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Development and Evaluation of a Comprehensive Atmospheric Emission Inventory for Air Quality Modeling in the Megacity of Bogotá

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Abstract: We built an emission inventory (EI) for the megacity of Bogotá, Colombia for 2012, which for the first time augments traditional industrial and mobile sources by including commercial sources, biogenic sources, and resuspended dust. We characterized the methodologies for estimating each source annually, and allocated the sources to hourly and 1 km^2 spatial resolution for use as inputs for air quality modeling purposes. A resuspended particulate matter (RPM) emission estimate was developed using the first measurements of road dust loadings and silt content for the city. Results show that mobile sources dominate emissions of CO_2 (80%), CO (99%), VOC (68%), NO_x (95%), and SO_2 (85%). However, the newly estimated RPM comprises 90% of total PM_{10} emissions, which are at least onefold larger than the PM_{10} emissions from combustion processes. The 2012 EI was implemented in a chemical transport model (CTM) in order to understand the pollutants' fate and transport. Model evaluation was conducted against observations from the city's air quality monitoring network in two different periods. Modeling results for O_3 concentrations showed a good agreement, with mean fractional bias (MFB) of +11%, and a mean fractional error (MFE) of +35% with observations, but simulated PM_{10} concentrations were strongly biased high (MFB +57%, MFE +68%), which was likely due to RPM emissions being overestimated. NO_x, CO, and SO_2 were also biased high by the model, which was probably due to emissions not reflecting current fleet conditions. Future work aims to revise emission factors for mobile sources, which are the main sources of pollutants to the atmosphere.

Keywords: emissions; Bogotá; mobile sources; resuspended dust; air quality model

1. Introduction

Air quality is of particular concern in developing countries, where a large portion of the population is exposed to high levels of pollutants [1]. In Latin America, it's estimated that over 100 million people are exposed to pollution levels, exceeding the World Health Organization (WHO) guidelines [2]. A recent report about the cost of environmental degradation in Colombia indicated that air quality in urban centers is responsible for 10,527 deaths and 67.8 million hospital visits, which together represent 5.1 billion USD (1.93% of Colombia's gross domestic product (GDP) for 2015) [3].

Bogotá, Colombia's largest city, is located in a plateau at approximately 2600 meters above sea level with an area of 1587 km^2. The region has a bimodal annual rain cycle, with increased rain in April and October (Figure S1). The city is home to approximately eight million people, with 1.8 million vehicles reported in 2012. Since 1997, the city has operated an air quality monitoring network; its Spanish acronym is RMCAB. Currently, RMCAB is composed of 13 sites measuring pollutants including PM_{10}, $PM_{2.5}$, CO, NO_x, SO_2, and O_3, along with the meteorological variables of wind speed and direction, temperature, solar radiation, precipitation, and relative humidity (Figure 1).

Most of the pollutants are generally below the Colombian air quality standards, but PM_{10} exceedance is still common during the first three months and the last two months of each year, especially in the south and southwest regions of the city [4]. Nevertheless, PM_{10} concentrations have significantly declined in the 2002–2012 period. The months of June, July, and August display a drop in ambient air concentrations of all pollutants, due to increased wind speed caused by the change of the Intertropical Convergence Zone (Figure S1) [4].

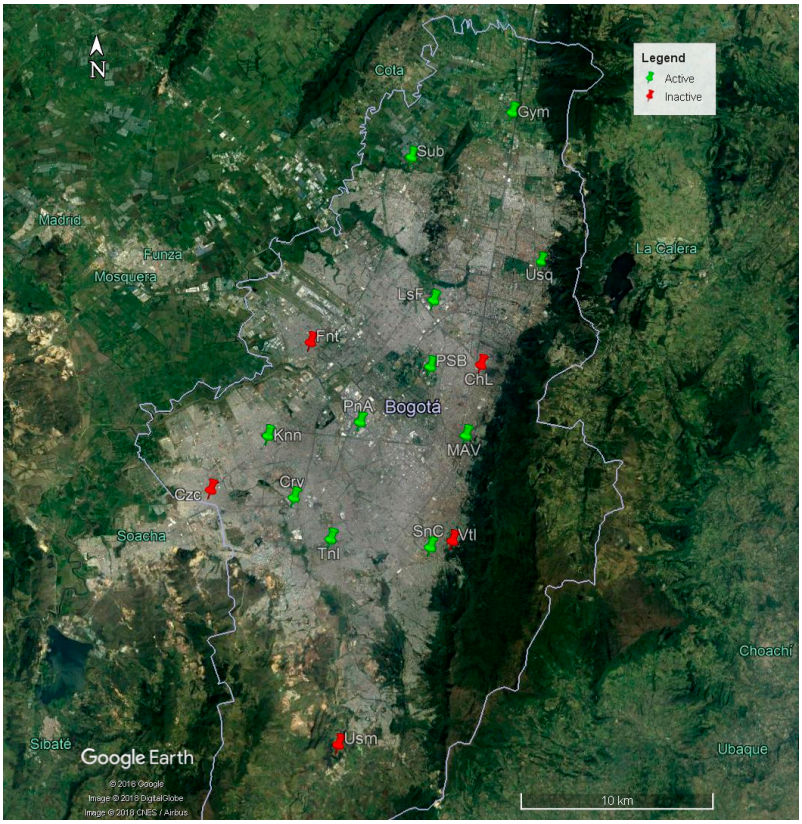

Figure 1. Map of districts in Bogotá and location of the sites of the Spanish air quality monitoring network, known as RMCAB (Gym–Guaymaral, Sub-Suba, Usq-Usaquen, LsF-las Ferias, Fon-Fontibon, PSB-Parque Simon Bolivar, ChL-Chico, MAV-Ministerio de Ambiente, PnA-Puente Aranda, Knn-Kennedy, Czc-Cazuca, Crv-Carvajal, Tnl-Tunal, Usm-Usme, Vti-Vitelma).

Emission inventories (EI) are a key component in the air quality management of urban areas, but they have been pointed out as the most uncertain input in the air quality modeling process [5]. Emissions are generally estimated with the use of emission factors (EFs) and activity data, which are both subjected to large uncertainties. While activity data are generally obtained from local information, EFs are usually estimated with emission models such as the Motor Vehicle Emission Simulator (MOVES) [6] or the International Vehicle Emissions (IVE) [7], or acquired from international databases, such as the US-EPA AP-42 document [8], the Core Inventory AIR emissions CORINAIR database [9], and the Intergovernmental Panel on Climate Change IPCC [10], among others.

