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Multispecies and high spatiotemporal resolution database of vehicular emissions in Brazil

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Abstract – In this article, we present the BRAzilian Vehicular Emissions inventory Software (BRAVES) database, a multispecies and high spatiotemporal resolution database of vehicular emissions in Brazil. We provide this database using spatial disaggregation based on road density, temporal disaggregation using vehicular flow profiles, and chemical speciation based on SPECIATE database from the United States Environmental Protection Agency. Our BRAVES database provides hourly and annual emissions of 41 gaseous and particle pollutants. Users can define the spatial resolution, which ranges from a coarse to very refined scale Spatial correlation analysis reveals that the BRAVES database reaches similar performance to the vehicular emissions inventory from Emissions Database for Global Atmospheric Research (EDGAR). A comparison with Modern-Era Retrospective analysis for Research and Applications, Version 2 (MERRA-2) surface concentration confirms the consistency and reliability of the BRAVES database on representing the spatial pattern of vehicular emissions. Compared to EDGAR, the BRAVES database brings more spatial, temporal, and chemical details. These additional features are crucial to understanding important atmospheric chemistry processes in Brazil. All codes and inputs are freely available, and the outputs are compatible with the input requirements of sophisticated chemical transport models. We envision that our database will enable the scientific and environmental community to gain new insights into vehicular emissions and their effects in Brazil, where emissions inventories are scarce and urgently needed.

Keywords: Vehicular emissions, temporal disaggregation, chemical speciation, spatial disaggregation, Brazil.

1. Introduction

Vehicular emissions threaten urban air quality (Brito et al. 2018; Sawyer, 2010) and cause several environmental damages from local to global scales. These emissions deleteriously affect human health (Anenberg et al. 2017; Krzyzanowski et al. 2005) and contribute to the increase in the concentration of greenhouse gases in the atmosphere (Shindell et al. 2011; Unger et al. 2010).

It is challenging to control vehicular emissions in developing countries where city growth is disorganized manner and vehicle population increases dramatically (Lyu et al. 2020; Sun et al. 2020). Furthermore, vehicular emissions inventories, an essential tool to control air pollution, are scarce and limited to wealthy cities (Huneeus et al. 2020) and developed countries. When available, an emissions inventory often does not provide the required data to design air quality management systems.

Brazil has experienced a rapid rise in its vehicular fleet (Carvalho et al. 2015) and transport volume. Even though the program to control vehicular emissions has reduced the emissions from the transport sector in Brazil (Andrade et al. 2017), vehicles are still potentially the dominant source of air pollution in several municipalities.

The impact of vehicular emissions in many Brazilian municipalities is still unknown (Ribeiro et al. 2021). Current inventories provide only annual emissions from national to municipality scales, not reaching the spatial and temporal resolution necessary for air quality modeling (Álamos et al. 2022), nor the concentration of chemical species that participate in chemical reactions in the atmosphere. For this reason, most regional air quality assessments in Brazil rely on global emissions inventories, which have been proved to be biased against local inventories. Also, global inventories do not present enough spatial and temporal resolution for regional and local studies (Ibarra-Espinosa et al. 2018). Even in the megacity of São Paulo, where the air quality network is well developed and multiple inventories have been developed, there is still room for improvement in emissions inventories (Andrade et al. 2017), especially regarding chemical species involved in the photochemical process in the atmosphere.

In this article, we present the first comprehensive multispecies high spatiotemporal resolution database of vehicular emissions for the entire Brazilian territory. The BRAzilian Vehicular Emissions inventory Software (BRAVES) database has spatial disaggregation based on road density, temporal disaggregation using vehicular flow profiles, and chemical speciation from the US EPA SPECIATE database. The BRAVES database provides hourly and annual emissions of 41 gaseous and particle pollutants. Users can define the spatial resolution from coarse to very refined scales. The emissions are derived from the BRAVES model (Vasques and Hoinaski, 2021), which uses a probabilistic approach that accounts for the fleet characteristics, fuel consumption, vehicle deterioration, and intensity of use, to calculate the vehicular emissions from the exhaust, tires, roads, brake wear, soil resuspension, refueling, and evaporative emissions. Here, we present methods and a comparison between the BRAVES database and independent databases. We also make all codes and inputs freely available.

2. Vehicular emissions data

Our database uses output data from the BRAzilian Vehicular Emissions inventory Software – BRAVES (Vasques and Hoinaski, 2021), which employs a probabilistic bottom-up method to estimate vehicular emissions aggregated by municipality (Figure 1). BRAVES estimates the vehicular emissions from the exhaust, tires, roads, brake wear, soil resuspension, refueling, and evaporative emissions. The software provides, by fleet category (i.e., commercial-light vehicles, motorcycles, light-duty and heavy-duty vehicles), annual emissions of carbon monoxide (CO), carbon dioxide (CO_2), methane (CH_4), hydrocarbons (HC), aldehydes (RCHO), non-methane volatile organic compounds (NMVOC), nitrogen oxides (NOx), particulate material (PM), nitrous oxide (N_2O), and sulfur oxide (SO_2). Throughout this article we call current database the BRAVES database. Codes and outputs from BRAVES are available by registering at https://hoinaski.prof.ufsc.br/BRAVES/ and https://github.com/leohoinaski/BRAVES, where users can access instructions to run the database and download the input files. The outputs are generated in netCDF format and with annual or hourly resolutions.

