Evaluation of OSPM against Air Quality Measurements in Brazil – The Case Study of Fortaleza, Ceará

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Abstract

The contribution of vehicle emissions to air pollution is considered a large environmental and health problem in big Brazilian cities caused, among others, by slow renewal of the old vehicle fleet. Brazilian studies usually only consider traffic-related issues in transportation analysis, with minor assessments of emissions and close to non-existent assessment of air quality. On this background, this research aimed to calibrate and evaluate the Operational Street Pollution Model (OSPM[®]) to Brazilian conditions by implementing Brazilian emission factors. The urban background concentrations were modelled with the Urban Background Model (UBM) as part of the air quality system (THOR – AirPAS). In this case, we used meteorological data from a ground meteorological station outside Fortaleza processed by meteorological pre-processor and regional background concentrations from the Integrated Forecast System (IFS) as input to UBM. New air quality measurements were collected in busy streets of the city of Fortaleza during the year of 2017. The study collected samples of daily NO₂ and PM₁₀ concentrations to evaluate OSPM daily estimations. In addition, a transportation travel demand model (TRANUS) has been calibrated to the case study area with observed traffic data collected, in order to provide Annual Average Daily Traffic (AADT) as inputs to OSPM[®]. Two sets of emission factors were

evaluated. Official Brazilian emission factors were applied to OSPM[®], as well as adjusted emission factors derived in the project based on calibration that were higher than the official emission factors. Data showed that concentrations are significantly influenced by meteorological factors (such as temperature, wind speeds, wind directions), and especially precipitation for PM₁₀ concentrations. OSPM[®] simulated results showed concentration levels and patterns close to air quality measurements with default emission factors and calibrated emission for UBM but large underestimations if official emissions were used for both UBM and OSPM.

Keywords: Air Pollution, dispersion modelling, Evaluation, OSPM.

1. Introduction

According to the latest air quality database, 97% of cities in low and middle income countries with more than 100 000 inhabitants do not meet WHO air quality guidelines (WHO, 2018). In Brazil, in the city of Rio de Janeiro, for example, 77% of the population is exposed to levels above the WHO guidelines (Coelho, 2006). Thus, when planning for improvements in urban mobility, not only indicators that result in improved traffic related issues should be observed, but also those that impact on human health, especially those related to air quality. In addition, HDV (Heavy Duty Vehicles) make up only 5% of the vehicle fleet in Sao Paulo city but have been found to contribute 40% of benzene and 47% of black carbon concentrations, showing the large contribution from HDV in urban areas (Brito et al., 2018). In newly industrialized countries, such as Brazil, traffic-related NO₂ and PM₁₀ air pollution is particularly high, due to old vehicle fleet technology with high emission factors from inefficient engines and control technologies (CETESB, 2017a).

Air pollution is a major cause of diseases. It is estimated that at least 3% of cardiopulmonary and 5% of lung cancer deaths are attributable to PM globally (WHO, 2013). NO₂ has been associated with adverse effects on hospital admissions for various diagnoses, decrements in measures of lung function and lung function growth, increases in respiratory symptoms, asthma prevalence and incidence, cancer incidence, adverse birth outcomes and mortality (US EPA, 2013; WHO, 2013). The most recent study on the Global Burden of Disease estimates that 7.5% of premature deaths globally were attributable to ambient air pollution in 2016. In the same year, 27.5% of

deaths due to Lower Respiratory Tract Infections and 26.8% of deaths due to Chronic Obstructive Pulmonary Diseases were linked to air pollution (Li and Mallat, 2018). All of these negative effects lead to an increase in the socio-economic costs due to health impacts while also influencing quality of life (Darcin, 2014; Vienneau et al., 2015).

To estimate air pollution concentrations of NO₂ and PM₁₀ in street levels, this research aims to calibrate and evaluate the Operational Street Pollution Model (OSPM) to Brazilian conditions with Fortaleza as case study to develop a model system for decision-support of traffic-related measures such as for example Low Emission Zones, which are still not implemented in Brazil (ICCT, 2019). Previous studies were successful in modelling air quality in Brazil (Andrade et al., 2015; Krecl et al., 2015, 2016, 2018, 2019). However, these previous approaches aimed to study regional concentrations or street concentrations with only one street observed, with different air pollutants. In addition, previous studies observe southern/south region of Brazil only. The new approach presented in this study combines travel demand modelling and air quality modelling, and is the first of its kind in Brazil. The study area is Fortaleza, Ceará, Brazil (03° 43' 02" south latitude and 38° 32' 35" west longitude, Northern Region), which suffers from high vehicle density, thus highly affected by traffic-related air pollution. Fortaleza is a typical big Brazilian city, which can represent traffic-related air pollution in general in developed cities in the country. OSPM Presents a more detailed mechanism for physical behavior in roads with presence of tunnels and urban canyons when compared to other street air pollution models (Martins et al., 2015), which is specifically accurate to observe in this study. In addition, OSPM is the final modelling step in air quality modelling system THOR-AirPAS, a sophisticated modelling system which integrates regional background concentrations (in this study performed with Integrated Forecast System, IFS), urban background concentrations (embedded in THOR-AirPAS system, Urban Background Model, UBM) and street levels air pollution concentrations (last step, Operational Street Pollution Model, OSPM) (DCE, 2014). OSPM is capable of disaggregate different types of vehicles, several different pollutants (some air pollution models are not capable of estimate NO₂ and PM₁₀) and different scenario configurations (Berkowicz et al., 1997; Berkowicz et al., 2003).

Air quality measurements with passive samplers for NO₂ and active samplers for PM₁₀ in selected streets were carried out for this research for air quality assessment and model evaluation as there is few air quality monitoring stations in Brazilian cities with reliable data. All required input data for the Operational Street Pollution Model (OSPM) was generated using different tools and models, and OSPM was calibrated and evaluated against the air quality measurements. The overall methodology for this study, results, discussion and conclusion are presented as follows.

