

On-road traffic emissions in a megacity

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ABSTRACT

A new annual bottom–up emission inventory of criteria pollutants and greenhouse gases from on-road mobile sources was developed for 2006 for the metropolitan area of Buenos Aires, Argentina, within a four-year regional project aimed at providing tools for chemical weather forecast in South America. Under the scarcity of local emission factors, we collected data from measuring campaigns performed in Argentina, Brazil, Chile and Colombia and compiled a data set of regional emission factors representative of Latin American fleets and driving conditions. The estimated emissions were validated with respect to downscaled national estimates and the EDGAR global emission database. Our results highlight the role of older technologies accounting in average for almost 80% of the emissions of all species. The area exhibits higher specific emissions than developed countries, with figures two times higher for criteria pollutants. We analyzed the effect on emissions of replacing gasoline by compressed natural gas, occurring in Argentina since 1995. We identified (i) a relationship between number of vehicles and a compound socioeconomic indicator, and (ii) time-lags in vehicle technologies between developed and developing countries, which can be respectively applied for spatial disaggregation and the development of projections for other Latin American cities. The results may also be employed to complement global emission inventories and by local policy makers as an environmental management tool.

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1. Introduction

Megacities are very large urban agglomerations with populations that exceed 10 million inhabitants, differing from urban areas not only in population size but also in the scale of their economy, infrastructure and associated environmental impacts (Gurjar and Lelieveld, 2005). Rapid urbanization has resulted in increasing air pollution emissions, typically arising from transportation, energy production and industrial activities, concentrated in densely populated areas (Gurjar et al., 2008) and surpassing the limits of the megacities' physically occupied area, thus contributing significantly to air quality on a global scale through the long range transport of air pollutants (Gurjar and Lelieveld, 2005; Butler et al., 2008; Butler and Lawrence, 2009).

This work was developed within the framework of the inter-American project "South American Emissions, Megacities and Climate" aimed at developing a number of tools for the chemical

weather forecast in the South American cities of Bogotá, Buenos Aires, Lima, Santiago and Sao Paulo. To the best of our knowledge this is the first inventory for the on-road transport categories of the metropolitan area of Buenos Aires that has estimated emissions of carbon dioxide (CO₂), methane (CH₄), nitrous oxide (N₂O), carbon monoxide (CO), nitrogen oxides (NO_x), non-methane volatile organic compounds (NMVOC), particulate matter (PM) and sulfur dioxide (SO₂) in two spatial disaggregation levels.

1.1. Emission inventories

Road transport became a significant source of air pollution in the last century and is currently one of the largest emission sources in megacities with subsequent adverse effects on human health (Faiz, 1993; Colville et al., 2001). In Argentina, road transport emissions accounted for 58% of total CO emissions in 2000 (Fundación Bariloche, 2005), share that is expected to rise because of the rising global demand for private mobility (Zachariadis et al., 2001). Real-world vehicle emissions are difficult to estimate because many factors are involved, and its accurate estimation is crucial for the formulation of efficient air quality management.

Emission inventories provide the necessary information required for air quality control policies, the development of future

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scenarios and modeling purposes (Parra et al., 2006; Zachariadis and Samaras, 2001; van Aardenne, 2002). There are two approaches for inventorying emissions: *top-down* and *bottom up*. Both methodologies share the same basic structure but present considerable differences regarding input data, assumptions and parameters (Reynolds and Broderick, 2000). The selection of the approach and the quality and consistency of the inventory depends almost entirely on the availability of the data and its degree of detail (Reynolds and Broderick, 2000; Tsiliniridis et al., 2002). It is futile to expect emission calculations to achieve results that are superior in accuracy to that of the original survey data (Sturm et al., 1996).

Differences are expected when comparing national emission inventories and specific megacity inventories. In the former, data on fuel consumption, technologies and installed abatement measures are applied uniformly over the country, while in the latter, the data employed is city-specific, the geographical units covered are smaller and the resulting emissions are distributed using more detailed maps of the emissions sources (Butler et al., 2008).

1.2. The metropolitan area of Buenos Aires

The metropolitan area of Buenos Aires (MABA) is composed of the city itself and 24 radially surrounding districts that are part of the province of Buenos Aires (Fig. 1), comprising an area of 3647 km². Being the 10th megalopolis in the world and the 3rd in Latin America it holds 32% of the total population in only 0.14% of the national territory, with a population density of 4600 inhabitants km⁻² and 2.4 million circulating vehicles. It is the greatest center of activities and the political, economical, and administrative center of the country, having the highest population density of the country, which diminishes from the center to its surrounding districts as poverty levels increase. In all its extent, the MABA is characterized by social and territorial inequities.

Three governmental levels coexist in the MABA (municipal, provincial and national) with different responsibilities and territorial limits within which each one may exercise its authority. Road transport regulation and management are key examples of this situation. For instance, emissions control for passenger vehicles is enforced at the municipal level in the city of Buenos Aires and at the provincial level in the 24 districts of the MABA; heavy-duty trucks are subject to national regulation, whereas light-duty trucks, being registered as passenger vehicles for private use, are also subject to municipal regulation; buses in the city of Buenos Aires are regulated at the national level while buses running in the provincial territory are subject to local regulation. This situation results in

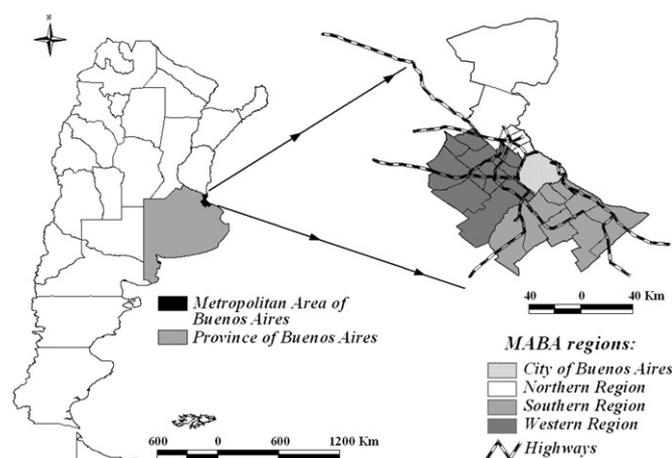


Fig. 1. Geographic location of the metropolitan area of Buenos Aires; Argentina (left). Regional subdivisions made for this study (right).

a juxtaposition of actors leading to diversity in sources of information and data. More importantly, this superimposing governmental levels combined with the fact that the MABA lies on a flat terrain with relatively good ventilation has been an excuse for a relatively poor air quality management in this highly populated urban zone, which lacks an official inventory of emissions of mobile sources. In addition, the availability of local and process-specific information on emissions represents a serious data gap, a common feature of many cities in developing countries (Guttikunda et al., 2005).

