Multispecies and high spatiotemporal resolution database of vehicular emissions in Brazil

Leonardo Hoinaski[1], Thiago Vieira Vasques[2], Camilo Bastos Ribeiro[2], Bianca Meotti[2]

[1]Department of Sanitary and Environmental Engineering, Federal University of Santa Catarina, Florianópolis, Santa Catarina, Brazil, leonardo.hoinaski@ufsc.br
[2]Graduate Program in Environmental Engineering, Federal University of Santa Catarina, Florianópolis, Santa Catarina, Brazil, vvthiago@hotmail.com, cb_ambiental@hotmail.com, meottibianca@gmail.com

Correspondence to: Leonardo Hoinaski (leonardo.hoinaski@ufsc.br)

Abstract. In this article, we present the BRAzilian Vehicular Emissions inventory Software (BRAVES) database, a multi-species 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 local studies and the SPECIATE database from the United States Environmental Protection Agency. Our BRAVES database provides hourly and annual emissions of 41 gaseous and particle pollutants~~. Users~~, where users can define the spatial resolution, which ranges from a coarse to a very refined scale. Spatial correlation analysis reveals that the BRAVES database reaches ~~similar~~better performance ~~to~~than 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~~in 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~~emission 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 local studies and 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 (road dust) 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~~).~~) by fleet category (i.e., commercial-light vehicles, motorcycles, light-duty, and heavy-duty vehicles). Throughout this article, we call the 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/leo-hoinaski/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.

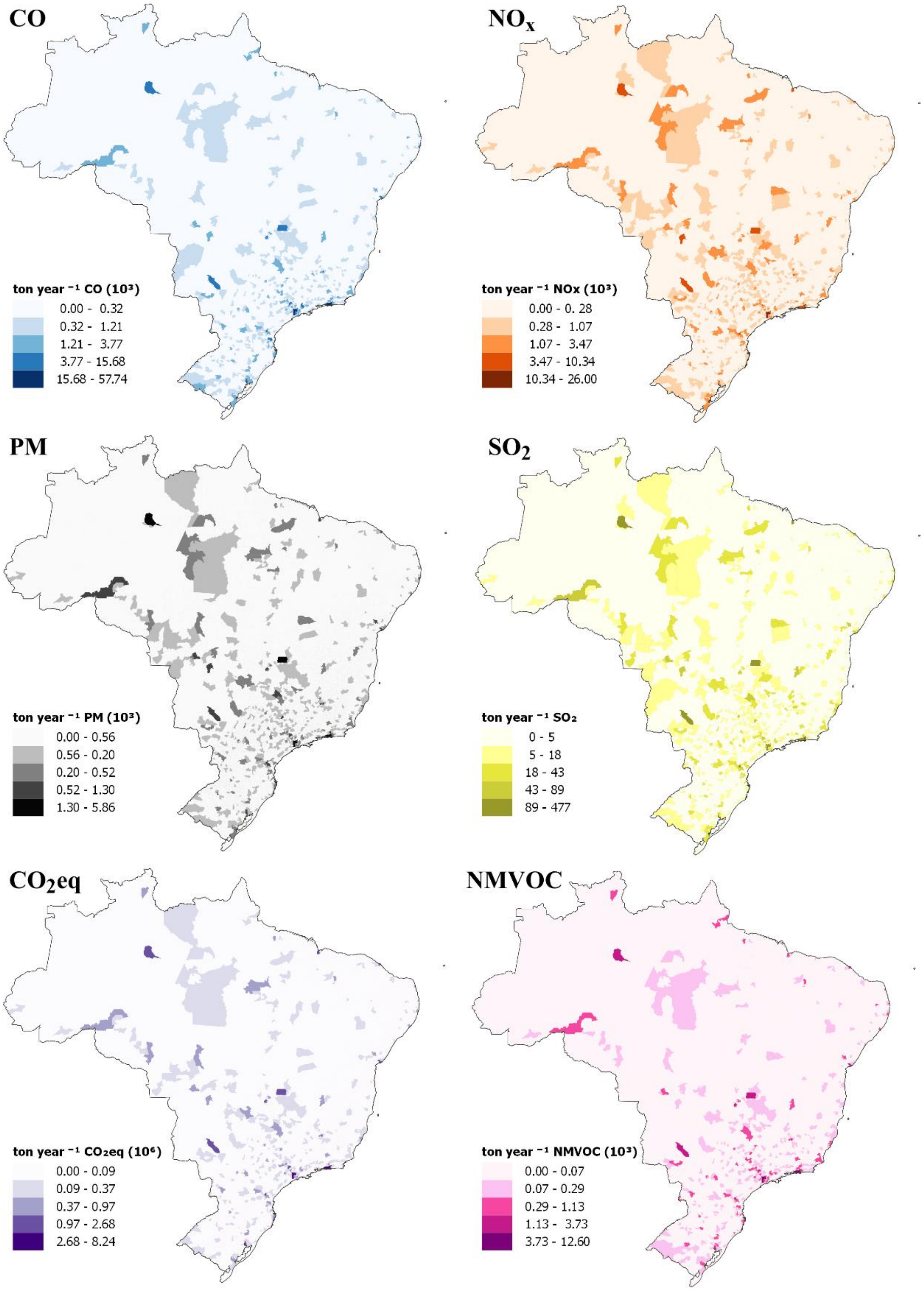


Figure 1. Vehicular emissions inventory in 2019 of a) CO, b) NOx, c) PM, d) SO_2, e) CO_2, and f) NMVOC provided by BRAVES (Vasques and Hoinaski, 2021).

3. Spatial disaggregation

Since vehicular emissions from BRAVES are aggregated by ~~municipality~~municipalities, we use a road density approach to distribute the emissions of each municipality in pixels with user-defined resolution. Previous work ~~of~~by 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, the 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.open-streetmap.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.

