



The chemistry CATT–BRAMS model (CCATT–BRAMS 4.5): a regional atmospheric model system for integrated air quality and weather forecasting and research

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Abstract

The Coupled Chemistry Aerosol-Tracer Transport model to the Brazilian developments on the Regional Atmospheric Modeling System (CCATT-BRAMS, version 4.5) is an online regional chemical transport model designed for local and regional studies of atmospheric chemistry from surface to the lower stratosphere suitable both for operational and research purposes. It includes gaseous/aqueous chemistry, photochemistry, scavenging and dry deposition. The CCATT-BRAMS model takes advantages of the BRAMS specific development for the tropics/subtropics and of the recent availability of preprocessing tools for chemical mechanisms and of fast codes for photolysis rates. BRAMS includes state-of-the-art physical parameterizations and dynamic formulations to simulate atmospheric circulations of scales down to meters. The online coupling between meteorology and chemistry allows the system to be used for simultaneous atmospheric weather and chemical composition forecasts as well as potential feedbacks between them. The entire system comprises three preprocessing software tools for chemical mechanism (which are user defined), aerosol and trace gases emission fields and atmospheric and chemistry fields for initial and boundary conditions. In this paper, the model description is provided along evaluations performed using observational data obtained from ground-based stations, instruments aboard of aircrafts and retrieval from space remote sensing. The evaluation takes into account model application on different scales from megacities and Amazon Basin up to intercontinental region of the Southern Hemisphere.

1 Introduction

To study and to forecast atmospheric chemistry, the type of models that is most commonly used is the Eulerian three-dimensional Chemistry Transport Model (CTM). Global CTMs are used for tropospheric (e.g. Wild et al., 2004; Bousserez et al., 2007) as well as for stratospheric researches (e.g. Lefèvre et al., 1994; Chipperfield, 2006)

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in particular on stratospheric ozone depletion. Regional CTMs are generally designed to study air pollution (e.g. Zhang et al., 2006; Honoré et al., 2007). Some of these models are used for operational purposes. In CTMs the dynamics is forced by off-line wind fields provided by analyses or forecasts computed by a meteorological General Circulation Model (GCM). Fairly detailed chemical mechanisms can be used in CTMs because of the low computing cost of the transport component. But in complex dynamic situations such as convective environments the use of off-line dynamics can be a major weakness (Grell and Baklanov, 2011).

Thanks to rapid advances in computing resources in the ten past years, models with fully coupled dynamics and chemistry have been developed: Climate Chemistry Models (CCMs) for climate studies at the global scale (e.g. Zeng and Pyle, 2003; Eyring et al., 2006) and mesoscale (resp. cloud resolving) meteorology-chemistry models for regional (resp. for local studies) (Grell et al., 2000; Wang and Prinn, 2000; Mari et al., 2000; Zhang et al., 2003; Grell et al., 2005; Fast et al., 2006; Arteta et al., 2006; Marécal et al., 2006; Barth et al., 2007a). Both types of coupled models need to compromise between the spatial resolution (and domain size for limited area models), the simulation length and the degree of complexity of the chemical mechanism. Simulation using fine resolutions, large domains and detailed chemistry over long duration is still too computing time consuming. CCMs usually use coarse horizontal and vertical resolutions with reasonably detailed chemistry mechanisms in order to run long periods of time. Limited-area model simulations at the regional scale or at the cloud scale generally use a reduced number of chemical species and reactions because of their fine horizontal and vertical resolutions (see examples of models used in the intercomparison exercise published by Barth et al., 2007b).

Here we present a new modeling tool devoted to local and regional studies of atmospheric chemistry from surface to the lower stratosphere that is designed for both operational and research purposes. This new model is the advancement of the limited-area model CATT-BRAMS (Coupled Aerosol-Tracer Transport model to the Brazilian developments on the Regional Atmospheric Modeling System, Freitas et al., 2009;

Longo et al., 2010), which includes a chemical module for both gaseous and aqueous phases. It also takes advantage of the BRAMS specific development for the tropics/subtropics (Freitas et al., 2009), of the recent availability of pre-processing tools for chemical mechanisms and of fast codes for photolysis rates. Similarly to BRAMS, this model is conceived to run for horizontal resolutions ranging from a few meters to more than a hundred kilometers depending on the scientific objective. The model computation is fully optimized in order to be used operationally. This specificity allows the use of chemical mechanisms with a large number of chemical compounds for research studies.

The full model description including the code optimization specificities is given in Sect. 2. Two examples of model applications for both regional and urban scales conditions, including the description of the simulation setups and model results are shown and discussed in Sect. 3. These two case studies illustrate the ability of the model to simulate the tropospheric chemistry in the tropical region. Section 4 is devoted to final remarks.

2 Model system description

The Chemistry-CATT-BRAMS (hereafter, CCATT-BRAMS) is an Eulerian atmospheric chemistry transport model coupled on-line with a limited-area atmospheric model. The atmospheric model is the BRAMS (Brazilian developments on the Regional Atmospheric Modeling System), which is based on the widely used RAMS model (Walko et al., 2000) with additional specific developments for the tropics and subtropics regions (Freitas et al., 2009). The RAMS model is a multipurpose, numerical prediction model designed to simulate atmospheric circulations spanning from hemispheric scales down to large eddy simulations of the planetary boundary layer. RAMS is a non-hydrostatic time-split compressible model (Tripoli and Cotton, 1982). It has a set of state-of-the-art physical parameterizations: surface-air exchanges, turbulence, convection, radiation and cloud microphysics. It has a multiple grid-nesting scheme solving

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the model equations simultaneously on two-way interacting computational meshes of differing spatial resolutions. For real case studies global meteorological analyses or forecasts are used to define the initial model state and the forcing at the boundaries that is applied during the simulation.

The radiation is based on the scheme of a modified version of the Community Aerosol and Radiation Model for Atmosphere (CARMA, Toon et al., 1988). The original CARMA version considered simultaneously an aerosol microphysics scheme and two-stream radiative transfer module for both solar and terrestrial spectral regions (Toon et al., 1989). The major modification from the original version refers to the prescription of aerosol intensive optical properties, specifically extinction efficiency, single scattering albedo and asymmetry parameter. These parameters are obtained from previous off-line Mie calculations. Specially for the South American continent, the prescription of smoke optical properties derives from the use of climatological size distribution and complex refractive index from several AERONET sites measurements in the southern of the Amazon Basin. These properties are used as input to an off-line Mie code to calculate the spectral optical properties required by CARMA (Procópio et al., 2003; Rosario et al., 2011). The spatial distribution of the optical models in the model is based on the concept of anisotropic areas of influence described by Hoelzemann et al. (2009). Elsewhere in the model domain not covered by the areas of influence, a reference optical model was adopted.

CATT is an Eulerian transport model coupled to BRAMS designed to study the transport processes associated with the emission of tracers and aerosols (Freitas et al., 2009; Longo et al., 2010). The tracer transport is run on-line with the atmospheric state evolution consistently with the BRAMS dynamical and physical parameterizations. The tracer mass-mixing ratio, which is a prognostic variable, includes the effects of sub-grid scale turbulence in the planetary boundary layer, convective transport by shallow and deep moist convection in addition to the grid scale advection transport. The advective transport of scalars can use the forward upstream of second order formulation (Tremback et al., 1987) or the monotonic formulation developed by Walcek (2000) and

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implemented by Freitas et al. (2012). In a form of tendency, the general mass continuity equation for tracers solved in the CATT-BRAMS model is:

$$\frac{\partial \bar{s}}{\partial t} = \left(\frac{\partial \bar{s}}{\partial t} \right)_{\text{adv}} + \left(\frac{\partial \bar{s}}{\partial t} \right)_{\text{PBL diff}} + \left(\frac{\partial \bar{s}}{\partial t} \right)_{\text{deep conv}} + \left(\frac{\partial \bar{s}}{\partial t} \right)_{\text{shallow conv}} + \left(\frac{\partial \bar{s}}{\partial t} \right)_{\text{chem}} + W + R + Q \quad (1)$$

where \bar{s} is the grid box mean tracer mixing ratio, the term adv represents the 3-d resolved transport (advection by the mean wind) and the terms PBL diff, deep conv and shallow conv are for the sub-grid diffusion in the planetary boundary layer (PBL), deep and shallow convection, respectively. In the original version of CATT-BRAMS, as described in Freitas et al. (2009), the chem term refers simply to the passive tracers' lifetime, the W was the term for wet removal applied only to aerosols, and R the term for the dry deposition applied to both gases and aerosol particles. Finally, Q was the source term that already included the plume rise mechanism associated with vegetation fires (Freitas et al., 2006, 2007, 2010). In addition, this model version already takes into account the interaction of aerosols with solar and long wave radiations and feedback to the atmospheric model in terms of heating rates (Longo et al., 2006). A detailed description and evaluation of the CATT-BRAMS can be found in Freitas et al. (2009), Longo et al. (2010) and Rosário et al. (2012). CATT-BRAMS has been used operationally by the Brazilian Center for Weather Prediction and Climate Studies (CPTEC, <http://meioambiente.cptec.inpe.br>) since 2003 and as a research tool by several groups around the world (e. g.: Marécal et al., 2007, 2010; Landulfo et al., 2009; Arteta et al., 2009a,b; Liu et al., 2010; Ramos, 2006; Gevaerd, 2005; Herrmann, 2004; Gácita, 2011; Munchow, 2008; Arroyo, 2007; Alonso, 2011), proving its robustness.

