An urban emissions inventory for South America and its application in numerical modeling of atmospheric chemical composition at local and regional scales

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SUMMARY

This work describes the development of an urban vehicle emissions inventory for South America, based on the analysis and aggregation of available inventories for major cities, with emphasis on its application in regional atmospheric chemistry modeling. Due to the limited number of available local inventories, urban emissions were extrapolated based on the correlation between city vehicle density and mobile source emissions of carbon monoxide (CO) and nitrogen oxides (NOx). Emissions were geographically distributed using a methodology that delimits urban areas using high spatial resolution remote sensing products. This numerical algorithm enabled a more precise representation of urban centers. The derived regional inventory was evaluated by analyzing the performance of a chemical weather forecast model in relation to observations of CO, NOx and O3 in two different urban areas, São Paulo and Belo Horizonte. The gas mixing ratios simulated using the proposed regional inventory show good agreement with observations, consistently representing their hourly and daily variability. These results show that the integration of municipal inventories in a regional emissions map and their precise distribution in fine scale resolutions are important tools in regional atmospheric chemistry modeling.

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1. Introduction

Urbanization has been one of the most significant trends in human activity throughout history. In the mid-19th century, only 1.7% of the global population lived in cities, but the urban population fraction grew to 50% by 2007. In South America, the data are even more striking: 82% of the population resides in urban areas, with an annual growth rate of 1.7% (UNFPA, 2007).

Population growth in large cities leads to air quality degradation at the local and regional scale, among other problems. Toxic trace gases emitted to the troposphere and their oxidation products represent a direct human health risk. In developing countries, this problem is generally aggravated by the age of the vehicle fleet, as well as intensive use of fossil fuels rather than low emission energy sources in both industry and the transport sector.

The local air quality effects of urban emissions and the regional impact of extensive remote urban areas have been the subjects of observational and modeling studies. The production of elevated near-surface levels of ozone is particularly worrisome. Clinical studies have related high ozone concentrations with reduced lung function (Abelson et al., 2002), and its phytotoxicity can compromise agricultural productivity and affect forest areas.

The air quality at a given city is driven by a combination of meteorological conditions and local emissions. To apply strategies for air quality improvement, it is necessary to quantify their impacts at local or regional scales. Some studies showed that regional and local air quality may also strongly depend on emissions at these scales (Schneider et al., 1997; Ponche and Vinuesa, 2004), therefore, the fine scale emissions inventories are very useful for air pollution prediction. It was shown, for the Expérience sur Site pour Contraindre les Modèles de Pollution atmosphérique et de Transport d’Emissions (ESCOMPTE) campaign over Marseilles city (France), that the use of an inventory with a higher resolution...
in space and time improve the primary and secondary pollutants estimation (Taghavi et al., 2005).

Characterizing and predicting local air quality and regional effects resulting from emissions in a large city requires a detailed survey of emissions sources. For example, for mobile sources, information about the vehicle fleet, traffic patterns, and street configuration must be considered. In addition, emission factors depend strongly on vehicle driving patterns, which can vary by location (Berkowicz et al., 2006). The methodology generally utilized to estimate mobile source emissions in large cities involves estimating the vehicular flux by counting vehicles or using transport models (Reynolds and Broderick, 2000). The difference between the development of municipal and national inventories lies in the level of detail of the input data, hypotheses, and analyzed parameters. National inventories require a broader approach for emissions estimation, as they encompass sources with larger geographic scales, including air and marine transport and the national energy grid. In South America, several efforts exist to survey these sources; particularly notable are the activities of source countries such as Argentina (Fundación Bariloche, 2005) and Brazil (http://www.mct.gov.br/index.php/content/view/17341.html) to construct national greenhouse gas inventories. However, these efforts are still preliminary, and the results have not been integrated.

With the increasing use of numerical atmospheric chemistry modeling on local, regional and global scales, the creation of inventories with wider coverage and analysis of the interaction among the various scales have become essential. In the past decade, various programs of international cooperation have emerged with the objective of providing integrated emission information on continental or global scales. Of note are the EMEP (Co-operative Programme for Monitoring and Evaluation of the Long-Range Transmission of Air Pollutants in Europe — http://www.ceip.at/) in Europe, and the Global Emissions Inventory Activity (GEIA) from the International Global Atmospheric Chemistry Programme (http://www.geiacenter.org/).