2. Methods

Overall methodology

Figure 1 shows the methodology applied in this research to evaluate OSPM modelled air pollutant concentrations against observed measurements at street levels.

Figure 1: Flowchart of methodology to evaluate OSPM air pollutant concentration estimations against observed concentrations at street level, Fortaleza, Ceará - Brazil

OSPM is used to calculate street concentrations for comparison and evaluation against collected air quality measurements at selected streets. OSPM requires inputs concerning traffic data and

street geometry, vehicle emissions, urban background concentrations and meteorology. Urban background concentrations are calculated with UBM. Urban Background Model estimate concentrations at a receptor by numerical integration of contribution from each of the individual area sources along each actual wind direction path upwind to 30 km from the receptor, where the rural background is used as boundary condition (in this case, results from the IFS model). The horizontal diffusion is assumed to follow a Gaussian function. UBM is set-up to calculate the following pollutants: NO_x (nitrogen oxides), NO₂ (nitrogen dioxide), O₃ (ozone), SO₂ (sulphur dioxide), CO (carbon monoxide), TSP (total suspended particulate matter), PM₁₀ (particles less than 10 µm in diameter) and PM_{2.5} (particles less than 2.5 µm in diameter) (DCE, 2014). UBM requires inputs on gridded emissions, regional background concentrations and meteorology. In this study the regional background concentrations were estimated using IFS (Integrated Forecast System) instead of DEHM since the latter only covers the Northern Hemisphere (Christensen, 1997). IFS is capable of estimating global Ozone (O₃), Nitrogen Monoxide (NO), Nitrogen Dioxide (NO₂), Carbon Monoxide (CO) and disaggregated particles, such as: seasalt; dust, hydrophilic organic matter, hydrophobic organic matter, hydrophilic black carbon and hydrophobic black carbon. IFS operates with a spatial resolution of about 80 km and three-hourly time resolution (Flemming et al., 2015).

Meteorological data for UBM and OSPM is based on processed meteorological data from a ground meteorological station outside Fortaleza using a meteorological pre-processor. THOR-AirPAS is an air pollution assessment system that has a user interface to OSPM and UBM to ease interaction with input data, air quality models and output data (Jensen et al., 2014).

In the following, the case study area and the air quality measurement campaign are described, and the models UBM and OSPM are described in brief together with the generation of model input data.

Case study area of Fortaleza

Fortaleza is the capital of Ceará State and the fifth biggest city of Brazil, located in Northeastern Brazil, with a population of approx. 2.6 mill. inhabitants in 2017 (Instituto Brasileiro de Geografia e Estatística [IBGE], 2018). Fortaleza is a tourist city. In addition, it is one of the main destinations for agricultural products from municipalities in Ceará and other states and also

supports these areas with public and private services. The total land area of Fortaleza is 315 km², which includes 119 districts in five regions throughout the city (Fortaleza, 2018). Fortaleza has a typical tropical climate, with a wet and dry season, high temperatures and high relative humidity throughout the year. The annual mean precipitation is approximately 1,448 mm, and the annual mean temperature (T) is 26.3 °C (FUNCEME, 2018). In general, Fortaleza is a typical, prosperous coastal city in Brazil. The air quality in this economically developed city is influenced by vehicle traffic, especially due to an old vehicle fleet (Lopes et al., 2018).

Air quality measurements

There were no permanent air quality monitoring stations in Fortaleza at the time of this study, only one monitoring station in the city of Pecém (metropolitan region of Fortaleza), which does not provide reliable data to assess air pollution at street levels of the case study area (SEMACE, 2016). Therefore, a methodology was developed to collect measurements of NO₂ and PM₁₀. Air pollutants data collection has been carried out in the case study area of Fortaleza on locations with high vehicle flow density and high population density. Locations were also chosen to reflect differences in heavy-duty traffic and urban canyon characteristics (different ratios of building height/street width). Passive samplers for NO₂ concentrations were developed especially for this research based on Palmes and Lindenboom (1979) method, which transfers the tested NO₂ gas through a tube of known dimensions using Fick's First law, adapting the NO₂ sampler with a device suitable for data collection in public lighting poles, in order to passively absorb NO₂ in street levels. Cases were constructed for use in street lighting poles (with a structure to resist rain, wind and small impacts), and samplers data collected NO₂ concentrations in street levels, observing data during 24 hours. The values have been obtained by means of spectrophotometry and chromatography techniques.

Active measurements of PM₁₀ concentrations have been performed based on gravimetric analysis to calculate the concentrations. The equipment used was an Ecotech HiVol 3000 (High Volume Particulate Sampler). PM₁₀ mass is accumulated on a filtering membrane, and filters were weighted before and after the data collection, through 24 hours, and divided by the total sampled volume, thereby reaching the PM concentrations (24 hours data collections for PM₁₀, in µg/m³).

The calibration and quality assurance (QA) of both data collection methods have been reported by Ribeiro et al., (2018) and Alcântara et al., (2019). Air quality measurements were collected in two campaigns to assess the unique seasonal variation in Northern Brazil which has a rainy season (March to May) and a dry season (rest of the year). Daily concentrations of NO₂ and PM₁₀ were measured for 24 hours in each day (usually 08:00 a.m. to 07:00 a.m. the following day, due to operational reasons).

First campaign was carried out between May 8 and June 1 and the second campaign from November 19 to December 3, 2017. Due to equipment availability, time for data collection and safety reasons we had to data collect NO₂ and PM₁₀ in different times and locations. In the first and second campaigns, data collection has been successfully carried out for all data collection points with the exception of NO₂ in the first campaign where two samplers missed out for unknown reasons. NO₂ were collected with passive samplers in both campaigns at 20 locations (201 to 220 in Figure 3), and PM₁₀ were collected with active method during both campaigns at 12 locations (101 to 112 in Figure 3). The sampling height was approx. 2.5 m to avoid destruction or other operational problems. Figure 2 shows the location of the case study area and Figure 3 displays the location of data collection points for air quality measurements within the highlighted studied area, and the concentrations observed in second campaign just for sake of illustration (second campaign chosen due to two missing NO₂ observations in first campaign).

