Atmos. Chem. Phys. Discuss., 7, S10009–S10017, 2009
www.atmos-chem-phys-discuss.net/7/S10009/2009/

Atmospheric Chemistry and Physics Discussions

Interactive comment on "The Coupled Aerosol and Tracer Transport model to the Brazilian developments on the Regional Atmospheric Modeling System (CATT-BRAMS) – Part1: Model description and evaluation" *by* S. R. Freitas et al.

S. Freitas

saulo.freitas@cptec.inpe.br

Received and published: 27 February 2009

Q) The authors implement an aerosol and tracer transport model in a regional atmospheric modeling system. The authors then apply this online tracer transport model to examine aerosol (PM2.5) and CO distributions over South America and compare the simulated results to corresponding measurements from satellite, aircraft, and in-situ for evaluation. Overall, the paper shows fairly detailed evaluations with the measurements and some interesting results were found. However, I think there are many areas that need clarifications or further analyses. I suggest the authors address the following

major comments before the paper is considered for publication in ACP.

A) We thank to the reviewer of this manuscript for his/her insightful and helpful comments. The paper was indeed improved by his/her comments and corrections. Q)General Remarks: 1. It is difficult to extract the primary objective of the manuscript. The authors’ ideas are interspersed into two topics; the regional Atmospheric Modeling System (BRAMS) and the tracer transport model (CATT). However the feedbacks which link the two parts (i.e. the “coupled” effects) are missing. For example, the authors devote a lot of effort in discussing the processes of BRAMS and evaluating the meteorological fields it simulates. However, there is no in depth discussion of how these processes and fields impact tracer simulations. On the other hand, the feedback of aerosol simulation on meteorological fields through radiation parameterization used in BRAMS is also not explored.

A) We changed the text in order to better describe our objective. In this paper we focus on the CATT-BRAMS system description and show its evaluation against near surface and airborne measurements and remote sensing derived data. Additional discussion was included in Section 2 addressing the improvements in BRAMS, its impacts on atmospheric modeling and the feedback between the atmospheric and the tracer transport models. As recommended by an anonymous reviewer of the companion paper (Longo et al., 2007), the feedback of aerosol simulation on meteorological fields and tracers through interaction with radiation is now included in that paper.

Q)2. It is still unclear which improvements (e.g. new convection parameterization, land use information, and soil type) in BRAMS are new features developed by this work and which were presented in previous studies but benefit the current study.

A) We now clearly stated that the described improvements were developed by the authors. In addition, mostly of the new features were motivated to improve the tracer’s simulations. Q)3. The authors indicated in the abstract that sources of tracer gases and aerosol particles contain the emissions from biomass burning and

ACPD

7, S10009–S10017, 2009

Interactive Comment

urban-industrial-vehicular activities. However, I observed that only biomass burning emission is considered in the study for both CO and PM2.5. How about other emissions, such as emissions from fossil fuel, biofuel, and biogenic for CO? How about CO chemistry production from CH4 oxidation? Are these other sources neglected because they are unimportant in comparison to biomass burning emission? If only biomass burning emission is considered in the study, the authors should give at least a rough estimation of the potential uncertainty due to neglecting the other sources.

A) In the areas were we focused model discussion and evaluation biomass burning is by far the most important source of both CO and PM2.5. In that region, urban-industrial-vehicular activities are not well developed and biogenic process do not compete with the biomass burning processes used for land use changes during the dry season. The Figure 1 (see pdf file uploaded at COSIS web site) shows CO emissions estimate from urban-industrial-vehicular activities using “REanalysis of the TROpospheric chemical composition” (RETRO, http://retro.enes.org) inventory, from biomass burning using Brazilian Biomass Burning Emission Model (3BEM, Freitas et al., 2005, Longo et al., 2007), and biogenic as described by GEIA-POET inventory (http://www.aero.jussieu.fr/projet/ACCENT/POET.php), respectively. The CO fluxes are daily mean of August and September (the months with the maximum fires counts) of 2002. In this case, CO biomass burning emissions are 10 to 100 higher than both biogenic and anthropogenic.