The spatial and temporal resolution of the global inventories is normally low, and therefore does not capture the specific characteristics of each region, principally with respect to the representation of urban centers (Gurjar et al., 2004). Through comparison of individual emissions estimates for the major megacities in the global inventories RETRO (REAnevalysis of TRopospheric chemical composition over the past 40 years — http://retro.enes.org), EDGAR (Emissions Database for Global Atmospheric Research — http://www.mnp.nl/edgar/) and IPCC-AR4 (Intergovernmental Panel on Climate Change — Fourth Assessment Report, Dentener et al., 2005), it was shown that large differences exist among these databases, principally due to methodological differences in the geographic distribution or the emissions aggregation. In certain cases, such as the city of Tokyo, the different inventories vary by a factor of two (Butler et al., 2008).

Even with the current initiative to construct finer resolution global emissions inventories, such as EDGAR 4.0 (http://edgar.jrc.ec.europa.eu/index.php), which has a spatial resolution of 10 km, it is necessary to develop methodologies to integrate local inventories into global emissions databases. This type of approach is even more necessary in regions which are typically poorly represented in global inventories due to scarcity of national inventories and measurement campaigns, such as is the case in the vast majority of countries in South America.

In 2006, the project SAEMC (South American Emissions, Megacities and Climate), financed by the Inter-American Institute for Global Change Research (IAI), established a collaboration network among Argentina, Brazil, Chile and Colombia, with the aims of creating updated emissions inventories, generating climate change scenarios for the South American continent with emphasis on the impact of megacities, and establishing a scientific basis for regional chemical weather forecasting. In this context, this work presents the consolidation of a regional urban emissions inventory for South America. This database integrates information from local inventories of vehicle emissions into existing global databases for the South American continent. The analysis procedures are described, including use of socio-economic data, extrapolation of emissions to cities lacking local inventories, and the geographic distribution of emissions at different spatial resolutions. Simulations were conducted with the operational chemistry model CCATT-BRAMS (Coupled Chemistry Aerosol and Tracer Transport model to the Brazilian developments on the Regional Atmospheric Modeling System, Freitas et al., 2009; Longo et al., in preparation) in order to evaluate the applicability of the regional urban emissions inventory to local and regional chemistry modeling.

2. Regional mobile source urban emissions inventory for South America

Although other anthropogenic sources of urban emissions exist, the focus of this work is vehicle emissions. In the metropolitan region of São Paulo, mobile sources are responsible for 95% of total carbon monoxide (CO) emissions and for 98% of total nitrogen oxides (NOx) emissions (CETESB, 2005). A similar relative contribution is found in other South American cities, such as Santiago—Chile (DICTUC, 2007) and Bogotá—Colombia (Zárate et al., 2007).

The regional scale mobile source urban emissions inventory presented here was developed in the CCATT-BRAMS emissions preprocessor (Freitas et al., 2010) by combining available local inventories, extrapolated values for cities without inventories, and the global inventories RETRO and EDGAR. The resulting inventory was then evaluated as described in Section 4.2. The RETRO inventory is a global emissions database with temporal resolution of 40 years (1960–2000), with monthly means evaluated on a grid with resolution of 0.5° × 0.5°. The anthropogenic sources include emissions from burning of fossil fuels and biofuels developed by the TNO (Netherlands Organization for Applied Research), combined with data on international maritime traffic from the VERITAS inventory (Endresen et al., 2003) and air traffic from the project ANCAT (http://www.mdcr.cz/en/Air+Transport/Environment/ecac.htm). The EDGAR inventory provides annual global emissions of greenhouse gases and precursors for the year 2000 on a 1° × 1° grid (Olivier and Berdowski, 2001). Both use national data from organizations such as the IEA (International Energy Agency), and thus are not completely independent (Butler et al., 2008). Their differences originate in estimates of emissions factors, the aggregation and spatial distribution methodologies, and the classification criteria for emissions sources.