In the new CCATT-BRAMS a chemical module is fully coupled to the meteorological/tracer transport model CATT-BRAMS to solve the term chem in Eq. (1), explicitly

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following:

$$\left(\frac{\partial \rho_k}{\partial t}\right)_{\text{chem}} = \left(\frac{d\rho_k}{dt}\right) = P_k(\boldsymbol{\rho}) - L_k(\boldsymbol{\rho}), \quad (2)$$

where $\boldsymbol{\rho} = \{\rho_1, \rho_2, \dots, \rho_N\}$ are the number density of the N reagent species considered, P_k and L_k correspond to the net production and lost of the species k . The equation uses in its solution the initial condition $\boldsymbol{\rho}(t_0) = \{\rho_1(t_0), \rho_2(t_0), \dots, \rho_N(t_0)\}$. The lost and production terms include gaseous photochemistry, kinetic and aqueous chemistry. Figure 1 illustrates the main sub-grid scale processes involved in the trace gas/aerosol distribution in the CCATT-BRAMS system, which will be described below.

The CCATT-BRAMS system was developed using advanced numerical tools in order to get a flexible multipurpose model that can be run for both operational forecasts and research simulations. Moreover, the model system is a fully comprehensive code package designed to allow the user for flexibility in the choice of the chemical species, the chemical mechanism, the emission types and databases. A schematic view of the model system, including its pre-processing tools is given in Fig. 2. The packed system comprises three preprocessors tools for emissions databases (PREP_CHEM_SRC, Freitas et al., 2011), chemical mechanisms, and boundary and initial conditions and the CCATT-BRAMS code itself.

The first step for any simulation is the choice of the gaseous species and of the gaseous kinetic and photochemistry reactions to account for. To make straightforward the use of different chemistry mechanisms in CCATT-BRAMS, it was chosen to use a modified version of the pre-processing tool SPACK (Simplified Pre-processor for Atmospheric Chemical Kinetics) developed by Djouad et al. (2002) following Damianlordache and Sandu (1995). The modified- SPACK (called hereafter M-SPACK) basically allow the passage of a list of species and chemical reactions from symbolic notation (text file) to a mathematical one (ODEs), automatically preprocesses chemical species aggregation and creates Fortran 90 routines files directly compatible to be compiled within the main CCATT-BRAMS code.

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The M-SPACK output code also feeds the emissions preprocessor tool (PREP-CHEM-SRC) in order to ensure consistency between the emissions database to be used in CCATT-BRAMS and the list of species treated in chemical mechanism. The PREP-CHEM-SRC provides the emissions from different sources (see Sect. 2.2 for details) interpolated onto the simulation grid chosen, which are then used as input for CCATT-BRAMS. It basically links the species that are in the emission databases to those in the chemical mechanism. When the species are the same, this is straightforward. But commonly in the mechanisms, there are species names representing a group of species. In this case, the PREP-CHEM-SRC converts the species from the database into the model species.

The M-SPACK output code also feeds the BC-PREP code that generates the initial and boundary fields for the chemical species to be used in the CCATT-BRAMS simulation.

2.1 Gaseous kinetic and photochemistry

In principle, CCATT-BRAMS can use any chemical mechanism provided by the user. In practice, the available computing resources limit the number of species and reactions. So far three widely used tropospheric chemistry mechanisms have been tested: RACM – Regional Atmospheric Chemistry Mechanism (Stockwell et al., 1997) with 77 species, Carbon Bond with 36 species (Yarwood et al., 2005), and RELACS – Regional Lumped Atmospheric Chemical Scheme, with 37 species (Crassier et al., 2000). The RACM mechanism is preferentially used for research purposes because it is more complete than Carbon Bond and RELACS. Since RACM and RELACS were used in the simulations presented in this paper, more details will be given on these mechanisms. RACM is based on the lumped molecule approach, in which the species are grouped according to their chemical nature, also using the magnitude of the emission rates and reactivity with respect to OH in the creation of the lumping from the species included. Originally designed to simulate the chemistry of the troposphere from urban to remote conditions, it includes 17 stable inorganic species, 4 inorganic intermediates, 32 stable

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organic species and 24 organic intermediates for a total of 237 reactions including 23 photolytic reactions. RELACS is a reduction of RACM, following a weighting by condensation reactivity, but modified so that the RACM organic species are grouped into new species according to its chemical nature and reactivity relative not only to OH but also to other oxidants. The result is a reduction to 37 species and 128 reactions. RELACS is currently used for operational air quality prediction at CPTEC because of its reduced mechanism leading to affordable computing times. For real-time forecasting RELACS fairly reproduces RACM response (Gácita, 2011).

Most of atmospheric chemistry models have used so far look-up tables of pre-calculated photolysis rates since their on-line determination by a radiative code is time consuming. Nevertheless, this approach does not account for the presence of aerosols and clouds, which is a frequent feature in meteorological simulations, in particular in the tropics. The development of fast radiative codes such as Fast-J (Wild et al., 2000; Brian and Prather, 2002) and Fast-TUV (Tie et al., 2003) provides the opportunity to couple on-line the photolysis processes calculations with a meteorological model in order to improve the estimation of the photolysis rates in aerosol polluted and cloudy environments. The Fast-TUV code chosen and introduced in the CCATT-BRAMS is based on the well-known TUV model (Madronich, 1989). It uses a reduced number of spectral bands (17) leading to an 8 times more rapid code than the original TUV with differences generally lower than 5 %.

2.2 Emissions

The PREP-CHEM-SRC tool provides emission fields interpolated onto the transport model grid. The preprocessor is coded using Fortran90 and C and is driven by a namelist allowing the user to choose the type of emissions, the databases and a chemical species specification file from the M-SPACK. The considered emissions are from the most recent databases of urban/industrial, biogenic, biomass burning, volcanic, biofuel use and burning from agricultural waste sources. The current version includes anthropogenic emission inventories provided by “REanalysis of the

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TROpospheric chemical composition over the past 40yr” (RETRO, <http://retro.enes.org>) for 26 species complemented by other species by the “Emission Database for Global Atmospheric Research – version 4.2” (EDGAR-4.2, <http://edgar.jrc.ec.europa.eu>, Olivier et al., 1996, 1999). This very recent version includes most of the direct greenhouse gases, 4 ozone precursor species and 3 acidifying gases, primary aerosol particles and stratospheric ozone depleting species. Specifically for the South American continent, it also includes urban/industrial based on local inventories (Alonso et al., 2010). For biomass burning, it includes emissions provided by the Global Fire Emissions Database (GFEDv2) based on Giglio et al. (2006) and van der Werf et al. (2006) or emissions can be also estimated directly from satellite remote sensing fire detections using the Brazilian Biomass Burning Emission Model (3BEM, Longo et al., 2010) included in the tool. In both cases, fire emissions are available for 107 different species. The biomass burning emission estimate is split into two contributions: smoldering, which releases material in the first model layer above the surface, and flaming, which makes use of an in-line 1-D plume rise model to determine the injection vertical layer (Freitas et al., 2007, 2010). Biogenic emissions are also considered via GEIA/ACCENT Activity on Emission Databases, (<http://www.aero.jussieu.fr/projet/ACCENT/description.php>) for 12 species and derived by the Model of Emissions of Gases and Aerosols from Nature (MEGAN, Guenther et al., 2006) for 15 species. Other emissions include: volcanic ashes (Mastin et al., 2009), volcanic degassing (Diehl, 2009; Diehl et al., 2011), biofuel use and agricultural waste burning inventories developed by Yevich and Logan (2003). A detailed description of the PREP_CHEM_SRC code and its functionalities can be found in Freitas et al. (2011).

2.3 Transfer to the aqueous phases and aqueous chemistry

In the presence of liquid clouds or rain, soluble species are partly transferred from the gas phase to the liquid phase. In the microphysical scheme, the mass transfer calculations between the phases follow Barth et al. (2001). At each time step, it is determined either by Henry’s law equilibrium or by mass transfers limitation calculations

if equilibrium is not attained in the time step. The cloud and the rain droplets transport the species in the liquid phase. They can be released to the gas phase if droplets evaporate or washout by rain sedimentation.