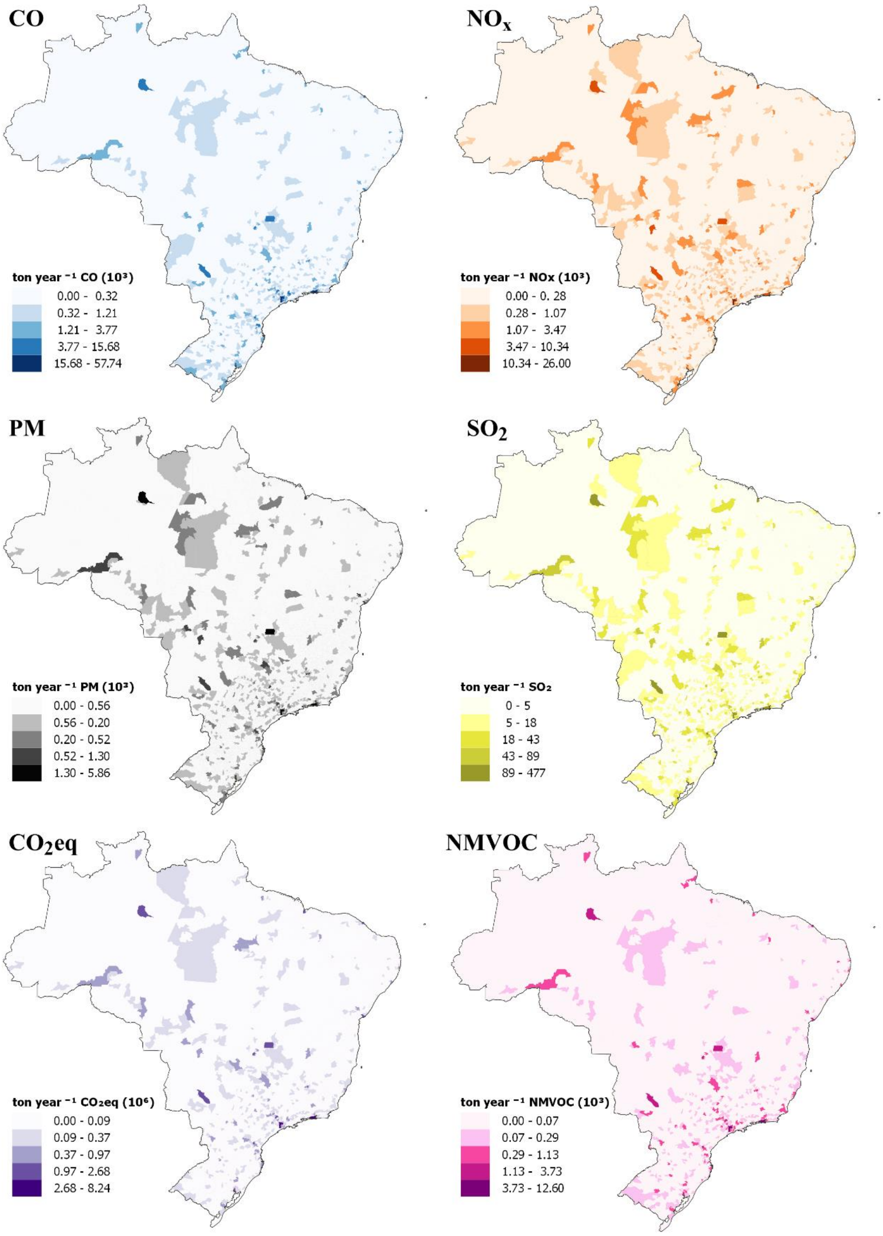

Figure 1. Vehicular emissions inventory in 2019 of a) CO, b) NOx, c) PM, d) SO_2, e) CO_2, and f) NMVOC provided by BRAVES (Vasques and Hoinaski, 2021).

3. Spatial disaggregation

Since vehicular emissions from BRAVES are aggregated by municipality, we use a road density approach to distribute the emissions of each municipality in pixels with user-defined resolution. Previous work of Tuia et al. (2007) and Gomez et al. (2018) shows that road density is one of the most reliable approaches to disaggregate vehicular emissions. In this article, road density factor ($RD_{p,m}$) is calculated by the sum of road lengths (L_p) on each pixel (p) divided by the total road length (L_m) inside a municipality (m) (Equation 1). Road shapefile data derived from OpenStreetMap (www.openstreetmap.org) can be downloaded at (https://download.geofabrik.de/south-america/brazil.html) for Brazilian territory. Figure 2 shows the spatial distribution of $RD_{p,m}$ in Brazil. Multiplying $RD_{p,m}$ by the vehicular emissions in each municipality derived from BRAVES provides the spatialized emission ($E_{p,m,c}$) of compound c in pixel p within a municipality m (Equation 2). We provide a parallelized method to estimate the road density in Brazil at [https://github.com/leohoinaski/BRAVES.](https://github.com/leohoinaski/BRAVES)