2. Methodology

A distance traveled approach was implemented to develop spatially disaggregated emission inventories for 2006. Annual emissions were estimated following the COPERT methodology (Ntziachristos and Samaras, 2000a,b), for which total emissions by category and species are estimated as the contribution of hot, cold and evaporative emissions. Hot emissions were calculated on the basis of Eq. (1).

$$E_{i,j,k} = N_j \times VKT_{j,k} \times EF_{i,j,k} \quad (1)$$

where, $E_{i,j,k}$: emissions of species i , vehicle category j , road type k , thermally stabilized engine; N_j : number of circulating vehicles, category j ; $VKT_{j,k}$: kilometers traveled, vehicle category j , road type k ; $EF_{i,j,k}$: hot emission factor, species i , vehicle category j , road type k . Cold emissions, occurring during warming-up, were also estimated on the basis of Eq. (1), incorporating the fraction of kilometers traveled with cold engines or catalyst operated below the light-off temperature and the ratio cold/hot of emission factors for species i . For details of the formulae used to estimate cold-start and evaporative emissions (comprising diurnal, hot soak contributions and running losses), the reader is referred to the COPERT methodology report.

We used the COPERT IV model for computing annual emissions. Data collection and main criteria for the selection of activity data and emission factors are discussed in Sections 2.1 and 2.2 respectively. A quality control system consisting of routine accuracy checks on data acquisition and calculations and the use of customary procedures used in our Laboratory for archiving information was implemented. Verification is dealt within Section 2.4.

The MABA was analyzed by its 25 districts and also according to four geographical zones, the city of Buenos Aires and the western, northern, and southern regions, grouping those districts that share similar socioeconomic characteristics (Fig. 1). Emissions of greenhouse gases and criteria pollutants were estimated. Our estimates of PM emissions account for exhaust and non-exhaust components.

2.1. Activity data

2.1.1. Vehicle fleet composition

Fleet was classified according to the structure of the COPERT methodology (Ntziachristos and Samaras, 2000a,b). The local fleet was arranged in seven major vehicle categories: light-duty – passenger cars (PC), sport utility vehicles (SUV), taxicabs (TX), and light-duty trucks (LDT) – and heavy-duty – composed by heavy-duty trucks (HDT), buses (B) and coaches (C) –. These categories were further subdivided according to fuel used, engine capacity, vehicle size, weight and emission control technology, resulting in 35 technology-based categories for which emission factors are directly linked to age and technology parameters (Zachariadis and Samaras, 2001).

The fuels employed in Argentina for on-road transport are compressed natural gas (CNG), diesel oil and gasoline. Unleaded

gasoline has been mainly used by light-duty vehicles since 1996 when leaded gasoline was banned from the Argentinean market. CNG has become increasingly used in the country since 1995, especially due to its lower price. Until December 2006, CNG was used by 1.4 million vehicles at national level and 330,000 at MABA level in originally manufactured gasoline light-duty vehicles that have been converted into bi-fuel vehicles (gasoline/CNG). Light-duty vehicles employ the three named fuels, whereas heavy-duty vehicles run only on diesel oil.

Different information sources were considered for the activity data. Number of vehicles per category is typically available from the sources listed in Table 1. At our request, staff of the Vehicle Registration Directory (DNRPA) compiled the number of PC, SUV and LDT disaggregated by fuel use for different registration periods (<1996, 1997–2000, 2001–2003 and 2004–2006). This age distribution reflects a technology distribution (Zachariadis et al., 2001) that accounts for the introduction of three-way catalyst (TWC) control. According to experts' assumptions, these devices have a lifetime of five years or 80,000 km. Since control and maintenance programs have not been thus far enforced, it was assumed that vehicles registered before 2000 no longer possessed the originally provided TWC in proper working conditions. Vehicles registered after year

2000 were considered to be equivalent to European vehicles following the EURO I emission standards and those registered after 2003, to EURO II. HDT were disaggregated in two categories representing the inclusion of emission control technologies; buses and coaches were all considered to be equivalent to European buses following the EURO II emission standards. The presented age structure is relevant in developing countries as Argentina, where a significant part of the fleet consists of relatively old and often poorly maintained vehicles (Faiz et al., 1996; Faiz and Sturm, 2000). It is common practice to apply a scrappage model to distinguish the real circulating fleets. However, the information sources used in our study removed non-circulating vehicles making unnecessary to model scrappage rates. Two-wheelers, which constitute a small percentage of the total fleet, were not included because of lack of information since the corresponding registration data has recently started to be organized.

2.1.2. Vehicle kilometers traveled (VKT)

As there is no traffic model presently available for the MABA and a lack of reliable information regarding travel surveys, traffic counts and questionnaires to drivers, central estimations of vehicle mileage were obtained from expert elicitation as the starting point.

Table 1
Vehicle classification for the metropolitan area of Buenos Aires and corresponding activity data for 2006.

Category	Vehicle type	Fuel	Emissions control	Period of registration	VKT	Fleet (2006)	Source of information	Classification Characteristics	
PC1	Passenger Cars	Gasoline	No	≤1996	7000	913,768	DNRPA	Vehicles of less than 2.5 ton with an engine size of 1.4–2.0 L that are employed by private owners for their own travel needs.	
PC2	Passenger Cars	Gasoline	No	1997–2000	7700	272,247			
PC3	Passenger Cars	Gasoline	Yes	2001–2003	8470	222,503			
PC4	Passenger Cars	Gasoline	Yes	2004–2006	9300	62,244			
PC5	Passenger Cars	Diesel oil	No	≤2000	20,000	170,826			
PC6	Passenger Cars	Diesel oil	Yes	2001–2003	22,000	81,822			
PC7	Passenger Cars	Diesel oil	Yes	2004–2006	24,200	10,695			
PC8	Passenger Cars	CNG	No	≤2000	29,000	158,141			ENARGAS
PC9	Passenger Cars	CNG	Yes	2001–2006	31,900	115,187			
TX1	Taxicabs	Diesel oil	No	≤2000	60,000	19,648	GCBA	Vehicles of less than 2.5 ton with an engine size of 1.4–2.0 L that are employed for passengers public transport. They were differentiated from PC as they meet different travel demands (higher VKT).	
TX2	Taxicabs	Diesel oil	Yes	2001–2003	60,000	6534			
TX3	Taxicabs	Diesel oil	Yes	2004–2006	60,000	1586			
TX4	Taxicabs	CNG	No	≤2000	60,000	5071			
TX5	Taxicabs	CNG	Yes	2001–2006	60,000	5559			
SUV1	Sport Utility Vehicles	Gasoline	No	≤1996	7000	56,542	DNRPA	Vehicles of less than 2.5 ton with an engine size > 2.0 L that are employed by private owners for their own travel needs.	
SUV2	Sport Utility Vehicles	Gasoline	No	1997–2000	7700	15,162			
SUV3	Sport Utility Vehicles	Gasoline	Yes	2001–2003	8470	12,204			
SUV4	Sport Utility Vehicles	Gasoline	Yes	2004–2006	9300	544			
SUV5	Sport Utility Vehicles	Diesel oil	No	≤2000	20,000	85,517			
SUV6	Sport Utility Vehicles	Diesel oil	Yes	2001–2003	22,000	22,647			
SUV7	Sport Utility Vehicles	Diesel oil	Yes	2004–2006	24,200	795			
SUV8	Sport Utility Vehicles	CNG	No	≤2000	29,000	20,363			ENARGAS
SUV9	Sport Utility Vehicles	CNG	Yes	2001–2006	31,900	11,698			
LDT1	Light-Duty Trucks	Gasoline	No	≤2000	18,000	31,603	DNRPA	Vehicles of less than 3.5 ton with an engine size < 2.0 L that are employed for the local transport of goods. They were differentiated from SUV as they meet different travel demands (higher VKT).	
LDT2	Light-Duty Trucks	Gasoline	Yes	2001–2003	19,800	4545			
LDT3	Light-Duty Trucks	Gasoline	Yes	2004–2006	21,780	357			
LDT4	Light-Duty Trucks	Diesel oil	No	≤2000	55,000	42,471			
LDT5	Light-Duty Trucks	Diesel oil	Yes	2001–2003	60,500	20,292			
LDT6	Light-Duty Trucks	Diesel oil	Yes	2004–2006	66,550	556			
LDT7	Light-Duty Trucks	CNG	No	≤2000	67,500	5989			ENARGAS
LDT8	Light-Duty Trucks	CNG	Yes	2001–2006	74,250	10,952			
HDT1	Heavy-Duty Trucks	Diesel oil	No	≤2000	75,000	24,486			
HDT2	Heavy-Duty Trucks	Diesel oil	Yes	2001–2006	82,500	5798	CNRT	Vehicles of weight >3.5 ton that travel long distances for the transport of goods at provincial, national or international levels.	
B	Buses	Diesel oil	Yes	≤2006	80,000	9654			
C	Coaches	Diesel oil	Yes	≤2006	13,500	10,560			
						2,438,569			