Figure 2: Zoom in on location of study area: Brazil, Ceará, Fortaleza, case study area.

Figure 3: Active (PM₁₀) and passive (NO₂) data collection points within highlighted studied area presented in Figure 2. PM₁₀ data collection points have the format 1xx and NO₂ data collection points 2xx. Second campaign concentrations are shown after the number of the collection point.

Urban Background Model (UBM)

Urban background concentrations have been calculated with the Urban Background Model (UBM) (Berkowicz, 2000b, Brandt et al. 2003) as part of the THOR-AirPAS system (Jensen et al. 2014). Concentrations are estimated by numerical integration contributions from each of the individual area sources along each actual wind direction path upwind to 30 km from the receptor point, where the regional background concentrations are used as boundary. The horizontal diffusion is assumed to follow a Gaussian function. The step size is 50 m in the numerical integration both along the wind direction and perpendicular to the wind direction, where horizontal diffusion is taken into account. In the present study a vehicle emissions database with a spatial resolution of 1 km x 1 km has been developed based on traffic data on a road network and vehicle emission factors as input for UBM. The concentrations of NO₂ and PM₁₀ are calculated with a 1 hour time resolution.

Operational Street Pollution Model (OSPM)

Street level air pollutant concentrations have been estimated with the Operational Street Pollution Model (OSPM) (Berkowicz, 2000a; Berkowicz et al., 2008; Kakosimos et al. 2010; Ketzel et al., 2012). OSPM is a semi-empirical atmospheric dispersion model for simulating the dispersion of air pollutants near streets with different building geometry, including open or half-open streets and so-called street canyons. Concentrations of traffic-emitted pollution are calculated using a combination of a plume model for the direct contribution and a box model for the recirculating part of the pollutants in the street. NO₂ concentrations are calculated taking into account NO-NO₂-O₃ chemistry and pollutants residence time in streets. The model is designed to work with input and output in form of one-hour averages. OSPM is capable of estimation of various pollutants provided emission data is available. In the present study hourly NO₂ and PM₁₀ concentrations were calculated and aggregated by day (24 hours), in order to compare with observed concentrations collected in the streets.

OSPM requires input on traffic and street configuration data, vehicle emissions, urban background concentrations and meteorological data. The collection and generation of input data are explained in the following.

Traffic and street configuration data for OSPM

The input data for the OSPM on traffic data and street configurations for the selected urban streets where air quality measurements were carried out are generated using the AirGIS system based on GIS maps of a road network with traffic data, building foot-prints with building heights and calculation points (Jensen et al., 2001;2017, Ketzel et al., 2011). Street configurations include e.g. street orientation, width of street, building heights in different wind sectors etc.

Traffic data for OSPM at air quality measurement locations

Traffic data has been collected with SCOOT (Split Cycle Time and Offset Optimization Technique) system. SCOOT uses inductive loop-type sensors and video cameras to monitor street traffic and to calculate the Annual Average Daily Traffic (AADT), data collected by Controle de Tráfego em Área de Fortaleza (Traffic Control in Fortaleza Area) (CTAFOR, 2015).

Traffic data has been collected for the same locations as the air quality measurements points as part of traffic data input to OSPM. Split Cycle Time and Offset Optimization Technique (SCOOT) system was developed to perform traffic control in real time, requiring constant data between field controllers and a central computer. This system allows monitoring the operation of all traffic lights: controllers, detectors, focus groups and lamps (Loureiro et al., 2002). This system works in real time with traffic signal programming determined dynamically by dedicated systems, based on traffic data collected by field inductive loop-type sensors in the streets (Oliveira, 1997). Monitored traffic data with a time resolution of 15 minutes was expanded to a whole day by SCOOT system, in order to calculate the Annual Average Daily Traffic (AADT) used as input for OSPM (Alves, 2014). Diurnal variations in this study considered SCOOT traffic data and real traffic data collection to validate SCOOT outputs (e.g. vehicle types, speed, flow). OSPM also requires input on vehicle distribution that was provided by SCOOT and traffic counts. Furthermore, OSPM requires input on the temporal variation of traffic in order to estimate hourly values. Here, standard diurnal profiles for the different vehicle categories were used.

Travel Demand Modelling of road network

In order to code and calibrate the travel demand model for the road network, data have been collected for the Municipality of Fortaleza in order to model gridded vehicle emissions for UBM. Data have been used as input to activity, land use and the transportation model TRANUS within the Municipality of Fortaleza. TRANUS is a transport and land-use model developed by de la Barra (de la Barra, 1982; de la Barra et al. 1984; de la Barra 1989, 1998), a transportation model which is based on a unifying principle for modelling and linking all transportation subsystems (Wegener et al. 1986). Travel modelling was based on zoning the city's 119 districts. The zoning was designed in order to build the transportation planning for Fortaleza city. It assumes that there are a small number of internal trips with origin and destination in the same district and that the transportation (road network and public transportation network) within each district is homogeneous in order to route assign all trips generated throughout defined traffic zones to the streets of Fortaleza. The travel demand modelling carried out in this research estimated vehicle

travel in Fortaleza and calculated AADT and Average Daily Travel Speed (ADTS). Travel speeds were also used for OSPM.

Vehicle distribution includes passenger cars, pickups, SUV's, vans, buses, trucks and motorcycles. Brazilian vehicle fleet uses gasoline, ethanol, LPG and diesel. In addition, vehicle technology distribution was considered. Technology evolution in this study considered Brazilian vehicle characteristics, such as: indirect injection (passenger cars); turbocharged vehicles (diesel fuelled) and Selective Catalytic Reduction (SCR) and Diesel Particle filter (DPF) for newer buses and trucks (EURO 5 equivalent in Brazilian standards).