Several projects that have aimed to construct EIs for the main urban centers in South America have been conducted, such as Bogotá (Colombia), Medellin (Colombia), Santiago (Chile), Buenos Aires (Argentina), and Sao Paulo (Brazil) [11–16], and consolidate a regional inventory combining local and global inventories (such as RETRO and EDGAR) [17,18].

EIs in Bogotá have traditionally included emissions for industrial and mobile sources. A compendium of studies is included in the Supplementary material (Table S1). An analysis of emission estimates prior to 2010 can be found in Pachón [19]. Briefly, emissions from point sources were consistent among different inventories due to the use of common databases of industrial activity and fuel consumption. Emission factors have been compiled from local measurements [20] and international references [8]. On the other hand, emissions from mobile sources have displayed large variability, especially for PM_{10}. This is mainly due to the use of different emission factors obtained from local measurements [21], international databases (Copert, Corinair, AP-42), and emission models such as IVE [7]. In general, mobile sources contribute more than 90% of CO and NO_x emissions, while PM_{10} from combustion activities can be almost equally emitted by mobile (46%) and point sources (54%). CO and NO_x emission estimates show a consistent and growing trend throughout the years, which is explained by the dramatic increase in vehicles from approximately 600,000 in 2002 to 1.7 million in 2012 [22].

Resuspended dust has been identified as a major component of PM_{10} in Bogotá, surpassing PM_{10} emissions from combustion sources by at least one order of magnitude [23,24]. This situation is also found in other cities [11,25,26]. Consistently, PM_{10} chemical composition studies conducted in Bogotá have observed dust fractions between 25–45% [27–29].

This project constructs a comprehensive emission inventory for Bogotá including many sources: mobile (top-down and bottom-up), industrial, commercial (restaurants, grilling, and street vendors), biogenic, gas stations and gas storage tanks, and resuspended dust (from paved and unpaved roads, material extraction, and building and road construction). This manuscript describes the process and results of constructing this emission inventory estimated for 2012 and its use in an air quality modeling platform implemented in the city. A preliminary modeling evaluation is also discussed.

2. Material and Methods

2.1. Mobile Sources (On-Road)

Mobile source emissions were calculated using two approaches: top-down and bottom-up, as described in Carmona et al. [30]. Briefly, in the top-down approach (Equation (1)), fleet size and vehicular activity were obtained from annual fleet registrations available at the city's government, while the emission factors were collected from different studies previously conducted in the city.

$$E_i = \sum_{j=0}^{n} EF_{ij} \times AF_j \times N_j \quad (1)$$

E_i (g/year): total emissions of pollutant i ($PM_{2.5}$, CO, CO_2, NO_x, SO_x, VOCs)
EF_{ij} (g/km): emission factor for pollutant i for vehicular category j
AF_j (km/year): average vehicle activity factor for vehicular category j
N_j (#): number of vehicles for category j

In the bottom-up approach (Equation (2)), vehicular activity was obtained from traffic counts available at 46 points throughout the city (Figure 2). Emission factors were the same as the ones used in the top-down approach.

$$E_i = \sum_{j=0}^{n} EF_{ij} \times F_{jk} \times L_k \quad (2)$$

E_i (g/h): total emissions of pollutant i ($PM_{2.5}$, CO, CO_2, NO_x, SO_x, VOCs)

EF_{ij} (g/km): emission factor for pollutant i for vehicular category j
F_{jk} (#/h): vehicular flux for category j in the road link k
L_k (km): length of road link category k

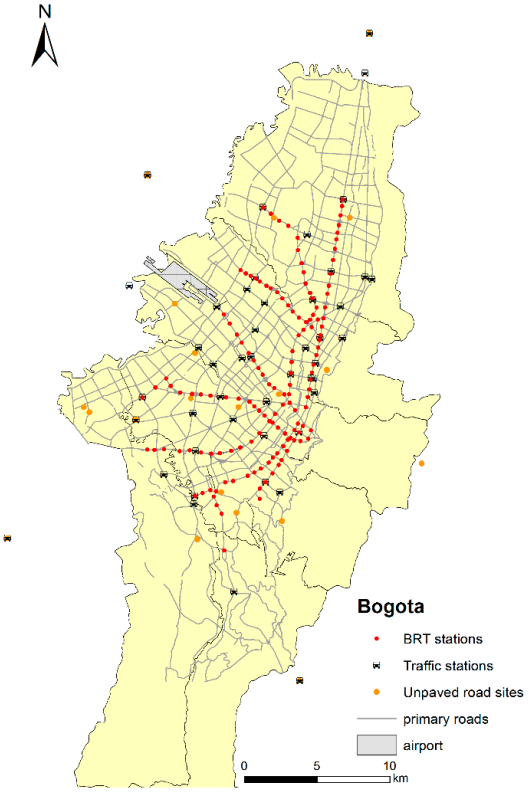

Figure 2. Traffic monitoring stations in Bogotá. Symbols outside the city's perimeter correspond to tolls. Sites for paved road dust sampling coincide with traffic monitoring sites. Sites for unpaved road dust sampling are distributed throughout the city. BRT is the Transmilenio bus system in Bogotá.

2.2. Point Sources (Industrial, Commercial)

The EI includes stationary source emissions from combustion processes, including boilers, heaters, and ovens used in industrial activities. For 2012, local databases reported 975 industries with 1681 stacks or emission points. Fuel consumption was distributed between natural gas (72%), coal (12%), fuel–oil (8%), LPG (4%), and others (4%). Emissions were estimated without emission controls (Equation (3)), due to the lack of information about control equipment. Emission factors were adopted from the AP-42 database [8] and local studies [16].

$$E_i = \sum_{j=0}^{n} EF_{ij} \times AF_j \times N_j \quad (3)$$

E_i (g/year): total emissions of pollutant i (PM_{10}, CO, CO_2, NO_x, SO_x, VOCs)
EF_{ij} (g/kg or g/m^3): emission factor for pollutant i for fuel j
AF_j (kg/year or m^3/year): fuel j consumption
N_j: number of point sources that consume fuel j

Commercial activities were also included as stationary emission sources in 2012 [31]. Briefly, a database of commercial sources was provided by the Bogota's environmental authority (SDA), with 843 establishments (51% restaurants, 44% grilling, and 5% street vendors), which is expected

to underestimate (at least four times) the real number of commercial sources. Fuels used in commercial activities are charcoal (75%), wood (13%), and a combination of both (12%). EFs for PM_{10}, NO_x, SO_2, CO, CO_2, and VOCs were given per amount of fuel consumed, with the exception that the EFs for charcoal are given as the mass of pollutant per mass of fuel and meat consumed (e.g., g CO/(kg fuel + kg meat)) [32]. EFs for wood combustion were obtained from the AP-42 database [8].