$$RD_{p,m} = \frac{L_p}{L_m} \qquad \text{(Eq.1)}$$

$$E_{c,p,m} = RD_{p,m} \times E_{c,m} \qquad \text{(Eq.2)}$$

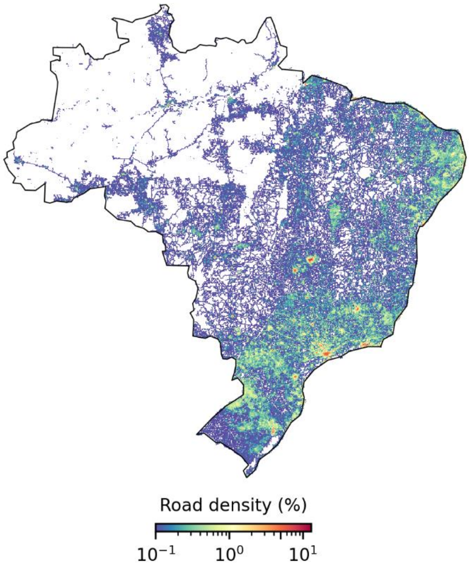

Figure 2. Road density factor in Brazil.

Figure 3 shows the annual emissions of aldehydes and CO from 2019 in Brazil by fleet category using spatial distribution based on the road density approach. Hotspots of vehicular emissions are concentrated in urbanized areas in Brazil (Figure 3). Among fleet categories, heavy-duty is the major emitter of aldehydes, while light-duty emits the most CO. Vasques and Hoinaski (2021) presents a full comparison of vehicular emissions from each fleet category in Brazil using BRAVES. Figures SM1 to SM5 demonstrate the BRAVES database by Brazilian state.

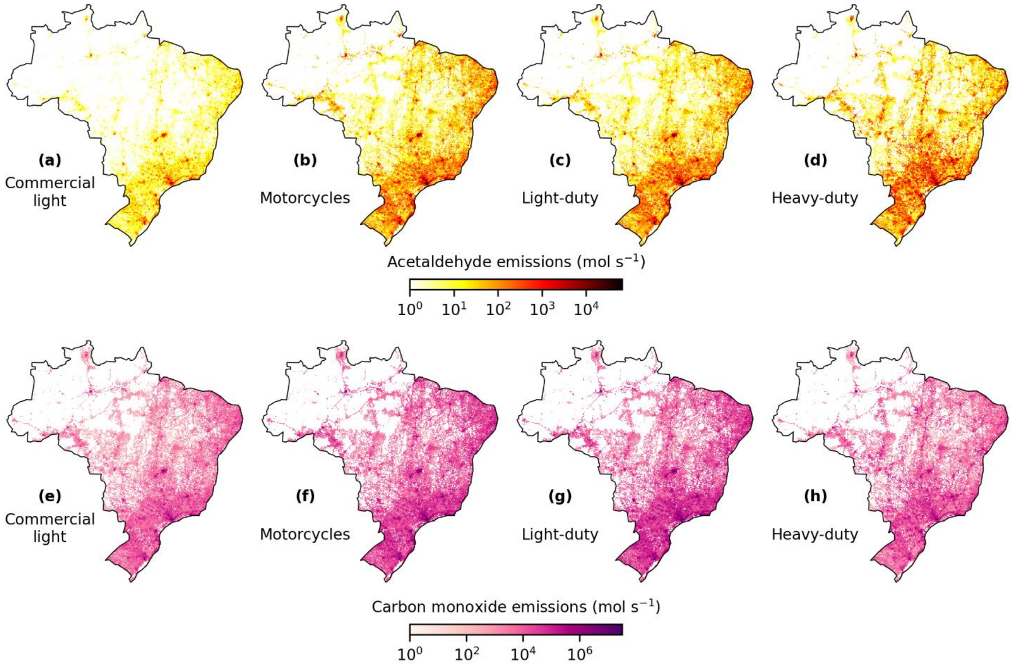

Figure 3. Vehicular emissions of aldehydes a-d) and carbon monoxide e-h) from commercial light, motorcycle, light, and heavy-duty fleets in 2019 in Brazil.