Evaluation of two sets of vehicle emission factors

OSPM requires emission factors and UBM requires gridded emissions as inputs. Calculations were carried out with two sets of vehicle emission factors. Firstly, with default emissions factors based on CETESB and secondly, with adjusted higher emission factors based on calibration as described below.

Default vehicle emissions are based on the Brazilian inventory emission factors calculated by CETESB (Companhia Ambiental do Estado de São Paulo) methodology (CETESB, 2017a), which is the most reliable Brazilian mobile emission database (for NO_x and Exhaust PM₁₀), and the European emission model COPERT IV (for Non-Exhaust PM₁₀ as the Brazilian database does not include non-exhaust) (Alam et al., 2017).

The adjusted higher emission factors were derived in the following way. UBM requires gridded emissions as inputs where vehicle emission factors are one of the parameters needed. In the THOR-AirPAS system it is possible to adjust the gridded vehicle emissions for UBM with single factors for each pollutant. This option was used to scale the vehicle emissions in a way that the modelled street concentrations using UBM and OSPM on average were close to measured street concentrations. This calibration was based on default vehicle emissions for OSPM, and for UBM, a factor of 1.5 was used for NO_x emissions and a factor of 4.0 for PM₁₀ emissions.

We carried out four scenario calculations:

Scenario 1. Default emissions for both UBM and OSPM (low scenario)

Scenario 2. Adjusted emissions for both UBM and OSPM (high scenario)

Scenario 3. Adjusted emissions for UBM and default emissions for OSPM (mixed scenario)

Scenario 4. Default emissions for UBM and adjusted emissions for OSPM (mixed scenario)

Scenario 1 with default emissions for both UBM and OSPM lead to a large underestimation of modelled street concentrations compared to measurements. On the other hand, scenario 2 with adjusted emissions for both UBM and OSPM resulted in a large overestimation of modelled street concentrations. Therefore we examined the mixed scenarios of scenario 3 with adjusted emissions for UBM and default emissions for OSPM, and scenario 4 with default emissions for UBM and adjusted emissions for OSPM. In this paper, we only present data for scenario 3 and 4. Few Brazilian studies have analyzed default CETESB emission factors in order to assess potential underestimations which will have high impacts on simulated concentrations. However, Krecl et al. (2018) assessed default emission factors and suggested adjustments due to methodological underestimations. Table 1 shows adjustment factors for default CETESB emission factors for the Brazilian vehicle fleet aggregating in the two categories of Light Duty Vehicles (LDV) and Heavy Duty Vehicles (HDV).

Table 1. Adjustment factors to account for underestimations in default CETESB emission factors, therefore Exhaust Emissions (Krecl et al., 2018)

Type of Vehicle	Factor NO _x	Factor PM ₁₀
LDV	1.8	4.3
HDV	3.0	3.6

The adjustment factors proposed by Krecl et al. (2018) are very close to the ones we use for our adjusted emission factors.

3. Modelling Inputs for OSPM

Average Annual Daily Traffic and Speeds

Annual Average Daily Traffic (AADT) was collected using the SCOOT system for the locations of the monitored streets in Fortaleza and used to calibrate the travel demand modelling TRANUS.

Vehicle fleet in Fortaleza was aggregated in groups of vehicle types to match emission data: (i) Passenger Cars: light duty vehicles, including cars and gasoline/flex compact SUV's (Brazilian legislation prohibits diesel fuelled small cars); (ii) Pickups, SUV's and Vans: all pickups, larger SUV's and Vans fuelled by diesel; (iii) Trucks: Light, Medium and Heavy Trucks, the major part of the Brazilian truck fleet is fueled by diesel (with very few exceptions and therefore disregarded in this study); (iv) Buses: Micro buses, urban buses and coaches are considered in this type, the majority is fueled by diesel (with very few exceptions that are disregarded) and; (v) Motorcycles: all motorcycles in the Fortaleza fleet are fueled by gasoline/flex. A flex car can run on a mix of gasoline and alcohol. Vehicle distribution in Fortaleza was observed specifically for this research. Several streets within the studied area in order to understand traffic proportions in 5 disaggregated vehicle classes, observed by traffic counting (manually) and with cameras in several different spots in our studied area. This information was crucial in order to use as inputs in OSPM. The vehicle distribution is shown in Table 2.

Table 2. Average vehicle distribution in Fortaleza, CE (%). Distribution calculated using the SCOOT data collection system and counts in street crossings in case study area.

Passenger Cars	Pickup, SUV, Van	Truck	Bus	Motorcycle
62.7	4.6	1.9	2.5	28.4

OSPM requires as inputs average traffic speeds in order to model air pollutants in street canyons (Kakosimos et al., 2010). TRANUS travel demand model was capable of estimate average travel speed in Fortaleza region and exported to shape-file, necessary as input to OSPM. In this regard, the road network with modelled average travel speeds of Fortaleza main streets is shown in Figure 4.

Figure 4. Road network with travel speeds (km/h) in all streets of the municipality – Fortaleza, CE.

Calibration of average daily traffic flows has been carried out with the inductive loop-type sensor for traffic data collection using the SCOOT system. TRANUS was calibrated to estimate AADT for all streets in Fortaleza and several parameters can interfere in traffic estimations, such as activity system and land use (de la Barra, 1982). Uncertainties in estimated AADT can be observed and influence air pollutant concentrations in street levels estimated by OSPM. The modelled traffic flow and observed traffic flow (used to calibrate travel demand model) of major streets inside and outside the case study area are shown in Table 3.

Table 3. Observed and modelled average daily traffic flows in major streets of Fortaleza.