2.3. Gas Stations and Gas Storage Tanks

VOC emissions were quantified for gas storage and distribution. Local databases accounted for 464 gas stations distributed throughout the city and three gas storage facilities located at the industrial zone in Bogotá. Activities prone to emit VOCs in gas stations are storage, distribution, the filling of underground containers, and spills. Storage emissions were estimated using the TANKS software [33]. For the other activities, EFs were obtained from the AP-42 database [8]. Information and conditions to execute TANKS are described in Fajardo et al. [31]. Briefly, information on gas stations was obtained from SDA local records, including the location in the city, number of tanks and capacity, type of fuels distributed, and fuel monthly sales. Information on tank material was obtained by consulting with tank suppliers. Meteorological information (i.e., temperature, atmospheric pressure, wind velocity) to run the model was obtained from RMCAB.

2.4. Biogenic Emissions

The estimation of biogenic emissions (i.e., isoprene, monoterpene, and other VOCs) was conducted using two models: the Global Biosphere Emissions and Interactions System (GloBEIS) and the Model of Emissions of Gases and Aerosols from Nature (MEGAN). The MEGAN's set-up and results are described in Nedbor-Gross et al. [34]. The GloBEIS model that was implemented here is similar to previous work [35]. Bogotá's vegetation density is significantly low in comparison to other Latin American cities. However, Bogotá is surrounded by dense forest at high and low altitudes that permanently emit VOCs, and can potentially be transported into the city.

The main inputs for the model were meteorological information (i.e., hourly temperature, relative humidity, cloudiness, and solar radiation) from Weather Research and Forecasting (WRF) modeling [36], land use (i.e., fraction of vegetation coverage) from local databases [37,38], vegetation type from Bogotá's botanical garden [39], and emission factors (default from GloBEIS). Vegetation types were processed to apply emission factors from GloBEIS, CORINAIR, EPA AP-42, and the European Environmental Agency (EEA) [9].

Urban and rural land cover layers were adjusted to the modeling domain, and the fraction of vegetation within each cell was estimated. Emissions were estimated as the product of the covered area and the emission factors (g/m^2). One typical day was modeled during a dry season (February) and one day during a rainy season (October) in 2012.

2.5. Resuspended Dust

Resuspended dust was estimated from three major sources: roads (paved and unpaved), building and road construction, and material (sands, clays) extraction. In this manuscript, we describe resuspended particulate matter (RPM) estimations only from roads, given the significance of emission from this source. The methods of the AP-42 database (Chapter 13.2) were applied for this purpose. Emissions are the product of an emission factor (g PM_{10}/VKT) multiplied by the vehicular activity (VKT). This method required conducting an extensive field campaign to quantify silt loadings, and use them to estimate EFs for paved (Equation (4)) and unpaved roads (Equations (5) and (6)).

$$EF = q \times SL^{0.67} \times W^{0.85} \quad (4)$$

where EF is the emission factor for paved roads (g/VKT), q is a particle size specific multiplier, SL is slit loading (g/m^2), and W is the average weight of vehicles (ton).

For vehicles traveling on unpaved surfaces at industrial sites:

$$\mathrm{EF} = g\left(\frac{s}{12}\right)^{a}\left(\frac{W}{3}\right)^{b} \quad (5)$$

and, for vehicles traveling on unpaved publicly accessible roads (generally light vehicles):

$$\mathrm{EF} = \frac{g\left(\frac{s}{12}\right)^{a}\left(\frac{v}{30}\right)^{d}}{\left(\frac{M}{0.5}\right)^{c}} - C \quad (6)$$

where EF is the emission factor for unpaved roads (g/VKT); g, a, b, c and d are empirical constants; s is the surface material silt content (%); W is the mean vehicle weight; v is the mean vehicle speed; M is the surface material moisture content, and C is the emission factor for brake and tire wear.

Field work was performed in 30 points on paved roads, and 15 points on unpaved roads (Figure 2). The points on paved roads were collocated with some of the traffic count sites. For unpaved roads, it was necessary to record traffic at the same time the sampling was conducted. Field samples were transported to the laboratory for humidity content and granulometry analysis. Silt loadings (SL) and silt content (s) were determined for each sample.

2.6. Spatial and Temporal Allocation

For air quality modeling purposes, emissions need to be processed on a temporal and spatial basis. A modeling domain was set up on a grid size of 55 × 55 km, with 1 × 1 km cells for two modeling periods: February and October 2012. Emissions for all of the sectors were distributed in the cells within Bogotá's political boundaries using different surrogates. Additionally, emissions were estimated under different temporal patterns: for all of the sectors, hourly disaggregation was conducted, and for some sectors, daily (weekday, weekend) or monthly disaggregation was also performed (Table 1).

Table 1. Temporal allocation factors and spatial surrogates used for emission disaggregation.

Sector	Temporal Disaggregation *					Temporal Allocation Factor (TAF)	Spatial Allocation Factor (SAF)
	H	T	W	N	M		
Mobile (bottom-up)	X		X	X		Traffic counts from 40 stations two days a week	Traffic volumes interpolated using Thiessen polygons
Point (Industrial)	X	X			X	Working schedule (24/7)	Geolocation of point source
Point (commercial)	X	X			X	Working schedule (peak and non-peak hours)	Geolocation of commercial source
Gas storage (tanks)	X	X			X	Daily temperature profile from observations	Geolocation of storage tanks
Gas distribution	X	X			X	Fuel sales and traffic allocation factors	Geolocation of gas stations
Biogenic	X	X				Daily temperature profile from meteorological model	Land cover maps
Resuspended particle matter (RPM) from roads	X		X	X		Traffic counts from 40 stations and six tolls	Dust loadings interpolated using inverse distance weighting

* H: hourly, T: typical day, W: weekday, N: weekend, M: monthly.

