Review of "Impact of vehicular emissions on the formation of fine particles in the Sao Paulo Metropolitain Area: a numerical study with the WRF-Chem model" by A. Vara-Vela et al.

This paper by Vara-Vela et al. aims to evaluate the impact of vehicular emissions on aerosol formation in the Sao Paulo Metropolitan Area (SPMA). To achieve this goal, a modelling study performed with a regional model over 3 domains (75, 15, and 3 km resolution) and 3 different simulations has been done, supported by aerosol and ozone observations from ground-based stations in the SPMA. Vehicular emissions were estimated by an emission model to take into account the traffic and specific emissions from SPMA vehicles mostly fuelled with ethanol based gas. The baseline simulation was run with vehicular emissions only as anthropogenic emissions whereas the second simulation was run without aerosol emissions in order to estimate the amount of particle secondary formed in the total PM amounts. Down-town SPMA, 20 to 30% of the PM2.5 amount were found to be due to secondary formation. The third simulation was run with aerosol feedbacks turned on to estimate the impact of aerosols on the ozone photochemistry. A decrease of 2% in the ozone concentration was found down-town SPMA due to the aerosol impact on ozone photochemistry.

The organisation of the paper needs to be revised and the English deserved improvements. Introduction is too short and need to be complete with a general context and background on the different aspects discussed in the paper. The model configuration description raises several issues which might affect significantly, depending on the answers of the authors to my comments, the results of this study. Then, the result and discussions section must be improved as well since results are just listed and figure described with only little discussions (sometimes inconsistent) on the outcomes of the study.

A list of the issues raised by the paper is given in the general comments whereas details are listed in the specific comments.

General comments:

- The introduction needs to be substantially improved. The general context of the study is not described and, since it's a rather new area of research, detailed informations on the emissions of ethanol/gasohol/flex-fuel fuelled vehicles are expected, as well as background on ozone formation.

- The methodology section deserved important improvements as well. The description of the observation techniques and datasets are incomplete and should be better organised. The explanation on the methods used for the calculation of anthropogenic emissions is confused and should be clarified. Non-vehicles anthropogenic, such as fire emissions are not included in the simulation despite industrial activity, heating and cooking in the SPMA and important fire activity in the simulation domains during the simulation period. This point is crucial and model results might be significantly affected by such missing inputs, and more important, this might affect scientific results as well since SOA formation is not linearly dependant to precursor concentrations.

- The result section is composed of several sub-sections which doesn't really connects between each others. In addition, the results are given but only little discussion is provided and, most of time, the organisation of the section is chaotic, which is really confusing for the reader. The authors are expected to reorganise more clearly this section and to discuss the results with the help of a context provide by references.

Specific comments:

Abstract

P. 14173, L. 9-10: the beginning of the sentence "The study period... " is unclear, please reword. Suggestion: The simulation has been performed on a 1 month period (7 August-6 September 2012) to cover the availability of experimental...

P. 14173, L. 13-16: the sentence "Results show that the emissions... in the downtown SPMA." is ambiguous and should be reword. State more clearly that 20-30% of the PM2.5 mass is due to secondary aerosols.

P. 14173, L. 16-17: Dust and Sea-salt contributed to 40-50% of the PM10 mass. Why not giving a percentage of the PM2.5 mass, in order to be able to compare it to the contribution of secondary aerosols ? Alternatively, contribution of secondary aerosols could be given as a percentage of PM10.

P. 14173, L. 23-24: "which simulates feedbacks...chemical species". This part of the sentence has no link with the following. It would better fit a the beginning of the abstract (L. 5-6).

Introduction

P. 14174, L1: the introduction should starts by a general paragraph describing the general context in which this study take place, e.g., why is it important to study PM2.5, ozone, SOA,... ? What are the impacts of vehicular emissions on air quality and climate, generally ?

P. 14174, L. 8-26: To support this significant part of the introduction, the authors referred to a report written by the Sao Paulo Environmental Protection Agency (CETESB) in Portuguese. At least a part of this information should be support by peer-review material – few papers in the reference list adequately match this scope.