$$RD_{p,m} = \frac{L_p}{L_m} \tag{1}$$

$$E_{c,p,m} = RD_{p,m} \times E_{c,m} \tag{2}$$

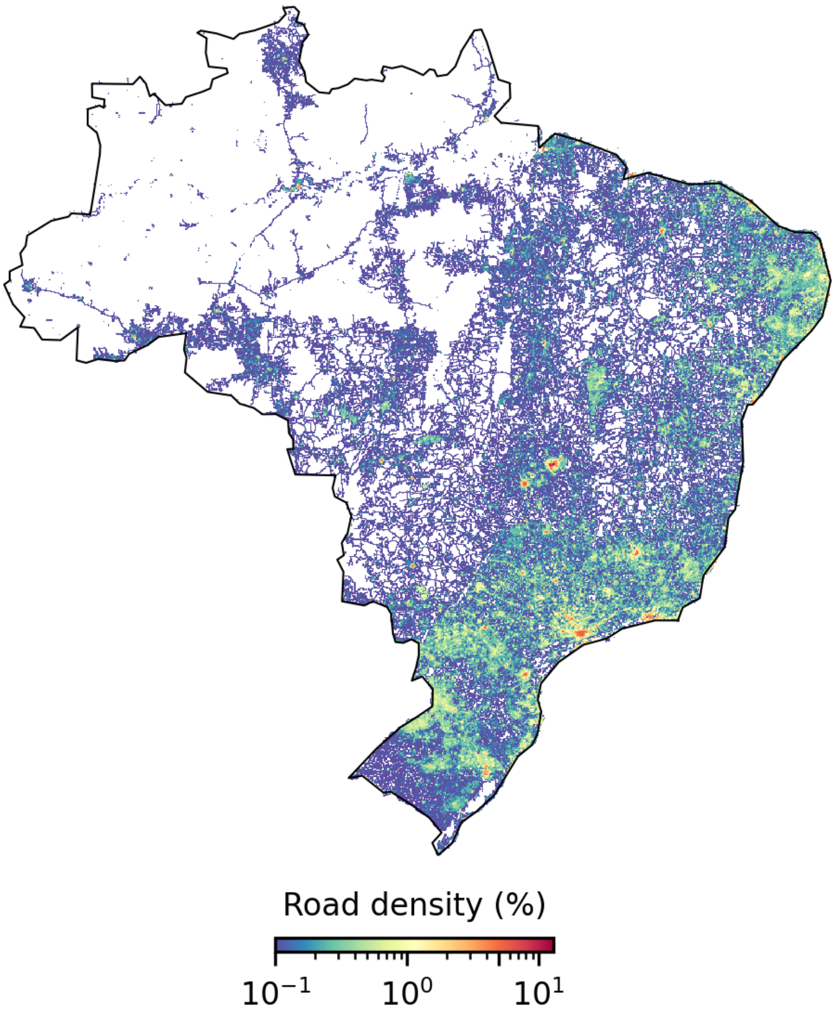
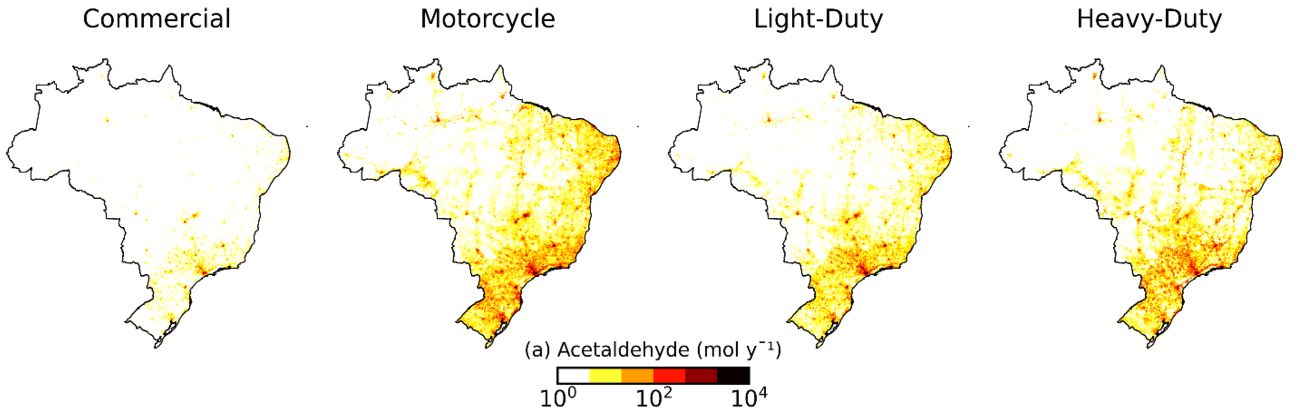
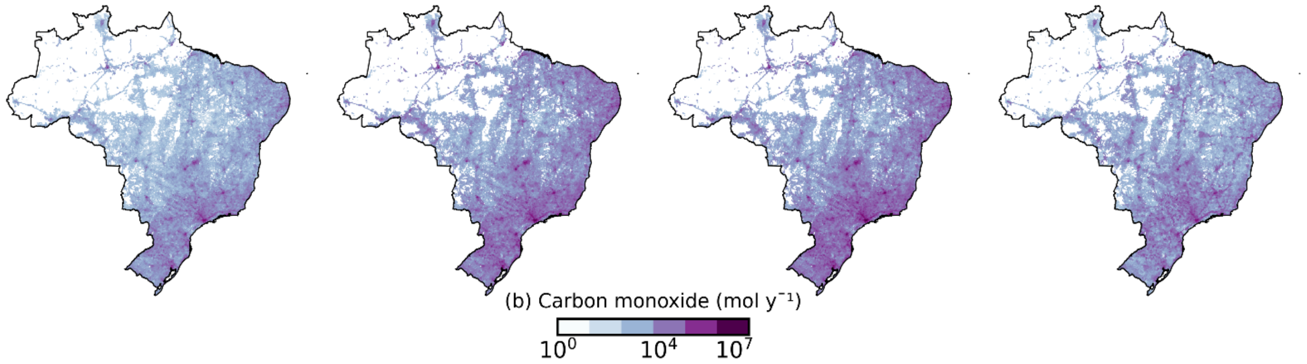


Figure 2. Road density factor in Brazil~~.~~ with spatial resolution of 0.05°×0.05°.

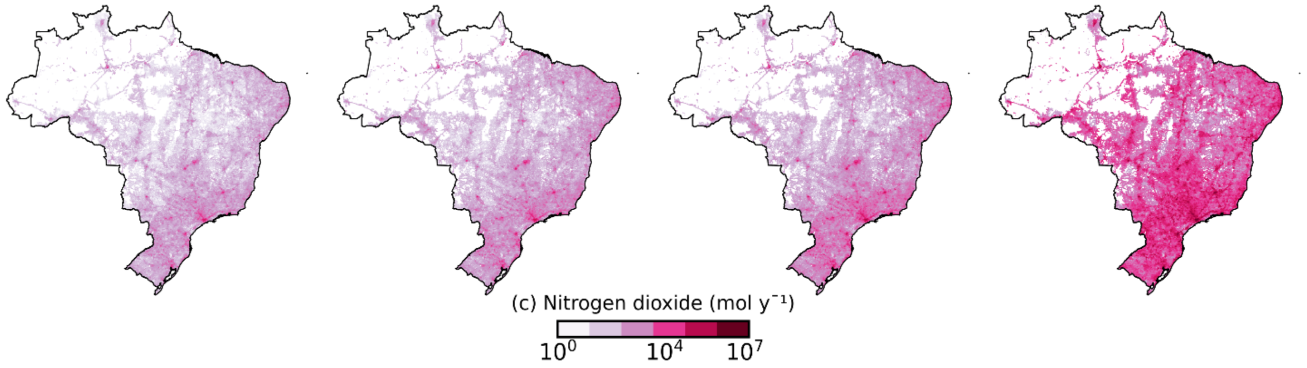
Figure 3 shows the annual emissions of ~~aldehydes~~acetaldehyde, CO, and ~~CO~~nitrogen dioxide from 2019 in Brazil by fleet category using spatial distribution based on the road density approach. Hotspots of vehicular emissions ~~are concentrated~~concentrate in urbanized areas in Brazil (Figure 3).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



(a) Acetaldehyde (mol y^{-1})



(b) Carbon monoxide (mol y^{-1})