2.1. Local vehicle emissions inventories

In order to evaluate the individual contribution of vehicle fleet emissions in large South American cities, information was used from local inventories for a few cities in the states of São Paulo and Rio de Janeiro in the Southeast Region of Brazil, the Metropolitan Region of Porto Alegre in the south of Brazil, the Metropolitan Region of Buenos Aires (Argentina), Santiago (Chile), and Bogotá (Colombia). The locations of the cities considered are shown in the map in Fig. 2, and the main characteristics of each inventory are shown in Table 1.

This study considers emission data from major South American cities with available inventories. For Brazil, the local inventories...
used are: the Metropolitan Region of São Paulo and neighboring cities (CETESB, 2005), which include Sorocaba, located 60 km to the west, São José dos Campos, with a population of about 540,000 inhabitants; the Metropolitan Region of Campinas, with approximately 970,000 inhabitants; the State of Rio de Janeiro (FEEMA, 2004), where the densest urbanized area of the Metropolitan Region of Rio de Janeiro encompasses approximately 700 km²; and the Metropolitan Region of Porto Alegre (Teixeira et al., 2008), located in the south of Brazil (Rio Grande do Sul state), which comprises 31 cities spread over an area of 9825 km².

Inventories were used from other South American cities, including the Metropolitan Area of Buenos Aires (MABA), comprised of the city itself and 24 surrounding districts that are part of the province of Buenos Aires, Argentina (D’Angiola et al., 2009); Santiago de Chile, located in the Chilean central valley, alongside the Andes (DICTUC, 2007); and Bogotá, Colombia’s capital and one of the largest urban centers in Latin America with approximately 8 million inhabitants (Behrentz et al., 2009).

### 3. Methodology

The construction of the regional mobile source urban emissions inventory consisted of three main steps: analysis of local inventories and correlation with socio-economic indexes, extrapolation of local information to other Brazilian cities without inventories and the spatial distribution of these emissions. The last two steps work automatically in the CCATT-BRAMS emissions preprocessor (see Fig. 1).

#### 3.1. Extrapolation of local information

Since vehicle emissions inventories exist for only a small fraction of medium and large South American cities, existing

<table>
<thead>
<tr>
<th>Inventory</th>
<th>Year</th>
<th>Reference</th>
<th>Methodology summary</th>
</tr>
</thead>
<tbody>
<tr>
<td>São Paulo, São José dos Campos, Campinas and Sorocaba</td>
<td>2005</td>
<td>CETESB (2005)</td>
<td>Bottom-up methodology. Some emission factors (EF) used are from the US Environmental Protection Agency (EPA) Compilation of Emission Factors (EPA, 1995), while the rest were obtained from laboratory analyses.</td>
</tr>
<tr>
<td>Porto Alegre</td>
<td>2004</td>
<td>Teixeira et al. (2008)</td>
<td>Bottom-up methodology. The EF were obtained from CETESB reports, which include EPA and laboratory data.</td>
</tr>
<tr>
<td>Rio de Janeiro</td>
<td>2004</td>
<td>FEEMA (2004)</td>
<td>Bottom-up methodology. The EF used are from the EPA.</td>
</tr>
<tr>
<td>Buenos Aires</td>
<td>2006</td>
<td>D’Angiola et al. (2009)</td>
<td>Bottom-up European COPERT methodology (Ntziachristos and Samaras, 2000). The EF were selected from different sources considering availability of data. Most were estimated using the COPERT IV model.</td>
</tr>
<tr>
<td>Santiago</td>
<td>2005</td>
<td>DICTUC (2007)</td>
<td>Bottom-up methodology (Corvalán et al., 2002). The EF used are from the COPERT III model (Ntziachristos and Samaras, 2000) and local measurements.</td>
</tr>
<tr>
<td>Bogotá</td>
<td>2006</td>
<td>Behrentz et al. (2009)</td>
<td>Bottom-up methodology. The EF were obtained from field campaign data, following the IVE project protocol.</td>
</tr>
</tbody>
</table>

![Fig. 1. Flow chart illustrating the regional mobile source urban emissions inventory construction methodology.](image)
inventories were extrapolated to other cities by identifying a correlation between socio-economic data and mobile source emissions of CO and NOx. Analyzed municipal data included the Human Development Index (HDI), Gross Domestic Product (GDP), urban population, and vehicle density, as well as other socio-economic indexes.