In the convection scheme, the species can be washout by rain sedimentation. The loss of gas species is proportional to the gas species concentration, to the effective Henry's constant of the species, to the mixing ratio of liquid cloud and rain and to the rain rates, following the work of Berge (1993).

The chemical transformations in the aqueous phase are resolved following the scheme proposed by Strader et al. (1999), which includes 28 species in the aqueous phase, related with 18 species in the gaseous phase, as described in Sartelet et al. (2007) and Mallet et al. (2007).

2.4 Dry deposition

Dry deposition of the gas species at the surface is taken into account in the CCATT-BRAMS. The deposition flux follows the resistance formulation and accounts for the aerodynamic, quasi-laminar layer and canopy resistances (Wesely, 1989; Seinfeld and Pandis, 1998) with updates from Weseley and Hicks (2000) and Zhang et al. (2003). Also, it is fully coupled to the RAMS surface parameterization, including the sub-grid land type patches within the parameterization.

2.5 Carbon cycle

The soil/vegetation model "Joint UK Land Environment Simulator" (JULES, Best et al., 2011; Clark et al., 2011) was also fully coupled to the CCATT-BRAMS model providing surface fluxes of momentum, latent and sensible heat, radiative as well as CO₂ and other trace gases. A detailed description of the coupling and applications to numerical weather forecasting and the CO₂ budget in South America is given in Moreira et al. (2012).

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2.6 Time integration of the chemical mechanism

The system of Eq. (2) is a stiff one because the chemical reactions rates occurring in the atmosphere vary by several orders of magnitudes, requiring implicit methods for non-prohibitive numerical solution. Furthermore, the typical chemical mechanisms suitable for the troposphere on a regional scale include a large number of species, for example, the Regional Atmospheric Chemistry Mechanism (RACM, Stockwell et al., 1997), has 77 species reacting via 230 kinetic and photolysis reactions. This implies a high computational cost involved in the solution of Eq. (2), which is by far the most expensive term in the solution of the continuity Eq. (1).

The numerical integrator of the chemical mechanism in this numerical modeling system is an efficient implicit and multi-stage solver based on Rosenbrock's method (Hairer and Wanner, 1991; Verwer, 1999). This method allows the exchange of the solution of a nonlinear differential equations system (Eq. 2) by a linear algebraic increment in terms of K_j , the solution being given by:

$$\rho(t_0 + \tau) = \rho(t_0) + \sum_{j=1}^s b_j K_j, \quad (3)$$

where s is the total number of stages, b_j are numerical constants depending on s and τ is the time step. The increments K_j are obtained sequentially throughout the solution of the linear algebraic system given by:

$$K_i = \tau F(\rho_i) + \tau \mathbf{J}(\rho_i(t_0)) \cdot \sum_{j=1}^i \gamma_{ij} K_j$$
$$\rho_i = \rho(t_0) + \sum_{j=1}^{i-1} \alpha_{ij} K_j \quad (4)$$

$$F(\rho_i) = P(\rho_i) - L(\rho_i)$$

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where $i = 1, s$, α_{ij} and γ_{ij} are constants that depend on s , ρ_i is an intermediate solution used to recalculate the net production on stage i given by the term $F(\rho_i)$ and \mathbf{J} is the Jacobian matrix of the net production at time t_0 . The first one is the fact that the Jacobian is extremely sparse, typically only about 10 % of the elements are non-zeros.

The second relevant aspect is that the matrix structure is invariant in time and space, which allows the prior recognition and mapping of non-zero elements on which the operations should be performed. The solution of the system of equations (4) follows the method proposed by Kundert (1986) and Kundert and Sangiovanni-Vincentelli (1988). In this method, with reference to fixed positions where the chemical reactions occur, one performs the LU factorization and solves the matrix system working with memory pointers. The method works on three matrices originated from the matrix to be solved: a diagonal matrix (\mathbf{D}), a lower triangular (\mathbf{L}) and another upper triangular (\mathbf{U}):

$$\mathbf{A} = \mathbf{D} + \mathbf{L} + \mathbf{U} \quad (5)$$

With the solution given by:

$$\mathbf{x}^{(k+1)} = \mathbf{D}^{-1} \left[-(\mathbf{L} + \mathbf{U})\mathbf{x}^{(k)} + \mathbf{b} \right], \quad (6)$$

where ($k = 1, 2, \dots$). The elements of the array are allocated dynamically only once and unnecessary iterations are eliminated. An index of pointers is previously created in order to relate the sequence of positions allocated in memory with its original position in the array. The factorization order of the rows and columns of a matrix is of outmost importance directly affecting the timeliness and accuracy of the result. The method solves the array into two parts, decomposition (or factorization) and the integrator uses the substitution forward-backward method.

Currently, ROS 2 (2nd order, 2 stages) and RODAS 3 (3rd order, 4 stages) Rosenbrock's methods are implemented. The time 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 (was used originally by the BRAMS model to

integrate the dynamics), sequential and sequential symmetric (Yanenko, 1971; McRae et al., 1981; Lanser and Verwer, 1998).

2.7 Model data structure

BRAMS original data structure uses Fortran 90/2003 derived data types to store related model fields. Each derived data type encapsulates the set of history carrying variables required by one model component. For example, the derived data type *radiation* contains radiation specific fields that are used and updated at each invocation of the radiation module. Prognostic fields are stored at a derived data type named *basic*, referenced by every module that requires prognostic field values. As a general rule, derived data types are used by every model component that requires their values, but modified only by its own model component.

This data structure keeps related fields in nearby memory positions, reducing cache line conflicts on current CPU memory architectures. Furthermore, it simplifies the introduction of new model parameterizations, by encapsulating at a derived data type all history carrying variables required by the new parameterization.

BRAMS multiple grids are implemented by arrays of these derived data types, indexed by grid number. As an example, suppose that variable *ngrid* contains the current grid number, suppose that *basic_g* is the array of the derived type *basic* that stores the prognostic fields of all grids and suppose that *u* is the component of *basic* that stores the x component of the wind velocity at all grid points. Then, *basic_g(ngrid)%u* stores the x component of the wind velocity of the current grid, and *basic_g(ngrid)%u(z,x,y)* addresses such variable at grid point (z, x, y).

CCATT-BRAMS data structure extends the original BRAMS data structure in a natural way. Derived data type *chem* contains history-carrying variables introduced by the chemistry. Multiple grids are represented by an array of type *chem*. But since each species requires its set of history carrying variables at each grid, the array has to be indexed by species number and grid number. Consequently, *chem_g(nspecie,ngrid)%sc_p(z,x,y)* stores field *sc_p* at grid point (z, x, y) of species

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*n*specie at grid *n*grid. This organization keeps at nearby memory addresses all fields of one species of one grid.

The (z, x, y) memory organization eases code development. Modelers can develop new code components for a single atmospheric column (the z direction) dealing with rank one arrays. Then, a wrapper is written to connect the new code component to the existing code. The wrapper loops over columns, invoking the code component with one column at a time. Unfortunately, most code components have vertical dependencies, preventing vectorization and increasing execution time. To reduce execution time by increasing the vectorization ratio, selected components are re-written to collapse the x and y dimension in just one dimension (xy) and to transpose the (z, x, y) organization into (xy, z) , following the work of Fazenda et al. (2006). The wrapper transposes the (z, x, y) memory organization into the (xy, z) and back, isolating the execution time optimization.

Chemistry is a grid point computation (that is, computation at one grid point is fully independent of any other grid point). Thus, the chemistry module could have been written on scalar variables, dealing with a single grid point at a time but that would prevent vectorization. To increase vectorization ratio, the chemistry module operates over a set of grid points at a time, represented by vectors of rank one. The driver breaks the (z, x, y) field organization into vectors of convenient size, invokes the module and stores back the results. Vector size is selected to keep the correct amount of data in cache in order to avoid cache misses.

2.8 Initial and boundary conditions

The initial and boundary conditions for the chemical species are treated in a similar way as for the meteorological variables. The RAMS/ISAN (ISentropic ANalysis package, Tremback, 1990) tool, originally developed to create initial and boundary conditions for the meteorological fields, was extended to include also chemical fields from global chemistry models. The model allows the boundary condition data be applied varying on time using Newtonian relaxation (nudging) technique.

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3 Examples of applications

The robustness of the CCATT-BRAMS system is explored throughout two case studies covering from the regional to local scale and from biogenic and fire emissions to urban emissions impacts.