2.7. Chemical Speciation

Air quality modeling requires the chemical speciation of emissions in addition to their spatial and temporal disaggregation. Chemical speciation for non-methane organic compounds (NMOC) and particulate matter (PM_{10}) was conducted for point and mobile sources following the US Environmental Protection Agency EPA methodology [40]. Chemical profiles from international databases and

publications including EMEP/EEA [9], and EPA's SPECIATE database [41] were applied for the industrial sector, vehicular category, and point sources [42]. As local profiles [41] become available, they can be incorporated in future modeling exercises.

The results were subsequently transformed to the categorization of defined species for modeling according to the chemical mechanism Carbon Bond 05 [42]. For PM_{10}, the species considered were elemental carbon (EC), organic carbon (OC), nitrates, sulfates, ammonia, and PM other. PM other is an aggregated fine mode of primary PM. For NMOC, speciation was conducted using the SpecDB tool [43].

2.8. Modeling Set-Up

The full description of the modeling system is found in Nedbor-Gross et al. [34]. Briefly, meteorological modeling has been performed for Bogotá using the Weather Research and Forecasting (WRF) model [44]. Four modeling domains have been set up centered over Bogotá (4.5° N, 74.1° W) and using a 3-to-1 parent to child nesting ratio, similar to Kumar et al. [45]. Meteorological simulations have been performed for two 25-day periods in 2012—one for February and one for October—coinciding with high pollution episodes that occurred for a dry season and a rainy season [36].

According to the RMCAB information considering all of the stations with available data for the identified periods, 8 h-ozone maximum values presented a wider variability in the observed period corresponding to the dry season, ranging from nearly 10 ppbv to 70 ppbv; for the wet period, these values are more stable, ranging from 20 ppbv to 60 ppbv. On the other hand, PM_{10} 24-h average concentrations at standard conditions varied for the dry season period from 70 $\mu g/m^3$ to 160 $\mu g/m^3$; meanwhile, the wet period developed smoother daily changes, with concentrations scaling from about 40 $\mu g/m^3$ to 120 $\mu g/m^3$. For both of the selected periods, the spatial distribution of observed O_3 tended to show greater mixing ratio values at the east and northeast of the city, following the mountain range (Figure 1). Measurements of PM_{10} concentration indicated that the higher values are mainly located at the southwest side of the city, where an important aspect of industrial and heavy diesel vehicle traffic is present.

Air quality modeling simulations were performed using the Community Multiscale Air Quality (CMAQ v5.0.1) model [46], using the same periods and domain configuration as the meteorological simulations. The model simulates aerosols using ISORROPIA (v2.1) for inorganic thermodynamic partitioning, and AERO6 treatment for organic aerosols [47]. The full description of the modeling system is found in Nedbor-Gross et al. [34]. The anthropogenic emissions for the outer domains were from the Emissions Database for Global Atmospheric Research (EDGARv3). Biogenic emissions were developed using the Model of Emissions, Gases, and Aerosols from Nature—MEGANv2 [48]. The local emissions inventories discussed previously were used to drive the innermost domain for the 2012 simulations. The CMAQ simulations were evaluated for pollutants of concern, including PM_{10} and O_3, using model performance metrics [49,50].

For an initial assessment of modeling performance using the emissions inventory, we evaluated the bias and error for the criteria pollutants for the combined simulation periods. The evaluation included qualitative analysis using graphics and examines the mean fractional bias (MFB) and error (MFE), which are considered the best indicators of model performance [49,50]. For each pollutant of concern, there is a criteria value and a goal value for the MFB and MFE, where meeting the criteria indicates that the model performance is suitable for regulatory applications, and meeting the goal indicates that the model is performance is ideal.

3. Results and Discussions

3.1. Mobile Sources (On-Road)

Local transport registrations for 2012 show approximately 1.8 million vehicles in Bogotá, from which 75% are passenger vehicles and pick-up trucks, 20% are motorcycles, 3% are taxis, 1.2% are

heavy trucks, and less than 1% are buses. In contrast, most of the motorized trips in the city are made using public transportation (30%), private vehicles (11%), taxis (3%), and motorcycles (2%). Non-motorized trips (pedestrian and bicycle) constitute approximately 50% of travel in the city [51]. This distribution of fleet and trips implies that there is an intensive use of passenger vehicles and motorcycles, with a subsequent impact on emissions.

For CO, CO_2, NO_x, VOC, and SO_2 emissions, the contribution from private vehicles constitute between 40–70% of mobile emissions. This large contribution is due to the rapid increase in the number of automobiles and pick-up trucks, from approximately 400,000 in 2002 to more than 1 million in 2012. Motorcycles (M) have also shown this growth trend (approximately 35,000 units in 2002 to 350,000 in 2012), which adds significant emissions of CO and VOCs. For $PM_{2.5}$, the largest emissions are associated with traditional public transportation and freight vehicles that operate with diesel engines (Figure 3). The Bus Rapid Transit (BRT) Transmilenio (TM) generates less $PM_{2.5}$ emissions than other buses. Details of the results for mobile sources can be found in Carmona et al. [30].

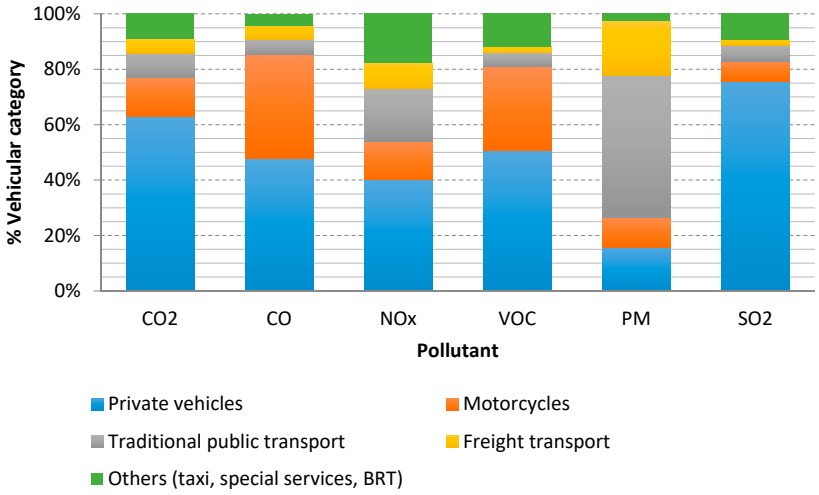

Figure 3. Mobile source emissions by category (top-down approach).

In comparison to other emission sources (e.g., industrial, commerce, biogenic), vehicles contribute 99% of the CO, 80% of the CO_2, 68% of the VOCs, 95% of the NO_x, and 85% of the SO_2, which is similar to other South American cities [52,53]. The contribution to PM_{10} emissions is 58% from only combustion processes (Table 2), but non-exhaust PM_{10} is much larger than any other source.