3.2. Spatial distribution of urban emissions

The high resolution spatial representation of the regional database was improved, including both available inventories updated with vehicle emissions information and the extrapolated emissions for other cities. The objective of this representation was to generate a consistent inventory for local and regional scales, as well as to enable evaluation of the impact of this representation on the regional scale.

In the original version of the CCATT-BRAMS emissions preprocessor, data from the global RETRO/EDGAR inventories are interpolated to the model grid using a "nearest neighbor" scheme, and the urban boundary is not represented with detail. In this work, a new scheme is proposed in which urban areas are identified with a numerical algorithm based on Graham (1972), which identifies points pertaining to a pre-defined range of values, making a sweep from a central point, generally defined as the center of the urban area. This algorithm was adapted to read the soil cover map derived by the GLCF (Global Land Cover Facility – http://glcf.umiacs.umd.edu/data/landcover) from images from the AVHRR (Advanced Very High Resolution Radiometer) instrument onboard the NOAA satellite, identifying urban areas using Band 13 (urban and construction). The algorithm developed by Graham (1972) is applied as a numerical solution for obtaining a convex polygon. Given a finite set \( S = \{S_1, \ldots, S_n\} \), a point \( P \) in the interior of this plane is identified. Each \( S_i \in S \) is expressed in polar coordinates, with origin in \( P \) and angle 0, and compared with its nearest neighbors, which are determined by their angle with relation to the point \( P \).

Depending on the resolution, distinct but extremely close cities can appear to be merged, and thus are represented as a single urban area. This can be resolved by applying a radius of influence which limits the application of the algorithm to the effective urban area of the municipality. Afterwards, the Ray Casting (Snyder and Barr, 1987) algorithm is applied to verify which grid points are located inside the obtained reference polygon, and emissions are then distributed within this area. This methodology, from now on refers as AREA DELIMITER scheme, also permits the distribution of emissions on areas defined by other geo-referencing processes, and thus is applicable at various resolutions.

3.3. Evaluation of the inventory

In order to evaluate the process of integration and distribution of mobile source emissions in the regional inventory, its implementation in a regional atmospheric chemistry model is analyzed. Numerical experiments were conducted using the modeling system CCATT-BRAMS (Longo et al., in preparation). CCATT is an Eulerian atmospheric transport model that predicts trace gas mixing ratios by solving the mass conservation equation, which includes terms for advection, turbulence in the Planetary Boundary Layer (PBL), wet and dry deposition, plume rise, and shallow and deep convection. In addition, the model includes chemical reactions and interaction of aerosols with solar and long wave radiation. The CCATT-BRAMS system can be configured with virtually any chemical mechanism using a modified version of the SPACK (Simplified Preprocessor for Atmospheric Chemical Kinetics, Djoud et al., 2002) preprocessor. The Regional Atmospheric Chemistry Mechanism (RACM, Stockwell et al., 1997), with 70 species and 237 kinetic and photolysis reactions, was used for this run. The numerical integrator of the chemical mechanism is based on the Rosenbrock method (Hairer and Wanner, 1991); this simulation used the third order version RODAS 3, with four iterations. Photolysis rates were calculated on-line using the FAST-TUV (Tie et al., 2003) model. Dry deposition follows the resistance formulation and accounts for aerodynamic, quasi-laminar layer and canopy resistances (Wesely, 1989; Seinfeld and Pandis, 1998).

In addition to anthropogenic emissions, the CCATT-BRAMS emissions preprocessor includes emissions from biogenic or natural sources according to GEIA/ACCENT Activity on Emission Databases (http://www.aero.jussieu.fr/projet/ACCENT/description.php), and from biomass burning as estimated by the Brazilian Biomass Burning Emissions Model (3BEM) (Freitas et al., 2005; Longo et al., 2007).