3.1 Regional scale: Amazonian case study

During the LBA/BARCA (Large-Scale Biosphere Atmosphere Experiment in the Amazon Basin / *Balanço Atmosférico Regional de Carbono na Amazônia*, Andreae et al., 2012; Beck et al., 2012; Bela et al., 2013) field experiment, measurements of several trace gases, including O₃, CO, CH₄, CO₂ and aerosols were made onboard the INPE “Bandeirante” aircraft. Flights from Manaus transected the Amazon Basin from the Tropical Atlantic Ocean shore to the northwestern part of the Amazon Basin, over passing a variety of ecosystems from the surface to about 4.5 km altitude (Fig. 3). BARCA Phase A was in November 2008, during the transition between the dry and wet season, with rainfall to the west and south of Manaus but much drier conditions and widespread vegetation fires in eastern and northern Amazonia. During this period, the ITCZ was north of the studied region, which therefore received predominantly Southern Hemisphere air masses, with a strong contribution of smoke pollutants from the African continent. Phase B occurred in May 2009 during the opposite seasonal transition, from wet to dry season, when the ITCZ was located over the equator, abnormally south of its climatological position, favoring events of Northern Hemisphere inflow to Amazonia. The precipitation was above average and scattered fires occurred mainly far to the south.

Here we explore the O₃ data measured during BARCA. For model CO evaluation we refer to Andreae et al. (2012), in which BARCA A CO measurements were already compared with several different models results, including CCATT-BRAMS with 3-BEM fire emission sources, together with simulations for the BARCA A period using 7 different models. Overall, all the limited area models, including CCATT-BRAMS with

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3BEM fire emissions, captured at least some of the features of the observed mixing ratios, including the general levels of CO enhancement and regional variations. The dominant sources were fire activities in the eastern and southern part of the Amazon, which were well captured by 3BEM with an estimated accuracy of about 20 %. This model comparison indicated that the use of 3BEM emissions in high resolution models, including CCATT-BRAMS, yielded results consistent with observations, versus those from GFED with a large underestimation for GFEDv2 and GFEDv3 emissions of factors of four and seven, respectively. Vertical transport was a more insubstantial aspect of the modeling because of its strong dependence on the ability of the model to simulate convection over tropical regions, plume rise and emissions. All the 6 considered models failed equally in accurately simulating the vertical distribution. More detailed CCATT-BRAMS model evaluation for biomass burning CO and aerosol over the Amazon area and central part of South America for different time periods have been shown by Freitas et al. (2009), Longo et al. (2010) and Rosário et al. (2012).

In this work, BARCA O₃ data were disaggregated following a similar procedure to that adopted by Andreae et al. (2012) for CO. Vertical average profiles within four regions were considered: E and W of Manaus, within 4° of the Equator, and N and S of Manaus, roughly on the 60° W meridian.

We also compared model results with mean tropospheric O₃ retrieved from the Aura Ozone Monitoring Instrument and the Microwave Limb Sounder measurements (OMI/MLS), available globally on a 1° × 1.25° (latitude by longitude) grid size (Chandra et al., 2003, 2004). For model comparison, the tropopause was calculated according to the WMO (World Meteorological Organization) 2 K km⁻¹ lapse rate definition by the National Center for Environmental Prediction (NCEP) Climate Diagnostics Center.

3.1.1 Model configuration

For this regional scale case study the model was run from 1 October to 31 November 2008 and 1 May 30 June 2009, with two nested grids. The external one had a 140 km resolution including both the South American and African continents and the internal

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one had a 35 km resolution including only South America. This configuration attempts to improve the description of long-range transport of pollutants from Africa to South America by providing better boundary conditions of chemical fields to the nested grid domain. However, it must be said that there is an unavoidable inconsistency between the fire data sets used for emissions estimations in Africa and South America. While in South America there are multi-sensor fire data available (for details, see Longo et al., 2010), for Africa only the MODIS fire count was available. In Fig. 4., the model grid domains for this case study are presented with the total emissions for carbon monoxide (CO) obtained from PREP-CHEM-SRC (Freitas et al., 2011), using 3BEM (Longo et al., 2010) fire emissions as well all the others described in Sect. 2.2.

The model parameterizations chosen for the simulations described in this subsection are the following. The surface scheme was LEAF (version 3, Walko et al., 2000) tuned for tropical areas. The vertical PBL diffusion parameterization of RAMS used in this simulation was based on the Mellor and Yamada (1982) formulation, which prognoses turbulent kinetic energy (TKE). All radiative calculations were done using CARMA (Toon et al., 1989) with aerosol optical properties prescribed accordingly to Rosario et al. (2012). For the microphysics, we use the single-moment bulk microphysics parameterization, which includes cloud water, rain, pristine ice, snow, aggregates, graupel and hail (Walko et al., 1995). It includes prognostic equations for the mixing ratios of rain, of each ice category of total water and for the concentration of pristine ice. Water vapour and liquid cloud mixing ratios are diagnosed from the prognostic variables using the saturation mixing ratio with respect to liquid water. The shallow and deep cumulus scheme is based on the mass-flux approach (Grell and Devenyi, 2002). For the chemical integration the RACM mechanism was used.

The CPTEC/INPE global model T213 analysis provided initial and boundary conditions for the meteorological integration and the MOCAGE global chemistry forecast (Josse et al., 2004; Bousserez et al., 2007) provided boundary conditions for chemistry fields. Initial soil moisture was taken from the Gevaerd and Freitas (2006) estimation technique and the soil temperature was initialized assuming a vertically homogeneous

field defined by the air temperature closest to the surface from the atmospheric initial data.

3.1.2 Results

The monthly mean tropospheric O₃ comparison between the model outer domain results and Aura OMI/MLS retrievals for both BARCA periods suggest that the model successfully captured the general pattern of O₃ distribution over the South Atlantic region, though with an underestimation of O₃ (see Fig. 5). In both cases, the reader must disregard noise in the OMI/MLS retrieval over higher latitudes (Jing et al., 2006; Froidevaux et al., 2006).

For the BARCA A period, in November 2008, the model results indicate the predominance of the inflow of southwest Atlantic air masses into the Amazon Basin, which can be seen also in OMI/MLS data (Fig. 5, top). This inflow brings enriched air masses with biomass burning products from South Africa. The east-west gradients of O₃ mixing ratio inside the Amazon Basin are similar for both model and OMI/MLS data, though with a bias of about 30 %. The mean O₃ mixing ratio from the model ranged from about 30 ppbv in the central part of the Amazon Basin to about 50 ppbv on the eastern border of the basin. On the other hand, OMI/MLS data ranged approximately between 45 and 65 ppbv. However, the underestimation over the basin seems to be associated with an underestimation of O₃ in the air masses inflow from the South Atlantic by a factor of 20 ppbv. The three SHADOZ (Southern Hemisphere Additional OZonesondes, Thompson et al., 2007) O₃ profiles (not shown) obtained during this period (07, 19 and 28 November, 2008) at Natal, Brazil site (5.4° S, 35.4° W) also indicate an inflow of enriched O₃ air masses above 700 hPa, with a typical vertical gradient going from about 25 ppbv below this level to 80 ppbv above it on average but reaching 100 ppbv. Although, the Natal SHADOZ profiles cannot be compared directly with BARCA measurements, they do give an indication of the inflow air masses from the South Atlantic. These O₃ rich air masses were not intercepted by BARCA flights, which were mainly

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below 700 hPa (Fig. 6). In Fig. 6, the typical O₃ values observed below 4.5 km were captured by the model compared to BARCA measurements.

The underestimation of O₃ in the mid-troposphere over the South Atlantic region is likely related to an underestimation of ozone precursors, CO in particular. According to Andreae et al. (2012), BARCA CO measurement comparisons with CCATT-BRAMS and WRF-CHEM model results, both using 3BEM fire emissions, indicate that fire activities in the eastern and southern part of the Amazon were well captured by 3BEM with an estimated accuracy of about 20 %. However, a much larger underestimation of 3BEM emissions is expected for Africa compared to South America, associated with the differences/inconsistency between the fire remote sensing data sets used. Based on the fire data input used by 3BEM for Africa, one should expect it to be more comparable to GFED, which, according Andreae et al. (2012), yield a large underestimation of a factor four and seven for GFEDv2 and GFEDv3, respectively.

For the BARCA B period, in May 2009, the events of air mass inflow from the Northern Hemisphere were mostly captured on a case-by-case basis, but smoothed in the monthly mean for both model and Aura data (Fig. 5, bottom). During this period ozone was measured primarily in the northern and eastern regions (not shown). Typically the model also captured the general vertical structure in both regions.

A more detailed discussion of BARCA CO and O₃ measurements and comparison with CCATT-BRAMS and WRF-CHEM results is given in Andreae et al. (2012) and Bela et al. (2013), respectively. The two models using the same emissions set performed with similar accurateness for this case study.

