

Multispecies and high spatiotemporal resolution database of vehicular emissions in Brazil

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Dear reviewers,

We greatly appreciate the comments and suggestions you provided, which are very constructive and have contributed to enhance the content of the revised manuscript. Please find below the reply to all the reviewer's comments.

Best regards

Leonardo Hoinaski and coauthors

Reply to comments by Reviewer #1, Dr. Sergio Ibarra:

Reply to major comments

It is important to check the validity of comparison of the emissions with MERRA. The thing is that, yes, it is possible to compare emissions with concentrations but under specific conditions, for instance, when the boundary layer is low. In this way, the air pollutant concentrations should be representative of the emissions, as shown by Gallardo et al., 2012. As the database provided by Hoinaski et al present hourly factors, my recommendation is to compare under a similar set of conditions such as the ones presented by Gallardo et al. Author can find similar research on literature. Regarding the air pollutant concentrations, authors could use the CAMS global reanalysis (EAC4) Copernicus <https://www.ecmwf.int/en/forecasts/dataset/cams-global-reanalysis>.

Reply: We appreciate your suggestions. We have improved this discussion and reinforced the limitations when using this approach. Indeed, a comparison between datasets in specific regions and hourly averaging time would be more accurate. We have included a comparison according to your suggestion. To compare EDGAR, BRAVES database, and MERRA on an hourly basis, we have multiplied both BRAVES database and EDGAR annual emissions by a temporal disaggregation factor. We will analyze these three databases on 01/01/2013 at 8:00, when the boundary layer is not fully developed, and the traffic has the first peak period.

Lines 55-58: “Current inventories provide only annual emissions not reaching the spatial ... resolution... nor the concentration of chemical species...” According the to these words, the authors are stating the one problem of the emissions inventories are not providing concentrations. This is conceptually wrong because emissions are mass and concentrations mass over volume. I think the author meant the inventories usually do not provide the required speciation, which would be an English problem, but need to check.

Reply: Yes, you are right. We have fixed this issue in the manuscript by replacing “nor the concentration of chemical species” with “nor the emission of chemical species”.

Line 85: According to the CETESB (2019, the same reference used by author), RCHO is actually the sum of aldehydes and formaldehyde. Furthermore, CETESB also provides NMHC - ETOH emission factors. In addition, as the road transportation fuel consumed in Brazil has a vast origin on bio-fuels, 27% of gasoline is ethanol and 7% of diesel is bio-diesel, this results in a unique chemical signature of the chemical composition of fuel, hence the emissions. Actually, there are literature mentioning the high number of carbonyls in the vehicular emissions (Nogueira et al., 2015). According to table SM7, the species C₂H₄O (ALD2), C₂H₆O (ETOH) and CH₂O (FORM PRIMARY) are present in the NMHC speciation. Then, in order to provide a correct speciation, the author must reply the following questions:

Which pollutant are they using to perform the speciation? NMHC or NMHC – ETOH? Do the NMHC emission factors already consider RCHO? If the authors are using NMHC, it is supposed that ETOH and RCHO is already part of NMHC. However, this would result in a different chemical composition signature. Then, the recommendation would be preserving the proportions of ALD, FORM and ETOH and recalculate the speciation for the other compounds. I would say that this is more important for ETOH than RCHO. Can the author comment on that? Is this part of an ongoing work?

Reply: Thank you for this very insightful comment. We have fully adapted the code to preserve the original estimates using CETESB's emission factors, which is better than the speciation factors from Speciate to represent the ETOH and RCHO emissions in Brazil.

The estimates of total aldehydes (ALDX) are based exclusively on CETESB emission factors. Now, we speciate the acetaldehydes (ALD2 and ALD2_PRIMARY) and formaldehyde (FORM and FORM PRIMARY) using data from local studies. We have considered that aldehydes represent 50% and 22% of ALDX, from light-duty and heavy-duty vehicles respectively. Formaldehyde emissions (FORM and FORM_PRIMARY) represents 39% (light-duty) and 69% (heavy-duty) of ALDX emissions

We have modified the code and database to include the ETOH emissions from Flex Fuel vehicles running with ethanol when CETESB's emission factor is available (since 2018). ETOH emissions from other vehicles will still be estimated using NMHC speciation factors from Speciate.

References:

DOI: [10.1016/j.fuel.2014.05.091](https://doi.org/10.1016/j.fuel.2014.05.091)

https://fapesp.br/eventos/2016/02/mc/Maria_de_Fatima.pdf

Lines 102-104: While this method is conservative, the emissions need to be considered are currently expressed as points in space and not mass flux. Then, the current format is not appropriate for air quality models. In other words, the temporal mass must be divided by the area, for instance, the gases inputs to the WRF Chem model are expressed as $\mu\text{g}/\text{km}^2/\text{h}$. Then, the authors must recalculate the NetCDF output or add the proper flag indicating that the user must do divide by the area.

