Response to Referees' comments

Angel Vara-Vela, M. F. Andrade, Prashant Kumar, R. Y. Ynoue, and A. G. Muñoz

We are attaching a new version of the manuscript trying to answer all the questions that were presented. We would like to thank both referees for all the suggestions that we believe improved the quality of the work clarifying the objectives and conclusions.

Anonymous Referee #1

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...

The sentence was reworded to "The simulation has been performed on a 1 month period (7 August – 6 September 2012) to cover the availability of experimental data…".

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.

The sentence was reworded to "Results show that emissions of primary gases, mostly from vehicles, led to a production of secondary particles between 20-30% in relation to the total mass concentration of PM2.5 in the downtown SPMA".

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.

The percentage of the PM2.5 mass was added in the text.

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).

That sentence was moved to the beginning of the abstract.

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 ?.

The introduction was modified including a paragraph describing the general context of emissions and their impacts on air quality in SPMA.

"The Sao Paulo Metropolitan Area, in the southeast region of Brazil, is considered a megalopolis comprised of Sao Paulo city and other 38 municipalities. One main concern is the occurrence of violations of air quality standards for ozone and fine particles at different air quality stations from the Sao Paulo Environmental Agency (CETESB). The air pollutant emissions in the SPMA are related to the burning of the fossil fuels ethanol, gasohol (gasoline with 25% ethanol) and diesel. They constitute the main cause of impairment to air quality in SPMA, but the number of air quality standard violations has decreased for almost all pollutants with the exception of $PM_{2.5}$ and O_3, that, similar to other big cities, impacted by the vehicular emissions, have experienced an increase in the number of violations of their air quality standards as discussed in depth by Carvalho et al. (2014). Perez-Martinez et al. (2015) have analyzed the monthly mean values for the regulated pollutants from 2000 to 2013 for the air quality stations in SPMA and have found that there was a decrease in the average concentration of NOx, CO, and PM_{10} by 0.65, 0.37, and 0.71 % $month^{-1}$, respectively, although the sales of the fuels gasoline, ethanol, and diesel had increased by 0.26, 1.96, and 0.38 % $month^{-1}$.".

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.

Some references published in peer-review journals were added, bringing studies regarding the air quality in SPMA: Carvalho et al. (2014) and Perez-Martinez et al. (2015).

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 ?.

Additional references discussing the role of emissions in SPMA were added in this part of the introduction

"Many studies regarding the air quality impact of the bio-fuels have been performed, especially in Brazil. Anderson (2009) in a review concerning ethanol fuel use in Brazil

highlighted that the atmospheric concentrations of acetaldehyde and ethanol are much higher in Brazil comparing to those in other areas of the world. Costa and Sodre (2010) showed that exhaust emissions of hydrous ethanol presented reduced CO and Hydrocarbons (HC), but increased CO_2 and NOx levels.".

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.

A sentence to reinforce the relation between ozone and aerosol particles was added in the text.

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 reorganized in order to provide a clear message to the reader.

The sentence was better reorganized. An additional NUANCE-SPS reference was also given.

<u>Methodology</u>

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.

The sentence was reworded to "Aerosol observation datasets used in this work were collected using a dichotomous sampler (Wedding et al., 1980) and a Micro-Orifice Uniform Deposit Impactor (MOUDI, model 100; MSP Corporation – Marple et al., 1986).".

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.

The reference was added in the text.

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

The expression was replaced by "subsequent stage" in the text.

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.

A sentence to clarify where filters and substrates come from was added in the text.

"The samples collected with the MOUDI impactor were deposited on a polycarbonate membrane filter with 0.4 µm porous and for the Dichotomous sampler the substrate was a teflon membrane filter with 2 µm porous. The after-filter in the MOUDI impactor is a 33 mm teflon membrane, that was not submitted to the reflectance analysis.".

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.

The text was clarified. The filters and membrane filters are related to the same material.

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).

The sentence was reworded to "The WRF-Chem model is a fully coupled online meteorological and chemical transport model (Grell et al., 2005), supported by…around the world".

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...".

The WRF-Chem was replaced by MADE in both sentences.

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 ?.

The formation of secondary aerosols in SPMA is very high, and thus, a high OM:OC ratio, probably higher than 1.54, is expected.

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

The model version was included in the text.

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.

The paragraph was reworded to "WRF-Chem simulation with coupled primary aerosol (dust, sea salt and anthropogenic) and gas (biogenic and anthropogenic) emission modules, together…".