DNRPA = National Vehicle Registration Directory.

ENARGAS = Gas Regulatory Board of Argentina.

GCBA = Government of the city of Buenos Aires.

CNRT = Transport Regulatory Commission of Argentina.

Part of this information, provided by local experts, was contrasted with reports by transport authorities (long-distance buses) and ad hoc questionnaires to drivers (TX and HDT) undertaken in this study. LDT represented the category with the largest uncertainty regarding VKT. Fuel use balance was employed to obtain a representative VKT for LDT and also to verify VKT estimations for the remaining categories. VKT by vehicle category were the optimization variables used to close the fuel balance. To obtain the representative VKT for MABA, whose districts share similarities between the city's and the province's consumption patterns, the results for the city and the province were weighed with the registered fleet belonging to the MABA. As the central estimations corresponded to vehicles registered in the period 2004–2006, an age-dependent annual mileage was assumed employing different mileage estimates for each model year group, as vehicles tend to be driven less as they grow older (Zachariadis, and Samaras, 2001). The final fleet classification with the corresponding VKT for the MABA is presented in Table 1.

2.2. Emission factors

Emissions factors (EFs) were selected from different sources considering availability of data and making an effort to best represent local conditions. Locally or regionally measured data exist only for hot-driving conditions as discussed below. Cold-driving and evaporative emissions were exclusively estimated

using the COPERT IV model, motivated by the large share of European models of the Argentinean fleet.

For hot-driving conditions, only a small number of emission factors has been measured in Argentina. Nevertheless, we decided to fully exploit the scarce local available data by combining it with regional information to estimate EFs that would better characterize the local fleet than those available in databases of developed countries. To this end, the two measurement campaigns undertaken in Buenos Aires were complemented with 12 studies undertaken in the following Latin American cities: Bogotá (Colombia), Mexico City (Mexico), Santiago de Chile (Chile) and Sao Paulo (Brazil). Table 2 provides a summary of the available data, indicating emissions test, fleet composition, measured compounds and reference study. In addition to the estimation of EFs for cold-driving and evaporative conditions, the COPERT IV model was also used to estimate EFs for (i) diesel light-duty vehicles for all compounds and (ii) CH₄ and N₂O EFs for all gasoline and diesel-fueled vehicle categories, for which the corresponding road transport emissions were practically not measured in Latin America. Methane and N₂O emission factors for CNG vehicles were obtained from the IPCC emission factor database (IPCC-EFDB, 2007). CO₂ and SO₂ emission factors were computed on the basis of the content of C and S in the fuels used locally.

The regional set of EFs for hot-driving conditions developed in this study was obtained by the following comparative assessment. For each vehicle category, the sets of EFs enumerated in Table 2

Table 2
Summary of the main characteristics of the emission factor measuring campaigns for hot conditions undertaken in Latin American cities.

City	Year	Emission test	PC	SUV	LDT	HDT	B & C	Fuel	CO ₂	CO	NO _x	PM	SO ₂	VOCs	Reference	
BA	2000	Dynamometer	3	–	1	–	–	G	–	X	X	–	–	X	Vasallo, 2000	
			3	–	1	–	–	CNG	–	X	X	–	–	X		
BA	2004	On-road	10	–	–	–	–	G	X	X	X	–	–	X	ARPEL, 2005	
			5	–	4	–	–	D	X	X	X	–	–	X		
			6	–	–	–	–	CNG	X	X	X	–	–	X		
SP	2004	2 road tunnels	2 × 10 ⁵	–	–	–	–	G*	X	X	X	–	X	X	Martins et al., 2006	
			–	–	4.3 × 10 ³	1.9 × 10 ⁴	–	D	X	X	X	–	X	X		
SP	2004	On-road	16	–	–	–	–	G*	X	X	X	–	–	X	ARPEL, 2005	
			2	–	–	–	–	CNG	X	X	X	–	–	X		
SP	2006	On-road	–	–	18	13	12	D	X	X	X	X	–	X	Lents et al., 2007	
SP	2001–2006	Dynamometer	NRS	NRS	NRS	–	–	G*	–	X	X	X	X	X	CETESB, 2007	
			NRS	NRS	NRS	–	–	D	–	X	X	X	X	X		
BR	1990–1994	Dynamometer	NRS	NRS	NRS	–	–	G*	X	X	X	X	X	X	MST, 2002	
			–	–	–	NRS	NRS	D	X	X	X	X	X	X		
B	2002	Inverse Modeling in Street Canyon	3.6 × 10 ⁵ (gasoline light-duty vehicles and diesel heavy-duty vehicles)				–	–	G*	–	X	X	X	X	–	Manzi et al., 2003
			D	–	–	X	X	X	X	–	–	–	–	X		
B	2007–2008	On-road	43	33	–	–	–	G	X	X	X	–	–	X	ARPEL, 2005	
			–	–	–	23	27	D	–	–	–	X	–	–		
			21	14	–	–	–	CNG	X	X	X	–	–	X		
SC	2004	On-road	17	10	–	–	–	G	X	X	X	–	–	X	SECTRA, 2007a,b	
			49	18	4	–	–	G	X	X	X	–	–	X		
SC	2006	On-road	15	12	13	36	–	D	X	X	X	X	–	X	CONAMA-RM, 2008	
			7	3	–	–	–	CNG	X	X	X	–	–	X		
			–	–	–	38	15	D	X	X	X	X	–	X		
SC	1997–1999	Dynamometer	127	–	39	–	–	G	–	X	X	–	–	X	Corvalán and Urrutia, 2000	
MX	2006	On-road	–	–	6	6	16	D	X	X	X	X	–	X	Lents et al., 2007	
Overall sample			265	61	44	–	–	G	–	–	–	–	–	–		
			20	12	41	116	70	D	–	–	–	–	–	–		
			39	17	1	–	–	CNG	–	–	–	–	–	–		

BA = Buenos Aires, Argentina.

SP = Sao Paulo, Brazil.

SC = Santiago de Chile, Chile.

MX = Mexico City.

B = Bogota, Colombia.

BR = Brazil.