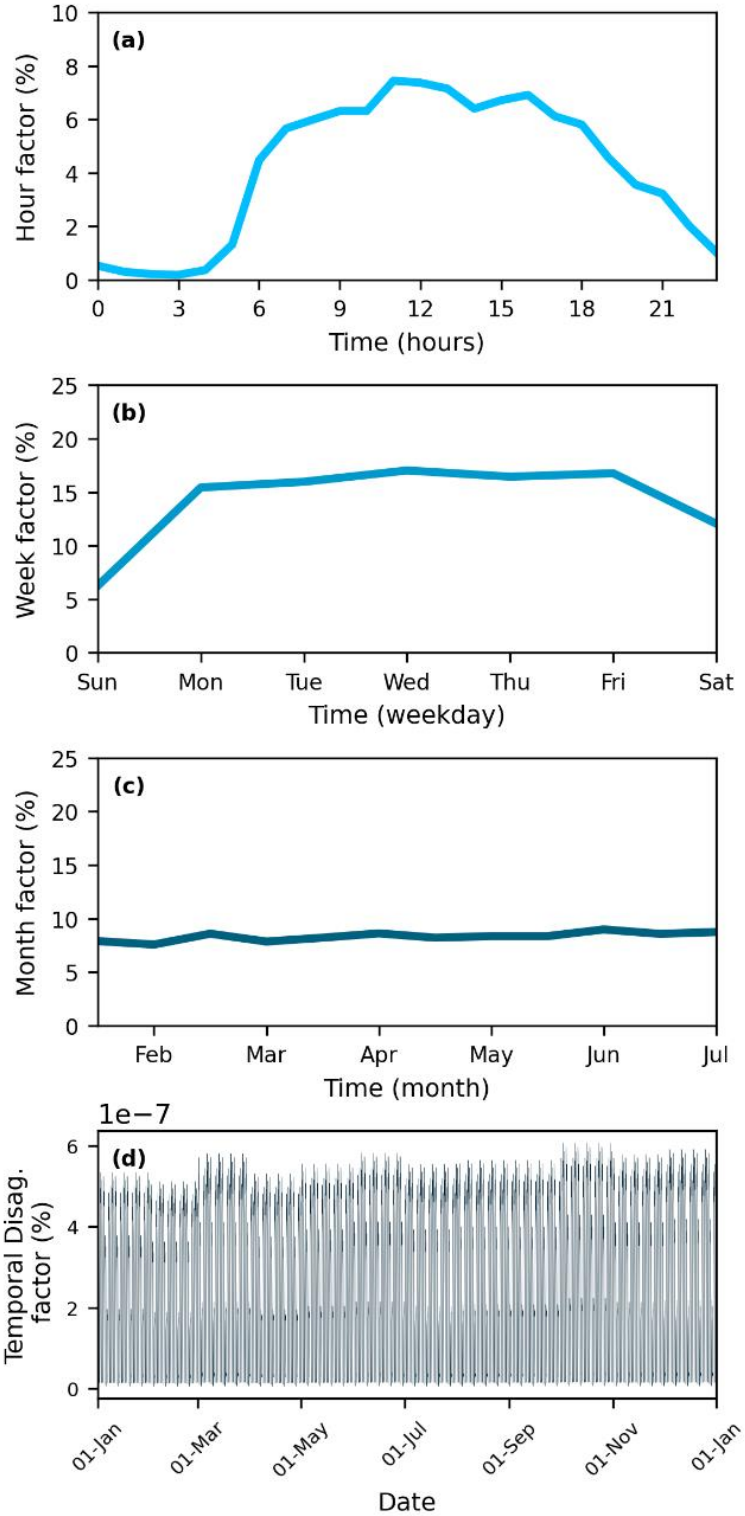
(c) Nitrogen dioxide (mol y^{-1})

Figure 3. Vehicular emissions of ~~aldehydes a-d) and~~ a) acetaldehyde, b) carbon monoxide ~~e-h)~~, and c) nitrogen dioxide 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).

$$E_{c,p,m,h} = T_f \times E_{c,p,m} \tag{3}$$



120 **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~~We have targeted the species required 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.

We speciate the acetaldehydes (ALD2 and ALD2_PRIMARY), formaldehyde (FORM and FORM PRIMARY), and aldehydes with 3 or more carbons (ALDX) using original RCHO estimates from BRAVES, which are based on local emission factor from *Companhia de Tecnologia de Saneamento Ambiental do Estado de São Paulo* - CETESB (CETESB, 2022). We have considered that acetaldehyde represents 50% of RCHO emissions from light-duty vehicles. Formaldehyde emissions (FORM and FORM_PRIMARY) represent 39% of RCHO emissions from light-duty (Nogueira et al. 2015). ALDX has been considered as 10% of RCHO emissions, while acetone (ACET) accounts for 8% of these emissions.

We have also kept the estimated using local emissions factor for ethanol (ETOH), which has been the best way to represent the particularities of biofuels in Brazil. CETESB has provided the ETHO emission factors since 2008, for light-duty vehicles running with ethanol and gasoline (CETESB, 2022). Since CETESBs' RCHO and ETOH emissions factors are available only for light-duty and commercial light vehicles, we have used percentage factors from Speciate to estimate aldehyde and ethanol emissions from NMHC for motorcycles and heavy-duties.

The Brazilian gasoline C, which has fueled light-duty vehicles, is a mixture of pure gasoline and 20 to 25% of anhydrous ethanol. Since 2008, heavy-duty vehicles have run with a blend of diesel and up to 15% of biodiesel. This unique chemical signature of the biofuels in Brazil reflected significantly in the vehicular emissions, especially those of carbonyls and ethanol (Nogueira et al. 2015; CNPE, 2018). These last compounds deserve attention since they are major precursors of tropospheric ozone (Atkinson, 2000, Jacob, 2000).

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

Specie	ID	Specie	ID
Acetone	ACET	Aluminum	PAL
~~total~~Total Acrolein	ACROLEIN	Calcium ion	PCA
~~total~~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~~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~~The table at https://github.com/leohoinaski/BRAVES/tree/main/ChemicalSpec summarizes the speciation factors used to build this database.