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

The sentence "The first seven days of each simulation were not analyzed and used for model spin-up." was added 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.

Yes, it is. In order to differentiate light and heavy vehicles, the LAPAt model applies the total fleet distribution to the total number of vehicles in each grid point within the domains. The total number of vehicles in each grid point of both domains is calculated from the normalization of the sum of individual intensities at each point (i.e. total road length for the 3-km modelling domain and night-time light for the 15-km modelling domain) by the total fleet, so that emissions in urban areas are mainly represented by emissions coming from their vehicles. A better explanation was added to the text.

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 ?.

Yes, I do. The expression "grid cells" was replaced by "modelling domains" throughout the manuscript. Basically, each grey stain on Fig. 2 (3-km modelling domain) represents an urban area, e.g., the second largest grey stain represents the Campinas Metropolitan

Area (CMA). The number of vehicles comes from DENATRAN (see P.14180.L.13-15). There were 3 metropolitan areas in Sao Paulo State until 2012. They were: Metropolitan Area of Sao Paulo (19683975 inhabitants in 2010 Census), Campinas (2797137) and Baixada Santista (1664136). In 2012, the MA of Vale do Paraíba e Litoral Norte was created and, in 2014, the MA of Sorocaba.

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 ?.

No, it doesn't. We have applied a diurnal cycle at all grid points where emissions have values greater than zero. The calculation of that diurnal cycle follows the approach used by Fast et al. (2006) in which gas and particle emission profiles were calculated from median diurnal variations on weekdays and weekends.

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.

Other anthropogenic emissions have not been considered because this study focuses on the impact of on-road vehicle emissions on the concentration of fine particles. The sentence "Anthropogenic emissions were not considered in the 75-km modelling domain." was added in the text.

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

We used the Guenther scheme (default option) which is based on Guenther et al. (1993; 1994).

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.

New back-trajectories were calculated using the suggestion on P.14184,L.21-25.

Results suggest that aerosols from either forest fires or biomass burning areas can be advected to SPMA impacting, somehow, on the concentration of aerosol particles over this region.