3.2 Local scale: urban conditions

This case study was conceived to test a configuration for a planned air quality forecasting procedure for the major urbanized region in Brazil, which includes the metropolitan areas of São Paulo and Rio de Janeiro, as well as the highly urbanized and industrial areas connecting them.

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This configuration was evaluated throughout a comparison with air quality measurements data from several stations operated by the Environmental Agency of the state of São Paulo (CETESB), which continuously operates 42 ground stations in the state of São Paulo, several inside São Paulo metropolitan area (CETESB, 2012). All of them measure O_3 and NO_x and most of the stations in the metropolitan area of São Paulo also measure CO.

3.2.1 Model configuration

The model was run with a 3 km horizontal resolution from 20 July to 30 August 2011, with an 11-day period, from 20 July to 31 July used for spin-up. The model domain covers a large area of the states of São Paulo and Rio de Janeiro and the southern part of the state of Minas Gerais. It was conceived with two purposes: to include Rio de Janeiro metropolitan area and its influence on the air quality of the state of São Paulo, and to provide air quality forecasting for the majority of the important cities in the state of São Paulo, considering the compromise in the increased computational requirements, necessary for operational forecasting. In Fig. 7, the model domain is presented with the urban emissions for carbon monoxide (CO) following Alonso et al. (2010), obtained from PREP-CHEM-SRC (Freitas et al., 2011). The location of the metropolitan areas of São Paulo, Rio de Janeiro, Campinas and Ribeirão Preto are also indicated in the same figure.

The same model parameterizations as for the previous case study were chosen, except for deep convective parameterization that was not used and for the RELACS chemical mechanism instead of RACM. For the simulations presented in this subsection, boundary conditions for both meteorological and chemistry fields are provided by CPTEC/INPE CCATT-BRAMS operational forecast data currently with 25 km horizontal resolution (CPTEC/INPE, 2012) using the one-way nesting approach.

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3.2.2 Results

The monthly average carbon monoxide (CO) mixing ratios at the first peak hour (9:00 a.m., local time) are presented in Fig. 8. The model results indicate high mixing ratios, ranging from 1000 to 2000 ppbv, in the city of São Paulo; and moderately high, ranging from 500–750 ppbv, in Rio de Janeiro, Campinas and other major cities in the state of São Paulo. A similar behavior is observed at 6:00 p.m., local time, though less intense and more concentrated around the cities. In São Paulo a second peak is observed around 09:00 p.m., local time, and mixing ratios stay elevated even at night. Nitrogen oxides (NO_x) mixing ratios follows a similar diurnal cycle and are also more elevated at these hours: monthly averages at 09:00 a.m., local time, are presented in Fig. 9. NO_x typically peaked above 150 ppbv in the city of São Paulo and around 100 ppbv in the smaller towns. Ozone mixing ratios peak around 3:00 p.m., local time, monthly averages for this time are presented in Fig. 10.

Figure 11 presents monthly average diurnal cycle for carbon monoxide mixing ratios at two stations, *Parque Dom Pedro II* and *Pinheiros*, both in downtown area of the city of São Paulo. The model tends to underestimate the nocturnal observed values of carbon monoxide mixing ratios at these São Paulo stations. This is due to the specified diurnal cycle, which is not able to reproduce the emissions cycles for São Paulo, where the traffic at night is less intense than during the day, but still not negligible. A known difficulty also evident from the results is that this cycle is not the same throughout the city, and the proportion of light/heavy vehicles and the traffic driving patterns are not constant as well, modifying the emissions. The model results tends to overestimate the CO for stations located inside large vegetated areas where observations indicate values typically lower than the average, specially at peak hours. Nevertheless, for all the 10 stations evaluated inside the São Paulo metropolitan area, the model represents the diurnal cycle during daytime hours, and adequately estimates the order of magnitude in most cases with model values within the standard deviation of the monthly mean.

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The monthly average nitrogen oxides and ozone mixing ratios for the *Parque Dom Pedro II* stations are presented in Fig. 12. It is noted that the model overestimates the NO_x mixing ratios at the peak hours. The reasons are very similar to those for carbon monoxide. Additionally, the emission inventories are probably not accurate enough, and their spatial distribution in the city, according to the number of vehicles registered for each region, also has uncertainties when the locations are considered together. Also, only emissions for NO_x and CO are available (Alonso et al., 2010), VOC emissions were updated from local inventories keeping the RETRO ratio. As a secondary pollutant, ozone higher values are expected far from the sources of its precursors (nitrogen oxides and non methane volatile organic compounds – NMVOC). For all cities, a VOC : NO_x ratio at peak hour of emissions below 4 was observed, in the VOC sensitive regime, where the ozone levels are controlled by the VOC levels and a decrease in NO_x mixing ratios results in higher ozone levels. This was indeed reproduced by the model, which showed higher ozone levels in the countryside of the state of São Paulo, corresponding to lower NO_x emissions. Modeled O_3 and NO_x mixing ratios are both much closer in magnitude to observed values for the stations within the countryside. Figure 13 illustrates the O_3 and NO_x diurnal cycles for *Sorocaba*. This is similar to all stations in the countryside evaluated, including *Ribeirão Preto*, which is a larger city compared to the others with more the 600 thousands inhabitants.

4 Conclusions

In this paper, we have described the functionalities of the new Chemistry-CATT-BRAMS modeling system, which has been developed using advanced numerical tools in order to get a flexible multipurpose model that can be run for both operational forecasts and research studies. The code package includes preprocessor tools for emissions databases, chemical mechanisms, and boundary and initial conditions and the CCATT-BRAMS code itself. The model has been developed focusing not only, but with special attention to tropical and subtropical regions. The model performance was

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explored throughout two different applications for regional and local scales and different emission source influences.

For the regional scale a period when BARCA airborne measurements were undertaken over the Amazonia was simulated with CCATT-BRAMS. In spite of fire emission underestimation for Africa, which biased the burden pollution over the South Atlantic region, the model was able to capture the O₃ distribution and transport patterns.

For the urban case, NO_x and O₃ mixing ratios are respectively over and underestimated, but in all cases, within the standard deviations. Even O₃ mixing ratios at night, which usually represent a greater challenge for modelers, is assessed accurately. However, air quality forecasting for South American cities is still challenging due to the lack of reliable urban emission inventories. Nevertheless, the model did reproduce the VOC sensitive regime, with VOC : NO_x ratios typically below 4 at peak hour of emissions. Coherently, showing higher ozone levels in the countryside of the state of São Paulo, corresponding to lower NO_x emissions.

The CCATT-BRAMS currently runs for daily operational air quality forecasts at CPTEC/INPE proving its robustness. It has already been used in research to study atmospheric chemistry in Marécal et al. (2012), Iacono-Marzano et al. (2012) and Krysztofiak et al. (2012). These studies were based on other chemistry schemes than discussed in the present paper using the CCATT-BRAMS capability to easily change the chemistry scheme.

The CCATT-BRAMS models system represents a set of numerical tools comprising state-of-the-art emission information as well as physical and chemical parameterizations coupled to a reliable dynamic core, which can cover scales from meters to hundred kilometers. Moreover the system is coded with updated FORTRAN 90. Also its data structure is compact with good readability, which makes the code easily understandable and lower load of work for maintenance. The CCATT-BRAMS code package is available upon request to the email address gmai@cptec.inpe.br.

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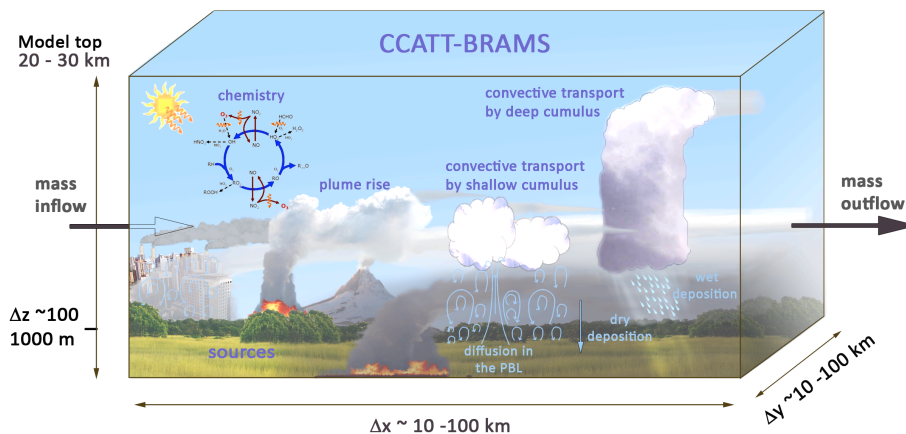