4. Temporal disaggregation

Temporal disaggregation based on traffic flow observations from Environment and Water Resources Institute from Espírito Santo state (IEMA ES, 2019) splits original annual emissions from BRAVES into hourly basis emissions. In this article, the temporal disaggregation factor (Figure 4) is composed of hourly, weekly, and monthly traffic factors. Hourly emissions ($E_{c,p,m,h}$) of each air pollutant are obtained by the multiplication of $E_{p,m,c}$ and the temporal disaggregation factor (T_f) (Equation 3).

$$E_{c,p,m,h} = T_f \times E_{c,p,m} \quad \text{(Eq.3)}$$

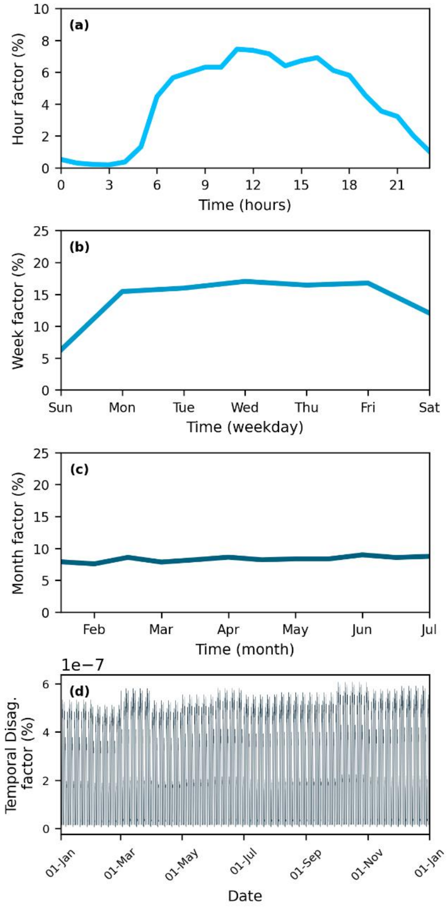

Figure 4. Temporal disaggregation factor and its components a) Hourly factor, b) Weekly factor, c) Monthly factor, d) Annual to hourly basis temporal disaggregation factor.

5. Chemical speciation

We use data from SPECIATE 5.1 (US EPA, 2020; Eyth et al. 2020) from the United States Environmental Protection Agency - US EPA (https://www.epa.gov/air-emissions-modeling/speciate) to speciate emissions of chemical constituents of Volatile Organic Compounds (VOC) and PM, which has not been previously estimated by BRAVES. The chemical speciation method also converts NOx to NO and NO_2.

Regarding the speciation procedures, in this article we group light duty and commercial light vehicles, and motorcycles together as light vehicles. We select profiles to speciate PM emissions from the exhaust of heavy and light vehicles, soil resuspension (road dust), tire wear, and brake wear. VOC emissions from the exhaust and evaporative process are also speciated. Table SM6 in the supplementary material summarizes the profiles from SPECIATE 5.1 used in the chemical speciation. We target the species required for Carbon Bond Chemical Mechanism (Yarwood et al. 2010ab) version 6 (CB06), which describe tropospheric oxidant chemistry in a concise manner suitable for use in complex 3-dimensional atmospheric models (Yarwood et al. 2010ab). Table 1 presents the VOC and PM compounds considered in the chemical speciation in this database.

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Table 1. List of chemical species included in the BRAVES database (Yarwood et al. 2010b).

Specie	**ID**	**Specie**	**ID**
Acetone	ACET	Aluminum	PAL
total Acrolein	ACROLEIN	Calcium ion	PCA
total Acetaldehyde	ALD2	Chloride ion	PCL
Benzene	BENZ	Elemental carbon	PEC
1,3-Butadiene	BUTA13	Iron	PFE
Ethanol	ETH	Potassium ion	PK
Ethane	ETHA	Magnesium ion	PMG
Ethyne	ETHY	Manganese	PMN
Formaldehyde	FORM	Sodium ion	PNA
Isoprene	ISO	Ammonium	PNH4
Naphthalene	NAPH	Nitrate	PNO3
Propane	PRPA	Silicon	PSI
Monoterpenes	TERP	Sulfate	PSO4
Toluene	TOL	Titanium	PTI
xylene	XYLMN		

The chemical speciation factors employed to split VOC and PM emissions are calculated by the average of the weighting percentage of the corresponding species from SPECIATE 5.1. We consider exhaust, evaporative, and particulate emissions of light and heavy vehicles. Figures 5 and 6 show the speciation factor used to generate the database. Multiplication factors of 0.495 and 0.505 derived from SPECIATE 5.1 convert NOx emissions to NO and NO_2, respectively. Table SM7 summarizes the speciation factors used to build this database.