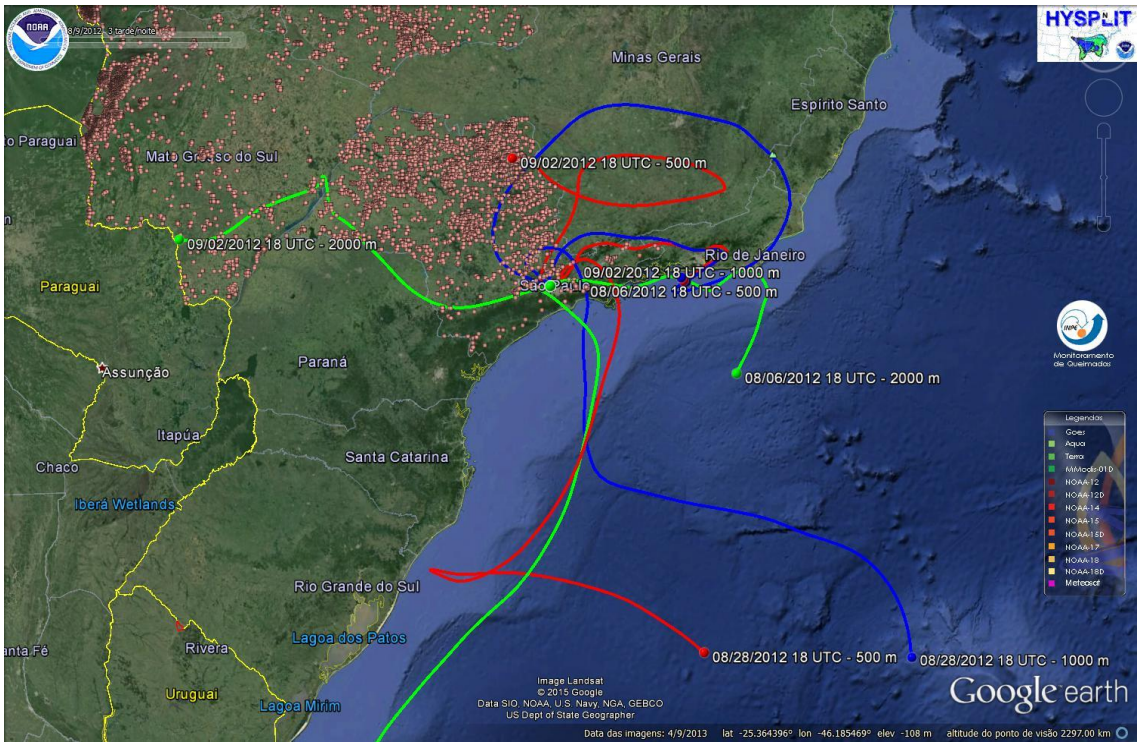

Figure 4. HYSPLIT three-day backward trajectories and locations of fires in Sao Paulo State and part of central-west region of Brazil. Pink markers represent the observed fire locations during the study period considering different satellite products (GOES, AQUA, TERRA, NOAA). The backward trajectories starting at IAG-USP were calculated for the days 9 and 31 August and 5 September 2012 at three different altitudes: 500 m (red lines), 1000 m (blue lines), and 2000 m (green lines).

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 ?

In SPMA, the observed precipitation rate was 38.6 mm lower than the climatological value for that month (August). This information is based on GrEC (2012a). The impact

of negatives anomalies, in terms of aerosols, implicates a less efficient removal of particles and gases which are able to form new particles.

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 ?

This information also comes from GrEC (2012a).

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.

The sentence has been reworded to "Fig. 3shows the hourly accumulated precipitation and relative humidity observed at the IAG-USP's climatological station".

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.

Yes, water content of aerosols means the remaining mass water. The expression "unexplainable concentration" was replaced by "remaining mass". A reference was added in the text.

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 ?

Another possible reason could be the transport of aerosol particles produced by forest fires in the central-west region of Brazil and the Sao Paulo State (see new Fig. 4).

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).

This sentence was removed.

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.

The expression "AQUA_M-T" was removed. The new caption of Fig. 4 (ex Fig. 5) mentions the forest fire products displayed in the legend.

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.

The HYSPLIT reference was added in the text. Fig. 4 shows three-day backward trajectories of air masses for the days 9 and 31 August and 5 September, when increases in the OC and EC concentrations were observed at IAG-USP. It is possible to note that some of trajectories have crossed forest fire areas before they come to SPMA, impacting on the concentration of aerosol particles over SPMA.

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.

The increasing organic matter may have two possible explanations. First, it should be noticed that the diurnal cycle of emissions is the same for every day, but sometimes there are traffic incidents which may rise the emissions. These specific incidents were not taken into account in the simulations. Second, if there are no traffic incidents, the increase of aerosol particles is only due to different meteorological scenarios. High-pollution episodes occur when winds are light and the PBL height is low and these meteorological conditions are met under post-frontal anticyclonic influence.

The expression "high vehicular emission events" was replaced by "traffic incidents which may raise the emissions". Additionally, a reference regarding the aerosol – meteorology relation was added in the text.

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.

