the vehicle distribution was slightly different: 99.8 (91.4 light and 8.4 heavy vehicles) vs. 106.9 106 VKT/weekday (100.0 light and 6.9 heavy vehicles).

The CO2 estimates in this manuscript are based on air quality measurements and segment activity data and reflect real-world characteristics and driving conditions in the urban environments of the Metro São Paulo and complement the exhaust emission dynamometer measurements of Brazilian vehicles performed by CETESB. Our model is geo-referenced and differentiates the total CO2 emissions and emission factors within the transport zones of the Metro São Paulo. Emission factors are higher inside the city center, delimited by the city’s marginals, because of heavier road transport activities and complex road system. Marginals are sections of urban express highways that run near the waterfront of the local rivers Tietê and Pinheiros connecting the East, North and West parts of the city and linking to other interurban highways and airports. Outside of the marginals there is an increasing trend in the CO2 emission factors, especially in the north and eastern sides of the Metro São Paulo, which originates by the construction of new road infrastructures and increment of the demography in the peripheral areas of this region. After studying time trends from 2007 to 2012, it is expected in the forthcoming years that the total emissions and emission factors in the transport zones outside the city’s marginals could increase those in the city central business district (CBD), if the demographical and road-construction trends continue, as it was shown by the air quality measurements at the CETESB’s sites (Fig. 4). The growth rates of vehicle-related pollutants are different comparing among sites at the central district and the peripheral regions during the period from 2007 to 2012. High-decrease rates (>20–30%/5 year) are measured in central areas for CO2, NOx and PM10 (oppositely to O3 high increments) due to slow change in vehicle activity.

In the city centre air-quality could had improved more than in the periphery of the Metro São Paulo because of stronger traffic restrictions, higher use of public transit and newest vehicle fleet. The investment in important mass transit electrified infrastructures and opening of new metro lines explain part of the change in the fossil fuel-driven vehicle activity that can justify the high decreasing ratios measured in CBD. However, the increasing use of transport network companies (TNCs) and the potential diversion of passengers from conventional public transport has to be researched more carefully in these regions. In general, central locations with a large decrease in heavy traffic showed the largest decreases in vehicle-related pollutant emissions after 2007 and the diesel trucks tended to be reallocated at the periphery of the Metro São Paulo. We show that our study to detect emissions using transportation mobility data and air quality concentrations identifies new transport area zones with higher growth in vehicle emissions. Growth rates are spatially heterogeneous indicating the need to continuously update the emission inventories. We have to take into account that data vary across our study and the fact of having more environmental monitoring stations in the central areas of the city could interfere the results. In the future, it is expected to enlarge the air quality network to other regions which could confirm our results about spatial patterns and CO2 emission enhancements in the periphery compared to the city centre.

3.1. Temporal patterns of CO2

Fig. 5 shows CO2 data from a site placed at JAG inside the Butantã Campus of the University of São Paulo USP (western part of the Metro São Paulo). The CO2 data, sampled from July to September 2014 (winter months), present a typical 24-hour model related to high mixing ratios at nighttime, corresponding to lower height of the city’s planetary boundary layer. During the day CO2 concentrations are lower due to the expanding PBL (Fig. 3a), solar heating of the surface and photosynthetic uptake of CO2 (McKain et al., 2012). Concentrations have a notorious peak during the early morning due to rush traffic, lower air turbulence and atmospheric stratification; CO2 concentrations decrease sharply afterwards as wind speed and boundary layer increase. The CO2 concentration pattern differs from transport activities during the afternoon and evening hours, explaining the greater influence of meteorological conditions above vehicular emissions. Fig. 5 also presents the daily profile of stable CO2 data from a background site at the northeastern part on a mountain range of 300 m over the mean altitude of the Metro São Paulo (Fig. 1). Background CO2 exceedances (CO2xs) fluctuate, in an hourly basis, from −5 to 20 ppm in the afternoon (12-18 h) to 15–55 ppm (1 SD = 35 ppm) in the night (22-6 h). CO2xs could be even negative in the afternoon due to CO2 consumption from the urban living organisms (Newman et al., 2013; O’Shea et al., 2014). During morning hours (7-10 h), CO2xs (10–35 ppm, 1 SD = 25 ppm) represent the combination of stable meteorological conditions.
and peak-hour traffic. CO₂ concentrations during winter months are higher than in summer. CO₂ concentrations at the USP site increase at a rate of 1.2 ppm/year between July 2013 and July 2015, lower than the continuous growth of 1.6-ppm/year presented at Manu Loa Research Observatory, Hawaii (http://www.esrl.noaa.gov/).

