

Interactive comment on “The Coupled Aerosol and Tracer Transport model to the Brazilian developments on the Regional Atmospheric Modeling System (CATT-BRAMS) – Part 2: Model sensitivity to the biomass burning inventories” by K. M. Longo et al.

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Questions and Answers to Reviewer 1:

Anonymous Referee #1

This manuscript presents an interesting and important study on the effect of using different biomass burning emission inventories in a regional transport model to improve the tropospheric simulation of carbon monoxide (CO) concentrations in South America. Comparing model simulation with in-situ and aircraft measurements, the study

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indicates the importance of accurate information within a model regarding timing and location of fires in the emissions inventories in order to simulate realistically the time variability of near-surface associated air pollution. Although the presented material is interesting and valuable, the approach adopted in the paper is not satisfactory. The reviewer recommends publishing the paper in ACP after some major corrections as suggested following.

(A) We thank the reviewer of this manuscript for his/her insightful and helpful comments. The paper is now much improved by his/her comments and corrections. His/her contribution also provides us a better understanding of limitations as well as the robustness of our modeling system.

General Remarks 1. My major concern is the methodology that the manuscript uses. The authors address CO simulations for the 2002 dry season using their real-time biomass burning inventories (3BEM) and two climatology inventories. Why do the authors only choose climatology inventories for their comparison for a specific study period? There are some referred year-specific biomass burning inventories available in our scientific community, such as Global Global Fire Emissions Database version 2 (GFED2) [van der Werf et al., 2005] and GWEM-1.3 inventory (GWEM) [Hoelzemann et al., 2004].

(A) The inventory “Global Fire Emissions Database version 2 (GFEDv2)” with 8-daily time resolution is now included in the comparison. GWEM-1.3 is not included because it already uses the 3BEM methodology for South America.

Q 2. The authors still need to clarify the CO module used in the simulation. The current study only considers two CO emissions from biomass burning and biofuel. Why does the study ignore the other emissions from fossil fuel and biogenic? How about CO produced by methane oxidation? If these processes are not important in comparison to biomass burning emission, the authors should give a general estimation of the

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simulated CO uncertainty due to missing these sources to justify the used approach.

A) In the areas where we focused our model discussion and evaluation, biomass burning is by far the most important source of both CO and PM_{2.5}. In this region, urban-industrial-vehicular activities are not well developed and biogenic processes do not compete with the biomass burning processes used for land use changes during the dry season. Figure 1 shows estimates of CO emissions from urban-industrial-vehicular activities using the REanalysis of the TROpospheric chemical composition (RETRO, <http://retro.enes.org>) inventory, from biomass burning using the Brazilian Biomass Burning Emission Model (3BEM), and biogenic sources as described by GEIA-POET inventory (<http://www.aero.jussieu.fr/projet/ACCENT/POET.php>), respectively. The CO fluxes are daily means for August and September (the months with the maximum fire counts) of 2002. In this case, CO biomass burning emissions are 10 to 100 higher than both biogenic and anthropogenic.

Figure 1. Carbon monoxide daily emission rate from anthropogenic (urban-industrial-vehicular), biomass burning and biogenic processes. Time mean for August and September, 2002.

The impact of neglecting CO emissions from urban-industrial-vehicular activities can be envisioned with Figure 2. This figure shows near-surface CO (ppb) temporal averages (Aug-Sep-Oct 2002) as simulated using only urban-industrial-vehicular and biogenic emissions (RETRO and GEIA-POET datasets, at left) and only biomass burning emissions (using 3BEM methodology, at right). Anthropogenic CO sources are mostly concentrated in the southeastern part of Brazil, Buenos Aires and Santiago areas, and in the northwestern part of the South American continent, composing the main South American megacities and economically developed areas. The impact of these emissions and biogenic sources on the central part of the continent and the border of the Amazon forest is between 10 and 30 ppb in the 3-monthly mean. On the other hand, biomass burning contributes CO that exceeds 100 ppb. Additionally, in the areas where the SMOCC-RACCI field campaign took place, this contribution is larger than 300 ppb.