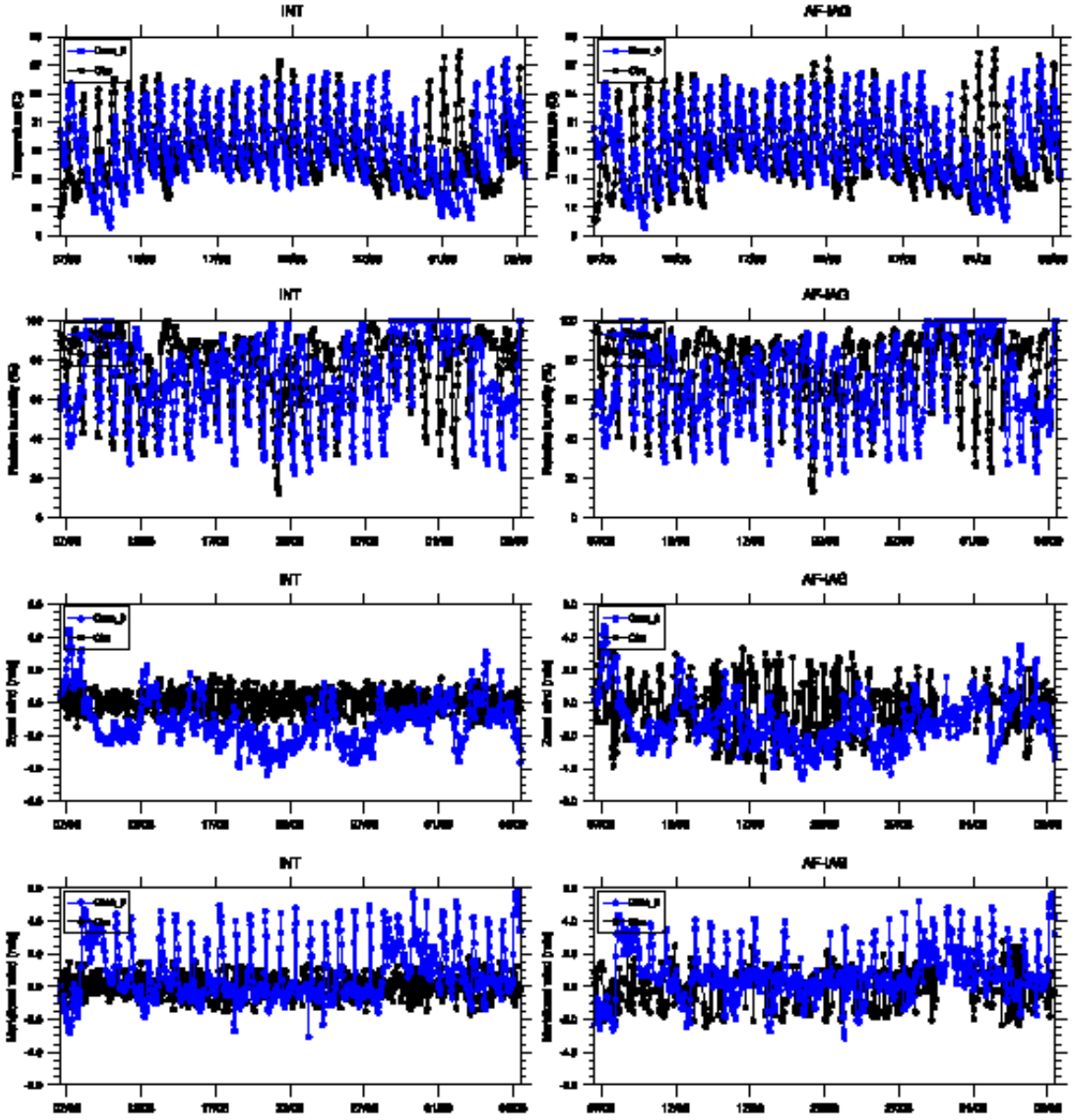

Figure 18. The observed and predicted hourly variations of temperature, relative humidity, and zonal and meridional wind components at INT and AF-IAG for the 3-km modelling domain.

We don't have PBL height observations to compare and evaluate the model performance. Mismatches in temperature and wind may lead to either underestimating or overestimating the PBL height calculation, increasing or decreasing, consequently, the amount of pollutants within this layer.

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.

The sentence was reworded to "Fig. 6 shows the predicted average of wind vectors at 10 m and temperature at 2 m for the whole study period in the 3-km modelling domain".

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 ?

The values 17.7 and 17.8°C are the observed average temperatures at AF-IAG and INT, respectively. These values are close to the predicted averages which range between 16 and 17 °C according to Fig. 6.

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 ?

The wrong sentence has been removed.

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

The term "point sources" was replaced by "industry".

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 ?

The sentence was modified. Traffic incidents may raise the emissions of aerosols (aerosol loadings) in the atmosphere. Lower PBL heights are commonly observed under post-frontal situations.

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 ?).

The sentence was modified, indicating the predicted average PBL height at the beginning and ending periods. All values of PBL height provided in this study come from model outputs.

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 ?

Aerosol particles between the coast and SPMA are expected to have high concentrations because of the influence of the ocean (sea salt aerosols) in addition to any coastal urban areas like Baixada Santista, whereas at the west edge, this contribution mainly comes from the biogenic emission and forest fires.

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.

From Fig. 13, higher PM2.5/PM10 ratios over offshore continental areas may be associated with a more efficient formation of fine particles from oxidation of biogenic VOCs as well as primary emission of biogenic POA, mostly small particles. Furthermore, it should be noticed that there is no other relevant sources of primary aerosols over these areas. So, at places where there is a high impact of resuspended soil, it is possible to observe that the coarse fraction explains most of the PM10 concentrations.

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

We have focused only on the days for which there are measurements of aerosol mass size distribution.

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

The sentence was reworded to "...mass with contributions of 55 and 40%, respectively".