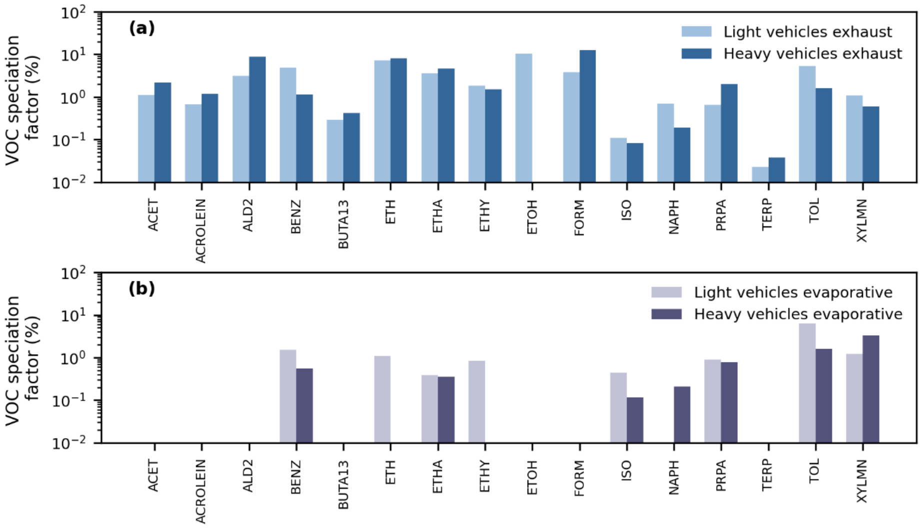


Figure 5. VOC chemical speciation factor for exhaust and evaporative emissions from light and heavy vehicles used in this article.

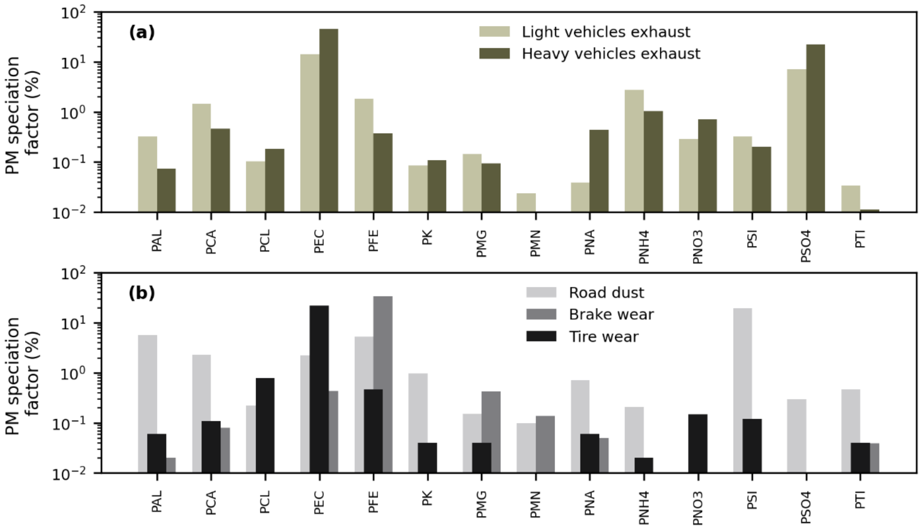


Figure 6. PM chemical speciation factors a) exhaust emissions from light and heavy vehicles b) road dust resuspension, brake wear, and tire wear.

6. The database and codes

The database contains hourly emissions of 41 chemical species, such as ACET, ACROLEIN, ALD2, BENZ, BUTADIENE13, CH_4, CO, CO_2, ETH, ETHA, ETHY, ETOH, FORM, ISO, N_2O, NAPH, NO, NO_2, PAL, PCA, PCL, PEC, PFE, PK, coarse mode primary PM (PMC), PMG, PMN, unspeciated $PM_{2.5}$ (PMOTHR), PNA, PNH_4, PNO_3, POC, PRPA, PSI, PSO_4, PTI, SO_2, TERP, TOL, VOC, and XYLMN. We provide a code to generate hourly resolved files with a user-defined grid for a single or whole group of species (https://github.com/leohoinaski/BRAVES). These files are compatible with the input requirements of sophisticated chemical transport models, such as the Community Multiscale Air Quality Model (CMAQ), the Weather Research and Forecasting (WRF) model coupled with Chemistry (WRF-Chem), the Comprehensive Air Quality Model with Extensions (CMAx), and others. Smaller domains and finer resolution can be easily created by modifying in the python codes. Figure 7 shows the vehicular emissions of Benzene in Brazil on January 1st, 2019 using the BRAVES database.