G = Gasoline.

D = Diesel oil.

CNG = Compressed natural gas.

G* = Gasoline with 22% anhydrous ethanol.

NRS = Not reported by the source.

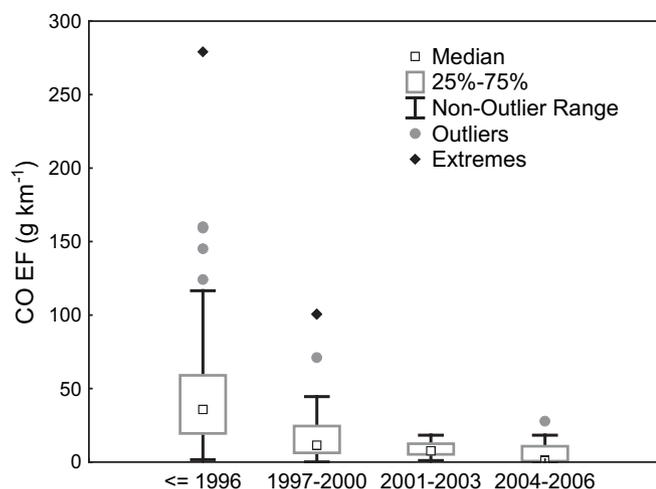


Fig. 2. Distribution of regional CO emission factors measured in Argentina, Brazil, Chile and Colombia for the four gasoline passenger cars categories considered in this study. White squares represent the median, the box indicates the 25–75% range of the distribution and the whiskers represent minimum and maximum values. Grey circles and black diamonds represent outliers and extremes respectively.

were used to estimate the corresponding regional EFs regarding the total number of vehicles composing the overall sample. To define the acceptable sample size, we followed the recommendations by CONAMA-RM (2008) indicating the required minimum number of tested units as 30 for light-duty vehicles and 15 for heavy-duty

vehicles. To estimate the most appropriate EF values by vehicle category and compound, we assessed the distribution of the corresponding data sets by means of probability–probability plots using the STATISTICA software. Right-skewed distributions provided the best fit; however the data could not be fitted by a log-normal distribution using the Kolmogorov–Smirnov test (Canavos, 1988). Medians were thus selected as a better indication of the central tendency of the data and quartile values as uncertainty range of the EFs (e.g., Fig. 2). The selected EFs by species and vehicle category are presented in Table 3.

The EFs in our regional data set account for different types of roads, driving patterns and climate conditions; their use implies some sort of average for these three underlying conditions. In consequence, only one type of compounded road was finally used in Eq. (1) ($k = 1$). For those EFs computed using the COPERT IV model, the parameters employed were urban road and the average speed of 35 km h⁻¹ reported for MABA (ARPEL, 2005).

2.3. Spatial resolution of the inventory

This inventory was computed by district and by region. Different approaches for spatial disaggregation were considered depending on the type of vehicle, route and source of information.

- Light-duty vehicles (PC, TX, SUV and LDT) travel where they were registered disregarding trips between districts.
- Bus-route was the basis for the spatial disaggregation of urban buses number.

Table 3

Emission factors (g km⁻¹) by category employed in this study. Regionally measured EFs are reported on the basis of the median values and accompanied with the lower and upper quartile values in brackets. Uncertainty ranges are not reported for those EFs obtained from international sources (COPERT IV and IPCC emission factors database); for the criteria adopted for these cases, see Section 2.5.

ID	Fuel	CO ₂	CH ₄	N ₂ O	CO	NMVOCS	NO _x	PM	SO ₂
PC1	G	299	0.184	0.013	53.51 (29.10–88.08)	4.87 (1.28–8.24)	1.43 (0.82–2.12)	0.024	0.1136
PC2	G	299	0.184	0.013	17.23 (9.42–36.66)	1.78 (0.87–2.86)	0.87 (0.39–1.44)	0.024	0.1136
PC3	G	213	0.038	0.032	20.46 (13.74–33.14)	1.55 (0.42–3.42)	1.16 (0.61–1.74)	0.024	0.0810
PC4	G	213	0.043	0.017	2.58 (1.31–23.64)	0.21 (0.02–0.54)	0.19 (0.06–0.98)	0.024	0.0810
PC5	D	213	0.034	0.000	0.82	0.18	0.58	0.306	0.2016
PC6	D	182	0.016	0.002	0.55	0.06	0.67	0.099	0.1720
PC7	D	182	0.009	0.005	0.46	0.04	0.73	0.086	0.1720
PC8	CNG	236	0.709	0.068	6.36 (2.00–18.23)	1.56 (0.24–2.79)	1.74 (1.05–3.70)	0.021	0.0002
PC9	CNG	168	0.709	0.068	6.36 (2.00–18.23)	1.56 (0.24–2.79)	1.74 (1.05–3.70)	0.021	0.0001
TX1	D	213	0.033	0.000	0.82	0.18	0.58	0.306	0.2016
TX2	D	182	0.015	0.002	0.55	0.06	0.67	0.099	0.1720
TX3	D	182	0.008	0.004	0.46	0.04	0.73	0.086	0.1720
TX4	CNG	236	0.709	0.068	6.36 (2.00–18.23)	1.56 (0.24–2.79)	1.74 (1.05–3.70)	0.021	0.0002
TX5	CNG	168	0.709	0.068	6.36 (2.00–18.23)	1.56 (0.24–2.79)	1.74 (1.05–3.70)	0.021	0.0001
SUV1	G	367	0.184	0.013	81.47 (27.46–146.58)	8.74 (5.03–13.16)	2.62 (1.43–3.70)	0.024	0.1392
SUV2	G	367	0.184	0.013	14.84 (9.94–33.14)	1.79 (1.50–6.56)	0.99 (0.65–3.32)	0.024	0.1392
SUV3	G	286	0.038	0.032	15.44 (6.81–22.85)	1.06 (0.28–1.77)	0.94 (0.29–1.59)	0.024	0.1085
SUV4	G	285	0.043	0.017	7.36 (3.21–23.19)	0.44 (0.17–0.70)	0.46 (0.21–0.80)	0.024	0.1082
SUV5	D	213	0.034	0.000	0.82	0.18	0.90	0.306	0.2016
SUV6	D	234	0.016	0.002	0.55	0.10	0.67	0.099	0.2209
SUV7	D	234	0.009	0.005	0.46	0.14	0.73	0.086	0.2209
SUV8	CNG	289	0.709	0.068	10.81 (4.03–19.67)	1.56 (0.24–2.79)	2.51 (1.46–3.92)	0.021	0.0002
SUV9	CNG	225	0.709	0.068	10.81 (4.03–19.67)	1.56 (0.24–2.79)	2.51 (1.46–3.92)	0.021	0.0002
LDT1	G	365	0.184	0.013	33.33 (20.90–53.29)	4.38 (2.85–7.23)	1.23 (0.92–1.43)	0.032	0.1387
LDT2	G	365	0.038	0.044	26.4 (22.89–32.79)	3.07 (0.09–8.07)	1.61 (0.62–2.41)	0.032	0.1387
LDT3	G	365	0.043	0.036	22.48 (19.49–27.92)	2.19 (0.05–5.78)	1.49 (0.58–2.24)	0.032	0.1387
LDT4	D	279	0.034	0.000	1.37	0.15	2.05	0.396	0.2638
LDT5	D	252	0.016	0.002	0.51	0.17	1.26	0.129	0.2378
LDT6	D	252	0.009	0.005	0.51	0.17	1.26	0.129	0.2378
LDT7	CNG	288	0.709	0.068	1.6 (0.60–4.60)	0.09 (/-0.64)	1.85 (0.75–3.50)	0.029	0.0002
LDT8	CNG	288	0.709	0.068	1.6 (0.60–4.60)	0.09 (/-0.64)	1.85 (0.75–3.50)	0.029	0.0002
HDT1	D	837	0.171	0.029	3.88 (2.11–7.13)	3.52 (1.98–8.10)	8.92 (4.37–12.14)	1.151 (0.527–1.977)	0.7906
HDT2	D	837	0.171	0.011	1.83 (1.11–5.84)	0.52 (0.35–1.18)	5.61 (3.50–8.37)	0.267 (0.160–0.583)	0.7906
B	D	771	0.111	0.012	6.09 (4.66–11.02)	1.32 (0.93–1.72)	15.98 (12.98–18.59)	0.287 (0.187–0.570)	0.7283
C	D	893	0.111	0.011	6.09 (4.66–11.02)	1.32 (0.93–1.72)	15.98 (12.98–18.59)	0.287 (0.187–0.570)	0.8437