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.

The section was renamed to "Contribution of dust, sea salt, and coarse anthropogenic aerosols to PM concentration".

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 ?

The second sentence was reworded to "The simulated average ratio between dust – sea salt aerosols and the total PM_{10} mass concentration is shown in Fig. 17b.". The average is calculated for the whole study period.

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

Although forest fires are not directly involved in the emission, it's important to indicate that MOZART's gas and particle background concentrations - used as initial and boundary chemical conditions in the simulations - were previously calculated using a biomass burning emission global inventory (FINNv1).

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.

The section was renamed to "Contribution of dust, sea salt, and coarse anthropogenic aerosols to PM concentration".

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.

The reference was added in the text.

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.

A sentence discussing OC and EC concentration peaks was added in the text.

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

It's an average for a given time, in this case 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) ?

According to model results, in downtown SPMA, aerosols have a less expressive impact on ozone photochemistry during the morning (~-1%), but can impact positively in other SPMA regions range between -1 – 1 %.

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

Even though there is no link between shortwave and longwave radiation, this sentence reinforces the fact that inclusion or neglect of the direct effect of aerosol particles can impact the predicted downward longwave radiation, impacting consequently on the predicted surface temperature.

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.

The paragraph "The impact of the fine particles has been discussed in previous works, with evaluation of the scattering and absorbing effects of the aerosol (e.g. Li et al., 2005; Real et al., 2011). Vehicular emissions of particulate matter in the SPMA have a high percentage of BC (Brito et al., 2013), which after emitted to the atmosphere can enhance the absorption coefficient and thus the attenuation rates" was moved to 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.

As mentioned in the section 3.3, mean biases for PM2.5, PM10 and O3 concentrations were -8.84 μg m^{-3}, -14.13 μg m^{-3} and -0.85 ppb, respectively (see Table 5). So, the sentence was reworded to "However, the predicted concentrations of PM2.5, PM10, and O3 (but in minor intensity) were lower than observed values.".

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

The sentence was reworded to "...O3 concentration by around 2% in the afternoon (16:00 LT) when the aerosol-radiation feedback..."

Tables and figures

Table 5: define UB (in "RMSE_UB")

RMSE_UB is the RMSE after a constant bias is removed. The Appendix A describes all the statistics used in this work to assess the model skill.

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.

The caption was reworded to "Downtown area of the 3-km modelling domain (d03) showing the locations of measurement sites and WRF topography in the vicinity of SPMA. Red dots represent sites with information on O_3 and aerosol. Yellow dots represent only sites with information on PM. Blue dot represents the location of the IAG-USP's climatological station.".

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

In our emission model, the emissions have the same diurnal cycle for every day which was calculated from median diurnal variations on weekdays and weekends.

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

The caption was reworded to "Hourly accumulated precipitation and relative humidity observed at the IAG-USP's climatological station during the study period.".

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.

The caption was reworded to "Daily (top), diurnal (bottom), and nocturnal (middle) mean concentrations for EC, OC, PM_{10}, $PM_{2.5-10}$, $PM_{2.5}$ (left panels), and Na, Fe_2SO_3, SiO_2, K_2O, and S (right panels). The $PM_{2.5-10}$ aerosol variable is defined as particulate matter with aerodynamic between 2.5 and 10 μm. The grey line indicates the WHO air quality standard for $PM_{2.5}$ (25 $\mu g/m^3$).".

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

The caption was reworded to "HYSPLIT three-day backward trajectories and locations of fires in the Sao Paulo State and part of central-west region of Brazil. Pink markers represent the observed fires locations during the study period considering different satellite products (GOES, AQUA, TERRA, NOAA). The backward trajectories starting at IAG-USP were calculated for the days 9 and 31 August and 5 September 2012 at three different altitudes: 500 m (red lines), 1000 m (blue lines), and 2000 m (green lines).".

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.

The caption was reworded to "The predicted average of 10 m - wind vectors and 2 m -

temperature from the baseline simulation (Case_0) for the whole study period in the 3-km modelling domain. Blue dots represent the location of the measurement sites, whereas cyan numbers represent the observed average temperature in those sites: 17.7 ^{o}C in AF-IAG and 17.8 ^{o}C in INT.".

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

The caption was reworded to "The observed and predicted hourly variations of O_3 concentrations at six sites in SPMA for the 3-km modelling domain.".

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

The reference was added in the text.