3.2. Point Sources (Industrial, Commercial)

PM_{10} emissions from industrial sources are comparable to those from mobile sources. The contribution of the gaseous pollutants coming from industrial sources is much less significant. Emissions from commercial sources constitute a small percentage of industrial emissions (4% NO_x, 11% SO_2, 12% PM_{10}), with the exception of CO, which exceeds industrial emissions by 337% (Table 2). This is due to the incomplete combustion conditions that are prevalent in restaurants, grilling, and street vendors.

Table 2. Comparison of emissions for point and mobiles sources (ton/year).

Pollutant	Mobile	Industrial	Commercial	Total
CO	661,495	674	2276	664,445
CO_2	7,379,087	862,810	32,415	8,274,312
PM_{10}	1521	992	115	2628
NO_x	51,777	1612	60	53,450
SO_2	11,982	1540	171	13,693

3.3. Gas Storage and Distribution

Fuel sales reported for the city were approximately 450 million gallons in 2012 (56% regular gasoline, 40% diesel, 4% premium gasoline). VOC emissions are associated with gas storage (three tanks) and gas distribution (464 stations). The gas storage represents 77% of VOC emissions, while the remaining 23% is associated with gas stations. The filling of underground tanks and gas distribution represent the activities with largest emissions. Spills are relatively low, because of the economic costs implied, should they occur. The analysis of fuels indicates that 67% of VOC emissions come from gasoline, and 33% come from diesel.

3.4. Biogenic

VOCs from biogenic sources were relatively low compared to anthropogenic emissions (Table 3). This can be explained by several factors: (i) at least 7% of the modeling domain had zero vegetation cover (i.e., zero biogenic emissions); (ii) vegetation within the urban area is located at elevated heights (2600 masl to 3200 masl) and low temperatures (15 $\pm$ 5 °C); (iii) meteorological patterns likely advect pollutants away from the city [36]; (iv) Bogotá's cloudiness is constant throughout the year, affecting direct solar radiation. Additionally, the EFs for local vegetation species used in MEGAN and BEIS have not been thoroughly validated in Colombia, which adds an element of uncertainty that should be addressed in future studies. The typical diurnal pattern for biogenic emissions indicate an increase of emissions during the daytime, with maximum emissions at midday.

Table 3. VOCs emissions from different sectors.

Source	Emission (ton/year)
Mobile	71,378
Point (ind + com)	2970 (2875 + 94)
Biogenic	2518
Gas storage and distribution	23,786
Waste facilities [&]	3127
Total	103,779

[&] PTAR: Bogotá's wastewater treatment plant El Salitre; RSDJ: Bogotá's solid waste landfill Doña Juana from [54].

Analysis of VOC emissions indicate that most of the VOCs are anthropogenic (only 2.5% biogenic), with 92% directly (tail pipe) or indirectly (gas storage and distribution) emitted by vehicles (Table 3). The VOC contribution from point sources and waste facilities is approximately the same (3%).

3.5. Resuspended Dust

Dust loadings (mass per area) estimated through field work according to AP-42 methods [8] in paved roads were very heterogeneous in the city, with a range 0.1–10.6 g/m^2. The largest values were observed towards Bogotá's surroundings given poor road conditions and the presence of unpaved roads and quarrying sites. The silt contents observed in unpaved roads were also diverse (range 3.0–30%) (Figure 4).

Estimates of RPM from paved and unpaved roads (Table 4) were significantly larger than the PM_{10} emitted from combustion processes (Table 2). This result highlights the magnitude of a fraction of PM_{10} that has not been considered in previous inventories, and has a potentially large impact in ambient PM_{10} concentrations.

In 2012, Bogotá had 7305 km of paved roads and 1300 km of unpaved roads. The contribution of unpaved roads to RPM is significantly larger than paved roads when emissions are expressed per km of road (Table 4). In effect, unpaved roads emit approximately five times more PM_{10} and twice as much $PM_{2.5}$ than paved roads. This result is consistent with the larger emission factors in unpaved roads (average 194 g/VKT PM_{10} and 19.4 g/VKT $PM_{2.5}$) than in paved roads (average 1.68 g/VKT

PM_{10} and 0.56 g/VKT $PM_{2.5}$). It is apparent that the pavement of roads can have a large effect on reducing RPM in the city.

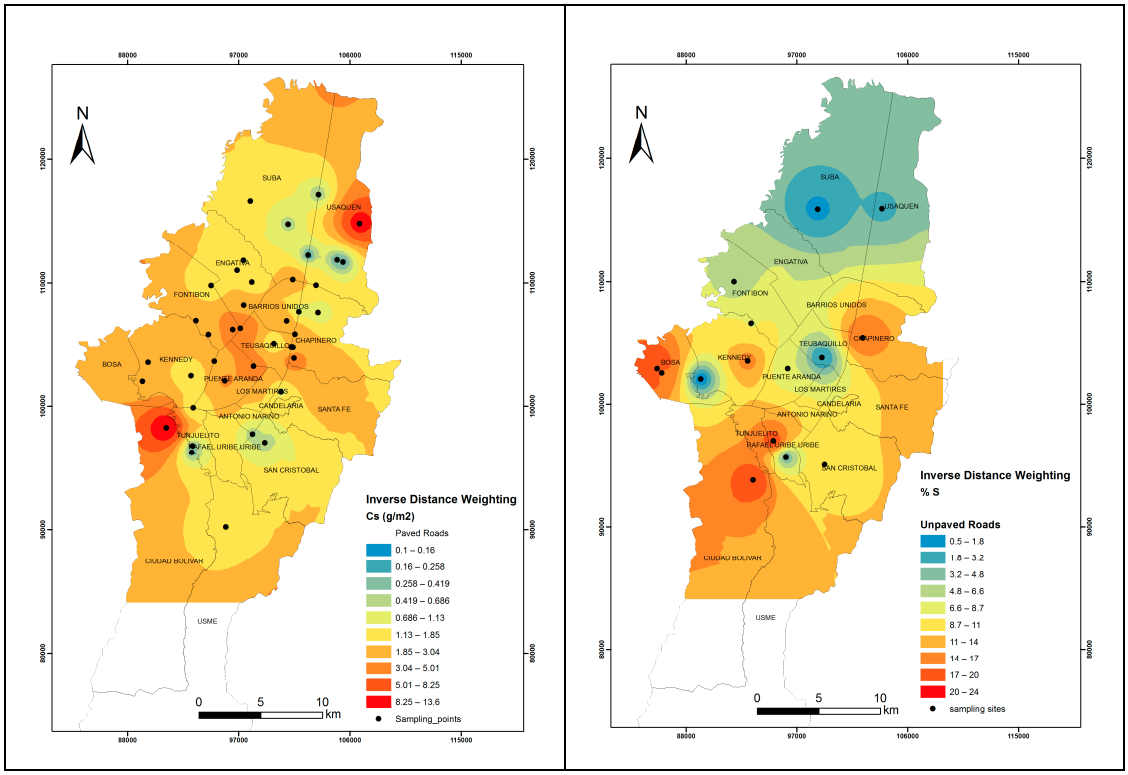

Figure 4. Interpolation of dust loadings in paved roads (**Left**) and silt contents in unpaved roads (**Right**).