Street/Avenue	Modelled	Observed	Difference	Relative Error (%)
Washington Soares	52625	50239	2386	+5
Santos Dumont	44671	42607	2064	+5
Antônio Sales	26934	34280	-7346	-21

Presidente Castelo Branco	46736	40439	6297	+16
Padre Antônio Tomás	39909	38745	1164	+3.
Padre Valdevino	27967	20936	7031	+34
Sen. Virgílio Távora	31637	30980	657	+2
Br. De Studart	25292	31501	-6209	-20
Rui Barbosa	15626	18484	-2858	-15
Ana Bilhar	22828	23006	-178	-1

The difference between modelled and measured traffic flows shows both under- and overestimations with modelled traffic flows within -21% to 34% of measured traffic flows due to uncertainties of AADT modelling in a city the size of Fortaleza.

Meteorological Data and Regional Background Concentrations

The characteristics of meteorological data in Fortaleza are shown in Table 4. The mean temperature (T) in 2017 was normal (mean $\pm$ SD: 28.3 ± 2.2 °C), while the relative humidity (RH) was high (76.1 ± 12.8 %). In addition, the precipitation intensity (PI) was normal for the city (0.2 ± 1.2 mm/h). The collected and calculated meteorological parameters were used in combination with regional background concentrations to estimate urban background concentrations using UBM.

Regional background air pollutant concentrations were available for the whole year of 2017, however, meteorological data for the air quality calculations for UBM and OSPM only consider the two measurement campaign periods (May, July and November) for NO₂ and PM₁₀.

Table 4. Characteristics of meteorological data collected and calculated hourly for year 2017. Temperature (T), Relative Humidity (RH), Precipitation Intensity (PI), Air Density (AD), Wind Speed (WS), Global Radiation (GR) were field data collected by FUNCEME. Mixing Height (MH), Monin-Obhukov Length (M-OL), Convective Velocity (CV) and Friction Velocity (FV) were calculated using OML-Highway meteorological Pre Processor.

Statistics	T (°C)	RH (%)	PI (mm/h)	AD (kg/m ³)	WS (m/s)	GR (W/m ²)	MH (m)	M-O L(m)	CV (m/s)	FV (m/s)
Mean	28.3	76.1	0.2	1.2	3.9	236.5	474.7	25.5	0.3	0.2
Standard Error	0.02	0.1	0.01	0	0.02	3.4	4.4	10.1	0.01	0
Kurtosis	-0.6	-0.5	328.4	-0.6	-0.8	-0.4	4.6	82.8	0.8	3.9
Skewness	0.3	-0.3	14.9	-0.1	0.04	1.0	1.9	1.9	1.4	1.7
Range	11.9	63.6	40.8	0.1	9.3	1071.1	3108	19998	2.9	1.5
Minimum	22.4	36.7	0	1.1	0	0	150	-9999	0	0.03

Maximum	34.3	100.3	40.8	1.2	9.3	1071.1	3258	9999	2.9	1.6
Count	8760	8760	8760	8760	8760	8760	8760	8760	8760	8760

Gridded vehicle emissions for modelling urban background concentrations

Gridded vehicle emissions in tons/year are needed as input to UBM to be able to model urban background concentrations. NO_x and total PM₁₀ emissions (exhaust and non-exhaust) are required. A grid with grid cells of 1 km x 1 km was defined for the case study area using GIS to be able to model vehicle emissions. The emissions of each grid cell were calculated based on data on road length, traffic flow, travel speed, vehicle distribution and emission factors of different emission technology layers (Pre-Euro to Euro 5) and further depending on vehicle type and fuel type. In Figure 5, the percentage of vehicle emissions in Fortaleza is shown broken down by vehicle type and fuel type and emission technology layer.

Variability in emission contributions for a given vehicle type are results of different reasons in this study: proportional fuel used (e.g. passenger cars using gasoline, ethanol, mixture of both or LPG; motorcycles using gasoline or mixture of gasoline and ethanol); age of manufacturing (which also change fuel used); vehicle engine sizes (e.g. trucks and buses with three different sizes mentioned in Figure 5, in addition to age of manufacturing). Variations in emission contributions among emission standards for a given pollutant and vehicle type are results of fleet age of manufacturing (real data used).

Passenger cars have been disaggregated by fuel type: gasoline, alcohol, flex and LPG. Pickups, SUV's and Vans use diesel. Trucks were sub-divided in less than 10 tons, between 10 and 15 tons, and plus 15 tons (all diesel). Buses are sub-divided in Urban buses, Coaches and Mini buses (all diesel). Motorcycles were divided in gasoline and flex fueled. Brazilian emission standards (PROCONVE standards) have been converted to equivalent EURO standards (ICCT, 2016; CONAMA, 2017). It is important to note that Flex vehicles are the most numerous in the Fortaleza fleet, however, around 90% of them uses gasoline, due to economic reasons as gasoline is cheaper than alcohol (FORTALEZA, 2015).

Figure 5. NO_x (a) and PM₁₀ (b) emission contributions (%) for traffic conditions in the Fortaleza vehicle fleet. PM₁₀ contributions includes exhaust and non-exhaust particles of mobile emissions.

Spatial distribution of gridded vehicle emissions (1 x 1 km) for NO_x is shown in Figure 6. The highest emissions are in the north part of Fortaleza due to high density of roads and high traffic flows, as observed by Oliveira et al. (2019) with 13 Urban Recreation Areas (URA's) data collection points experiment.

Figure 6. NO_x emissions from traffic in Fortaleza in 2017 based on adjusted emission factors (ton/year/km²). The same spatial pattern is also observed for PM₁₀ emissions.

4. Results

Modelling urban background concentrations

Urban background concentrations were modelled with UBM based on the gridded vehicle emissions, regional background concentrations and meteorological data for the whole year of 2017, and hence also for the two campaign periods. Urban background concentrations are used as input to model street concentrations with OSPM. Below we show results based on adjusted emission factors.