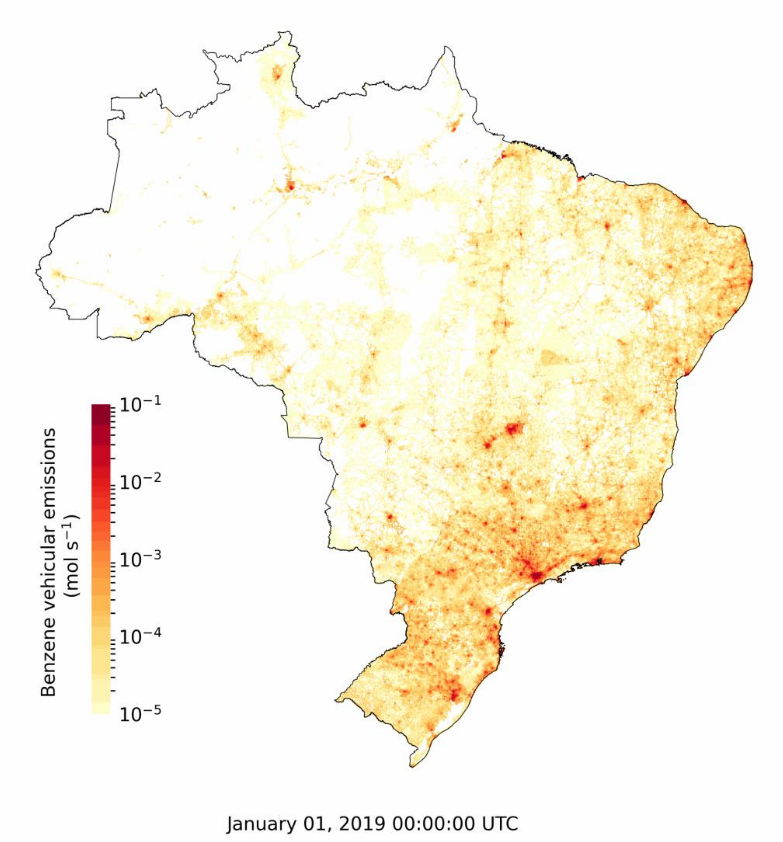


Figure 7. Vehicular emissions of Benzene in Brazil on January 1st, 2019 using the BRAVES database.

We provide the BRAVES database annual speciated emissions with 0.05°x0.05° of resolution covering the entire Brazilian territory at: https://doi.org/10.5281/zenodo.6141109

7. Comparison with independent databases

We analyze the spatial correlation and bias between the BRAVES database and the Emissions Database for Global Atmospheric Research (EDGAR – version 5.0 https://edgar.jrc.ec.europa.eu/dataset_ap50) annual gridmaps (Crippa et al. 2018; European Comission 2022). We performed the comparison using the "Road Transportation" emissions from EDGAR for the Brazilian territory, including soil resuspension emission rates of PM_{10} from EDGAR. The BRAVES database emission rates in tons per year from 2015 were regridded to the same spatial resolution of EDGAR. The Spearman coefficient estimates the spatial correlation, while the difference in absolute emissions calculates the bias between the datasets. We compare the disaggregated emissions of CO, PM_{10}, NOx, and COV from BRAVES and EDGAR.

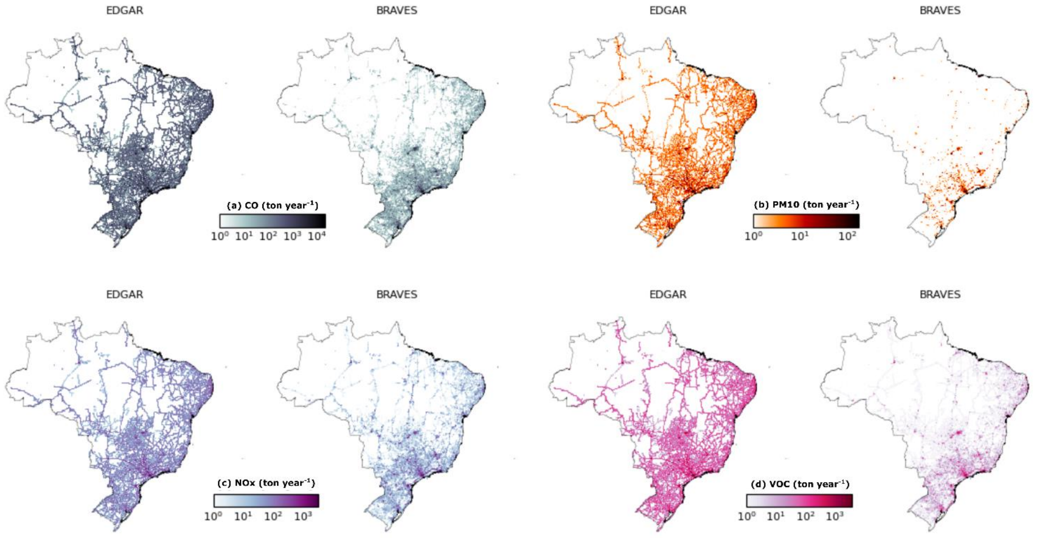


Figure 8. Comparison of a) CO, b) PM_{10}, c) NOx, and d) VOC spatial distribution (log scale) provided by the EDGAR and BRAVES databases.