- Information from tolls of the four highways that interconnect the MABA (Fig. 1) was employed for HDT. These emissions were disaggregated by district taking as the weighing indicator the distance traveled within each district.
- For coaches, we used the shares departing from the city's main bus terminal towards CBA, northern, southern and western regions. For the district approach, the same methodology as for HDT was employed.

2.4. Validation of emission inventories

We compared our results with those reported in the national greenhouse gas emission inventory (NGHGEI) (Fundación Bariloche, 2005) and the 1×1 degree grid MABA cell ($-59, -35$) of the EDGAR¹ global emission database (van Aardenne et al., 2005; Olivier et al., 2005). Since these were computed for the year 2000, we recalculated our inventory only accounting for vehicles registered until 2000. The NGHGEI emissions were downscaled to the MABA using the ratio between vehicle numbers in the two geographical areas, as specified by Eq. (2).

$$E_{ij|MABA} = \frac{N_{j|MABA}}{N_{j|NGHGEI}} \times E_{ij|NGHGEI} \quad (2)$$

where, E_{ij} : emissions of species i , vehicle category j and N_j , vehicle number.

2.5. Uncertainty analysis

Emission uncertainties were estimated using Approach 1 of the 2006 IPCC guidelines based upon error propagation (IPCC, 2006). Our quantitative uncertainty analysis dealt with random components of measurement errors and systematic errors associated with data representativeness. The uncertainties of emission factors, number of vehicles and VKT were combined by multiplication and finally aggregated for the computation of the overall uncertainty to the total inventory by species. This approach works well for relatively small uncertainties but as the uncertainty becomes larger, the error propagation approach systematically underestimates the uncertainty. For those cases for which the uncertainty exceeded 100% the correction factor proposed in the 2006 IPCC guidelines was employed.

Different approaches were employed to estimate EF uncertainties. For those pollutants and vehicle categories for which appropriate samples of local or regional measurements were available, asymmetric confidence intervals between the 25 and 75 percentiles were employed, as previously discussed. A 2% value was adopted for CO₂ and SO₂ EF uncertainties, as local data of fuel consumption and carbon and sulfur contents were employed, and a factor of 3 for CH₄ and N₂O EFs, reflecting its lower degree of accuracy (IPCC, 2006). For the EFs obtained using the COPERT IV model, a 100% uncertainty was considered assuming a poorer representativeness of these European-based data for the local circumstances.

Regarding the number of vehicles, an uncertainty of 2% was assigned to PC, TX, SUV, and LDT, assuming a high level of confidence in the sources of information. An additional 1% was adopted for these types of vehicles running on CNG accounting for

a potential misallocation in the districts surrounding the city of Buenos Aires. For freight vehicles a 10% uncertainty was chosen to represent the misallocation of these categories in the MABA region.

Regarding the uncertainties associated with VKT by category, an asymmetric confidence interval was considered using the difference between the computed VKT on the basis of fuel balance and those proposed by national experts and a 2% uncertainty for the opposite range, considered of higher confidence.

2.6. Reconstruction of activity data

The regional EFs compiled in our study can be applied not only to the MABA but also to other cities of the region. To extend the applicability of this methodology to other Argentinean cities, we assessed the existence of the necessary activity data. For instance, number of vehicles and fuel consumption are reported only at the provincial level by official institutions. We explored suitable underlying variables to infer number of vehicles in Argentina, considering: population, population density, gross geographic product (GGP), the percentage of the population with unsatisfied basic needs (UBN) and the human development index (HDI). The UBN index, computed by the Economic Commission for Latin America and the Caribbean, indicates the percentage of the population with poor housing and crowding conditions, sanitation, school attendance, and subsistence capacity, computed at district level for all Argentinean provinces, whilst the HDI acts as an indicator of life expectancy, level of acquired knowledge, and gross domestic product per capita, being only computed at provincial level by country. A correlation analysis between both, number of vehicles (NV) and number of vehicles per capita (NVPC), and the named socioeconomic variables was performed at country and MABA levels.

3. Results

3.1. Emission inventories

Annual emissions for the metropolitan area of Buenos Aires for 2006 from on-road mobile sources are presented as follows, indicating the level of uncertainty between brackets (Gg): CO₂ 11,524 (10,071–12,195), CH₄ 10.6, N₂O 1.04, CO 569 (406–829), NO_x 81.9 (63.4–97.6), NMVOC 69.8 (43.8–96.8), PM 6.37 (3.27–9.11) and SO₂ 6.60 (5.38–6.90). As the level of uncertainty associated with CH₄ and N₂O emissions was 140% the corresponding range was not reported above because of its negative lower bound. Uncertainty levels for criteria pollutants were in the following order: PM (50%) > CO ~ NMVOC (40%) > NO_x (20%) > CO₂ ~ SO₂ (10%). The computed uncertainties were compared with those reported for the EDGAR database. While the uncertainties for CH₄ and N₂O resulted almost three times higher in the present study, those for criteria pollutants were found to be lower; being the uncertainties associated with CO₂ and SO₂ emissions equal.

Fig. 3 presents the distribution of emissions according to fuel use and major vehicle type. The four main groups contributed about equally to CO₂ emissions. Although heavy-duty vehicles were small in number compared to the total fleet (2%), their emissions represented 23% of total CO₂ emissions, nearly equaling those of gasoline and diesel vehicles, because of their high number of VKT and the associated high fuel consumption values. Gasoline light-duty vehicles were the main emitters of CO and NMVOC, accounting for 85% and 65% of emissions respectively, while they only represented a small fraction (~20%) of CH₄, N₂O, NO_x and SO₂ emissions. Together, diesel light and heavy-duty vehicles were responsible for almost all PM emissions, 80% of SO₂ emissions and 55% of NO_x emissions, while they accounted for only 4% of CO

¹ The Emission Database for Global Atmospheric Research (EDGAR) is a joint project of MNP, Bilthoven (NL), TNO, Apeldoorn (NL), JRC-IES, Ispra (IT) and MPIC-AC, Mainz (D) that stores global emission inventories of direct and indirect greenhouse gases from anthropogenic sources both on a per country and region basis as well as on a grid.