P. 14174, L. 15-18: It is claimed that "SPMA has a significant fleet... gasoline and ethanol).". In this section, informations on these specific fuels should be given – Are emitted pollutants the same as for classical fuel ? Does previous studies exist ?

P.14175, L. 14-15: "One of the most important aspect of this work is the quantitative analysis of the formation of PM2.5 and ozone". Please add a paragraph introducing ozone formation mechanisms and explaining why it's relevant in the context of your study. In addition, the statement that "primary pollutants have rather increased in the past years" is vague and should be support by a reference.

P 14175, L. 26-29: More specific informations needed, how many measurement sites ? Is there a reference describing the NUANCE-SPS project and/or the field campaigns. Also, the sentence "In order to achieve these goals, aerosol measurements were taken..." don't fit with the previous sentence in which a numerical simulation is mentioned... this part should be reword and reorganised in order to provide a clear message to the reader.

Methodology

P. 14176, L. 14-15: The beginning of this sentence should be reword, e.g.: "Aerosol observation datasets used in this work were collected using..." If you really want to keep mentioning the PM2.5 and PM2.5-10 acronyms, please define PM2.5-10.

P. 14176, L. 15-16: Please provide references which describe the dichotomous sampler and the MOUDI impactor used in the study. If references doesn't exist, more detailed descriptions of the instrument measurement techniques, as well as their efficiency in collecting particles is necessary.

P. 14176, L. 18-19: the expression "after filter" is rather vague and should be replace by a more specific one.

P. 14176, L. 19: "The collected filters and substrates". It is unclear so far, where filters and substrates are coming from ? Are filters from the MOUDI impactor and substrates from dichotomous sampler ? Such informations should be stated clearly when instruments are described.

P. 14176, L. 20-24: To which samples (filters, substrates, both ?) X-ray, gravimetric, reflectance and thermo analysis were applied ? Ion chromatography is applied to material collected "on the membrane filters". Are membrane filters the same as filters mentioned above ? In addition, the analysis techniques should be support with references.

P. 14177, L. 7-12: The sentence "The WRF-Chem model (Grell et al. 2005) is an online mesoscale

meteorological model ...." is incomplete. WRF-Chem is a fully coupled (as mentioned later in the text) online meteorological and chemical transport model and this information should appear first in this section. Then, the WRF acronym should have been define earlier (p. 14176, L. 1).

P. 14178, L. 6 and 18-19: "It is parametrized in WRF-Chem..." Such parametrisation is not included in all the WRF-Chem aerosol scheme, thus WRF-Chem should be replace by MADE-SORGAM. The same applies for "The primary organic aerosol (POA) in WRF-Chem..."

P. 14178, L. 22-27: In which category would SPMA fit the best ? Low amount of SA formation or area with a significant wood smoke influence ? What would be the expected OM:OC ratio in SPMA ?

P. 14179, L. 1: Indicate the model version

P. 14179, L. 16-23: This paragraph is inconsistent with Table 4, at least anthropogenic aerosol emissions are not mentioned in the text. Or, does the model feed with dust and sea salt only as primary aerosols ? This paragraph needs to be clarified.

P. 14179, Model description: The spin-up period used to initiate the model should be given somewhere in this section.

P. 14180, L. 3-9: Is the LAPAt model able to differentiate, e.g., light vehicles as basic cars and heavy duty trucks ? The answer to that question should be given in the text in addition to an explanation of how the difference is made or why the model can't differentiate the different vehicles.

P. 14180, L. 23-28: This paragraph is confusing and needs clarification. "...areas inside both grid cells", do you mean in both domains ? What are the others metropolitan areas ? How many inhabitants vs SPMA ? Where are they located in the 3 and 15 km domains ? Where does "the number of vehicles in each one of the main urban areas" comes from ?

P. 14181, L. 14-17: Does the sentence "Furthermore, due to the complexity... for distributing the emissions during the day in both grid cells." means that a constant value has been used for vehicle emissions during the day ? If true, why not applying a diurnal cycle as observed in many megacities ?