Fig. 6 shows the temporal pattern of road CO₂ emissions, sum of light and heavy vehicles for all road types in terms of tons of C/h. Light vehicles constitute 73% of total daily emissions vs. 27% corresponding to heavy vehicles. Cars and trucks have different emission profiles. Cars have their emissions mainly during daytime (≈ 70%). The VKT corresponding to trucks occurs also during nighttime and the share of the trucking CO₂ emissions are maintained high during night (12–6 h, Fig. 5). The temporal patterns are also different considering the main road-types due to different real-word driving characteristics and conditions. Therefore, emissions at freeways correlate better to the rush traffic hours than emissions at arterials and residential roads. Arterial and residential segments are less affected by the driving characteristics (i.e. acceleration, deceleration and congestion events) than freeway segments, since they represent interrupted traffic-flow conditions and could peak just after rush hours, when traffic activities are still high. Fig. 6 also compares the temporal patterns of CO₂ emissions with variations in explanatory variables, as boundary layer heights, dilution rates $D$ and air quality levels (CO and PM$_{10}$). The variations of vehicle CO₂ emissions correlate well with the variations of the CO and PM$_{10}$ levels, corresponding to the traffic flows (VKT of Fig. 5). These emissions are modulated by the boundary layer heights and dilution rates, especially during afternoon and evening hours. The hourly variations of the emissions are slightly different than the variation of the traffic flows. This difference is due to dependent factors affecting the driving conditions, as speed and acceleration, but also to independent factors as meteorological and topographical conditions.
3.2. Gridded inventory and high-resolution modeling

In Fig. 7 estimated spatial patterns of on-road vehicle emissions (hot-stabilized) of CO$_2$ by hour of the weekday in the Metro São Paulo are shown for the 460 transport area zones. The inventory estimates used model data based on air quality and traffic flow and estimated traffic counts from the origin/destination mobility survey. The maps are for the year 2012 and the air quality CETESB's sampling sites are marked with blue crosses and squares for reference. Fig. 7 shows fossil fuel driven on-road CO$_2$ emissions, gasoline light vehicles (cars, motorcycles and vans) and diesel heavy vehicles (buses and trucks), from the Metro São Paulo represented in a 0.5 km resolution gridded map. The map represents the spatial patterns of road CO$_2$ emissions during morning, afternoon and evening traffic hours. Higher emissions match with the main highways and intersections and with the major arterials inbound and outbound the city centre.

Northern and eastern areas have higher emissions related to stronger traffic activities than southern areas. The map differentiates the main urban centres and highways; CO$_2$ emissions are higher in the urban centres (>600–1000 kg C/0.25 km$^2$/h) than in the suburban periphery (<400 kg C/0.25 km$^2$/h). In the city centre low road emission spots are identified at the Ibirapuera Central Park and at the Congonhas and Campo de Marte Airports; other areas with more traffic restrictions, as the USP Campus, have lower emissions as well (Fig. 1). Vehicle CO$_2$ emissions correlate well with land-use patterns of the Metro São Paulo as the population density.

Hourly values of road emission fluxes of $\approx$ 382 $\mu$g CO$_2$/m$^2$/s (1500 kg C/0.25 km$^2$/h) and $\approx$ 1273 $\mu$g CO$_2$/m$^2$/s (5000 kg C/0.25 km$^2$/h) have been estimated differentiating near highway network sites and residential areas. These average values are closed to the estimates reported in other cities (Grimmond et al., 2002; Mays et al., 2009; Nemitz et al., 2002; Velasco et al., 2005): Chicago (440–1670 $\mu$g CO$_2$/m$^2$/s, nocturnal to diurnal

Fig. 5. Measured CO$_2$ hourly concentrations during winter months (July–September 2014) at the University of São Paulo (USP) site (left y-axis) and at the Jariaguá Peak (background site at the Metro São Paulo) vs. hourly traffic vehicle kilometers travelled (light and heavy vehicles) estimated from the Metro’s Mobility Survey (2012) for the Metropolitan Area of São Paulo (right y-axis). Note: error bars of CO$_2$ concentrations represent standard deviations (SD) for the means across the two sites.

Fig. 6. Averaged daily profiles of vehicle CO$_2$ emissions ($\pm$ C/h); pollutant concentrations (CO, PM$_{10}$; Maximum value Max = 1.0); planetary boundary layer (PBL; Max = 1.0); dilution rates (D; Max = 1.0).
variations), Indianapolis (840 μg CO₂/m²/s), Mexico City (410 μg CO₂/m²/s) and Edinburgh (750–1670 μg CO₂/m²/s, nocturnal to diurnal variations during autumn).

The city centre of the Metro São Paulo concentrates most of the economy and private transport activities but not all the demography, residential areas are also located in the periphery. The motorization rates and incomes are much higher in the city centre. People of the periphery, mostly living in the north and east residential transport zones, use the public transport more than the people living in the city centre (METRÔ-SP, 2012). There are large inward and outward public transport flows during morning (7–9:00 h) and evening rush-hours (17–19:00 h) to and from the city centre, respectively. Due to demographical and economic characteristics, in the Metro São Paulo the spatial patterns do not fluctuate much during the day and the vehicle CO₂ emissions are always concentrated in the city centre and the main highways.