Table 4. Resuspended particulate matter (RPM) emissions from paved and unpaved roads.

Emission (ton/year)	PM_{10}	$PM_{2.5}$	PM_{10} (ton/year/km)	$PM_{2.5}$ (ton/year/km)
Paved roads	32,847	10,786	4.5	1.5
Unpaved roads	32,680	3268	25.1	2.5
Total	65,527	14,054		

3.6. Temporal and Spatial Disaggregation of Emissions

$PM_{2.5}$ emissions from mobile sources are ubiquitous in the modeling domain, with large emission levels and "hot spots" in areas with heavy-duty fleet predominance, such as industrial and commercial zones, food transfer stations, and construction sites (Figure 5a).

Unlike mobile sources, PM_{10} from industrial sources is more localized towards the south and western parts of the city, where fuels are burned in ovens, heaters, and industrial processes (Figure 5b). PM_{10} from commercial sources is spread throughout the city, since restaurants and street vendors are present everywhere (Figure 5c).

Spatial plots for CO, NO_x, SO_2, and VOCs for mobile and stationary sources are available in the Supplementary Information (Figures S2 and S3).

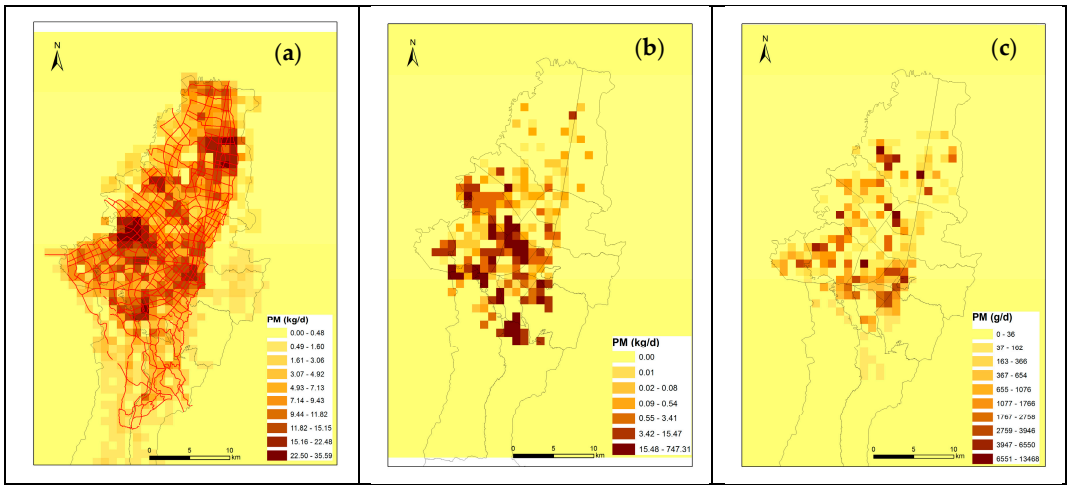

Figure 5. Spatial distribution of particulate matter (PM) emissions from (**a**) mobile, (**b**) industrial, and (**c**) commercial sources. Scales are different given the level of emissions. PM in mobile sources represent $PM_{2.5}$, while industrial and commercial sources include PM_{10}.

RPM emissions from paved and unpaved roads are distributed in a different way. The emissions from paved roads tend to follow traffic distribution as well as mobile source emissions (Figure 6a), but they are also affected by dust loadings (Figure 4). RPM emissions are particularly large towards the south and western parts of the city (Figure 6a) for several reasons: (i) heavy-duty vehicle transit is dense, because roads in this area of the city exchange freight with the rest of the country; (ii) heavily populated areas are observed; (iii) dust loadings observed in field campaigns are large in these areas; (iv) unpaved roads and roads without proper sidewalks in the area contribute to dust in the streets; and (v) the cleaning and sweeping of roads is practically non-existent. The emissions from unpaved roads are denser towards the city's surroundings, where most of the roads are still unpaved (Figure 6b).