The numerical experiments are representative of the summer (month of January), winter (month of July) and spring (month of October) seasons in the Southern Hemisphere, for the year 2005. Two simulations were conducted per period, the first with anthropogenic emissions based on the RETRO/EDGAR inventories, called CONTROL, and the second using the regional vehicle emissions inventory generated in this work, called SA-INV. Three grids with 80, 20 and 5 km horizontal spacing were used in a nested configuration with 2-way interaction. The 80 km grid covers an extensive area of South America, between latitudes 145 and 325 and longitudes 37W and 55W. The 20 km grid covers primarily the Southeast and Center-West regions of Brazil. The finest grid covers the states of Minas Gerais and São Paulo, including their respective capitals, Belo Horizonte and the Metropolitan Region of São Paulo. The experiments were conducted with 35 vertical grid levels. The thickness of the first level was 100 m, increasing in the above levels in a geometric progression with a rate of 1.1. Both simulations use initial and boundary conditions from the global model T26L28 (CPTEC/INPE). An average vertical profile for CO, NO, NO2, Ozone, HNO3 and PAN was used as the chemistry initial condition, based on a climatological analysis of the global model MOCAGE (Multi-scale Chemistry and Transport Model – Josse et al., 2004; Teyssèdre et al., 2007) for the South American continent.

For evaluation of simulation results, observations of O3, CO and NOx from the Metropolitan Regions of São Paulo (CETESB Air Quality Information System – http://www.cetesb.sp.gov.br/Ar/ar_qualar.asp) and Belo Horizonte (Minas Gerais State Environment Foundation – http://www.steam.br) were used.

4. Results and discussion

4.1. Urban emissions inventory for South America

Among the various socio-economic indexes that were analyzed (Table 2), the CO and NOx emission fluxes correlate best with vehicle density,\(^1\) with Pearson correlation indexes of 0.92 and 0.93 and determination coefficients of 0.88 and 0.95, respectively. Thus, emissions for other Brazilian cities were extrapolated as a function of vehicle density according to the correlation curve shown in Fig. 2. CO and NOx are present in all the local inventories and could be analyzed in more detail. Although some cities with inventories made available total hydrocarbon emissions, there is not sufficient information about how these species were grouped, making it

\(^1\) Data on the 2004 vehicle fleet for Brazilian cities were obtained from the municipal database of the IBGE (Brazilian Institute of Geography and Statistics – http://www.ibge.gov.br/). For the Metropolitan Region of Buenos Aires, the total vehicle fleet was estimated with the database of D’Angiola et al. (2009). For the city of Santiago, the information is from the INE (National Institute of Statistics – http://www.ine.cl) and for Bogotá, the information estimated by Behrenz et al. (2009) was used.
difficult to compare them and extrapolate to the other cities. Also, the RETRO and EDGAR global inventories use different treatments for non-methane hydrocarbons (NMHC); while the EDGAR database aggregates them in a single group, RETRO divides them into 22 distinct species categories. Therefore, for hydrocarbons and other chemical species, correction factors were applied for each grid point based on the ratio between the original and updated CO emissions. The ratios of emission of non-methane volatile organic compounds (NMVOC) with relation to those of NOx from the original global inventories presented a difference on the order of 15% with relation to the local emissions. Thus, the values of NMVOC were adjusted by a 1.15 factor in the regional database proposed in this work.

The impact of the inclusion of information from local inventories in the regional map and extrapolation to Brazilian cities without inventories is more evident in medium and large urban areas in the Central, South and Southeast regions of Brazil, which contribute the most to GDP and thus have a higher vehicle density (Fig. 3). Another factor that contributes to this difference, principally in the Southeast region, is the discrepancy between the local inventory information and the global RETRO and EDGAR databases (Table 3). For example, NOx emissions for São Paulo in the CETESB inventory and the global databases vary by a factor of 2. Total CO emissions for Buenos Aires and Bogotá do not differ much among the inventories, but present a very distinct spatial distribution in the two cities. Due to their size of their urban areas, the urban area detection methodology can be applied at horizontal resolutions of 20 km. In finer grids, the spatial distribution methodology based on the urban area analysis results in a more realistic representation of the urban contribution. Fig. 4 shows the updated pre-processed emissions on the 5 km grid and the satellite image with a resolution of 1 km. The level of detail of the representation of the urban area will depend on the spatial resolution of the grid and the number of points used in the construction of the convex polygon over which the emissions will be distributed.