Adjustments in UBM have been carried out similarly to research presented in Valencia et al., (2019), where emission inputs can be used to calibrate both NO_x and PM_{10} . Ideally, these factors must have the value of one, higher values increase the emissions and produce higher concentrations and the opposite is valid. In this study, UBM adjustments considering OSPM default emission factors have been calculated with $\text{NO}_x = 1.5$ and $\text{PM}_{10} = 4.0$. In scenario with OSPM adjusted emission factors (Krecl et al. 2018), UBM factors were calculated with value of one with satisfactory good results.

Results are illustrated with average over the campaign period and daily for urban background concentrations and compared to regional background data for the first campaign in Figure 7.

Meteorological data shows that first campaign is rainy season in Fortaleza and second campaign shows dry season in studied area, however, in order to illustrate underestimations in street pollution modelling during rainy days, the first campaign has been highlighted in data and figures.

Temporal variation of background pollution concentrations is influenced by the meteorological parameters. Regional NO_x , NO_2 and O_3 concentrations showed variations probably due to wind direction changes and higher wind speeds in month of July (reduction or increasing of source areas), PM due to precipitation days during the month of May (PM washout) (Akyuz et al., 2009; Barmpadimos et al., 2011). Urban concentrations follow regional concentration patterns, with urban mobile sources increment.

As expected the urban background concentrations are much higher than the regional background concentrations for NO_x and NO_2 as traffic is a major source of NO_x emissions. It is also seen that regional background ozone concentrations are higher than urban background ozone

concentrations as NO_x emissions in the city deplete ozone. For PM₁₀ the regional background concentration is relatively high compared to the urban background concentration of PM₁₀, due to seasalt contributions in Fortaleza (city close to the sea). PM₁₀ regional background concentrations are proportionally much higher when compared to NO_x and NO₂ due to the same reasons previously mentioned.

Figure 7. Daily and average NO_x, NO₂, O₃ and PM₁₀ urban background concentrations in Fortaleza based on UBM (µg/m³), calculated with adjusted emission factors, presented in (a), (c), (e) and (g). Regional background concentrations presented to compare with urban background concentrations (b), (d), (f) and (h).

Evaluation of Modelled Street Concentrations with OSPM against Air Quality Measurements

Street concentrations were modelled with OSPM for selected streets based on traffic data, street configuration data, modelled urban background concentrations and meteorological data.

Calculations were carried out with two sets of vehicle emission factors for OSPM. Firstly, with default emission factors based on CETESB and with adjusted emissions for UBM (scenario 3), secondly, with adjusted emission factors for OSPM and default emissions for UBM (scenario 4).

Based on default vehicle emission factors for OSPM and adjusted emissions for UBM (scenario 3)

Figure 8 and 9 show OSPM estimated concentrations of NO₂ and PM₁₀ compared to observed air quality data (24 hours) collected during the two campaigns (2017). Table 5 shows that mean simulated NO₂ concentration was higher in the first campaign in July (mean ± SD: 27.5 ± 6.2) compared to the second campaign in November (23.9 ± 6.5). Observed concentrations showed the same pattern in the first campaign (28.9 ± 17.8) compared to the second campaign (19.8 ± 9.9). In the case of PM₁₀, the mean simulated concentration was lower in the first campaign in May (22.2 ± 3.7) when compared to the second campaign in November (27.3 ± 4.4), and the same pattern was also seen for measured concentrations in first campaign (19.7 ± 6.5) compared to the second campaign (24.7 ± 4.1). Meteorological and traffic factors show large influence on measured and simulated results. Streets with high AADT and days with lower wind speeds show higher concentrations, and days with precipitation show lower concentrations, especially for PM₁₀. In the second campaign (November) there was almost no rains in Fortaleza and hence no washout of PM in the atmosphere. This may be the reason why PM₁₀ is better in agreement with the measurements in the second campaign compared to the first campaign as OSPM does not take into account PM washout. There was a good correlation between the mean of all observations and simulations although the model overestimated for NO₂ in the second campaign.

The good agreement between the mean of all observations and simulations is a result of the calibration of the emission factors for UBM to fit modelled street concentrations.

Figure 8. Scenario 3. Simulated and observed NO₂ concentrations during two campaigns (July and November, 2017). (a) NO₂ in first campaign. (b) NO₂ in second campaign. (c) Scatter Plot of simulated and observed NO₂ data, 1^o Campaign. (d) Scatter Plot of simulated and observed NO₂ data, 2^o Campaign. (e) Boxplot of NO₂ concentrations in first campaign. (f) Boxplot of NO₂ concentrations in second campaign.

Figure 9. Scenario 3. Simulated and observed PM₁₀ concentrations during two campaigns (May and November, 2017). (a) PM₁₀ in first campaign. (b) PM₁₀ in second campaign. (c) Scatter Plot of simulated and observed PM₁₀ data, 1^o Campaign. (d) Scatter Plot of simulated and observed PM₁₀ data, 2^o Campaign. (e) Boxplot of PM₁₀ concentrations in first campaign. (f) Boxplot of PM₁₀ concentrations in second campaign.

Table 5. Characteristics of simulated and observed concentrations during the two campaigns for NO₂ and PM₁₀. Simulated concentrations have been calculated using default emission factors from CETESB.

Statistics	1 ^o Campaign (NO ₂) (µg/m ³)		2 ^o Campaign (NO ₂) (µg/m ³)		1 ^o Campaign (PM ₁₀) (µg/m ³)		2 ^o Campaign (PM ₁₀) (µg/m ³)	
	Simulated	Observed	Simulated	Observed	Simulated	Observed	Simulated	Observed
Mean	27.5	28.9	23.9	19.8	22.2	19.7	27.3	24.7
Standard Error	1.5	4.2	1.5	2.2	1.1	1.9	1.3	1.8
Kurtosis	0.7	-0.5	-0.01	-0.3	0.1	1.0	0.4	1.2
Skewness	0.8	0.7	0.8	0.9	0.7	0.2	0.9	0.3
Range	23.6	59.8	23.3	34	12.7	24.4	14.5	16

Minimum	19.8	5.4	16	9.1	17.2	8.9	21.9	17.2
Maximum	42.8	65.2	39.3	43.1	29.9	33.3	36.5	33.2
Count	18	18	20	20	12	12	12	12
RMSE	15.2		9.0		5.2		4.5	

Based on adjusted vehicle emission factors for OSPM and default emissions for UBM (scenario 4)

Figure 10 and 11 and Table 6 show comparisons between simulated and observed street concentrations based on adjusted emission factors for OSPM and default emissions for UBM.