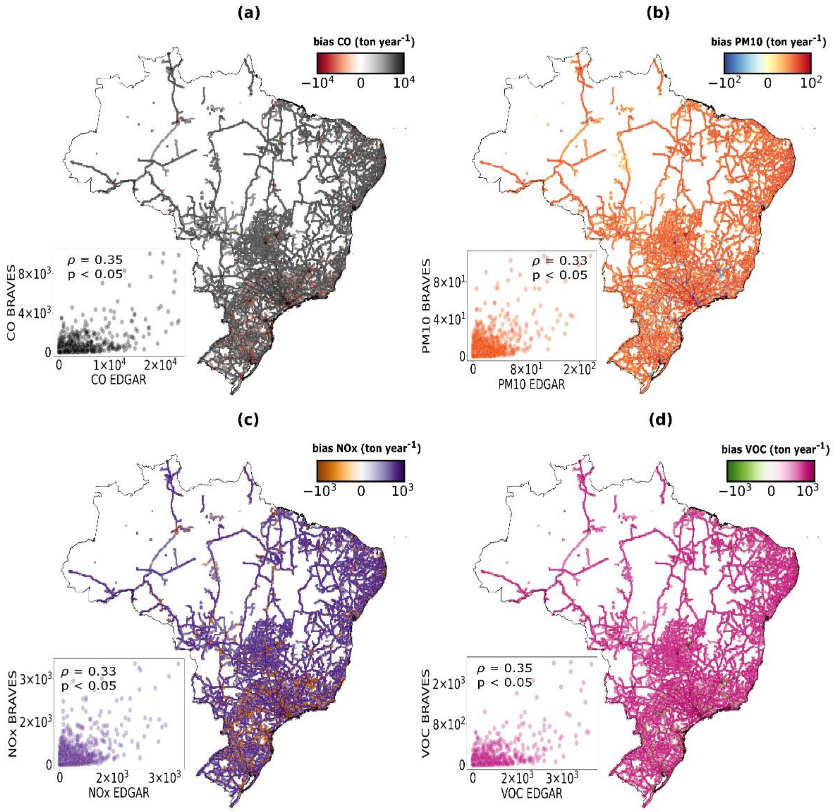


Figure 9. Bias (log scale) and scatter plots of a) CO, b) PM_{10}, c) NOx, and d) VOC emission rates provided by the BRAVES database and EDGAR in 2015.

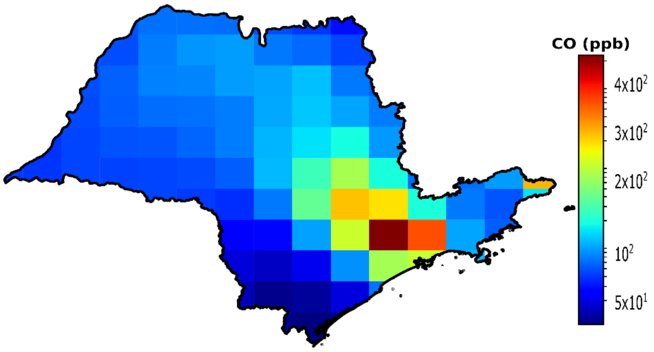
Emission from BRAVES and EDGAR presents overall spatial correlation ($p<0.05$) of $\rho = 0.35$ for CO, $\rho = 0.33$ for PM_{10}, $\rho = 0.33$ for NOx, and $\rho = 0.35$ for VOC (Figure 9). Emissions from EDGAR are consistently higher (Figure 9) than from BRAVES, as also reported by Vasques and Hoinaski (2021). The largest differences are observed in CO emissions, followed by VOC, NOx, and PM_{10}. Madrazo et al. (2018) explains that most of the road transport Emission Factors are overestimated in EDGAR, while Huneeus et al. (2020) found discrepancies between EDGAR and local/national city emissions. Álamos et al. (2022) also reported an overestimation of EDGAR emissions.

We analyze the spatial correlation between vehicular emissions of CO from the BRAVES database, EDGAR, and surface concentration of CO from the Modern-Era Retrospective Analysis for Research and Applications - MERRA-2. The Global Modeling and Assimilation Office (GMAO), managed by NASA, provides MERRA-2 reanalysis products in a spatial resolution of 0.625°×0.5°, covering from 1980 to the present (Gelaro et al. 2017; Randles et al., 2017). We calculate the annual average concentration in 2015 from monthly data in netCDF files available at GES-DISC platform (https://disc.gsfc.nasa.gov/datasets/M2TMNXCHM_5.12.4/summary). All grids are realigned to match the MERRA-2 spatial resolution. We analyze the spatial correlation by Brazilian state since the vehicular emission has more influence in urbanized ones. We assume that those cells which have the vehicular emissions as the major source of air pollutant also have higher surface concentration. However, this assumption has several limitations and should be carefully evaluated since it does not account for the dispersion process and other source types (i.e., industrial, biomass burning, biogenic sources).