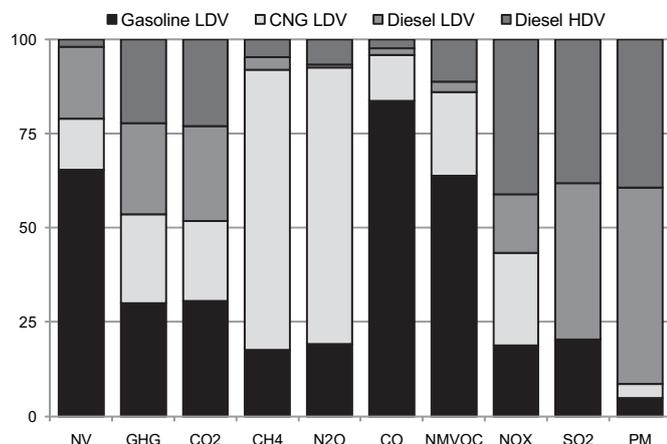


Fig. 3. Emissions distribution by fuel type (compressed natural gas (CNG), diesel oil and gasoline) and main vehicle category (light and heavy-duty vehicles). GHG represents the sum of CO₂, CH₄ and N₂O emissions in terms of global warming potential (CO₂: 1, CH₄: 21 and N₂O: 310); NV stands for number of vehicles.

emissions. CNG light-duty vehicles were the main emitters of CH₄ and N₂O, accounting for more than 70% in both cases.

Further disaggregation by vehicle categories allowed the analysis of the effect of fleet categorization on the computation of final emissions. Gasoline PC registered before 1997 were the highest emitters of CO, NMVOC and CO₂, accounting for 60%, 45% and 17% of total emissions respectively. This category was composed of old vehicles with neither emission control devices nor maintenance, making them very difficult to characterize with a non-local EF. The incorporation of regional EFs was key for improving accuracy of the estimated emissions of this category. CNG passenger cars accounted for 55% of CH₄ and N₂O emissions representing 10% of the total fleet. Emissions of diesel HDT were calculated using data with an important level of uncertainty, largely affecting the emission estimates of CO₂, NO_x, PM and SO₂ where HDT were responsible for 17%, 23%, 39% and 28% of total emissions respectively.

Overall specific emissions for the MABA were computed by TJ of consumed fuel. Due to the lack of information for other megacities, these values were compared with emissions at country level, employing the reported data by the Parties to the United Nations Framework Convention on Climate Change. Time series of overall CO specific emissions in ton TJ⁻¹ (1990–2006) for Australia, Japan, United States and EU15² are shown in Fig. 4 together with the corresponding value for the MABA for 2006. The MABA's overall CO specific emissions corresponded to the 1990 EU15 value, showing a 16-year lag in the incorporation of the European fleet technologies. A similar time lag of 10 years is observed with Australia and the United States. The large difference between MABA's specific emissions and those from Japan (11 times) is mainly driven by the different vehicle technologies present in the two countries. The same behavior was observed for NO_x, N₂O and CH₄. These results highlight the importance of acquiring regionally measured EFs as the best proxy for local representativeness. The use of EFs from developed countries implied the underestimation of the emissions as follows: CO (35–90%), NO_x (33–70%), CH₄ (65–95%) and N₂O (15–58%).

² EU15 stands for European Union members prior to 2004: Austria, Belgium, Denmark, Finland, France, Germany, Greece, Ireland, Italy, Luxembourg, Netherlands, Portugal, Spain, Sweden, and United Kingdom.

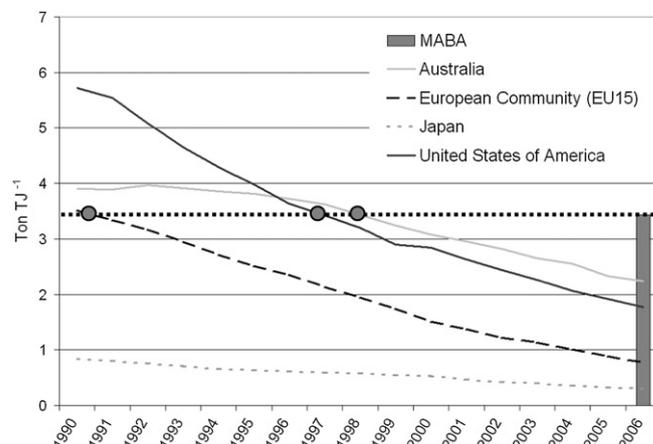


Fig. 4. Comparison of overall CO specific emissions (ton TJ⁻¹) between the metropolitan area of Buenos Aires (2006) and selected developed countries (1990–2006).

3.2. Validation of emission inventories

Our estimates of CO₂ and criteria pollutants emissions and those from the NGHGEI are roughly within the range of maximum and minimum values of EDGAR, except for SO₂ (Fig. 5). Apparent overestimation of SO₂ emissions in EDGAR may arise from considering fuels with higher sulfur content than the country specific content used in NGHGEI and our study. NGHGEI reported lower emissions of CO₂ and SO₂ than those computed in this study. As these emissions depend directly on fuel consumption, the difference is based on the use of nation-wide average consumption patterns that enclose urban and rural conditions in the NGHGEI that are smaller than the urban consumption factors employed in this work. Emission levels of criteria pollutants computed in this study are similar to those obtained from the downscaling of the NGHGEI. EDGAR seems to underestimate CH₄ and N₂O emissions, possibly because of the misallocation of CNG bi-fuel vehicles in the corresponding MABA grid cell, which accounts for ~50% of the CNG national fleet. Conversely, CH₄ emissions may be overestimated by the NGHGEI, which employed the 1996 IPCC EF for CNG vehicles that is almost 5 times higher than that presented in the 2006 guidelines employed in the present study. The NGHGEI may have underestimated N₂O emissions by employing lower EFs for gasoline vehicles, which represent ~65% of the total fleet, and did not compute emissions from CNG vehicles, which have the highest N₂O EF. New vehicles with TWC are the main emitters of this compound, thus emissions of N₂O increased 80% since 2000–2006 due to the incorporation of this technology to the registered fleet. As reported by Olivier (2005), EDGAR CH₄ and N₂O emission estimates are associated with substantial uncertainty mainly driven by the inaccurate international activity data and the selected EFs for calculating country emissions (Olivier et al., 1999, 2001).