The impact on neglecting CO emissions from urban-industrial-vehicular activities can be envisioned with Figure 2(see pdf file uploaded at COSIS web site). This figure shows near surface CO (ppbv) timely mean (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 is 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 economical

ACPD

7, S10009–S10017, 2009

Interactive Comment

developed areas. The impact of these emissions and biogenic on central part of the continent and border of Amazon forest is between 10 and 30 ppbv at 3-monthy mean. On the other hand, biomass burning contributes with CO that exceeds 100 ppbv. Additionally, in the areas where SMOCC-RaCCI field campaign took place, this contribution is larger than 300 ppbv. These model results corroborate a report of methane and CO airborne measured in both fresh smoke plumes and regional haze dominated by smoke in Brazil, which did not show any statistically significant difference in the ration 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.

To provide a robust estimation of the potential uncertainty due treating CO as a passive tracer with lifetime of 30 days (according to Seinfeld and Pandis, 1998), we re-run this case using the newly version of the model that includes chemistry reactivity. As this system is not yet published, we provide here a short description. The new CATT-BRAMS system might be virtually configured with any chemical mechanism using a modified version of 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 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-step 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 on-line using 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 PM2.5, Henry’s law for gaseous 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

ACPD

7, S10009–S10017, 2009

and photolysis reactions. Chemistry fields were initialized using horizontally 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, 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. The one where chemistry was turned OFF, a lifetime of 30 days for CO was included in mass continuity equation. To quantity the error associated to considering CO as a tracer with the lifetime above, we calculate the percent difference expressed as follows (see Figure 3, pdf file uploaded at COSIS web site),

diff=100*abs(CO_racm - CO_lifetime30days)/CO_racm

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

Q)Specific comments: 1. Page 8527 line 10-12 (abstract): Which aerosol data was used in radiation parameterization? Is it the one produced by the simulation of this study?

A) The sentence was reformulated and now appears as “The radiation param-

ACPD

7, S10009–S10017, 2009

eterization takes into account the interaction between the simulated biomass burning aerosol particles and short and long wave radiation.”

Q)2. The introduction is somewhat weak. I suggest augmenting it by highlighting the new features of the model and the objectives of this study. A) The introduction was substantially augmented and now resembles better the objectives of the paper. Thanks. Q)3. Page 8530 line 16-17 and page 8531 line 22-26 (model description): How about other types of emissions? How to treat PM2.5 in the model? Please clarify whether the study differentiates aerosol composition for PM2.5 in simulation. How does the model treat dry and wet depositions for PM2.5?

A) For biomass burning this modeling system is able to estimate emission of 110 species using 3BEM (Freitas et al, 2005 and companion paper Longo et al., 2007) or GFED inventories and emission factors compiled by Andreae and Merlet (2001). In this work, we use only PM2.5 from biomass burning since the background is very clean (˜10 μg m-3) according to Artaxo et al., 2002). A generic smoke particle (without elemental composition specification) was assumed, with an average mass density of 1.35 g cm-3 (according Reid et al. [1998]) and spectral optical properties following the AERONET Amazonian climatology from Procopio et al. (2003). Dry deposition uses the resistance formulation following Wesely (1989) and Seinfeld and Pandis (1998) and is coupled to the surface parameterization. Wet deposition is parameterized following Berge (1993) and is described in Freitas et al. (2005) and is coupled with the deep convective parameterization. This information is now in the paper.

Q)4. Page 8530 last line: Fine mode aerosol usually refers to particles with a radius of less than 0.5μm.

A) The qualification ‘fine’ was removed.

Q)5. Page 8533 line 9: What is CPTEC? Does CPTEC T126 provide initial field for CO and aerosols?

ACPD

7, S10009–S10017, 2009

Interactive Comment

A) CPTEC stands for Brazilian Center for Weather Forecasts and Climate Studies. CPTEC analysis data contains horizontal wind, geopotential height, air temperature and water vapor mixing ratio. CO and PM2.5 fields were initialized using horizontally homogeneous profiles associated with a background situation and the model ran during a period of 15 days with sources for the spin-up. Lateral boundary condition was defined as has constant inflow and radiative variable outflow. This information is now in the paper.

Q)6. Page 8533: Very clear and detailed descriptions for land surface data over South America. However, the link of this data to CO and aerosol surface fluxes in this study is missing. How does land surface situation influence CO and aerosol emissions studied in the paper?