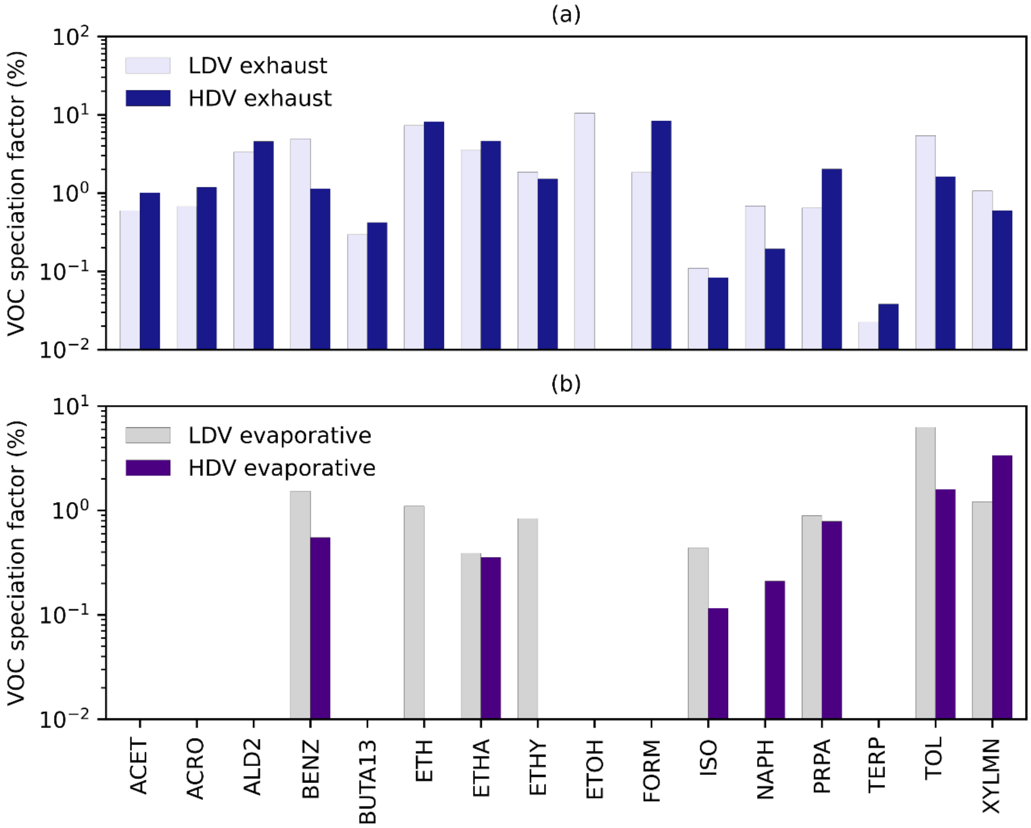


Figure 5. VOC chemical speciation factor for (a) exhaust and (b)evaporative emissions from light and heavy ~~vehicles used in this~~vehicle article. Data derived from US EPA SPECIATE 5.1 (US EPA, 2020; Eyth et al. 2020).

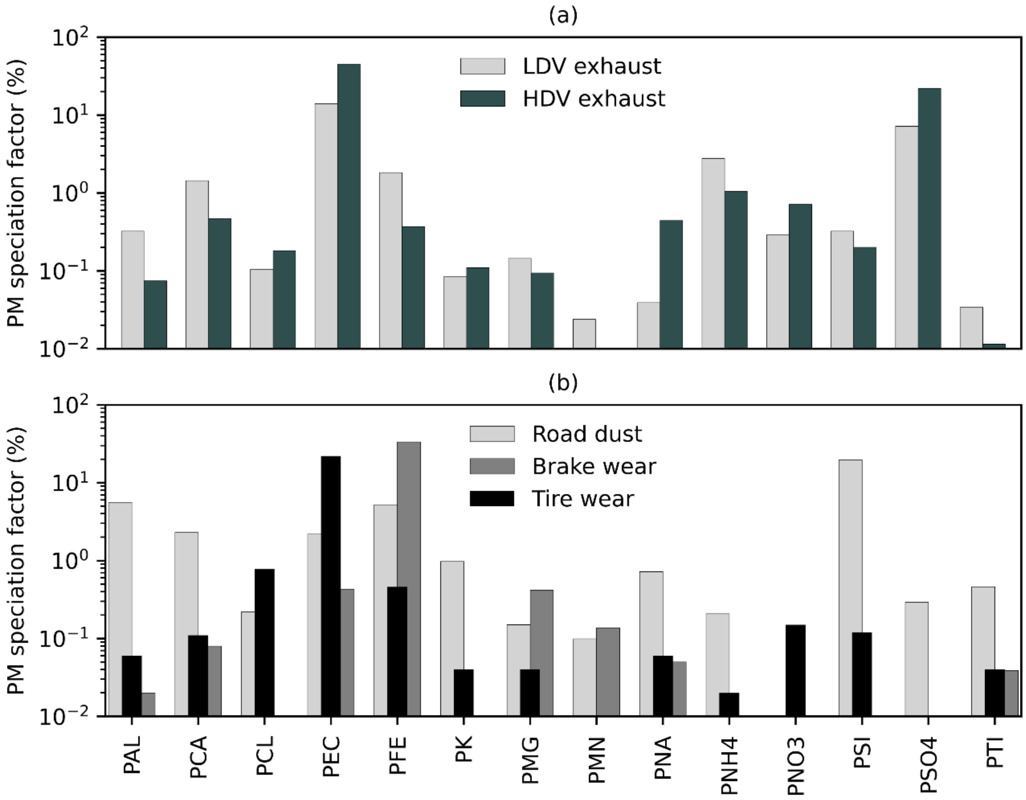


Figure 6. PM chemical speciation factors (a) exhaust emissions from light and heavy vehicles (b) road dust resuspension, brake wear, and tire wear. Data derived from US EPA SPECIATE 5.1 (US EPA, 2020; Eyth et al. 2020).

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. Flags have been included in the netCDF files to provide the area and time zones of each pixel, so users can choose the option to generate ready-to-use hourly input files for CMAQ (in mass or mol per second) or WRF-CHEM (in mass or mol flux per area).

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.

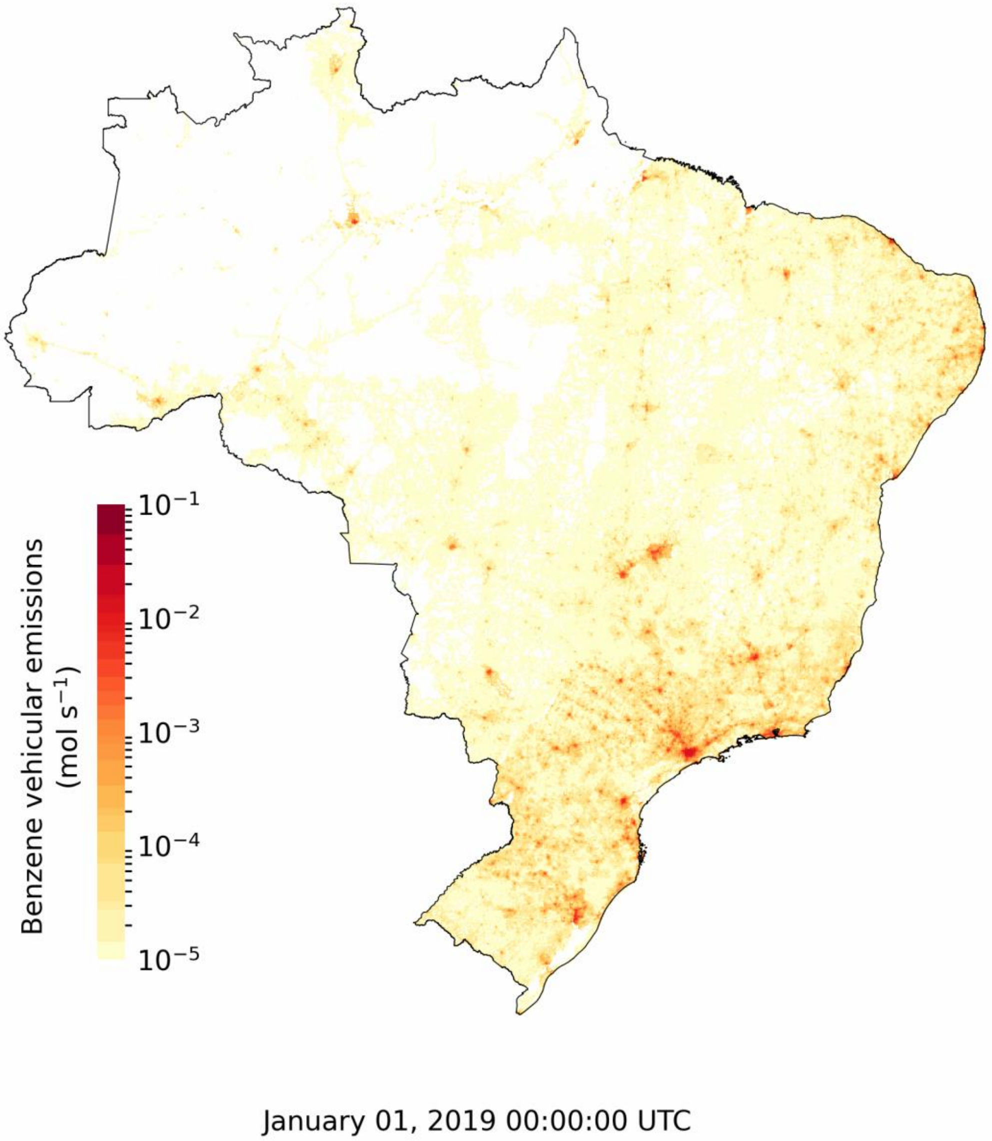


Figure 7. Vehicular emissions of Benzene in Brazil on January 1st, 2019 using the BRAVES database.