Figure 10. Scenario 4. Simulated and observed NO₂ street concentrations in two campaigns (July and November, 2017). Simulated data with adjusted emission factors for OSPM and default emissions for UBM. (a) NO₂ in first campaign. (b) NO₂ in second campaign. (c) Scatter Plot of

simulated and observed NO₂ data, 1° Campaign. (d) Scatter Plot of simulated and observed NO₂ data, 2° Campaign. (e) Boxplot of NO₂ concentrations in first campaign. (f) Boxplot of NO₂ concentrations in second campaign.

Figure 11. Scenario 4. Simulated and observed PM₁₀ street concentrations in two campaigns (May and November, 2017). Simulated data with adjusted emission factors for OSPM and default emissions for UBM. (a) PM₁₀ in first campaign. (b) PM₁₀ in second campaign. (c) Scatter Plot of simulated and observed PM₁₀ data, 1° Campaign. (d) Scatter Plot of simulated and observed PM₁₀ data, 2° Campaign. (e) Boxplot of PM₁₀ concentrations in first campaign. (f) Boxplot of PM₁₀ concentrations in second campaign.

Table 6. Characteristics of simulated and observed concentrations calculated and collected in the two proposed campaigns, NO₂ and PM₁₀. Simulated concentrations have been calculated using adjusted emissions for OSPM and default emissions for UBM.

Statistics	1° Campaign (NO ₂) (µg/m³)		2° Campaign (NO ₂) (µg/m³)		1° Campaign (PM ₁₀) (µg/m³)		2° Campaign (PM ₁₀) (µg/m³)	
	Simulated	Observed	Simulated	Observed	Simulated	Observed	Simulated	Observed
Mean	29.8	28.9	26	19.8	25.5	19.7	29.4	24.7

Standard Error	2.7	4.2	2.7	2.2	2.4	1.9	1.9	1.2
Kurtosis	0.3	-0.5	-0.3	-0.3	3.3	1.0	-1.2	1.2
Skewness	0.7	0.7	0.7	0.9	1.6	0.2	0.2	0.3
Range	42.3	59.8	42	34.0	31.1	24.4	20.1	16.0
Minimum	14	5.4	11.1	9.1	15.6	8.9	19.6	17.2
Maximum	56.3	65.2	53.0	43.1	46.7	33.3	39.7	33.2
Count	18	18	20	20	12	12	12	12
RMSE	15.5		13.5		9.3		7.6	

Figures 10 and 11 show similar correlations between observed and simulated concentrations as Figure 8 and 9. However, the simulated street concentrations are higher in scenario 4 compared to scenario 3. The reason is that the street concentration is dominated by the street contribution (difference between street concentrations and background concentrations). In scenario 4 we use adjusted emission factors for OSPM (default emissions for UBM) and hence the street contribution becomes much larger than in scenario 3 (default emission for OSPM and adjusted for UBM).

Figure 12 shows source apportionment of PM_{10} street concentrations in selected streets based on OSPM with adjusted emission factors and UBM with default emissions. It is seen that the urban background concentrations are a major contributor to PM_{10} street concentrations where about half originates from the regional background (Figure 7). Additionally, non-exhaust particles contribute much more than exhaust particles.

Figure 12. Source apportionment of PM_{10} street concentrations. PM_{10_back} is the urban background concentrations and $PM_{10}Exh$ is the exhaust part and $PM_{10}NonExh$ the non-exhaust part calculated with OSPM.

5. Discussion

It was possible to modify the emission module of OSPM to match all five vehicle types used in this study for Brazilian conditions. Air quality calculations were carried out with two sets of vehicle emission factors. Firstly, with official Brazilian default emissions factors based on CETESB and secondly, with adjusted higher emission factors based on calibration of emissions for UBM to fit the average measured street concentrations. The adjusted emission factors were close to adjusted emission factors proposed by Krecl et al. (2018). Non-exhaust PM₁₀ emissions are not included in the official Brazilian emission factors and hence non-exhaust emission factors were taken from the European COPERT IV emission model (EEA, 2016).

On average, modelled street concentrations showed relatively good agreement with measurements when using the default vehicle emission factors for OSPM and adjusted emission for UBM (scenario 3). This is also expected, as it is a result of the calibration of emissions for UBM to fit measured street concentrations. It is also expected that modelled street concentrations are overestimated in scenario 4 with adjusted emissions for OSPM and default for UBM since the street contribution becomes overestimated. In scenario 1 with default emissions for both UBM and OSPM, street concentrations were largely underestimated and in scenario 2 with adjusted emission for both UBM and OSPM, street concentrations were largely overestimated (data not shown). This indicates that the derived adjusted emission factors are too high although they are similar to those proposed by Krecl et al. (2018). However, based on the modelled street concentrations and observations in this study it is not possible to firmly conclude which vehicle emissions represent real emissions due to lack of measurements for validation of the modelled urban background concentrations and modelled regional background concentrations. It could also be that the regional background concentrations used as input to UBM are too low or too high. Regional background concentrations showed high concentrations of ozone (O₃) and particles (Figure 6) due to transport of anthropogenic emissions (industrial and transportation) as well as seasalt, organic matter and black carbon from the Fortaleza region (Flemming and Huijnen, 2011). In addition, low regional concentrations of NO_x (NO + NO₂) have been observed. It has not been possible to validate the modelled regional background concentrations against measurements as no air quality monitor station exists that represents regional background concentrations to Fortaleza. Hence, it is not possible to assess if regional background concentrations are under- or overestimated. The same is the case for modelled urban background concentrations as no air quality monitor station exists to represent urban background.