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These model results corroborate a report on methane and CO airborne measurements in both fresh smoke plumes and regional haze dominated by smoke in Brazil, which did not show any statistically significant difference in the ratio of methane to CO between fresh and aged smoke (Reid et al., 1998ab). According to these authors, this is a good indicator that the regional hazes studied (which were in an area consistent with the model domain) were not significantly affected by urban anthropogenic sources.

Figure 2. At left, near surface CO (ppb) from only urban-industrial-vehicular (RETRO) and biogenic (GEIA-POET) emissions. At right, only biomass burning emissions (Aug-Sep-Oct, 2002 time mean).

To provide a robust estimation of the potential uncertainty due treating CO as a passive tracer with a lifetime of 30 days (according to Seinfeld and Pandis, 1998), we re-ran this case using the new version of the model that includes chemical reactivity. As this system is not yet published, we provide here a short description. The new CATT-BRAMS system can be configured with any chemical mechanism, using a modified version of the SPACK (Simplified Preprocessor for Atmospheric Chemical Kinetics, Djouad et al., 2002) pre-processor. The solver of the chemical mechanism is an implicit and multi-stage algorithm based on Rosenbrock's method (Hairer and Wanner, 1991). Currently are implemented ROS 2 (2nd order, 2 stages) and RODAS 3 (3rd order, 4 stages). The integration may use manual, splitting, or dynamic time-steps for the chemistry. The operator splitting used to solve the mass continuity equation may be defined as parallel, sequential, and sequential symmetric. Photolysis rates are calculated online using the FAST-TUV model. Dry deposition follows the resistance formulation, and accounts for the aerodynamic, quasi-laminar layer and canopy resistances (Wesely, 1989, Seinfeld and Pandis, 1998). Wet deposition is parameterized following Berge (1993) for PM_{2.5} and Henry's law for gaseous species, and is fully coupled with the convective scheme. In the case presented here, we used the Regional Atmospheric Chemistry Mechanism (RACM, Stockwell et al., 1997), with 70 species and 237 kinetic and photolysis reactions. Chemistry fields were initialized using horizon-

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tally homogeneous profiles associated with a background situation, and the model ran during a period of 15 days for the spin-up. Boundary condition was defined as constant inflow and variable outflow. CPTEC analysis data provided initial and boundary conditions for the meteorological integration. Emission sources are defined for anthropogenic, biogenic and biomass burning as prescribed by RETRO, GEIA-POET and 3BEM inventories, respectively. The total length of the time integration was 75 days, starting at on 15 July 2002 at 00 UTC, but only results for the last 60 days (August and September) are considered. Two simulations were performed using exactly the same dynamics, physics and sources, but including chemistry reactivity or not. In the run where chemistry was turned OFF, a lifetime of 30 days for CO was included in mass continuity equation. To quantify the error associated with considering CO as a tracer with the above lifetime, we calculate the percent difference expressed as follows (see Figure 3):

Figure 3. Difference of CO between two simulations (one includes RACM chemical mechanism, the other one treats CO as a tracer with lifetime of 30 days) in terms of monthly means for August and September 2002.

In those areas where we focused our discussion and model evaluation, the difference is typically less than 5%, being about 1-2 % in areas with intense biomass burning and/or downwind thereof. The error increases towards the domain borders, because the CO mixing ratio in these areas is too small, and the boundary conditions are not very well prescribed. Model results would be improved in these areas by using more realistic boundary conditions from data assimilation and/or global chemistry models. Therefore we can state that it is reasonable to treat CO as a tracer with a lifetime of about 1 month for limited area models with open lateral boundaries.

Specific comments

1. Page 8572 line 23: What is the fraction of biomass burning emission of aerosol particles in South America to global total? This answer tells readers how important

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South America biomass burning emission is on the perspective of the global scale.