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

The captions were reworded to "The predicted average surface distribution of $PM_{2.5}$ concentrations for the whole study period in the 3-km modelling domain. Red dots represent the location of the measurement sites with information on $PM_{2.5}$, whereas cyan numbers represent the observed average $PM_{2.5}$ concentration in those sites: 23.4 μg m^{-3} in IPEN-USP, 21.3 μg m^{-3} in IAG-USP, and 22.2 μg m^{-3} in CON.", "The predicted average surface distribution of PM_{10} concentrations for the whole study period in the 3-km modelling domain. Red dots represent the location of the measurement sites with information on PM_{10}, whereas cyan numbers represent the observed average $PM_{2.5}$ concentration in those sites: 49.5 μg m^{-3} in IAG-USP and 38.7 μg m^{-3} in CON.", and "The predicted average surface distribution of the $PM_{2.5}/PM_{10}$ ratio for the whole study period in the 3-km modelling domain. Red dots represent the location of the measurement sites with information on both $PM_{2.5}$ and PM_{10}, whereas cyan numbers represent the observed average $PM_{2.5}$:PM_{10} ratio in those sites: 0.43 in IAG-USP and 0.57 in CON." for the figures 11, 12, and 13, respectively.

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

The caption was reworded to "The observed and predicted average aerosol mass size distribution for SO_4, NO_3, NH_4, Na, Cl, and other PM_{10} constituents at IAG-USP. The observed aerosol distributions were collected in ten size classes using a rotated impactor (MOUDI) and joined adequately according to the three modes used by the MADE aerosol module: Aitken (<0.1 μm), accumulation (0.1–1 μm) and coarse (>1 μm). The five inorganic ions carried in MADE are only calculated for the Aitken and accumulation modes. The WRF's PM_{10} aerosol variable does not include neither OC nor

EC for this comparison.”.

Technical comments

Introduction

P.14174,L.10: Corrected.

P.14174,L.11: Corrected.

P.14174,L.12: Corrected.

P.14175,L.14: Corrected.

P.14176,L.1: “...the Weather Research and Forecasting with Chemistry (WRF-Chem) model...”.

Methodology

P.14176,L.9: Corrected.

P.14176,L.13: Corrected.

P.14176,L.16: Corrected.

P.14177,L.13-14: Corrected.

P.14178,L.23: SD (Standard Deviation)

P.14179,L.11: Corrected.

P.14179,L.12: Corrected.

P.14179,L.12: “...the Model for Ozone and Related chemical Tracers, version 4 (MOZART-4; Emmons et al., 2010)”.

P.14180,L.28: Corrected.

P.14181,L.11: Corrected.

P.14181,L.26: Corrected.

P.14181,L.26: Corrected.

P.14182,L.16: Corrected.

Results

P.14182,L.25: Corrected.

P.14183,L.4: Corrected.

P.14183,L.7: Corrected.

P.14183,L.8: “comes” was replaced by “coming”

P.14183,L.9: Corrected.

P.14183,L.11: “western State” was replaced by “western Sao Paulo State”.

P.14183,L.11-12: “Precipitation areas” has been replaced by “Precipitation events”.

P.14183,L.12: Corrected.

P.14184,L.16: Corrected.

P.14184,L.25: The sentence has been removed.

P.14185,L.12: Corrected.

P.14185,L.13: Corrected.

P.14185,L.21: Corrected.

P.14187,L.6: Corrected.

P.14187-14188,L.29-1: Yes. The sentence has been reworded to "...Tuccella et al. (2012) found simulated SOA:OM ratios in the range between 5-40% against 50-80% observed...".

P.14188,L.13: Corrected.

P.14189,L.2: Corrected.

P.14189,L.16: Corrected.

P.14190,L.14: Corrected.

P.14190,L.18-19: Corrected.

P.14190,L.19-20: Corrected.

P.14191,L.1: Corrected.

P.14191,L.9-11: The sentence has been reworded to "...ground level O_3 concentrations in a few ppb.".

P.14191,L.15: Corrected.

Summary and conclusions

P.14192,L.8: Corrected.

P.14192,L.8: Corrected.

Anonymous Referee #2

Comments

1) The introduction should be extended with more background in general regarding traffic and its impacts on aerosol levels and the radiative impacts of the aerosols on photochemistry. The motivation and the aim of the study should be more clear and can be organized in a paragraph rather than scattering throughout the introduction section.