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~~ https://doi.org/10.5281/zenodo.6588692

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~~https://edgar.jrc.ec.europa.eu/dataset_ap50) annual ~~gridmaps~~grid maps (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. Table SM7 also shows a comparison of the total vehicular emissions aggregate in Brazilian territory, considering BRAVES, EDGAR, and others available national inventories.

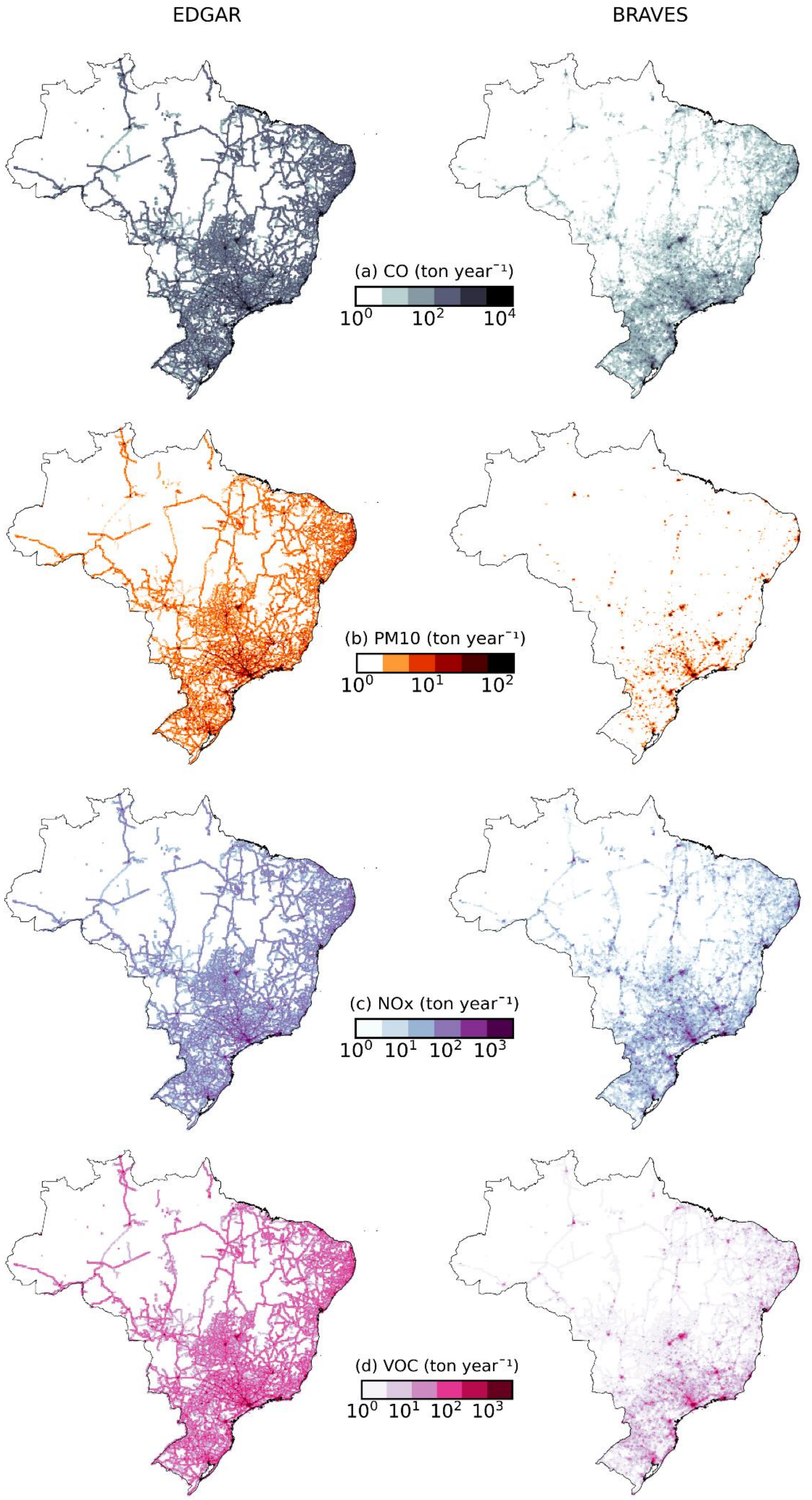


Figure 8. Comparison of a) CO, b) PM_{10}, c) NOx, and d) VOC spatial distribution (log scale) provided by the EDGAR and BRAVES databases.