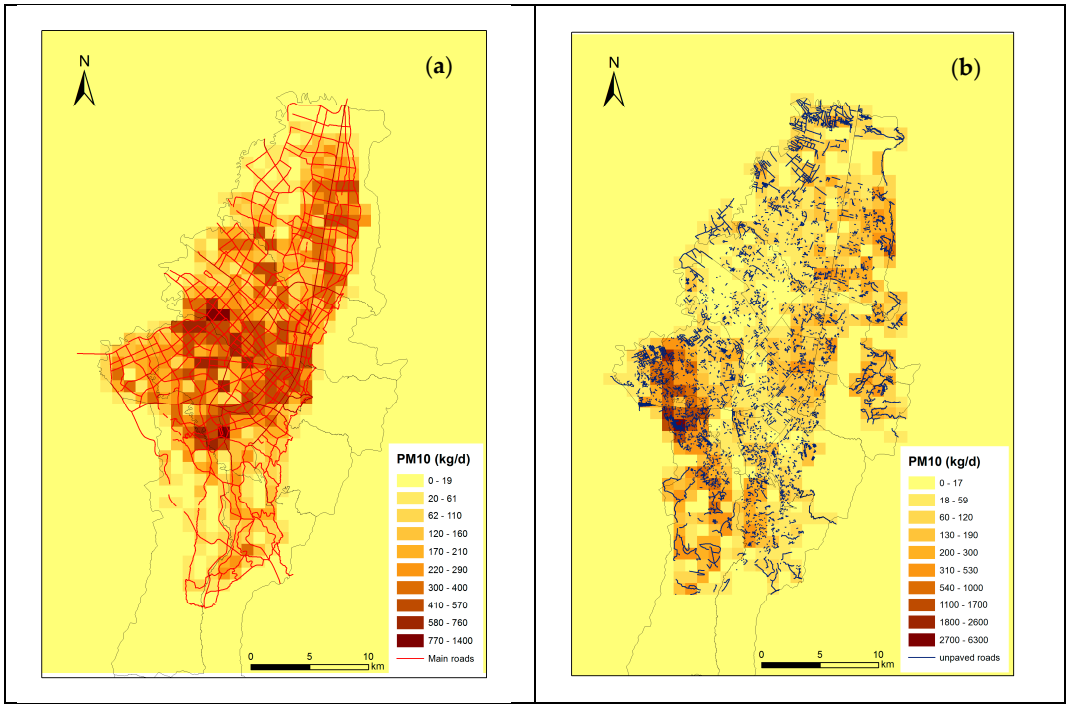

Figure 6. Spatial distribution of RPM emissions from (**a**) paved roads and (**b**) unpaved roads.

VOC emissions from gas storage and distribution are particularly large at the industrial zone, where storage tanks are located. The rest of the emissions correspond to gas stations distributed throughout the city (Figure S4a). The VOCs from biogenic sources are large in areas with vegetation cover (Figure S4b). Given that both anthropogenic and biogenic emissions are represented in the same scale, it is apparent how VOCs from gas storage dominate over biogenic VOCs.

3.7. Chemical Speciation

$PM_{2.5}$ speciation in Bogotá is dominated by carbonaceous material (EC, OC), followed by PM other and secondary material (Figure 7). For mobile sources, EC represents about 70% of the $PM_{2.5}$ mass, which is mainly associated with diesel engines operating in heavy-duty trucks and buses. Figure S5 in the Supplementary Information displays the $PM_{2.5}$ chemical composition by vehicular category. In point sources, OC comprises a larger mass fraction (50%) of the inventory (Figure 7).

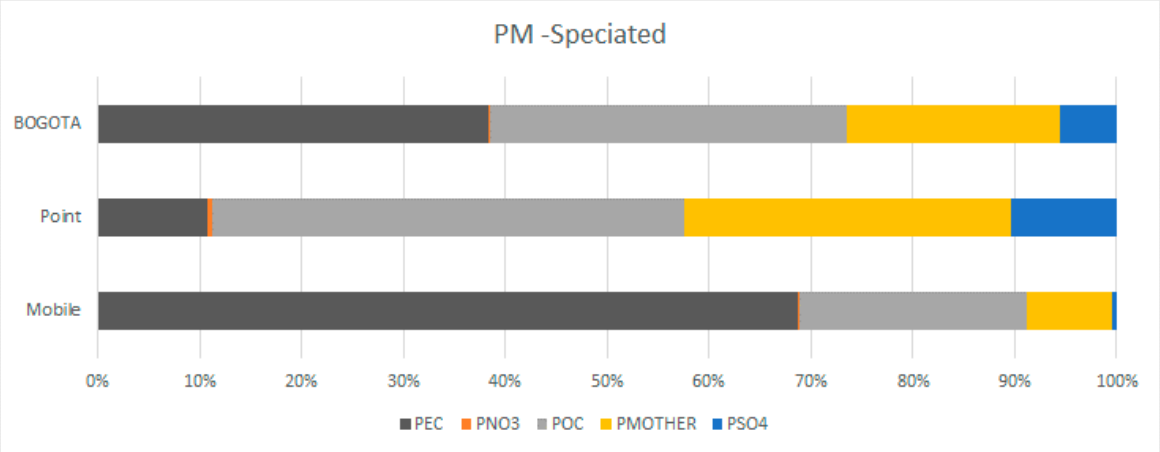

Figure 7. Comparison of PM modeling speciation by mobile and stationary sources. PM in mobile sources represent $PM_{2.5}$, while industrial and commercial sources include PM_{10}.

For point sources, emissions of PM_{10} are composed mostly of organic carbon (OC), PM other, elemental carbon (EC), sulfate (SO_4), and nitrate (NO_3) (Figure S6 in the Supplementary Information).

3.8. Evaluation of Modeling Results

The model evaluation for the combined dry–wet periods shows that PM_{10} concentrations are drastically over predicted. The model evaluation for each simulated period is available elsewhere [34]. Figure 8a shows a quantile–quantile (QQ) plot for 24-h average PM_{10} concentrations. A QQ plot demonstrates the model bias by pairing the low and high values and plotting them. For perfect model performance, all of the points in a QQ plot would fall along a 1-to-1 line. Figure 8a shows that the modeled values for PM_{10} are up to three times higher than what is observed. Figure 9a shows the spatial aspect of the modeled and observed average PM_{10} concentrations, and includes "donuts" at the RMCAB monitor locations, with the observational values as the outer ring and modeled values on the inside. Figure 9a also shows that at RMCAB monitor locations, PM_{10} concentrations are overpredicted on average. Table 5 shows that the overall MFB and MFE for the model compared to RMCAB observations are just barely within the criteria values for suitability for regulatory applications [49,50]. However, it is clear from the QQ plot and spatial plot that emissions for PM_{10} are too high. Given that RPM constitutes 90% of PM_{10} emissions, it is likely that RPM emissions are overestimated using the AP-42 (Chapter 13) methodology. Further research is underway to design and apply an adjustment factor that considers the effects of precipitation, relative humidity, and land use in the reduction of RPM emissions [34,55]. Additionally, the model evaluation of the specific PM_{10} components is desirable, but currently, no PM_{10} chemical composition is available for the city.

Table 5. Overall mean fractional bias (MFB) and mean fractional error (MFE) for the combined periods for PM_{10} and O_3.

	PM_{10}	O_3	CO	NO_x	SO_2
Overall MFB (%)	56.84% <60%	10.38% <20%	39.06% <60%	49.86% <60%	96.74% <60%
Overall MFE (%)	68.54% <75%	34.67% <30%	63.56% <75%	54.07% <75%	108.15% <75%

Criteria values are provided below the evaluation metrics MFB and MFE from Boylan et al. [49] and Simon et al. [50].