3.3. Caveats and difficulties of emission patterns

Determination of road real-world emissions has several caveats and difficulties because the complex processes involved, as the meteorological conditions, vehicle fleet characteristics and traffic conditions. The uncertainties of the parameters used in the Eqs. (1) to (6), as the height of boundary layer and road segment VKT, represent the spatial and temporal variability of real-world emissions. The first difficulty tried to understand why the instantaneous bottom up emissions in this manuscript are higher (≈16%) than the top-down emission simulated from the dynamometer.

Fig. 7. Estimated high-resolution CO₂ emissions from fuel-based vehicles in the Metropolitan Area of São Paulo during 2012, in terms of kg C/0.25 km²/h (on unit grid cells of 0.5 km × 0.5 km). (a) 6:00–7:00 h, (b) 13:00–14:00 h, (c) 18:00–19:00 h, using meteorological parameters, model Eqs. (1) to (6) and interpolation Kriging. Note: All vehicles, 0.5 km grid by transport area zones TAZs (n = 460) in kg/0.25 km²/h for an average weekday over the year.
measurements of CETESB, since we used the same vehicle fleet. Therefore, the difference lay down in the emission factors and vehicle activity data used. Second, we gathered accurate vehicle activity data; high scale spatially distributed VKT were determined for each road segment (320,087) of the 40,590 km road network; vehicle activity data complemented vehicle fleet data (vehicle types and technological timing) and studied their air quality impacts. Integration with the empirical traffic flow measurements of the CET was also done.

Third, we developed vehicle activity data based on mobility surveys, which could be extrapolated to other Brazilian urban environments with deficient transport statistics. Fourth, we established real-world emission factors at the Metro São Paulo by vehicle types (light and heavy vehicles) which complement other databases based on laboratory regional measurements (CETESB) or international emission standards, such as European and United States emission rates (COPERT, MOVES and IVE models). After performing additional vehicle tests in the future, based not only on laboratory engine cycles but also based on vehicle driving tests, the gap between our estimates and the CETESB’s inventory data could be closed (Huo et al., 2010).

Performing additional vehicle tests in the future, based not only on laboratory engine cycles but also based on vehicle driving tests, the gap between our estimates and the CETESB’s inventory data could be closed (Huo et al., 2010; Franco et al., 2013).

4. Conclusions, implications and further work

In this manuscript we have shown a new combined framework capable of estimating changes in vehicular CO2 emissions from the Metropolitan Area of São Paulo on a spatial and temporal resolution of 0.5 km and 1 h. The approach consists on a transport demand model based on a high-resolution stochastic user equilibrium and coupled to the boundary layer height estimates coming from the mesoscale meteorological model coupled with chemistry (WRF-Chem) and to the ground observations of pollutants from the CETESB’s monitoring stations and LAPAt measurements. Vehicle emission factors by vehicle types were estimated by intermediate pollutant dilution rates, giving central and confidence intervals by time of the day and metropolitan regions (transport area zones TAZs). Finally, urban CO2 emissions have been reported by means of carbon fluxes on unit grid cells and hourly basis. In the Metro São Paulo there was an increase of CO2 concentrations during morning/afternoon related to the vehicle kilometers travelled (from light and heavy vehicle transportation activities). Evening/night concentrations came from other sources such as the combustion of natural gas and the biosphere. Overall changes until 15% in fossil fuel driven emissions were estimated during a five-year period (2007–2012), oppositely to changes of air pollutants (high-decrease rates \( \approx 25\% / 5 \) year).

The estimations of accurate vehicle emissions are still challenging because they depend on many parameters and can change rapidly over time and space. Our model approach intends to complement other worldwide and regional approaches, developed for different aims, as the complexity of the methodology used does not guarantee the accuracy of the road vehicle source estimates. Our bottom-up approach estimates reflect CO2 emissions at a road-segment level that agree with air quality and meteorological measurements and estimates. Our results could be useful for policy makers to establish ad-hoc measures to reduce air quality impacts of vehicles in the Metro São Paulo. They can also serve as base study to other atmospheric and chemistry models to explain the apportionment of on-road sources to air pollution and quality. Once the dimensions of the problem have been assessed over a broad period of years in the region, it will be possible to prospect future vehicular emissions and establish mitigation scenarios on past trends analysis, based on the development of technological measures and policies. The importance of the relationships observed in the manuscript may serve to underlie the necessity to establish new urban planning, transport policies and pollution control measures to reduce concentrations of pollutants in other megacities worldwide.

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