(A) Our methodology is applied only for South America, because WF_ABBA covers only the Americas. In this way, we are not able to estimate that fraction.

2. Page 8574 line 11-16: The road map is messed up. It does not correspond to the following text.

(A) It was rewritten.

3. Page 8574 line 18-19: What are these new improvements for biomass burning emission parameterization? Why do the authors introduce these improvements? What are the improvements of simulated CO concentrations due to using the upgraded emission parameterization?

(A) The main improvement was the combined use of GOES WF_ABBA with MODIS and AVHRR fire products, which improved the model performance over savanna and pasture areas; however we did not show it in this paper.

4. Page 8576 line 11-13: Please give a rough estimation of the potential influence of ignoring emissions other than biomass burning and biofuel.

(A) Please refer to answer to the General Remark 2

5. Page 8576 line 14: Add climatology; before biomass burning emission.

(A) Done.

6. Page 8577 line 21-23: The simulation started on July 15, 2002 and the results used for analysis started on August 1, 2002. The fifteen days spin-up time is too short for CO simulation since CO lifetime in South America could be a couple of months.

(A) During the Austral winter in South America, mid-latitude cold fronts approaching the central part of Brazil happen every week, typically. These systems are very efficient for the dispersion of biomass burning pollutants (Freitas et al., 2005). Also Freitas

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et al. (2000) and Longo et al. (1999) have shown using trajectory calculations that most air parcels departing from typical fire locations, take 5–8 days to fly out of South America. On the other hand, July 15 is a too early a stage of the burning season and the atmosphere still has pristine conditions over most areas of Amazon basin. So, we feel that 15 days spin-up time, starting on July 15, is enough to set a realistic air pollution condition. Especially, because our comparisons with near surface and aircrafts observations are for late September and October.

7. Page 8578 line 5-6: How do the authors treat CO dry deposition? CO dry deposition is small and was ignored by many previous studies.

(A) Dry deposition processes are simulated using the resistance approach following Wesely (1989) and Seinfeld and Pandis (1998). We refer to Freitas et al. (2005, 2009) for the model description.

8. Section 3: I suggest splitting subsection 3.1 into two subsections; one for model comparison with in-site surface measurement and one for comparison with aircraft measurement. This way, the manuscript has a structure for section 3 which is consistent with the task summary outlined in the prelude of section 3.

(A) Done

9. Page 8580 line 18-19: Figure 5 shows that model CO using biomass burning emissions of EDGAR and D2003 is much lower than the model CO simulated with 3BEM emission; however, this is contrary to what I observe from Figure 3. Figure 3 indicates that EDGAR emission is consistently higher than that of 3BEM. Adding the exact location and time of each flight presented in the Figure 5 derived from the experiment SMOCC/RaCCI would help readers understand the results better.

(A) Figure 3 indicates the total amount of CO mass emitted over the entire South America for each inventory. A description of the location of SMOCC/RaCCI flights is now in the text. Thanks for the suggestion.

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10. Page 8581 line 11-12: No. MOPITT retrievals are reported at 7 pressure levels, not 3. The authors only use 3 layers of MOPITT data.

(A) This information is now included.

11. Page 8581 line 19: Add a space between D2003emissions;.

(A) Done.

12. Page 8581-8582: Two sentences are repeated; one is on line 17-19, page 8581 and the other is on line 25, page 8581 to line 2, page 8582.

(A) Removed one sentence.

13. Page 8582 line 3-4: Figure 7 and 8 also indicate that CO simulated by burning emissions from EDGAR and D2003 is lower than that of 3BEM. This is contradictory with the emission information revealed in Figure 3.

(A) New discussion is included.

14. Page 8583 line 6-9: Rephrase the sentence.

(A) Done.

15. Figure 4: For the scatter plot, where is the CODPSH data? Should the black dots be the red dots? Also the black number of $R^2=0.66$; may be $R^2=0.66$; in red.

(A) Corrections done.

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