Fig. 1. Sub-grid processes involved in trace gases emission, transport, transformation and deposition simulated by the Chemistry–CATT-BRAMS system

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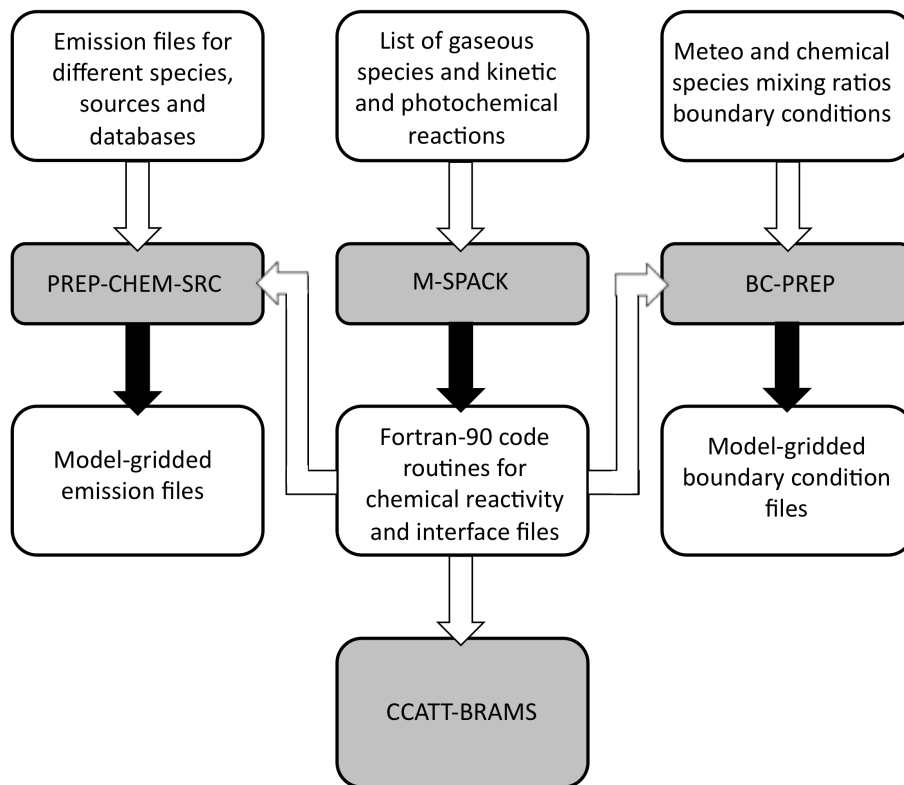


Fig. 2. Schematic of the CCATT-BRAMS system. The darker gray blocks and the black arrows indicate the codes that compose the CCATT-BRAMS System and their outputs, respectively. The lighter gray blocks indicate either the input files for the pre processing (first line) as the pre processing outputs (third line), which are also input files for pre processing emissions and boundary conditions and routines to compose CCATT-BRAMS model.

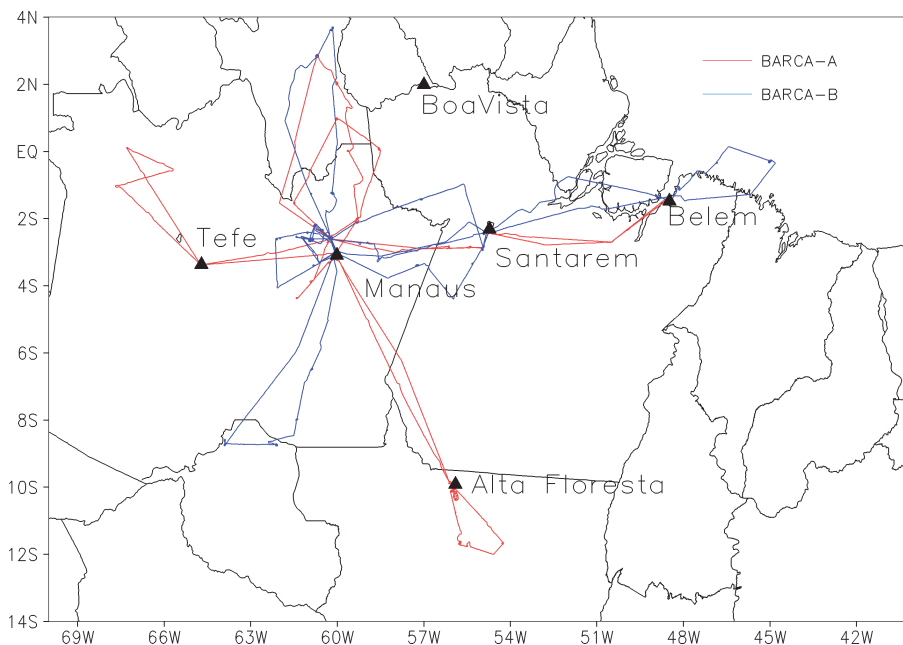


Fig. 3. Flight tracks of the BARCA A (red) and B (black) aircraft campaigns.

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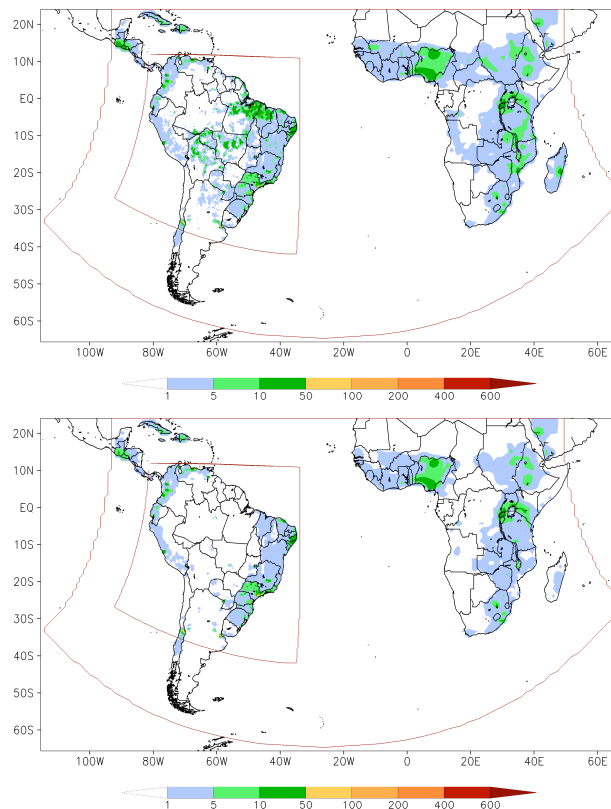


Fig. 4. CO emission rates ($10^{-5} \text{ kg m}^{-2} \text{ d}^{-1}$) for BARCA A (top) and BARCA B (bottom) periods from PREP-CHEM-SRC in the model grid domains.

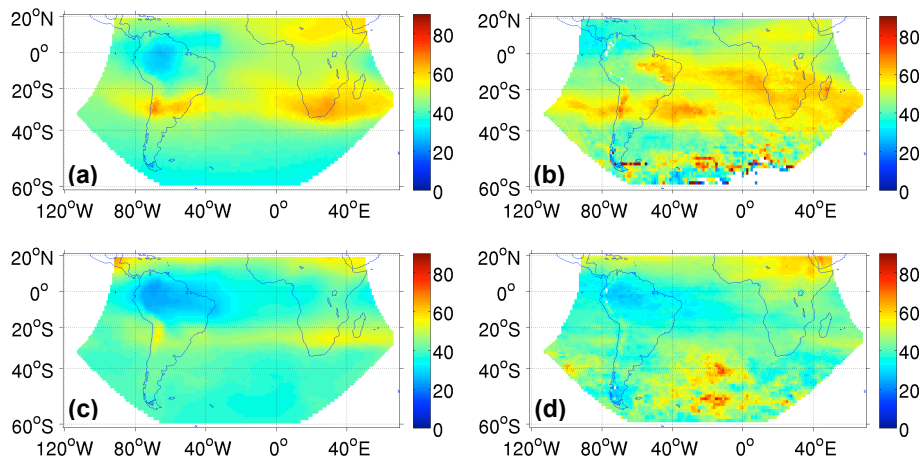


Fig. 5. Mean tropospheric O₃ (ppbv) for November 2008 (top) and May 2009 (bottom) from the model (a, c) and from OMI/MLS (b, d).