3.3. Emissions disaggregation

3.3.1. By district

Fig. 6 shows the 2006 annual emissions. The City of Buenos Aires accounted for ~40% of emissions, being six times higher and ten times higher than the second and third maximum emitters respectively. Emission distribution by pollutant followed the main emitting categories in each district. CO emissions correlated with the registration of gasoline light-duty vehicles, accounting for their type and number. PM emissions correlated with the circulation of diesel heavy-duty vehicles. CH₄ and N₂O emissions followed the

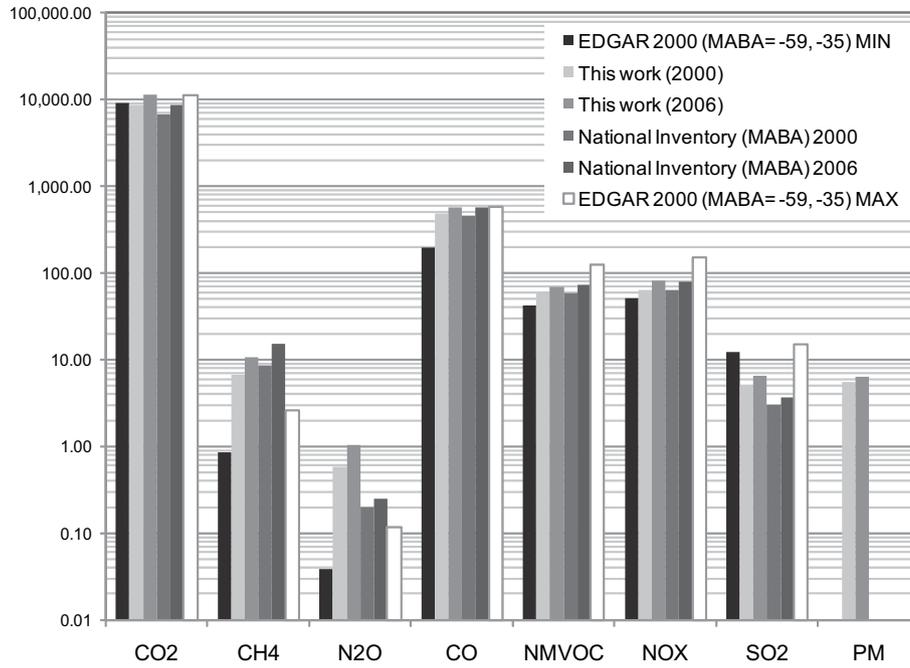


Fig. 5. Comparison between the results for the metropolitan area of Buenos Aires obtained in this study with the corresponding emissions downscaled from the national greenhouse gas emission inventory (Fundación Bariloche, 2005) and the EDGAR international emissions database (Olivier, 2005).

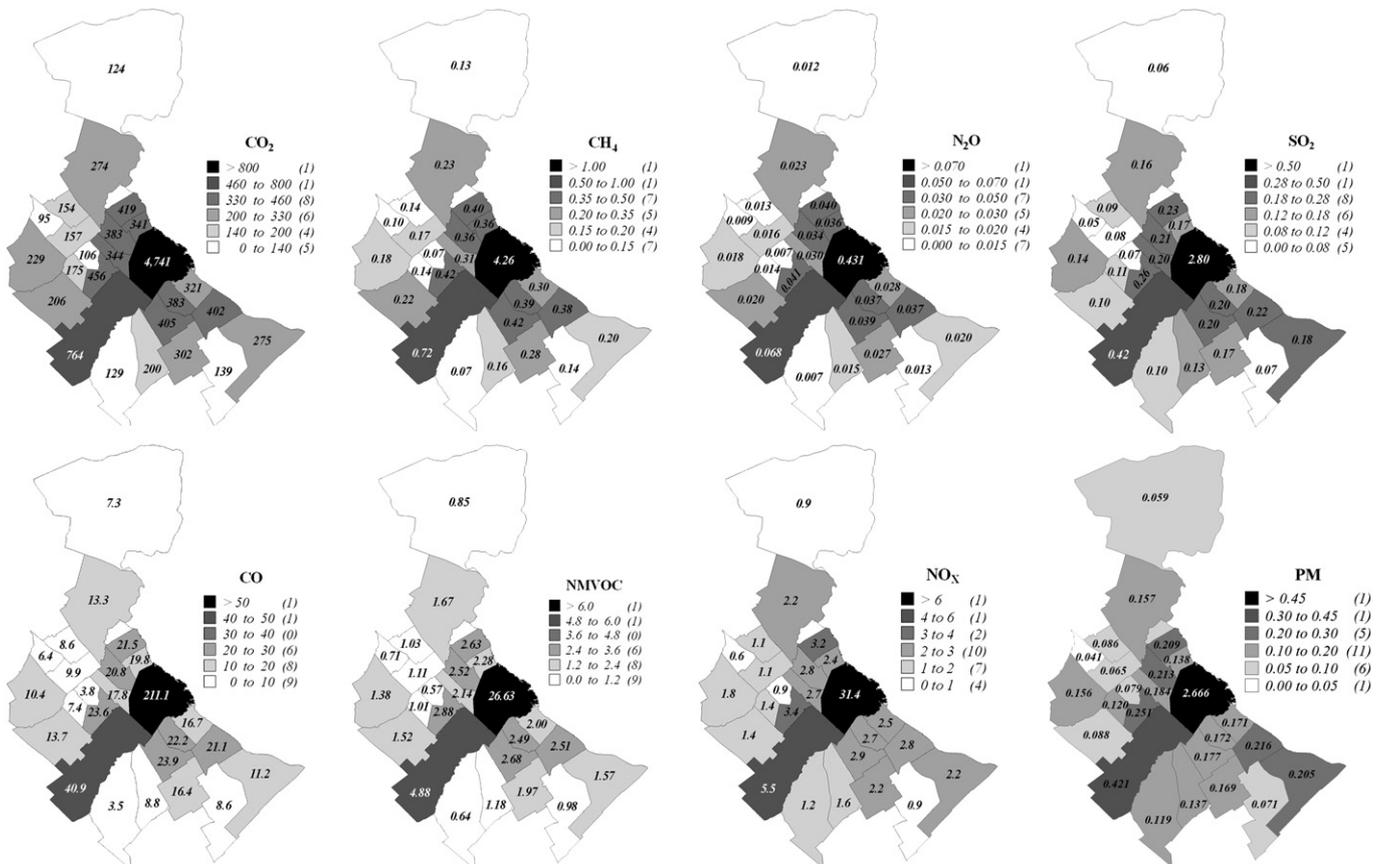


Fig. 6. Emissions in Gg of CO₂, CH₄, N₂O, CO, NMVOCs, NO_x, PM and SO₂ disaggregated by district of the metropolitan area of Buenos Aires (2006).

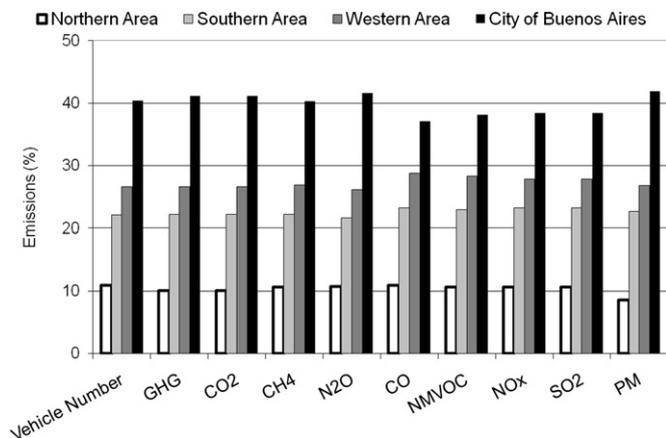


Fig. 7. Emissions distribution by main region for all considered species.

registration of CNG light-duty vehicles and NO_x emissions followed the circulation of diesel heavy-duty vehicles.