Contributions from industrial emissions were not analyzed. However, the contributions to Fortaleza from stationary sources are expected to be relatively low due to the location of the industrial sources outside the city and the dominant wind directions, reported by Silva et al., (2017).

Modelled contributions to PM₁₀ exhaust, PM₁₀ non-exhaust and urban background concentrations of PM₁₀ are comparable to other related PM contribution studies (Wang et al., 2010; Harrison et al., 2012; Lutz, 2013; Cyrus et al., 2014; and Holman et al., 2015). However, all in all, the default and also adjusted vehicle emissions are likely to remain uncertain. While non-exhaust emission are relative constant over the last years, in contrary exhaust emission have been substantially reduced by e.g. introduction of particle filters and improved engine technology. As a result, the relative contribution of non-exhaust emission compared to exhaust emissions has been increasing over time and today non-exhaust are dominating over exhaust emissions.

With relative errors in traffic flows of -22% to +34% in comparison with observations, as also mentioned in another reported study (Sousa, 2016). Under- and overestimations in modelled traffic flows are due to uncertainties in measurements of traffic data collected by CTAFOR and simulations with the travel demand model. Vehicle distributions were based on traffic counts and travel speed on travel demand modelling. In summary, the uncertainties on traffic data are assessed to be modest compared to other elements in the model chain.

Meteorological parameters estimated by the IFS model were consistent with the official Fortaleza meteorological data collection (FUNCEME, 2018). Parameters obtained, such as temperature, precipitation, wind speed and wind direction have low uncertainties whereas calculated parameters, such as mixing height have higher uncertainty although it was shown to directly correlate with NO₂ and PM₁₀ as mentioned in previous studies (Banerjee and Srivastava, 2009; Zheng et al., 2015; Seo et al., 2017).

Observed NO₂ street concentrations showed higher values in the first campaign when compared to the second campaign and the same was seen in the simulated NO₂ concentrations (Figure 7 and Table 4). One of the potential reasons for higher NO₂ concentrations in July was the higher concentrations of ozone (O₃) leading to formation of NO₂ through reaction with NO during this period (Fowler et al., 1998, 1999). Lower mixing height was observed in the first campaign, that

will slightly increase NO₂ concentrations, as mentioned in previous studies (Dieudonné et al., 2013).

Observed PM₁₀ concentrations showed higher values in the second campaign also reproduced in the simulated PM₁₀ concentrations (Figure 8 and Table 4). One of the potential reasons for this increase in concentrations could be the effect of temperatures on gas-to-particle conversion (Strader et al., 1999; Sheehan and Bowman, 2001). Furthermore, lower temperatures in the rainy season could be observed in the meteorological parameters leading to lower mixing heights (convective boundary layer depth), also confirmed in a previous study (Pal et al., 2012). Lower mixing heights will, other things equal, result in higher concentrations of atmospheric particles (Tiwari et al., 2014).

In addition, PM₁₀ measurements showed outlier concentrations at data collection points 1 and 9 (lower concentrations). These outliers could be observed due to strong rainfall during the data collection days in the rainy season in May. Many studies on precipitation washout of PM in the air are reported with comparison between particulate matter concentrations under conditions of precipitation and non-precipitation (Akyuz et al., 2009; Barmpadimos et al., 2011). However, OSPM simulated concentrations overestimate PM₁₀ concentrations under such conditions as washout of PM is not modelled in OSPM. Outliers in simulations at data collection points 7 and 11 (higher concentrations) were seen due to low calculated mixing height during most of the simulated day, what can differ from the actual mixing height. However, in general simulated PM₁₀ concentrations followed the variation in observed concentrations.

There are also limitations concerning measurements. Firstly, the locations, number and days of data collected were relatively low due to lack of budget and free public data. Secondly, the measurements of NO₂ may be uncertain as it was based on passive sampler methodology. Thirdly, the absence of air quality monitoring data for regional and urban background concentrations made it impossible to validate modelled regional background concentrations and modelled urban background concentrations.

6. Conclusion

This study presents the comparison between modelled street concentrations and measurements of NO₂ and PM₁₀ at selected streets in Fortaleza. The wider aim is to develop a model system for decision-support estimating the effects of potential traffic-related measures such as, introduction of a Low Emission Zones.

In one of the tested model set-up scenarios, Scenario 3 with adjusted emissions for UBM based on calibration and default emissions for OSPM, the system agreed well with observations on average. In Scenario 4 with default emissions for UBM and adjusted emissions for OSPM, the model overestimated observations as expected. However, based on the modelled street concentrations and observations it is not possible to conclude which vehicle emissions represent the most realistic emissions due to a lack of measurements at urban background and regional background levels. Hence, it is not possible to assess whether or not regional and urban background concentrations are under- or overestimated. This illustrates the difficulties to disentangle the uncertainties on the different contributions (regional, urban background and street) when observations only are available for street concentrations. In a future research concentration data should be collected simultaneously at kerbside, urban background and regional background location. Such data could help to further estimate the contributions of vehicle emissions on many roads with different traffic volumes and vehicle type composition in street concentrations.

However, given the scenario calculations in relation to observations there are indications that official Brazilian default emissions are too low but also that the adjusted emissions derived in this project are too high. The findings in this research could contribute to strengthening the management of traffic-related air pollution in Fortaleza. Furthermore, based on the findings of this study, the local governments of other developed cities in Brazil, such as São Paulo, Rio de Janeiro, and Curitiba, could take advantage of the developed methodology to evaluate the effect of potential measures to reduce air pollution.

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