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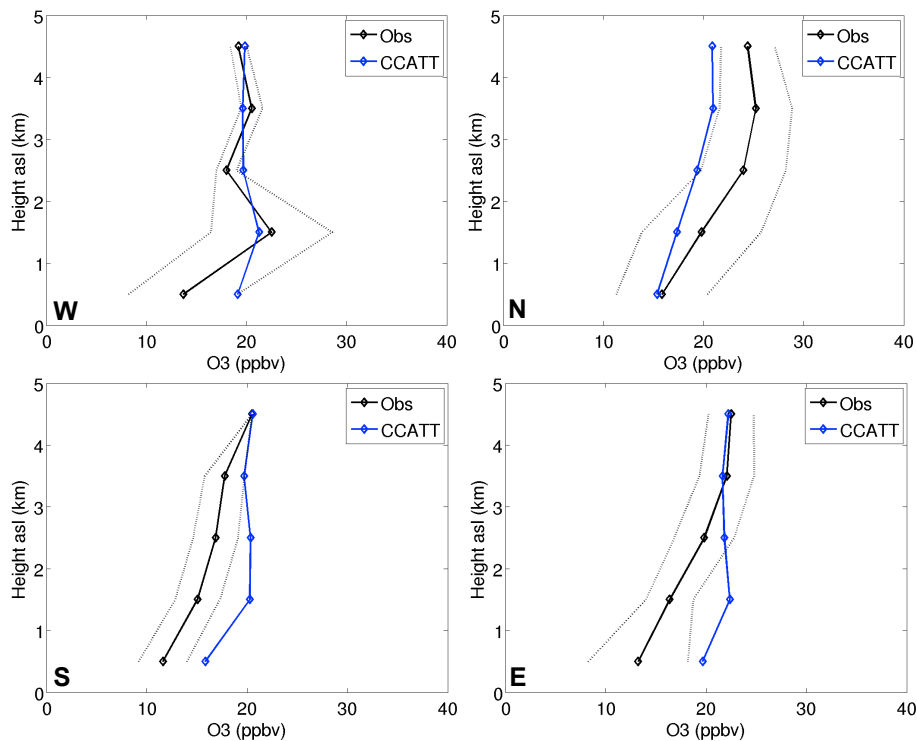


Fig. 6. Vertical profiles (regional medians, 1000 m blocks) for O_3 from CCATT-BRAMS model, compared to observations disaggregated by E, W, N and S regions of Manaus.

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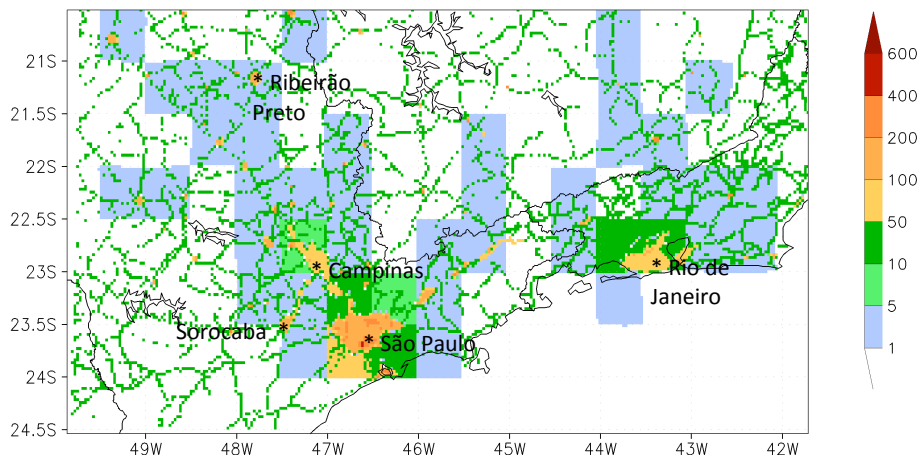


Fig. 7. CO emission rates ($10^{-5} \text{ kg m}^{-2} \text{ day}^{-1}$) from PREP-CHEM-SRC in the model domain.

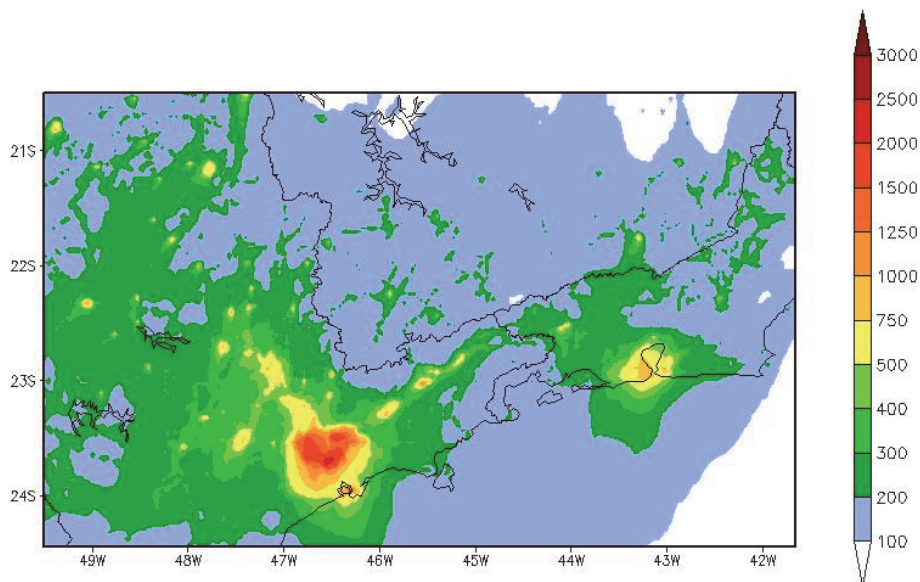


Fig. 8. Monthly average spatial distribution of CO mixing ratio (ppbv) at 09:00 a.m. local time from the model.

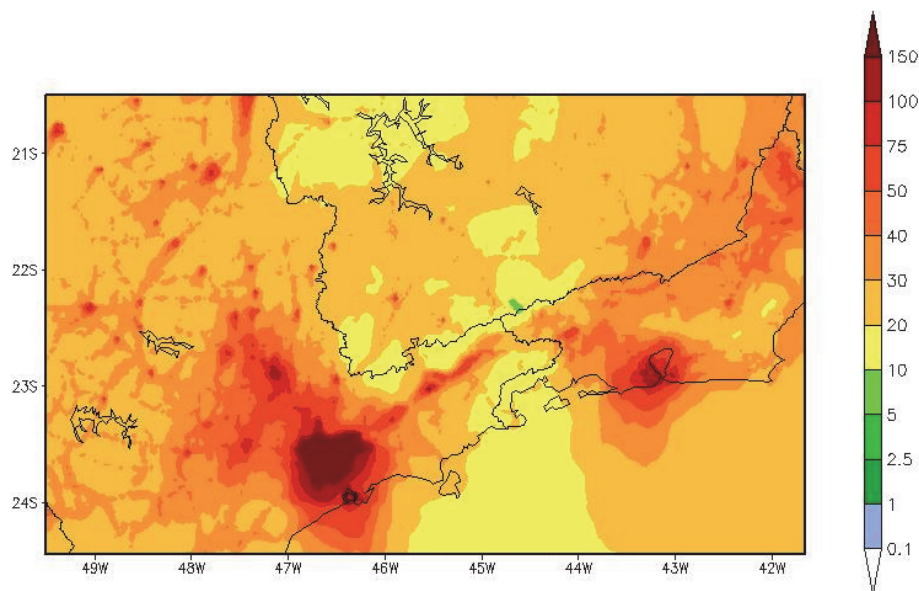


Fig. 9. Monthly average spatial distribution of NO_x mixing ratio (ppbv) at 9:00 a.m. local time from the model.

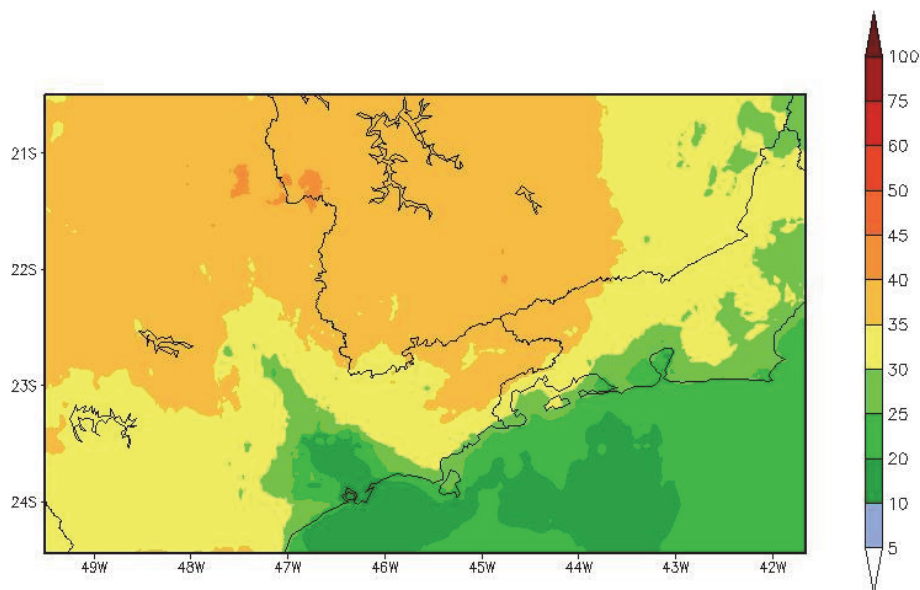


Fig. 10. Monthly average spatial distribution of O_3 mixing ratio (ppbv) at 03:00 p.m. local time from the model.