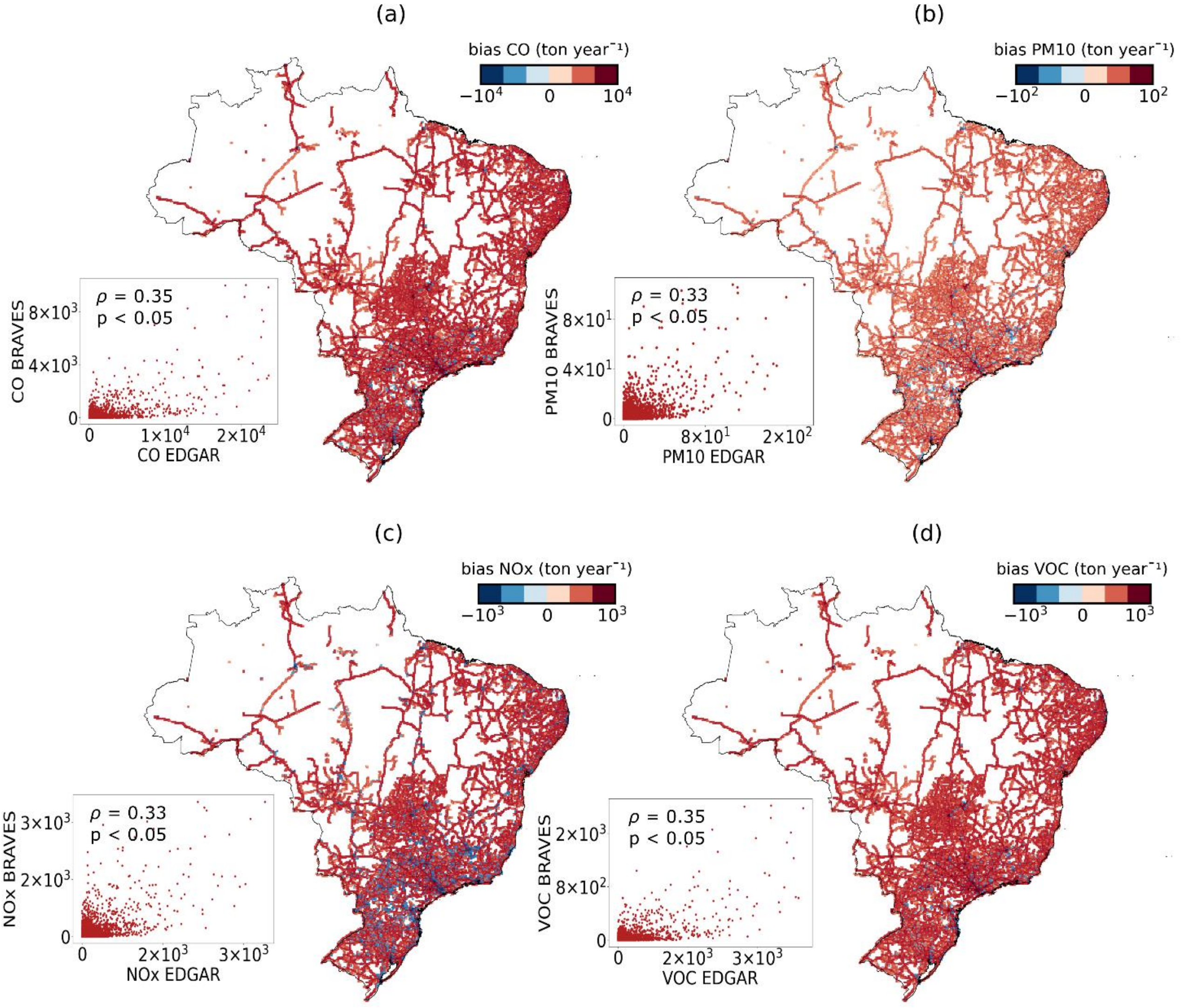


Figure 9. Bias (log scale) and scatter plots of a) CO, b) PM_{10}, c) NOx, and d) VOC emission rates provided by the BRAVES database and EDGAR in 2015.

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~~overall 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~~explain that most of the road transport ~~Emission Factors~~emissions 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. Compared with the present database, we have observed higher estimates from EDGAR in pixels with low road densities and less populated areas, while in high population areas EDGAR has estimated lower values. A similar pattern has been also observed by Ibarra-Espinosa et al. (2018) when comparing EDGAR with VEIN.

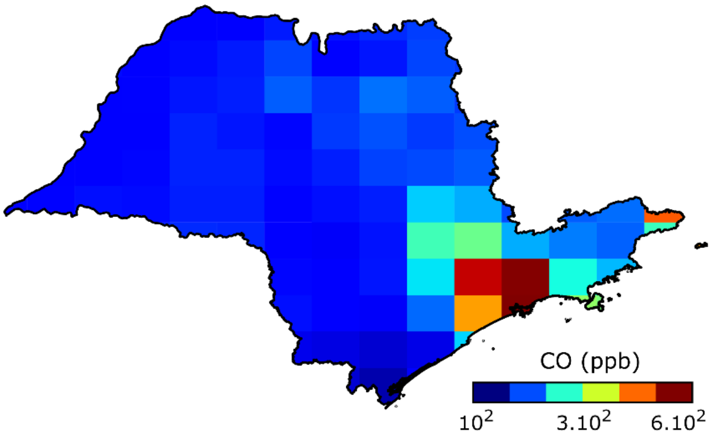
We analyze the spatial correlation between CO vehicular emissions ~~of CO from the~~estimated by BRAVES ~~database,~~and EDGAR, and CO surface concentration ~~of CO from~~estimated by the Modern-Era Retrospective Analysis for Research and Applications - MERRA-2. The Global Modeling and Assimilation Office (GMAO), managed by NASA, provides MERRA-2 reanalysis products in a spatial resolution of 0.~~625~~5°×0.~~5~~625°, 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).~~the GES-DISC platform (https://disc.gsfc.nasa.gov/datasets/M2TMNXCHM_5.12.4/summary). MERRA-2 hourly dataset used in Figure 10 can be downloaded at https://disc.gsfc.nasa.gov/datasets/M2T1NXCHM_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~~pollutants also have higher surface ~~concentration~~concentrations. 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).

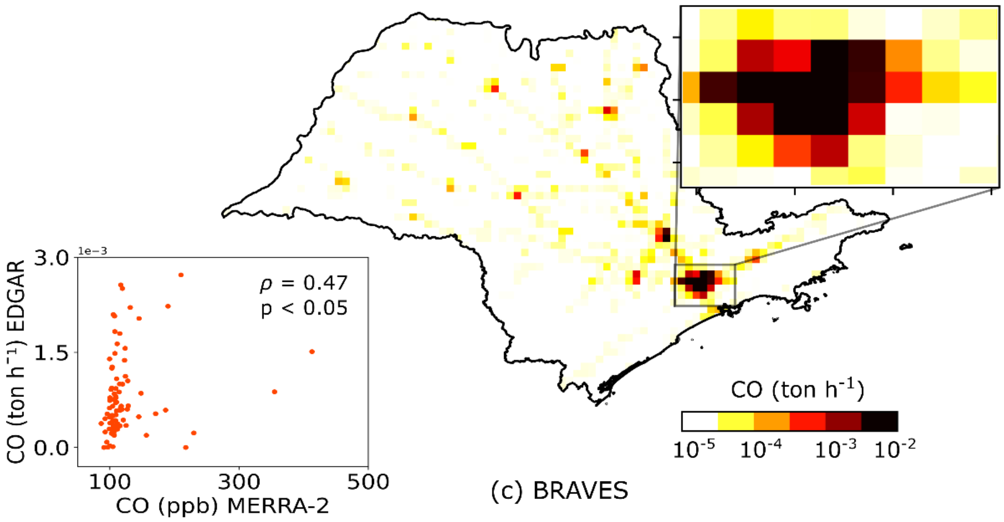
Figure 10 shows the ~~(~~a) CO concentrations from MERRA-2 in the São Paulo (SP) state, ~~(~~b) vehicular emission of CO from EDGAR, and ~~(~~c) vehicular emission of CO from the BRAVES database. ~~We highlight the state of SP since it has approximately 7~~In 2021, ~31 million vehicles were registered in SP state, being considered the state with the highest vehicular emission in Brazil (SENATRAN 2021; Vasques and Hoinaski, 2021). The BRAVES database and EDGAR reach a similar spatial correlation with MERRA-2 when using annual averages (Figure ~~10). However, the~~SM8). The zoom-in quadrant in the São Paulo metropolitan region in Figure 10 reveals ~~the~~a greater level of details from the BRAVES database compared to EDGAR. In addition, BRAVES has higher temporal resolution and chemical speciated emissions~~.~~ and has presented a better correlation with MERRA-2 when comparing hourly averages (Figure 10). In Figure 10, we have compared MERRA-2 and emissions on 01/01/2015 at 8:00, when the boundary layer is low and the concentrations are representative of the emissions, as shown by Gallardo et al. (2012). It is worth emphasizing that the straightforward comparison between emission and concentrations from monitors or reanalysis data must be made carefully and under specific conditions.