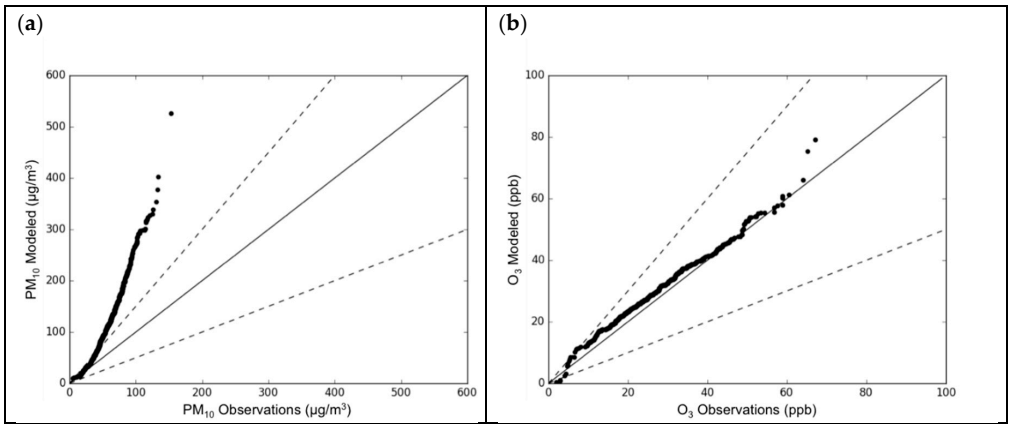

Figure 8. Quantile–quantile plots for all of the stations and the combined simulated periods for (**a**) 24-h average PM_{10} concentrations ($\mu g/m^3$) at standard conditions and (**b**) maximum daily 8-h O_3 concentrations (ppb).

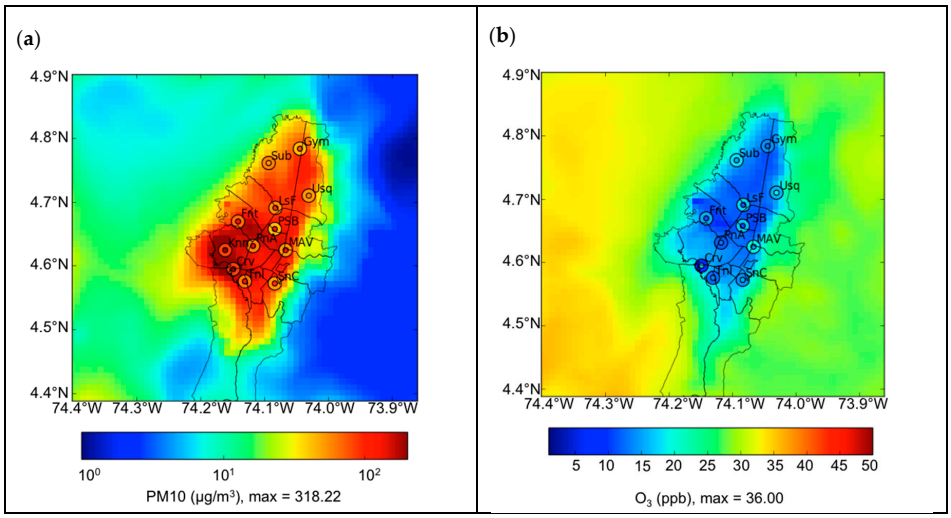

Figure 9. Spatial plots of the mean for the combined dry–wet periods of (**a**) 24-h average PM_{10} concentrations ($\mu g/m^3$) and (**b**) maximum daily 8-h O_3 concentrations (ppb). Values at the RMCAB monitor sites are shown as "donuts" with the observational values as the outer ring and the model values on the inside dot.

Ozone estimation by CMAQ successfully reproduced ambient observations (Figures 8b and 9b); evaluation metrics can be found in Table 5. The QQ plot in Figure 8b shows how the model is relatively unbiased for maximum daily 8-h O_3 concentrations, as the points nearly fall perfectly along a 1-to-1

line. The spatial plot in Figure 9b shows that at the RMCAB monitor locations, the model represents the O_3 concentrations well. The overall MFB shown in Table 5 meets the goal value (±10%); however, the MFE is just outside of the criteria (30%). Therefore, on the basis of the MFB strength and closeness to the criteria for the MFE, we consider the O_3 model performance strong. The ozone in Bogotá has been identified as a VOC-limited regime.

For CO, NO_x, and SO_2, model values are biased high (Table 5). For CO and NO_x, the MFB and MFE meet the criteria value, but for SO_2, they do not. The overestimation of these gas-phase species is likely due to using emission factors that do not reflect current fleet and fuel conditions. In effect, the sulfur content in diesel was lowered to 50 ppm in 2008, a situation that is not contemplated in EFs for SO_2. At the same time, the passenger fleet has been rapidly renovated in the last few years, and therefore, CO and NO_x emissions are expected to decrease. This situation is not considered in the EFs for CO and NO_x.

4. Conclusions

The authors have constructed a comprehensive emission inventory for air quality modeling purposes in Bogotá for 2012 that includes more emission sectors and processes than have historically been available. Commercial sources, biogenic sources, and resuspended dust were included in the inventory, in addition to traditional industrial and mobile sources. The newly estimated RPM constitutes 90% of total PM_{10} emissions, and they are at least onefold larger than PM_{10} emissions from combustion processes in mobile and stationary sources.

The emissions are chemically speciated and disaggregated in space and time. This EI was used in the implementation of the WRF–CMAQ modeling system. Model results suggest a high bias for PM_{10}, SO_2, NO_x, and CO ambient concentrations, while O_3 values were successfully predicted. The large PM_{10} values estimated by the model are likely due to RPM emissions being overestimated. This first RPM inventory for the city is prone to high uncertainties.

The areas of the city with the largest emission levels observed are the south and the southwest, where industrial and commercial activities are concentrated, as well as important highways that connect with the rest of the country. In addition, resuspended dust is of great magnitude due to large dust loadings and unpaved roads. The model successfully captures large concentrations of PM_{10} in these areas.

Traditionally, EIs in Bogotá were only compared among themselves, with a relative consistency in emissions from stationary sources, and some variability in mobile source estimates. This work advances in the evaluation of emission levels using air quality modeling results as an additional quality assurance step.

Future works include the revision of the RPM inventory considering the effect of precipitation, relative humidity, and land use in the resuspension of PM_{10}. Emissions of SO_2, NO_x, and CO need also to be revised in order to consider recent changes in fuel composition and fleet renovation. PM_{10} chemical compositions are also necessary to validate modeling results.

Supplementary Materials: The following are available online at www.mdpi.com/2073-4433/9/2/49/s1.

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