P. 14181, anthropogenic emissions section: The authors choose to force the model with vehicle emissions only. However, one would expect the presence of other anthropogenic emissions, e.g., industrial activities, able to impact SOA formation and PM concentrations. Have these emissions been evaluated ? How this lack is addressed in the study ? There is no mention of the emission used for the 75 km domain, please provide this information.

P. 14182, L. 5: Update the MEGAN reference to Guenther et al. (2006)

P. 14182, Other emissions section: No fire emissions are used in this study. However, as visible on online tools (e.g., https://firms.modaps.eosdis.nasa.gov/firemap/), and as claimed in this study (section 3.2 and Fig. 5), important fires occur in Brazil at the period of this study. The validity of the back-trajectory presented in Fig. 5 is discussed further in this review, but missing fire emissions may induce important bias in the model outputs and thus in this study results.

Results and discussions

P. 14183, L. 3-6: "According to the climate reports... intensification of the South Atlantic Subtropical high (SASH)". How much observed precipitation rates are lower than climatological values ? A reference is necessary to support the statement that SASH is responsible for precipitation anomalies. Which impact, if any, such precipitation anomalies would have on the study ?

P. 14183, L. 7: "These conditions established an easterly wind anomaly pattern at the 850 hPa level". Where this information comes from ? Which kind of anomaly ?

P. 14183, L. 17-19: "Figure 3 shows the accumulated daily precipitations and humidity". It looks like precipitation are not daily accumulated, please check and reword accordingly. In addition, the (relative ?) humidity is presented in Fig. 3 but not discussed in the text.

P. 14184, L. 1-2: "Part of the unexplainable concentration is related to the water content of aerosols". "remaining mass" should be used instead of "unexplainable concentration". What do you mean by "water content of the aerosols" ? Is the remaining mass water ? Please be more specific and support this assumption with a reference.

P. 14184, L. 3-9: "On the other hand... SASH system is moved away from the continent." Why is the semi-stationary front situation not described in section 3.1 ? The main information from this paragraph is that the meteorological situation is the main driver of the PM concentration, how is it compatible with this study ? It is claimed that an aerosol increased could be due to "an increase in relative humidity", however, such increase of relative humidity in not noticeable in the observed relative humidity time series in Fig. 3. Is there another reason which could explain it ?

P. 14184, L. 9-10: "Aerosols coming from forest fires... during this period." This sentence belongs to the short discussion on forest fires (L. 21-24).

P. 14184, L. 19: "AQUA_M-T" M-T needs to be defined. More informations on the satellite and the product(s) used to detect fire locations should be provide, as well as references. In Fig. 5 legends, it appears that an other satellite was used (Terra), as well as MODIS and NOAA products.

P. 14184, L. 21-25: "Figure 5 shows... reaching the measurement site." First a reference to the back-trajectory model, i.e. HYSPLIT, should be given (as requested on the HYSPLIT website). On how many days the back-trajectory has been calculated ? How would be back-trajectories for other peaks ? As mentioned previously, important fires occurred in Brazil during the study period and it is crucial to be sure that fire emissions can be ignored.

P. 14184, L. 26-29: "The increasing organic matter...efficient formation of secondary particles." The statement on a possible impact of PBL height on a "more efficient formation of secondary particles" must be supported by references. The expression "high vehicular emission events" is unclear and should be reworded.

P. 14185, L. 15-16: The correlation coefficient between model results and observations are not very high, especially for temperature. Is the model nudge in the boundary layer with NCEP FNL data ? How the rather low correlation coefficient would impact the simulation of PBL and pollutant concentrations ? An additional figure showing the T, RH, WS, WD time series would help a lot in understanding where and why the model partly fails in reproducing observations.

P. 14185, L. 17: "temporal variations" is not an appropriate expression to describe average wind and temperature fields. The period on which the fields have been averaged should be given.

P. 14185, L. 19: "(i.e 17.7°C at AF-IAG and 17.8°C at INT)" How does these values compare with observations ?

P. 14185, L. 20: "the predicted wind direction was easterly in SPMA" this sentence is in contradiction with the sentence p. 14184, L. 25-26 "The predicted average surface wind was predominantly from southeast (see Fig. 6)". Please modify the wrong sentence. How this wind direction compares with observations ?