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~~SM9). It shows that both databases consistently capture ~~he~~the 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

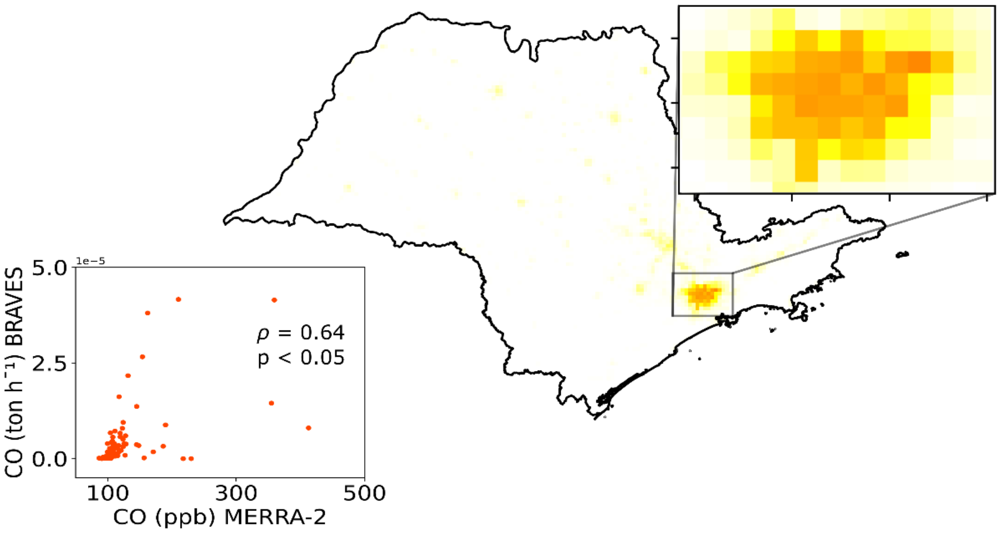


Figure 10. CO spatial distribution provided by ~~(~~a) ~~MERRA2, (~~MERRA-2, b) EDGAR, and ~~(~~c) BRAVES. Scatter plots of CO vehicle emission and CO hourly surface concentrations in SP on 01/01/2015 at 8:00.

8. Data availability

The BRAVES database is freely available at ~~https://doi.org/10.5281/zenodo.6141109~~https://doi.org/10.5281/zenodo.6588692 (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 of 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~~with 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~~on 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~~.~~ or by including monthly fuel consumption data. An evaluation using BRAVES database as input in air quality models would bring important information about the model's errors and representativeness. As reported by Nogueira et al. (2021), the emission factors from CETESB used in this work would require future corrections to better represent field measurements.

Acknowledgments

Authors would like to thank the Fundação de Amparo à Pesquisa e Inovação de Santa Catarina - FAPESC, for financial support of project number 2018TR499.

References

Andrade MF, Kumar P, Freitas ED, Ynoue RY, Martins J, Martins LD, Nogueira T, Perez-Martinez P, de Miranda RM, Albuquerque T, Gonçalves FLT, Oyama B, Zhang Y (2017). Air quality in the megacity of São Paulo: evolution over the last 30 years and future perspectives. Atmospheric Environment 159:66–82. https://doi.org/10.1016/j.atmosenv.2017.03.051

Álamos N, Huneeus N, Opazo M, Osses M, Puja S, Pantoja N, Denier van der Gon H, Schueftan A, Reyes R, Calvo R (2022). High-resolution inventory of atmospheric emissions from transport, industrial, energy, mining and residential activities in Chile. Earth System Science Data 14:361–379. https://doi.org/10.5194/essd-14-361-2022

Anenberg S, Miller J, Minjares R, Du L, Henze DK, Lacey F, Malley CS, Emberson L, Franco V, Klimont Z, Heyes C (2017). Impacts and mitigation of excess diesel-related NO_x emissions in 11 major vehicle markets. Nature 545:467–471. https://doi.org/10.1038/nature22086

Atkinson, R (2000). Atmospheric chemistry of VOCs and NOx. Atmospheric Environment 34:2063-2101. https://doi.org/10.1016/S1352-2310(99)00460-4

Brito J, Carbone S, Santos DAM, Dominutti P, Alves NO, Rizzo LV, Artaxo P (2018). Disentangling vehicular emission impact on urban air pollution using ethanol as a tracer. Scientific Reports 8:10679. https://doi.org/10.1038/s41598-018-29138-7

Carvalho VSB, Freitas ED, Martins LD, Martins JA, Mazzoli CR, Andrade MF (2015). Air quality status and trends over the Metropolitan Area of São Paulo, Brazil as a result of emission control policies. Environmental Science & Policy 47:68–79. https://doi.org/10.1016/j.envsci.2014.11.001

CETESB, 2022. Emissões veiculares no estado de São Paulo 2022. São Paulo (in Portuguese). Accessed February 2022. Available online: https://cetesb.sp.gov.br/veicular/wp-content/uploads/sites/6/2022/03/Relatorio-Emissoes-Veiculares-2020.pdf.

CNPE, 2018. Resolução CNPE nº 16, de 29 de outubro de 2018, DOU, de novembro de 2018. Seção 1, página 2, Brasil (in Portuguese).

Crippa M, Guizzardi D, Muntean M, Schaaf E, Dentener F, Van Aardenne JA, Monni S, Doering U, Olivier JGJ, Pagliari V, Janssens-Maenhout G (2018). Gridded emissions of air pollutants for the period 1970–2012 within EDGAR v4.3.2. Earth System Science Data 10:1987–2013. https://doi.org/10.5194/essd-10-1987-2018

European Commission, Joint Research Centre (JRC). Emission Database for Global Atmospheric Research (EDGAR), v5.0. Accessed January 2022. Available online: https://edgar.jrc.ec.europa.eu/index.php/dataset_ap50, https://doi.org/10.2904/JRC_DATASET_EDGAR

Eyth A, Strum M, Murphy B, Shah T, Shi Y, Beardsley R, Yarwood G (2020). Speciation Tool User's Guide Version 5.0. Available online: https://www.cmascenter.org/speciation_tool/documentation/5.0/Ramboll_sptool_users_guideV5.pdf.

Gallardo L, Escribano J, Dawidowski L, Rojas N, Andrade, MF, Osses M (2012). Evaluation of vehicle emission inventories for carbon monoxide and nitrogen oxides for Bogotá, Buenos Aires, Santiago, and São Paulo. Atmospheric Environment 47:12-19. https://doi.org/10.1016/j.atmosenv.2011.11.051

Gelaro R, McCarty W, Suárez MJ, Todling R, Molod A, Takacs L, Randles CA, Darmenov A, Bosilovich MG, Reichle R, Wargan K, Coy L, Cullather R, Draper C, Akella S, Buchard V, Conaty A, Silva AM, Gu W, Kim G, Koster R, Lucchesi R, Merkova D, Nielsen JE, Partyka G, Pawson S, Putman W, Rienecker M, Schubert SD, Sienkiewicz M, Zhao B (2017). The Modern-Era Retrospective Analysis for Research and Applications, Version 2 (MERRA-2). Journal of Climate 30(14):5419-5454. https://doi.org/10.1175/JCLI-D-16-0758.1

GLOBAL MODELING AND ASSIMILATION OFFICE (GMAO) (2015). MERRA-2 tavg1_2d_chm_Nx: 2d,1-Hourly, Time-Averaged, Single-Level, Assimilation, Carbon Monoxide and Ozone Diagnostics V5.12.4, Greenbelt, MD, USA, Goddard Earth Sciences Data and Information Services Center (GES DISC). 10.5067/3RQ5YS674DGQ

GLOBAL MODELING AND ASSIMILATION OFFICE (GMAO) (2015). MERRA-2 tavgM_2d_chm_Nx: 2d, Monthly mean, Time-Averaged, Single-Level, Assimilation, Carbon Monoxide and Ozone Diagnostics V5.12.4, Greenbelt, MD, USA, Goddard Earth Sciences Data and Information Services Center (GES DISC). 10.5067/WMT31RKEXK8I