4.2. Evaluation of the regional inventory

Large quantities of chemical compounds are emitted to the atmosphere by anthropogenic and biogenic processes. In the atmosphere, these trace gases are subject to various transport mechanisms and complex physical—chemical transformations, the latter being responsible for the formation of secondary pollutants. Therefore, it is important to evaluate the dynamics and

<table>
<thead>
<tr>
<th>INDEX</th>
<th>CO (kg m$^{-2}$ s$^{-1}$)</th>
<th>NOx (kg m$^{-2}$ s$^{-1}$)</th>
</tr>
</thead>
<tbody>
<tr>
<td>Human Development Index</td>
<td>0.42</td>
<td>0.34</td>
</tr>
<tr>
<td>Gross Domestic Product (dollars)</td>
<td>0.64</td>
<td>0.74</td>
</tr>
<tr>
<td>Urban population (millions inhabitants)</td>
<td>0.62</td>
<td>0.58</td>
</tr>
<tr>
<td>Population density (inhabitants km$^{-2}$)</td>
<td>0.83</td>
<td>0.89</td>
</tr>
<tr>
<td>Vehicle density (vehicles km$^{-2}$)</td>
<td>0.88</td>
<td>0.95</td>
</tr>
</tbody>
</table>

Table 2

Relation between CO/NOx emissions and socio-economic index (determination coefficients).
thermodynamics of the model, through its representation of micro- and meso-scale meteorological phenomena. The resulting emissions in the updated inventory are evaluated by comparison of the mixing ratios of CO, NOx and O3 simulated by the model with direct observations.

4.2.1. Meteorological conditions and evaluation of model performance in the simulation of general atmospheric conditions

January 2005 was marked by three active episodes of the South Atlantic Convergence Zone (SCAZ), triggering an increase in precipitation in some states, including São Paulo and southern Minas Gerais. In July, the average monthly precipitation was low in Southeast Brazil, particularly in the North and Center-West of the state of Minas Gerais. Values slightly above the mean were observed in the north of São Paulo, due to the approach of cold fronts originating in the South Pole. The month of October was marked by above average precipitation in the state of São Paulo, but precipitation was below historical values in the states of Rio de Janeiro and Minas Gerais (CPTEC — http://climanalise.cptec.inpe.br/%7erclimanl/boletim). In general, the simulations represented well the spatial distribution of precipitation, but overestimated its intensity. The monthly total precipitation simulated for the month of January (Fig. 5B), principally in the Center-South of Brazil, showed good agreement with the observed total (Fig. 5A).

The experiments also show good performance in simulating maximum and minimum temperatures. Fig. 6 shows daily minimum and maximum temperatures for January 2005 as observed at the Campo de Marte airport (SBMT) in the city of São Paulo and simulated in the experiment AS-INV. Since pollutant removal and dispersion processes are strongly affected by the behavior of the Planetary Boundary Layer (PBL), the PBL height simulated on the finest spatial resolution grid is also evaluated. The PBL height was estimated from potential temperature and humidity radio-sounding profiles conducted every 12 h at the SBMT station. In general, the simulations represented well the daily variability of the PBL. The average height simulated for the month of January was on the order of 950 m, while observed values varied between 800 and 1500 m, with an average of 1087 m.

4.2.2. Comparison of simulated CO, NOx and O3 mixing ratios with observations in urban areas

CO and NOx mixing ratios simulated by the model were compared with observed values. In general, CO and total NOx = NO + NO2 are little influenced by chemistry on a time scale of a few hours and are therefore good indicators for evaluation of emissions (Zárate et al., 2007). The new regional inventory clearly affected the simulated CO and NOx mixing ratios, and resulted in a significant improvement in model performance in relation to the observations (Figs. 7 and 8). In Figs. 7 and 8 the error bars represent the standard deviations of the mean observed and simulated values, which were calculated differently for each of the analyzed cities. For São Paulo the standard deviation measures the spatial variability, that is, represents the average values of all the stations in the metropolitan area. For the city of Belo Horizonte, which has only one surface station, the standard deviation represents the variability of the average hourly values.