P. 14186, L. 2: What is the term "point sources" uses for ? More specific words are expected here.

P. 14186, L. 3-5: "The high concentration of PM2.5, ... establishment of lower PBL heights". There is no higher ozone concentration neither at the beginning or at the end of the study period (as seen in Fig. 9). "reasonably well captured" is vague and certainly don't apply to the PM time series presented in Fig. 7 and 8 where simulation can miss up to 50% of the observed PM concentrations. Why "the emission of high aerosol loadings" and "lower PBL height" would happened ? Emission rates and PBL height are available parameters, emissions because the authors created them and PBL heights because it's a basic output of the model (it can alternatively be calculated with basic meteorological parameters). Why not showing variations of such parameters as a figure to support these statements ?

P. 14186, L. 6-7: In relation with the previous comment, the PBL height value given here is not sufficient. To support the reasoning, it should be completed with PBL height of the beginning and ending periods as well as informations on the origin of this value (from the model ? observations ?).

P. 14186, L. 13-18: Figures 11 and 12 are not described nor used as support for discussions. However, high concentration patterns are visible on both figures, between the coast and SPMA and at the west edge of the domain. Are such patterns expected ? What are their origins ?

P. 14186, L. 18-20: A high PM2.5/PM10 ratio, meaning that most of the mass is due to particles with diameter smaller than 2.5 µm, would be expected in high vehicular emission areas, why isn't it the case ? Impact of vehicular emissions is the main focus of this study, in that regard discussions should be focusing on it.

P. 14187, L. 11-13: Why only focusing on 16 days instead of the entire simulation period ?

P. 14187, L. 22: "around 55 and 40%" please remove around and give the exact values.

P. 14188, L. 5: Rename the section as "Contribution of dust and sea-salt to PM concentration" or approaching since this is more relevant to describe the actual content of the section.

P. 14188, L. 8-9: "The simulated average ratio...concentration is shown in Fig. 17b." This sentence is redundant with the previous one. On which period the average is calculated ?

P. 14188, L. 15: How can forest fires be involved in emissions if they are not provided as input ?

P. 14188, L. 18-24: this paragraph has no connexion with the purpose of the section, it should be move elsewhere, or the section should be renamed.

P. 14189, L. 7-8: "But for the SPMA, the importance of SOA... transport sector was noted." A reference is needed to support that statement.

P. 14190, L. 2-6: Since it's the focus of the study, OC time series in Fig. 14 should be discussed in this paragraph.

P. 14190, L. 17-19: Is it an average or at a given time (16:00 LT) ?

P. 14190, L. 22-24: How aerosols impact ozone formation in the SPMA morning ? How does it compares to Li et al. (2011a) ?

P. 14191, L. 5-7: There is no link between shortwave and longwave radiation, this sentence should be removed.

P. 14191, L. 11-16: Results from this study should be compared to these from the references given, otherwise, if references are given to acknowledge previous work, they belong to the introduction section.

Summary and conclusions

P. 14191, L. 27: Ozone concentrations are not lower than observations, at least, this is not obvious on Fig. 9. This statement should be reinforced with numbers or modified adequately.

P. 14192, L. 10-12: The 2% might be higher when looking at the morning values ? The afternoon context should be given in the text.

Tables and figures

Table 5: define UB (in "RMSE_UB")

Figure 1: Is topography from the model ? Add the information in the caption. Replace "with information of ..." by "with information on ..." twice in the caption.

Figure 2: Is this figure for a week day or a week-end day ?

Figure 3: This doesn't seem to be daily precipitation data, please check and modify the caption accordingly.

Figure 4: The meaning of the 6 panels should be describe in the caption. The 4 bottom panels are not discussed within the text. Remove "some" or replace it by the list of aerosol constituents.

Figure 5: Mention the sources of fire back-trajectory data.

Figure 6: Mention the period on which data are averaged. This not surface temperature and wind but 2 m temperature and 10 m wind speed and direction, please modify the caption accordingly.

Figure 9: This is obviously not daily data, please modify the text accordingly.

Figure 10: A reference to the Taylor (2001) paper should be given, either here or in the text.