A) We better describe this link now in the paper. The main point we raise up is that during the dry season, the two main biomes of SA (cerrado and amazon forest) presents markedly differences. For forest, the Bowen ratio during the dry season is similar of the wet season (˜ 1/3) and mixing boundary layer (Zi) peaks ˜1200 m, however for cerrado/pasture areas this number is about 4 with Zi ˜ 2500 m. So, to properly simulate aerosols and CO distribution is essential that the model is able to simulate realistically the contrasts presented by these biomes. Inappropriate Bowen ratio simulation for forest, e. g., might cause too deep, warm and dry mixing layer with strong dilution of the simulated tracers and inhibit cumulus convection, affecting the transport to the free troposphere.

Q)7. Page 8534 line 1-9: The authors indicate here again that only biomass burning emission is considered in the study.

A) Please, see our answer for the question “General Remarks # 3”.

Q)8. Page 8539 line 22-23: Please elaborate on the sentence “The diurnal evolution of the boundary layer contributes to this high variability.”

ACPD

7, S10009–S10017, 2009

A) We want to point out that is not only the fire occurrence/intensity that is responsible for the observed time variability of near surface CO concentration observations, but the planetary boundary layer time evolution itself also plays a role. Q)9. Page 8541 line 21-22: How to perform the condition of ‘using retrievals with $<$ 50% a priori contribution’ . A) As recommend by MOPITT data provider: filtering out MOPITT CO data set for CO Mixing Ratio Percent Apriori variable higher than 50%. Q)10. Page 8555 figure 4: Change AUG/SEP/OCT to percent persistence. A) Done. References:

STOCKWELL W. R.; KIRCHNER F.; KUHN M.; SEEFELD S. A: new mechanism for regional atmospheric chemistry modeling. J. Geophys. Res., vol. 102, noD22, pp. 25847-25879, 1997.

SAATCHI, S. S.; HOUGHTON, R. A.; DOS SANTOS ALVAL, R. C.; SOARES, J. V.; YU, Y. Distribution of aboveground live biomass in the Amazon basin. Global Change Biology, 13, 816-837, 2007.

HAIRER, E. AND WANNER, G. Solving Ordinary Differential Equations II. Stiff and Differential-Algebraic Problems, Springer-Verlag, Berlin, 1991.

DJOUAD, R., SPORTISSE, B., AUDIFFREN, N. Numerical simulation of aqueous-phase atmospheric models: use of a non-autonomous Rosenbrock method. Atmospheric Environment, vol. 36- 5, 873-879(7), 2002.

Wesely, M. L.: Parameterizations of surface resistance to gaseous dry deposition in regional scale numerical models, Atmos. Environ., 23, 1293-1304, 1989.

Seinfeld, J. H., and Pandis, S. N.: Atmospheric Chemistry and Physics, from Air Pollution to Climate Change, John Wiley, New York, 136 pp., 1998.

Berge, E.: Coupling of wet scavenging of sulphur to clouds in a numerical weather prediction model, Tellus, 45B, 1-22, 1993.

Poet: Granier, C., J.F. Lamarque, A. Mieville, J.F. Muller, J. Olivier, J. Orlando, J. Peters,

ACPD

7, S10009–S10017, 2009

Interactive Comment

G. Petron, G. Tyndall, S. Wallens, POET, a database of surface emissions of ozone precursors, available on internet at http://www.aero.jussieu.fr/projet/ACCENT/POET.php , 200

Procópio, A. S.;, Remer L. A., Artaxo P., Kaufman Y. J., Holben B. N.: Modeled spectral optical properties for smoke aerosols in Amazonia. Geophys. Res. Let., 30, 24, 2265 - 2270. doi: 10.1029/2003GL018063, 2003.

Reid, J. S., Hobbs, P.: Physical and optical properties of young smoke from individual bio-mass fires in Brazil, J. Geophys. Res., 103, D24, 32,013-32,030, 1998a.

Reid, J. S., Hobbs, P.: Physical and optical properties of regional hazes dominated by smoke in Brazil, J. Geophys. Res., 103, D24, 32,059-32,080, 1998b.

Interactive comment on Atmos. Chem. Phys. Discuss., 7, 8525, 2007.