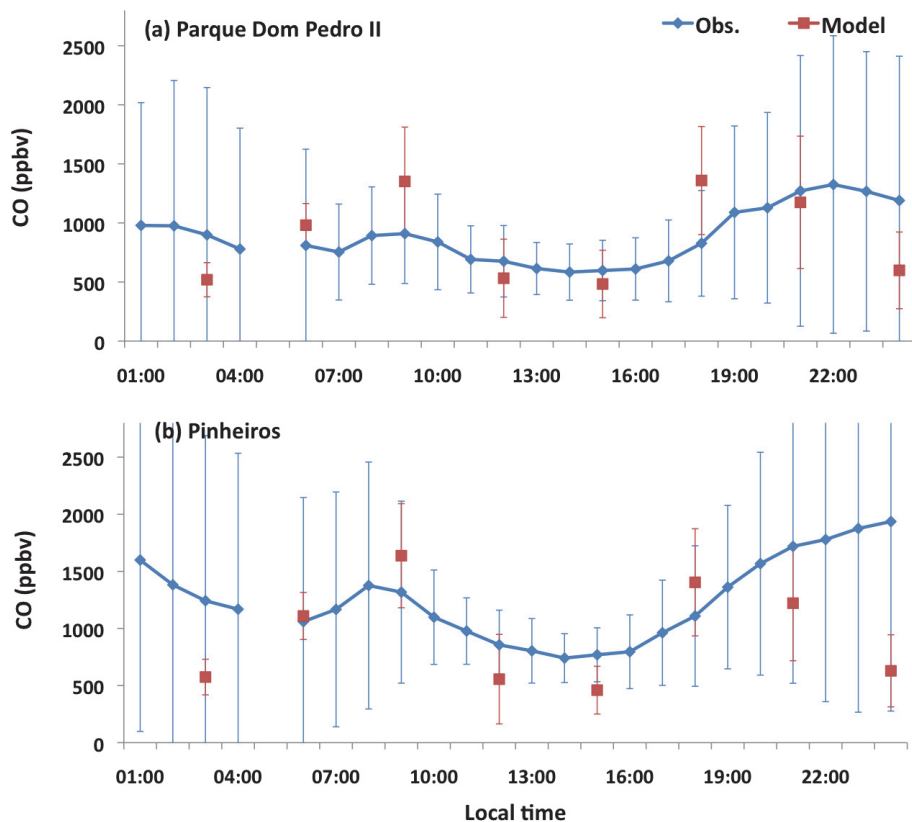


Fig. 11. Monthly average CO mixing ratio (ppbv) from two CETESB ground stations in São Paulo city, **(a) Parque Dom Pedro II** and **(b) Pinheiros**, and from the model.

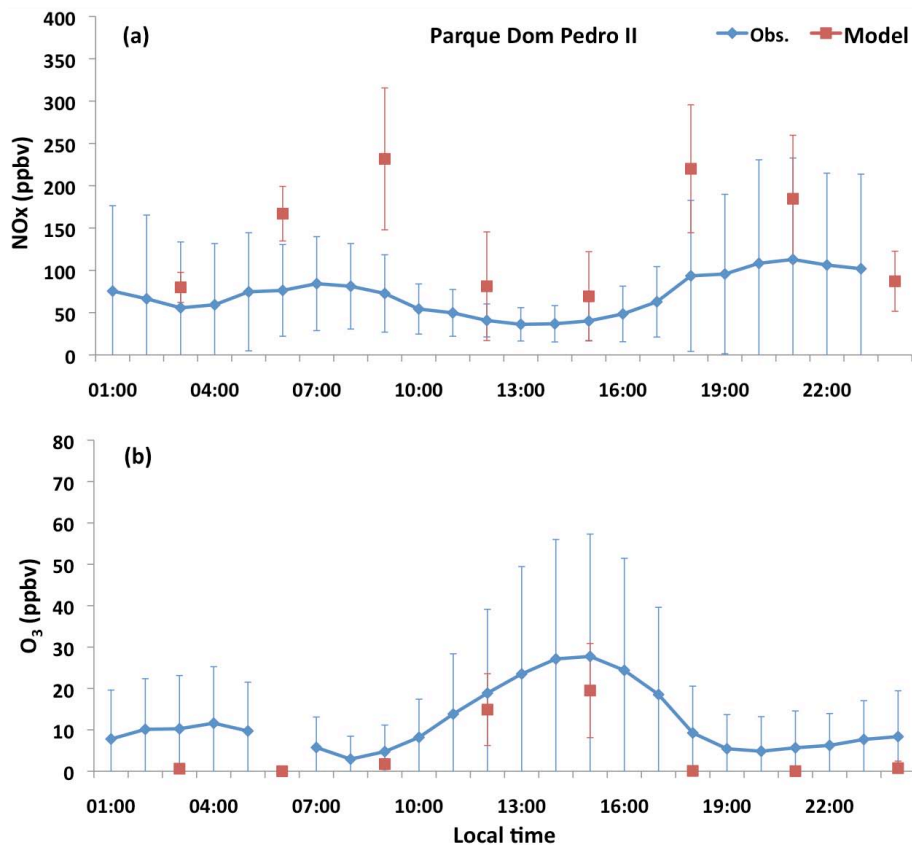


Fig. 12. Monthly (a) NO_x mixing ratio (ppbv–top) and (b) ozone (ppbv – bottom) from a CETESB ground station (*Parque Dom Pedro II*) in São Paulo city and from the model.

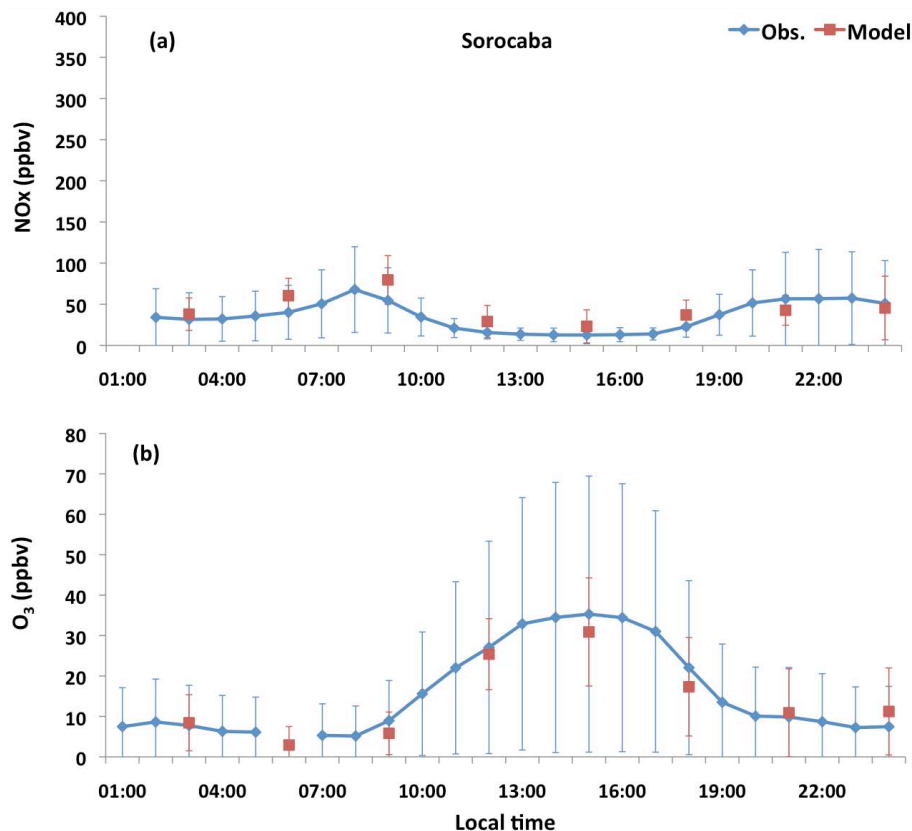


Fig. 13. Monthly (a) NO_x mixing ratio (ppbv–top) and (b) O_3 (ppbv – bottom) from a CETESB ground station in a small city in the countryside of the state of São Paulo (*Sorocaba*) and from the model.