3.3.2. By region

Fig. 7 presents the distribution of 2006 emissions together with the number of vehicles. The City of Buenos Aires exhibited the highest emissions for all compounds, followed by the western, southern and northern regions. The three neighboring regions presented higher shares of the byproducts of incomplete combustion (CO, NMVOC) than the City, compared with CO_2 shares, because of different technology distribution between regions, where the City concentrated a major proportion of newer, more efficient vehicles. NO_x emissions presented a similar situation based on the higher ratio of older gasoline vehicles of the surrounding regions. Regarding PM emissions, the City, and the southern and western regions, presented a greater PM/ CO_2 ratio than the northern region, driven by the main emitting categories conventional diesel light and heavy-duty vehicles.

3.4. Replacement of gasoline by CNG: impact on emissions

Argentina is the second country in the world after Pakistan with the highest number of CNG vehicles amounting in 2006–1430 thousands that are served by 1600 refueling stations. We have assessed the effect of fuel switching from gasoline to CNG occurring since 1995 in light-duty vehicles by comparing the estimated 2006 emissions from CNG used in bi-fuel vehicles (gasoline/CNG) with those that would have occurred if only gasoline had been used by this type of vehicles. The use of CNG implied an increase of CH_4 and N_2O emissions by 62% and 49% respectively and a decrease of 6% in CO_2 emissions. The combined results of GHG gases would have implied a decrease of $3 \pm 2.8\%$ in terms of global warming potential. Regarding criteria pollutants, the conversion of gasoline vehicles to CNG produced an increase of 6% in NO_x emissions and decreases of the other pollutants in the following order: CO (64%) > NMVOC (35%) > SO_2 (18%) > PM (1%).

3.5. Reconstruction of activity data

The UBN index, which exhibited a symmetrical distribution and a high correlation with NVPC, was identified as the most suitable indicator for all regions under analysis (Fig. 8). The best fit is indicated by the following equation

$$\text{NVPC}_l = 0.424e^{-0.054\text{UBN}_l} \quad (3)$$

where, l denotes the geographical area of interest.

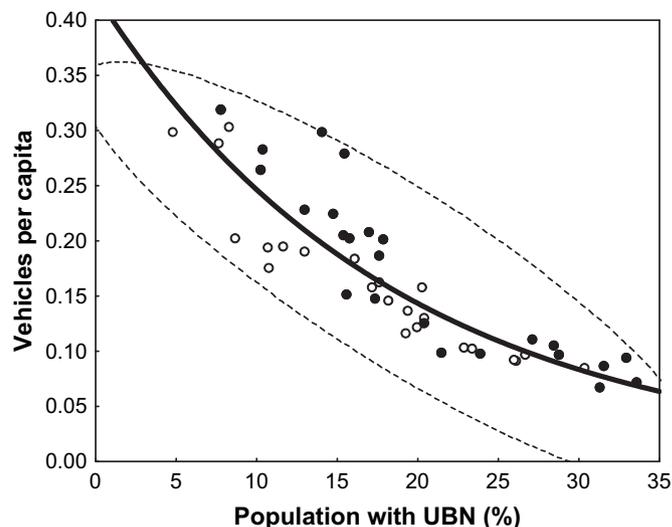


Fig. 8. Correlation between the number of vehicles per capita and the Unsatisfied Basic Needs index for the districts of the metropolitan area of Buenos Aires (black dots) and the provinces of Argentina (circles). The 95% ellipse confidence is shown in dotted line.

Regarding the other indicators, GGP by province correlated with NV ($r^2 = 0.98$). However, its low availability for smaller regions impairs its usage as a driving variable. In general, population, population density and HDI did not correlate with either NV or NVPC.

4. Conclusions

The on-road transport emission inventory for the metropolitan area of Buenos Aires for greenhouse gases and criteria pollutants was developed as part of an inter-American project aimed at chemical weather forecasting in South American megacities. Our inventory, which is presented for two spatial disaggregation levels, is to the best of our knowledge the first study with these characteristics for this megacity.

Within the scope of this research the relationship between the MABA emissions and those downscaled from the national inventory was established. Emissions of criteria pollutants computed at national level could be downscaled to the MABA region only by knowing the activity data. This approach may be applied to back-cast MABA emissions but also to calculate the present and past emissions for other cities in the country.

Our results highlight the role of older technologies accounting for almost 80% of the emissions of all species. As a consequence, the MABA exhibits higher specific emissions (ton TJ^{-1}) than developed countries, with figures two times higher for criteria pollutants. This finding indicates the need of systematically updating this inventory as the replacement of most polluting technologies would certainly have a major impact on the emission levels. Specific emissions (ton km^{-2}) confirm the MABA as a concentrated emission focus, as emissions at megacity level are 2–4 orders of magnitude higher than those at country level.

Regional EFs that better represent the higher emissions of the local fleet and the estimated uncertainties are an added value of this work. In most cases regional EFs differed significantly from those reported by developed countries that are usually employed as the basis to compute inventories in Latin America, thus conveying a significant underestimation of the emissions. Although our results could be improved with new measurements of local vehicles, the regional EFs of CO, NMVOC and NO_x for all categories of gasoline and CNG-fueled vehicles and the EFs of the mentioned gaseous pollutants and PM for diesel heavy-duty vehicles can be

used as reference for estimating emissions in Latin American cities. Our compilation indicates the need of acquiring local EFs of diesel light-duty vehicles and of non-CO₂ greenhouse gases for all vehicle categories. Our findings of time-lags in vehicle technologies between developed and developing countries may be useful for emission projections.

The environmental implications of the use of CNG are usually argued by many authors. Our scenario analysis of the replacement of gasoline by CNG showed that although CH₄ and N₂O emissions greatly increased, the decrease of CO₂ emissions implied an overall reduction of $3 \pm 2.8\%$ in terms of global warming potential. This result's uncertainty indicates the need of developing CH₄ and N₂O EFs to better estimate the impact of this fuel switch. The CNG introduction has been beneficial reducing emission levels of CO, NMVOC and SO₂; PM exhibited a small decrease and NO_x an increase.

An innovative approach for the determination of the registered vehicles by district was developed, showing a high correlation between the unsatisfied basic needs index with the number of vehicles per capita. This finding shows that the latter does not directly follow the income pattern of the population but a combination of socioeconomic indicators that is better reflected by this index.

The uncertainties associated with our results pave the road for future studies that may consider improving EFs data, particularly considering old, high-emitting vehicles that are very difficult to characterize with non-local data, together with the enhancement of activity data regarding km traveled, number of heavy-duty trucks and vehicular flow.

The relatively coarse spatial resolution and their static nature are the main drawbacks of our inventories. These features are a consequence of the lack of a traffic model for the MABA whose development was beyond the scope of this study. Such model would be crucial to better represent the dynamic effects of traffic activity on emissions and serve to generate appropriate temporal and spatial disaggregation schemes, which applied to our inventories would make them useful as input for regional dispersion modeling. The analysis performed in this study allows the computation of emission inventories for different cities in Argentina by generating a regional EF database, providing methodologies to infer the number of vehicles where it is not readily available and downscaling country to city level emissions. These tools may be directly used or adapted to compute inventories in other Latin American cities. They may also be employed to complement global emission inventories and for local policy makers that thus far have been lacking of such an environmental management tool.

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Appendix. Supplementary material

Supplementary data associated with this article can be found in the online version at doi:10.1016/j.atmosenv.2009.11.004.

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