Gómez CD, González CM, Osses M, Aristizábal BH (2018). Spatial and temporal disaggregation of the on-road vehicle emission inventory in a medium-sized Andean city. Comparison of GIS-based top-down methodologies. Atmospheric Environment 179:142–155. https://doi.org/10.1016/j.atmosenv.2018.01.049

Hoinaski L, Vasques ~~T V~~TV, Ribeiro ~~C B~~CB, Meotti B~~.~~ (2022). BRAVES database Version 1~~:~~.1 (REVISED): multispecies and high spatiotemporal resolution database of vehicular emissions in Brazil. ~~https://doi.org/10.5281/zenodo.6141109~~[Data set]. In Earth System Science Data - https://doi.org/10.5194/essd-2022-74 (BRAVES database version 1.1 (REVISED)). Zenodo. https://doi.org/10.5281/zenodo.6588692

Huneeus N, Van der Gon HD, Castesana P, Menares C, Granier C, Granier L, Alonso M, Andrade MF, Dawidowski L, Gallardo L, Gomez D, Klimont Z, Janssens-Maenhout G, Osses M, Puliafito SE, Rojas N, Sánchez-Ccoyllo O, Tolvett S, Ynoue RY (2020). Evaluation of anthropogenic air pollutant emission inventories for South America at national and city scale. Atmospheric Environment 235:117606. https://doi.org/10.1016/j.atmosenv.2020.117606

Ibarra-Espinosa S, Ynoue R, O'Sullivan S, Pebesma E, Andrade MDF, Osses M (2018). VEIN v0.2.2: an R package for bottom–up vehicular emissions inventories. Geoscientific Model Development 11:2209–2229. https://doi.org/10.5194/gmd-11-2209-2018

Instituto de Meio Ambiente e Recursos Hídricos do Espírito Santo (IEMA-ES) (2019). Inventário de Emissões Atmosféricas Região da Grande Vitória – Ano Base 2015 (in Portuguese). Available online: https://iema.es.gov.br/qualidadedoar/inventariodefontes/2015

Jacob, DJ (2000). Heterogeneous chemistry and tropospheric ozone. Atmospheric Environment 34:2131-2159. https://doi.org/10.1016/S1352-2310(99)00462-8

Krzyzanowski M, Kuna-Dibbert Birgit, Schneider J (2005). Health effects of transport-related air pollution. World Health Organization. Regional Office for Europe. https://www.euro.who.int/en/publications/abstracts/health-effects-of-transport-related-air-pollution

Lyu M, Bao X, Zhu R, Matthews R (2020). State-of-the-art outlook for light-duty vehicle emission control standards and technologies in China. Clean Technologies and Environmental Policy 22(4):757–771. https://doi.org/10.1007/s10098-020-01834-x

Madrazo J, Clappier A, Belalcazar LC, Cuesta O, Contreras H, Golay F (2018). Screening differences between a local inventory and the Emissions Database for Global Atmospheric Research (EDGAR). Science of The Total Environment 631–632:934-941. https://doi.org/10.1016/j.scitotenv.2018.03.094

Nogueira T, Kamigauti LY, Pereira GM, Gavidia-Calderón ME, Ibarra-Espinosa S, Oliveira GL, Miranda RM, Vasconcellos PC, Freitas ED, Andrade MF (2021). Evolution of Vehicle Emission Factors in a Megacity Affected by Extensive Biofuel Use: Results of Tunnel Measurements in São Paulo, Brazil. Environmental Science & Technology 55:6677-6687. https://doi.org/10.1021/acs.est.1c01006

Nogueira T, Souza KF, Fornaro A, Andrade MF, Carvalho LRF (2015). On-road emissions of carbonyls from vehicles powered by biofuel blends in traffic tunnels in the Metropolitan Area of Sao Paulo, Brazil. Atmospheric Environment 108:88-97. https://doi.org/10.1016/j.atmosenv.2015.02.064

Randles CA, Silva AM, Buchard V, Colarco PR, Darmenov A, Govindaraju R, Smirnov A, Holben B, Ferrare R, Hair J, Shinozuka Y, Flynn CJ (2017). The MERRA-2 Aerosol Reanalysis, 1980 Onward. Part I: System Description and Data Assimilation Evaluation. Journal of Climate 30(17):6823-6850. https://doi.org/10.1175/JCLI-D-16-0609.1

Ribeiro CB, Rodella FHC, Hoinaski L (2021). Regulating light-duty vehicle emissions: an overview of US, EU, China and Brazil programs and its effect on air quality. Clean Technologies and Environmental Policy. https://doi.org/10.1007/s10098-021-02238-1

Sawyer R (2010). Vehicle emissions: progress and challenges. Journal of Exposure Science & Environmental Epidemiology 20:487–488. https://doi.org/10.1038/jes.2010.44

SENATRAN, Secretaria Nacional de Trânsito, Ministério da Infraestrutura. 2021 (in Portuguese). Available online: https://www.gov.br/infraestrutura/pt-br/assuntos/transito/conteudo-denatran/frota-de-veiculos-2021

Shindell D, Faluvegi G, Walsh M, Anenberg SC, Van Dingenen R, Muller NZ, Austin J, Koch D, Milly G (2011). Climate, health, agricultural and economic impacts of tighter vehicle-emission standards. Nature Climate Change 1:59–66. https://doi.org/10.1038/nclimate1066

Sun S, Jin J, Xia M, Liu Y, Gao M, Zou C, Wang T, Lin Y, Wu L, Mao H, Wang P (2020). Vehicle emissions in a middle-sized city of China: current status and future trends. Environmental International 137:105514. https://doi.org/10.1016/j.envint.2020.105514

Tuia D, Ossés de Eicker M, Zah R, Osses M, Zarate E, Clappier A (2007). Evaluation of a simplified top-down model for the spatial assessment of hot traffic emissions in mid-sized cities. Atmospheric Environment. 41(17):3658–3671. https://doi.org/10.1016/j.atmosenv.2006.12.045

Unger N, Bond TC, Wang JS, Koch DM, Menon S, Shindell DT, Bauer S (2010) Attribution of climate forcing to economic sectors. Proceedings of the National Academy of Sciences 107(8):3382-3387. https://doi.org/10.1073/pnas.0906548107

United States Environmental Protection Agency (US EPA) (2020). Addendum Speciate Version 5.1 Database Development Documentation. Available online: https://www.epa.gov/air-emissions-modeling/speciate-51-and-50-addendum-and-final-report

Vasques TV, Hoinaski L (2021). Brazilian vehicular emission inventory software – BRAVES. Transportation Research Part D: Transport and Environment 100:103041. https://doi.org/10.1016/j.trd.2021.103041

Yarwood G, Jung J, Whitten GZ, Heo G, Mellberg J, Estes M (2010a). Updates to the Carbon Bond Mechanism for version 6 (CB6). 9th CMAS Conference, Chapel hill, NC. Avaliable online: https://www.cmascenter.org/conference/2010/abstracts/emery_updates_carbon_2010.pdf

Yarwood G, Whitten GZ, Jung J (2010b). Development, Evaluation and Testing of Version 6 of the Carbon Bond Chemical Mechanism (CB6). Texas Commission on Environmental Quality. Available online:https://www.tceq.texas.gov/assets/public/implementation/air/am/contracts/reports/pm/5820784005FY1026-20100922-environ-cb6.pdf