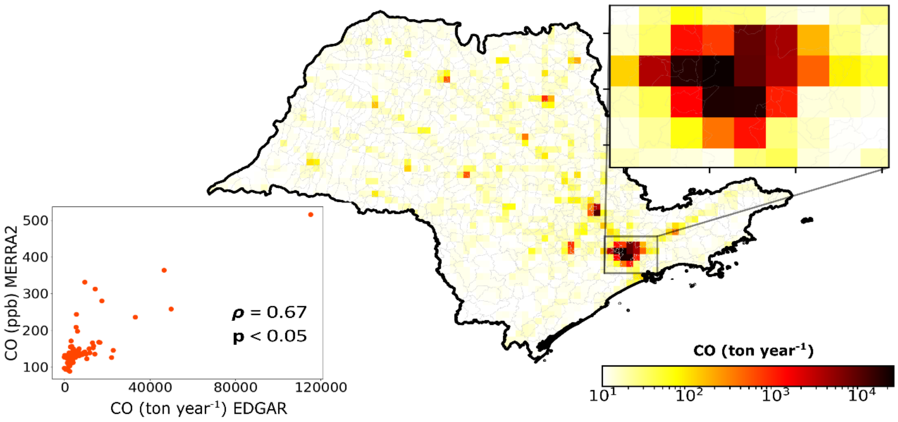
Figure 10 shows the (a) CO concentrations from MERRA-2 in São Paulo (SP) state, (b) vehicular emission of CO from EDGAR, and (c) vehicular emission of CO from the BRAVES database. We highlight the state of SP since it has approximately 7 million vehicles, being considered the state with the highest vehicular emission in Brazil (Vasques and Hoinaski, 2021). The BRAVES database and EDGAR reach similar spatial correlation with MERRA-2 (Figure 10). However, the zoom-in quadrant in the São Paulo metropolitan region reveals the greater level of details from the BRAVES database compared to EDGAR. In addition, BRAVES has higher temporal resolution and chemical speciated emissions. In other Brazilian states, such as Minas Gerais (MG) and Rio Grande do Sul (RS), there is also a positive correlation between vehicle emissions and surface concentrations of CO

(Table SM8). It shows that both databases consistently capture he spatial variability of vehicular emissions and the BRAVES database brings additional features for air quality studies in Brazil.

(a) MERRA-2



(b) EDGAR



(c) BRAVES

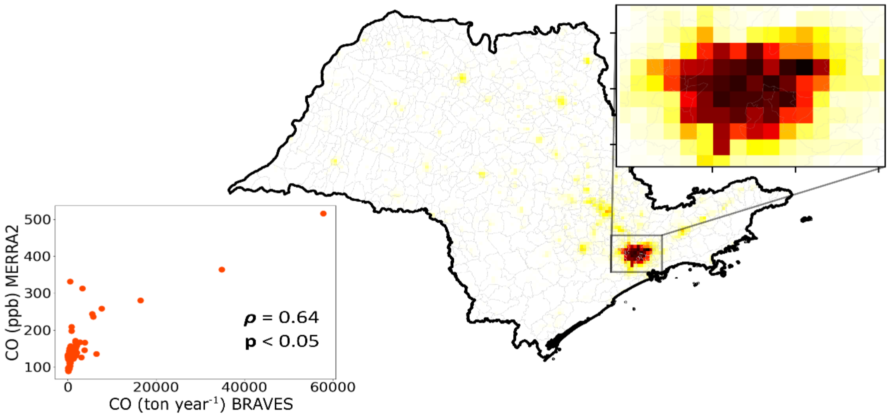


Figure 10. CO spatial distribution provided by (a) MERRA2, (b) EDGAR, and (c) BRAVES. Scatter plots of CO vehicle emission and CO surface concentrations in SP.

8. Data availability

The BRAVES database is freely available at https://doi.org/10.5281/zenodo.6141109 (Hoinaski et al., 2022). We provide annual speciated emissions with 0.05°x0.05° of resolution covering the entire Brazilian territory. Codes to generate the database are available at: https://github.com/leohoinaski/BRAVES. Using the annual files, users can derive hourly basis emissions through the available codes.

9. Conclusions

Here, we introduce the BRAVES database, the first high-resolution and chemical speciated database of vehicular emissions covering the entire Brazilian territory. The BRAVES database contains emissions of 41 air pollutants, from annual to hourly basis temporal resolution and user-defined spatial resolution. The attributes of this emission database are fully compatible with sophisticated air quality models. Moreover, the emissions of multiple chemical species presented here provide essential information to understand important atmospheric chemistry processes in Brazil. We also provide python scripts for users who want to create their custom gridded inventory.

Even though detailed emission inventories are required to control air pollution, vehicular emissions are scarce in most developing countries. So far, Brazil has lacked a comprehensive and easily accessible database of vehicular emissions, and, creating gridded inventories in South America is urgently needed. This work contributes to overcoming this gap.

The spatial correlation analysis reveals that the BRAVES database agrees with the vehicular emissions from EDGAR, even though EDGAR emissions are consistently higher than those BRAVES ones. We conclude that this database can be a better alternative to represent the spatial variability of vehicular emissions in Brazil. The BRAVES database has a similar performance representing the spatial pattern of vehicular emissions, with more spatial, temporal, and chemical details when compared with EDGAR. Moreover, the BRAVES database is in closer agreement to local and very detailed emissions inventories. A comparison with MERRA-2 surface concentration confirms the consistency of the BRAVES database.

Even though the present database is a step forward for air pollution research in Brazil, there are several opportunities for expanding and improving this work. Most heavy-duty emissions occur in high flow and high-speed limit roads, such as expressways. Future versions could improve the spatial disaggregation in pixels containing roads with high traffic flow and/or high-speed traffic through the optimization of the disaggregation factors. Different criteria for light and heavy vehicles would also be needed. Moreover, the chemical speciation could include profiles to consider the Brazilian reality as biofuels, fleet motorization, and regionalized soil resuspension properties. Temporal variability would also be improved by regionalizing the profiles to account for the traffic flow in each location.

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