Reply: We have adapted the code and database to include the flag with the area of each pixel. Also, we provide a new option to generate ready-to-use WRFChem input files in hourly basis.

Reply to minor comments

There are many paragraphs consisting in less than two phrases. Each paragraph should have at least three parts, intro, body and conclusion. Please fix.

English needs to be revised

Line 83: "The software provides..." Improve English.

Reply: We have fully revised the English in the manuscript.

Line 84: Given that there are fuel consumption data by month available in Brazil, why the authors provide monthly emissions instead of calculating annual emissions with monthly profiles? Are you planning to improve this database?

Reply: Yes, indeed. We have been planning to generate the database using monthly fuel consumption. However, the National Petroleum Agency has not provided this data until now.

Figure 2: Include the resolution in the figure, also, increase the size of the legend and fire itself.

Reply: Ok! We will fix the resolution and size from figure 2.

Lines 120-121: can you a plot of the emission factors of RCHO and CO for heavy-duty and light-duty vehicles?

Reply: We will provide this figure as supplementary material.

Figure 4: present the first three plots horizontally and below the expanded hourly temporal factors.

Reply: Ok! We will plot them horizontally and enlarge this figure.

Lines 181-184; I think the author is being ambiguous. One thing is having the emission files according CB6 and another is that the files are according to these models. Each one of these models have emissions input files with different NetCDF characteristics. Then the authors must be more careful choosing the right words.

Reply: We have fixed this issue in the manuscript. We have targeted the species from CB06 to elaborate the input files. Each file has a different NetCDF structure and flags.

Figure 8: Make figure bigger, reduce space between Brazil for each comparison, use more pages if needed.

Reply: Thanks. We have fixed figure 8 following your recommendations.

Figure 9: Correlation figures seems distorted. These figures could be a) EDGAR, b) BRAVES, c) bias with the same color legend. For instance, negative could be blue, positive red, centered at 0, with degradation.

Reply: We have fixed this issue in figure 9.

Line 217. Why EDGAR is higher than BRAVES? On a recent paper (Nogueira et al., 2021) it was found that the CETESB emission actors need to be corrected to represent tunnel emission factors. The correction based on this publication is shown below. This correction is already available in the VEIN model (Ibarra-Espinosa et al., 2018).

Reply: EDGAR is higher than BRAVES because most of the road transport EFs are overestimated in EDGAR, as reported by Madrazo et al. (2018). Also, large discrepancies were found by Huneus et al. (2020) between EDGAR and local/national city emissions data

for the same domain. In addition, Huneus et al. (2020) showed that transportation emissions from EDGAR were higher than local city inventory in Rio de Janeiro.

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

Huneus, N., Denier van der Gon, H., Castesana, P., Menares, C., Granier, C., Granier, L., Alonso, M., de Fatima Andrade, M., Dawidowski, L., Gallardo, L., Gomez D., Klimont, Z., Janssens-Maenhout, G., Osses, M., Puliafito, S. E., Rojas, N., Ccoyllo, O. S., Tolvett, S., Ynoue, R. Y., 2020. Evaluation of anthropogenic air pollutant emission inventories for South America at national and city scale. *Atmospheric Environment*, 235, 117606. 10.1016/j.atmosenv.2020.117606

Line 238: registered or circulating fleet?

Reply: Circulating fleet. We have included this information in the manuscript.

Reply to comments by Reviewer #2:

1. there are local estimations regarding the composition and speciation of particles and VOCs which could be considered in the model or at least discussed why they were not considered.

Reply: We have adapted the code and database to include local data for better representing the Brazilian reality.

2. The legend in Figures 5 and 6 should include that the speciation of VOC and PM is based on SPECIATE (from EPA).

Reply: Thank you. We have included “Speciation factors from US EPA Speciate” in figures 5 and 6 captions.

3. The authors need to discuss the role of the biofuels used in Brazil in the total emission of VOC and the speciation considering this fleet.

Reply: We have included a new discussion in the manuscript to provide a baseline to understand the biofuels in Brazil and the role of ethanol.

4. the segregation of the streets is considering the type of vehicles or only an average of the composition with homogeneous distribution.

Reply: We have calculated the road density considering all roads in the OpenStreetMaps shapefile. We have not segregated by fleet category or road type.

5. the graphical presentation of the difference between inventories is a good visual form but I suggest a table with the total emissions comparing the different inventories, including the Brazilian one.

Reply: Ok! We will provide this table as supplementary material.