Figure 11-12-13: Mention the period on which the data are averaged.

Figure 15: Mention in the caption that no model data are available for > 1µm, as well as the intruments used to measure the concentrations.

Additional references

Guenther, A., T. Karl, P. Harley, C. Wiedinmyer, P. I. Palmer, C. Geron (2006) Estimates of global terrestrial isoprene emissions using MEGAN (Model of Emissions of Gases and Aerosols from Nature), *Atmos. Chem. Phys.,* 6, 3181-3210

Taylor, K. E. (2001), *Summarizing multiple aspects of model performance in a single diagram*, *J. Geophys. Res.*, 106(D7), 7183–7192, http://dx.doi.org/10.1029/2000JD900719

Technical comments:

Introduction

P. 14174, L. 10: missing "(" between "PM10" and "< 10 µm..."

P. 14174, L. 11: typo "emissions" instead of "emssions".

P. 14174, L. 12: replace "as per the estimates of" by "as estimated by".

P. 14175, L. 14: replace "aspects" by "aspect".

P. 14176, L. 1: "The WRF-Chem model" + define the acronym.

Methodology

P. 14176, L. 9: replace "is" by "was".

P. 14176, L. 13: remove "the".

P. 14176, L. 16: "...collected mass of particle..." this sounds weird, maybe remove "mass of" .

P. 14177, L. 13-14: "atmosphere compressible", please switch the two words.

P. 14178, L. 23: define SD

P. 14179, L. 11: the initial and boundary chemical conditions are not global, remove the word "global"

P. 14179, L. 12: replace "are obtained" by "were obtained"

P. 14179, L. 12: For clarity, add the acronym MOZART-4 for "Model for Ozone and Related chemical tracers, version 4"

P. 14180, L. 28: replace "at" by "in"

P. 14181, L. 11 and 17: replace "grid cells" by "domains"

P. 14181, L. 26: replace "is used" by "was used".

P. 14181, L. 26: replace "to assimilate emission rates to WRF..." by "to scale emission rates on WRF..."

P. 14182, L. 16: replace "of" by "on"

Results

P. 14182, L. 25: replace "Firstly, it is" by "It is first"

P. 14183, L. 4: replace "lesser" by "lower"

P. 14183, L. 7: replace "anomalies" by "anomaly"

P. 14183, L. 8: replace "comes" by "transport"

P. 14183, L. 9: replace "Tropicial" by "Tropical"

P. 14183, L. 11: What is the Western State ? Reword

P. 14183, L. 11-12: "precipitation areas" is unclear, "precipitation events" ?

P. 14183, L. 12: replace "observed at the second half" by "observed during the..."

P. 14184, L. 16: replace "BC" by "EC"

P. 14184, L. 25: "southeast", isn't it "northeast" ?

P. 14185, L. 12: replace "explore" by "evaluate"

P. 14185, L. 13: remove "some"

P. 14185, L. 21: remove "is" in "as is examined"

P. 14187, L. 6: remove "among"

P. 14187-14188, L. 29-1: "...ratios were underestimated in the range 5-40% against 50-80% observed." This sentence is unclear. Does it mean that the model simulates a ratio of 5-40% whereas the observed ratio was 50-80% ?

P. 14188, L. 13: replace "would be associated with the emissions of primary..." by "would be the primary..."

P. 14189, L. 2: remove "the" from "(the MADE/SORGAM)"

P. 14189, L. 16: replace "performed" by "calculated"

P. 14190, L. 14: remove "on"

P. 14190, L. 18-19: the sentence " are shown by Case_0 and Case_2, respectively (Fig. 17c)" has no sense and should be replace by "are shown in Fig. 17c."

P. 14190, L. 19-20: replace "transported" by "transport"

P. 14191, L. 1: typo "shortwave"

P. 14191, L. 9-11: The end of the sentence "... shorwave radiation what...concentrations to a few ppb." is unclear and should be reworded.

P. 14191, L. 15: "absorption"

Summary and conclusions

P. 14192, L. 8: replace "come" by "coming"

P. 14192, L. 8: replace "has" by "have"