The diurnal cycle of CO and NOx is also well represented, with maximum concentrations corresponding to the hours of most intense traffic, both in the Metropolitan Region of São Paulo and the city of Belo Horizonte. However, the removal of these species during the daytime, which is the period of maximum convective activity, was overestimated, principally during the month of October. The experiments using the original emissions showed low agreement with the observations for two primary reasons: the poor spatial representation of emissions at finer resolutions, and

### Table 3

<table>
<thead>
<tr>
<th>Cities</th>
<th>CO (Gg year⁻¹)</th>
<th>NOx (Gg year⁻¹)</th>
</tr>
</thead>
<tbody>
<tr>
<td></td>
<td>莉</td>
<td>RETRO</td>
</tr>
<tr>
<td>São Paulo</td>
<td>1464</td>
<td>1039</td>
</tr>
<tr>
<td>São José dos Campos</td>
<td>43</td>
<td>25</td>
</tr>
<tr>
<td>Campinas</td>
<td>282</td>
<td>151</td>
</tr>
<tr>
<td>Porto Alegre</td>
<td>196</td>
<td>158</td>
</tr>
<tr>
<td>Sorocaba</td>
<td>53</td>
<td>40</td>
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<tr>
<td>Rio de Janeiro</td>
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<td>Buenos Aires</td>
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<td>463</td>
</tr>
<tr>
<td>Bogotá</td>
<td>459</td>
<td>457</td>
</tr>
</tbody>
</table>

Fig. 3. Emissions of CO (× 10⁻⁶ kg m⁻² day⁻¹) from original (A) and extrapolated (B) inventories on a 20 km grid covering South America.
underestimated emissions values. The diurnal cycle of ozone was well represented in both experiments, with maximums around 14:00 local time in the two analyzed cities. Although CO and NOx mixing ratios are systematically underestimated in the CONTROL experiment, O3 values are in general well represented, except for slight overestimation during the nighttime (Fig. 9).

The O3 mixing ratios in both experiments are of the same order because of the non-linearity of the impact of NOx on the ozone concentration. Ambient NOx and NMHC mixing ratios are directly related to the instantaneous rate of production of O3, but its exact dependence varies with the assumptions and conditions used in generating the isopleth plot. With the increasing of NOx mixing ratio above a threshold, which depends on the NMHC mixing ratio, ozone production decreases.

The comparison of observed maximums and minimums with values simulated by the experiment SA-INV, shown in Figs. 10–12,
demonstrates that the model consistently represented the daily variability of concentrations in the Metropolitan Region of São Paulo.

4.2.3. Impact of the inclusion of local emissions on the regional distribution of CO, NOx and Ozone

The addition of vehicle emissions from local South American inventories in the global database caused a significant impact on the simulated spatial distribution of CO and NOx emissions from large urban centers and on the ozone generated from chemical reactions. The regional and global scales impact of megacities has been much discussed recently (ex. Guttikunda et al., 2003; Lawrence et al., 2007; Butler and Lawrence, 2009) and a good spatial representation of emissions is of extreme importance in the study of these impacts. Fig. 13 shows the percentage difference in

![Fig. 7. Hourly average CO mixing ratios observed and simulated in the experiments CONTROL and SA-INV in the Metropolitan Region of São Paulo and in the city of Belo Horizonte.](image)

![Fig. 8. Hourly average NOx mixing ratios observed and simulated in the experiments CONTROL and SA-INV in the Metropolitan Region of São Paulo and in the city of Belo Horizonte.](image)
the monthly average maximum concentration of CO, NOx and ozone during peak hours for the month of October, as simulated by the experiments CONTROL and SA-INV on the 80 km grid, superimposed on the average monthly wind field. The percentage difference is more evident near the major cities of the Central, South, and Southeast regions. The inclusion of local inventories and extrapolation to other Brazilian cities impacted the spatial distribution of CO and NOx concentrations by more than 25% in extensive areas around the large urban centers. For O3, the percentage difference was most evident in the Southeast region of Brazil,

![Graphs showing CO mixing ratios](image)

**Fig. 9.** Hourly average O₃ mixing ratios observed and simulated in the experiments CONTROL and SA-INV in the Metropolitan Region of São Paulo and in the city of Belo Horizonte.