The introduction was modified including a paragraph describing the general context of emissions and their impacts on air quality in SPMA.

"The Sao Paulo Metropolitan Area, in the southeast region of Brazil, is considered a megalopolis comprised of Sao Paulo city and other 38 municipalities. One main concern is the occurrence of violations of air quality standards for ozone and fine particles at

different air quality stations from the Sao Paulo Environmental Agency (CETESB). The air pollutant emissions in the SPMA are related to the burning of the fossil fuels ethanol, gasohol (gasoline with 25% ethanol) and diesel. They constitute the main cause of impairment to air quality in SPMA, but the number of air quality standard violations has decreased for almost all pollutants with the exception of $PM_{2.5}$ and O_3, that, similar to other big cities, impacted by the vehicular emissions, have experienced an increase in the number of violations of their air quality standards as discussed in depth by Carvalho et al. (2014). Perez-Martinez et al. (2015) have analyzed the monthly mean values for the regulated pollutants from 2000 to 2013 for the air quality stations in SPMA and have found that there was a decrease in the average concentration of NOx, CO, and PM_{10} by 0.65, 0.37, and 0.71 % $month^{-1}$, respectively, although the sales of the fuels gasoline, ethanol, and diesel had increased by 0.26, 1.96, and 0.38 % $month^{-1}$.".

2) Omission of anthropogenic emissions other than the traffic sources should be mentioned clear in the Emissions section. Additionally, the anthropogenic emissions used in the coarser domains should be described clearly.

Anthropogenic emissions of trace gases and particles in both 3 and 15-km modelling domains were considered to include emissions only coming from on-road vehicles through the use of a vehicular emission model developed by the IAG-USP's Laboratory of Atmospheric Processes (LAPAt). Anthropogenic emissions were not considered in the 75 km modelling domain.

3) As the on-road vehicular emissions are emitting on the surface, a vertical distribution is not necessary. However, the authors should explain if they have used any vertical distribution for the emissions in the coarser domains. The impact of these missing sources on the model results and related discussions on the impact of traffic on air quality levels should be discussed.

There was no vertical distribution for emissions in all domains, since only on-road vehicles emissions is considered. A vertical distribution would be important for industrial sources and biomass burning. This last source is somewhat regarded due to the boundary conditions. Inside the urban area, the vehicular source is the most important one, but in rural areas upwind urban regions, these missing sources would result in lower air pollutant concentrations.

4) What are the spatial and temporal resolutions of the boundary conditions?.

The initial and boundary meteorological conditions are from the National Center for Environmental Prediction's Final Operational Global Analysis with 1^o of grid spacing,

26 vertical levels and are available every six hours: 00, 06, 12 and 18 UTC (http://rda.ucar.edu/datasets/ds083.2/). The chemical initial and boundary conditions are from MOZART at a horizontal resolution of 1.9^{o}x2.5^{o}, 56 vertical levels and are also available every six hours.

5) The HYSPLIT configuration should be described in the methodology section with a reference to the model.

The HYSPLIT configuration and reference were added in the text.

6) Section 3.1: While it is true that the behavior of the main meteorological systems should be analyzed in order to understand the spatial and temporal variability of aerosols, the authors lack to discuss how these conditions would affect the levels. As the measurements would have a temporal variability, the authors could compare the measurements with the meteorology to show how meteorology (e.g. precipitation) can influence aerosol levels.

The Figure 18 (page 10) shows comparisons between measurements and WRF outputs for temperature, relative humidity, and zonal and meridional wind components at INT and AF-IAG measurement sites. In general, both wind components were overestimated on intensity, what may have led to a dilution of aerosol particles in SPMA.

7) Section 3.2: Page 14, line 28: As the WRF model calculates wind speeds and PBL height, it would be straightforward to show if low PBL heights or wind speeds caused the high levels of PM.

The model results show that overall the predicted PBL heights have a regular diurnal variation in the downtown SPMA with averaged daily values around 500 m at both the beginning and the end of the study period, when higher concentrations of aerosols were observed, whereas values of up to 700 m in the middle of the study period, when lower concentrations of aerosols were observed. In addition, aerosol particles transported from other regions (e.g. biomass burning areas; see Fig. 4) may have led to an increase of aerosol concentrations in SPMA.

8) As the statistics are made for all the sites, can the authors also comment on how the model performance differs among individual stations? For example correlation coefficients and Normalized mean biases (NMB) can be shown for each station in Figures 7-9.

Individual statistics are shown in Fig. 10. In general, most of evaluated WRF-Chem parameters present good correlation coefficients (mainly those for PM_{10}), but with

negative biases (except for O_3 at NSO and PDP) and standard deviations lower than those for observations.

9) Section 3.3: It would be good to see more discussion on Figure 15 as I think it is an important figure showing the size distribution capability of the model for the different aerosol species. For instance, while the model underestimates SO4, it over estimates NO3 and NH4, meaning that the model simulates more NH4NO3 aerosols compared to (NH4)2SO4 aerosols. Correlation coefficients of observed and simulated NH4 and SO4 levels can give important information on why the model behaves as such (see for example Im et al., AtmEnv, 2012 and references therein).

Correlation coefficients for both chemical species show a good WRF-Chem simulation performance, especially for SO4.

Abstract

Line 9: Remove "during a month,"

The expression was removed.

Lines 21-27: I would move this part to line 9, before the sentence starting with "The study period. . ."

The sentence was adequately relocated.

Introduction

Page 5, Line 16: Add a reference for the increase in PM2.5 and O3.

The reference was added in the text.

Section 2.1: More detail (urban/traffic/background etc. . .) is needed for the characterization of the sampling site in the NUANCE_SPS project.

The following sentence "All these samplings were performed on the roof of the main building of the Institute of Astronomy, Geophysics and Atmospheric Sciences of the University of Sao Paulo (IAG-USP) (hereafter also referred as IAG-USP measurement site or simply IAG-USP), which is inside a small green-park (approximately 7.4 km^2), with local traffic during the day and surrounded by major roads with intense traffic by light and heavy-duty vehicles (Nogueira et al., 2014)." was added in the text.

Section 3.1:

Page 13, Line 4: Change "lesser" to "lower".

Corrected.

Page 13, Line 8: Change "comes" to "coming".

Corrected.

Section 3.2:

Figure 4: The figure caption of Figure 4 should be more explanatory.

The caption was reworded to "Daily (top), diurnal (bottom), and nocturnal (middle) mean concentrations for EC, OC, PM_{10}, $PM_{2.5-10}$, $PM_{2.5}$ (left panels), and Na, Fe_2SO_3, SiO_2, K_2O, and S (right panels). The $PM_{2.5-10}$ aerosol variable is defined as particulate matter with aerodynamic between 2.5 and 10 μm. The grey line indicates the WHO air quality standard for $PM_{2.5}$ (25 $\mu g/m^3$).".

Figure 10: The figure caption of Figure 10 should be more explanatory, showing that these statistics are for the individual 11 stations for example.

The caption was reworded to "Taylor diagram showing the individual correlation coefficients, bias, and normalized standard deviations for the $PM_{2.5}$, PM_{10}, and O_3 concentrations.".

Figures 11-13: The figure caption of Figure 11-13 should explain what red dots and cyan numbers are although they are described in the text. This comment goes for all relevant figures in the manuscript.

The captions were reworded to "The predicted average surface distribution of $PM_{2.5}$ concentrations for the whole study period in the 3-km modelling domain. Red dots represent the location of the measurement sites with information on $PM_{2.5}$, whereas cyan numbers represent the observed average $PM_{2.5}$ concentration in those sites: 23.4 μg m^{-3} in IPEN-USP, 21.3 μg m^{-3} in IAG-USP, and 22.2 μg m^{-3} in CON.", "The predicted average surface distribution of PM_{10} concentrations for the whole study period in the 3-km modelling domain. Red dots represent the location of the measurement sites with information on PM_{10}, whereas cyan numbers represent the observed average $PM_{2.5}$ concentration in those sites: 49.5 μg m^{-3} in IAG-USP and 38.7 μg m^{-3} in CON.", and "The predicted average surface distribution of the $PM_{2.5}/PM_{10}$ ratio for the whole study period in the 3-km modelling domain. Red dots represent the location of the measurement sites with information on both $PM_{2.5}$ and PM_{10}, whereas cyan numbers represent the observed average $PM_{2.5}$:PM_{10} ratio in those sites: 0.43 in IAG-USP and 0.57 in CON." for the figures 11, 12, and 13, respectively.

Section 3.5:

Page 20, Line 2: Change to ". . .and measurements at IAG-USP shown in Figure 14 include the Case_1 simulation.

Corrected.