![Graphs showing CO mixing ratios](image)

**Fig. 10.** Maximum and minimum daily average CO mixing ratios observed and simulated in the experiment SA-INV in the Metropolitan Region of São Paulo.
Fig. 11. Maximum and minimum daily average O₃ mixing ratios observed and simulated in the experiment SA-INV in the Metropolitan Region of São Paulo.

Fig. 12. Maximum and minimum daily average NOx mixing ratios observed and simulated in the experiment SA-INV in the Metropolitan Region of São Paulo.
where emissions of ozone precursors in the new inventory are significantly higher than in the RETRO and EDGAR global inventories. The patterns of percentage differences for the months of July and January were similar to those of the month of October.

5. Conclusions

In general, the global scale emissions inventories are lacking in detailed representation of local scale emissions, primarily in regions with little available data, as is the case in South America. It is therefore an important task to incorporate local inventories and data into the global databases. In the present study, a methodology is discussed for the construction of a regional inventory for the South American continent, with the aim of incorporating information from the municipal inventories in the global RETRO/EDGAR emissions maps. The first challenge lies in the low number of municipal inventories, due to the rapid urban expansion in South America. This problem was resolved by extrapolating the available information to municipalities without inventories, with the aim of constructing a more complete and consistent regional database. Vehicle emissions of CO and NOx from available local inventories were extrapolated to locations without inventories based on correlations with vehicle densities.

Nevertheless emission inventory at national level also depends upon many factors such as vehicle technology, socio-economic characteristics, transport policy etc... This information is intrinsically included in each respective local inventory. Therefore, the fitting function of the local inventories points should contain this information at least in a first approach. Also, at this stage, the extrapolation was performed only for the Brazilian cities, because of the easier data accessibility and since a reasonable homogeneity of the local characteristics is expected. Meanwhile, this work has been extended to extrapolate these data to other South American cities without inventories, and maybe some factors will have to be added in order to better represent the local characteristics for different countries.

A comparison between vehicle emissions from local and global inventories in the analyzed South American cities showed considerable differences, confirming what has been discussed in other studies.

A method for detection and analysis of the urbanized area of a city was proposed and incorporated in the emissions preprocessor of the CCATT-BRAMS model, which allowed a more precise distribution of emissions at fine resolutions. An algorithm was applied to obtain a convex polygon based on an analysis of the urban class of a soil coverage remote sensing product, which was then used to distribute vehicle emissions. This tool is automatic and applicable at any resolution. The radius of influence prevents neighboring cities from being represented as a single area, which is extremely important in densely populated areas. This method permits the consistent use of the same emissions inventory from the local to large scale.

The results of the evaluation of the regional inventory, through analysis of CO, NOx and O3 mixing ratios in the cities of São Paulo and Belo Horizonte, showed a significant gain in the numerical representation of the local atmospheric chemistry composition, suggesting that the updating of the global database with local inventories is essential in order to obtain a good coupling between local and regional scales. The overestimation of simulated NOx and O3 values for January (which are summer holidays with reduced vehicle traffic in São Paulo) in the experiment AS-INV suggest the need to include monthly and weekly variability factors, influenced primarily by the patterns of urban traffic on these time scales. It is noted that for Belo Horizonte, the emissions values in the regional inventory are extrapolated, because no local inventory is available for this city. The good performance of the experiment suggests a good effectiveness of the extrapolation technique.

Regional modeling is an important tool in the analysis and prediction of the chemical composition of the atmosphere. The use of emissions databases that integrate specific information about urban centers into broader databases is shown to be relevant for a good representation of emissions in chemical weather forecasting models on local and regional scales. It is demonstrated that the inventory proposed in this work improves the performance of atmospheric chemistry simulations on the local scale and significantly affects the regional spatial distribution of tropospheric O3 and its precursors. The tool proposed in this work enables a good distribution of emissions at various levels of detail, and can be consistently applied to a wide range of spatial resolutions. In the future, the extrapolation will be expanded to additional South American cities without inventories that were not included in this first version.

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