

Interactive comment on “Gas-phase chemistry in the online multiscale NMMB/BSC Chemical Transport Model: Description and evaluation at global scale” by Alba Badia et al.

Anonymous Referee #2

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Review of

“Gas-phase chemistry in the online multiscale NMMB/BSC Chemical Transport Model: Description and evaluation at global scale” by Badia et al.

Overview:

The paper is a description and an evaluation of a one-year (2004) global simulation of what is called the NMMB/BSC Chemical Transport Model. The model results for CO, O₃ and NO₂/PAN/HNO₃ are compared against surface observations, profile observations and satellite retrievals. The comparison shows that the NMMB/BSC model gives acceptable results but both CO and ozone are overestimated on the global scale.

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General remarks:

My two main concerns with the presented model run for 2004 is (i) that NO_x emissions from lightning were not considered in the model run and (ii) that no specific biomass burning emissions for 2004 were used. A parameterisation of lightning emissions is scientific standard in global CTMs and there is no good reason, why such an important contribution to global tropospheric chemistry can be omitted. Getting daily or 8-day-mean 2004 biomass emission data (GFED, GFAS etc.) would not have been difficult. Also the lack of seasonality of the anthropogenic emissions is an unnecessary simplification. Against the backdrop of these omissions it becomes difficult to draw conclusion from the model results and it severely undermines the scientific credibility of the paper.

On the other hand, the presented model had two advanced properties, namely the on-line calculation of VOC emissions using the MEGAN model and the fact that the presented model is an on-line coupled chemistry – meteorological model (or Chemistry-GCM). The term CTM, which the authors choose, is commonly used for off-line model without the simulation of meteorology (see Baklanov et al. 2014, ACP). I therefore recommend not to use the term CTM in the name of the model because it is an on-line coupled model. Unfortunately, it is a missed chance that these two new aspects were not explored further in the paper.

The evaluation is carried out with a well-balanced choice of observations but the results are too often only described with the words such as “good agreement” etc. I think this is not very meaningful, instead the results should be quantified in a better way, i.e. a bias of 10 ppb, 20% etc.

It is a thought-provoking result that both CO and ozone are overestimated because an overestimation of the oxidation capacity is often linked with CO underestimation (see Strode et al. 2015, ACP) It is something which can not be found in other models using similar emission data, and especially for models that also use the CB05 chemical mechanism. I think this result deserves a more thorough investigation.

Without carrying out sensitivity studies, it is in general problematic to come to valid conclusion on the reasons for certain aspects (bad or good) of the model performance. The authors predominately only argue (without doing sensitivity studies) that (i) deficiency in the emissions and/or (ii) the lack of considering aerosol in the photolysis rates are the reasons for identified model deficiencies. While there is consensus in the scientific community that emissions can be very uncertain, there is no evidence given in the paper, why the aerosol impact should be so important as the authors claim. (I am happy to be convinced otherwise by a sensitivity study or a reference to it).

The authors should discuss other aspect of their model setup in more detail. If there is the feeling that photolysis rates play a role, then cloud cover would be the first suspect. The cloud cover should be checked for biases since the model simulates clouds itself. Also worth checking are the ozone total columns used in the photolysis scheme because they are not constrained by observations.

Another potentially important aspect is the fact the emissions are injected uniformly in the lowest 500m (anthropogenic) or 1300 m (biomass burning). This could have a large impact on dry deposition, which depends on the surface level concentration, and ozone titration by NO during the night. The 500 m seems to be an exaggeration of the extent of the mixed layer during the night over land and the choice needs to be better motivated. One would expect that the diffusion scheme of the model simulates the vertical mixing in the PBL. Also, the 1300 m for the biomass burning injection would need to be justified, as the fire injection height can vary substantially (see for example. Remy al., 2016, ACP)

The paper would greatly benefit from proofreading for English language.

Specific comments:

P1

Title: consider not calling the model a CTM as CTM's are understood as "off-line"

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Abstract - Spell out NMMB/BSC and other acronyms - No need to specify all the network and instruments in the abstract

P2

L 27: a better reference for IFS-MOZART is Flemming et al. 2009, GMD

L 33: Please clarify if the non-hydrostatic option was used in the run.

P3

L 26: better “in detail”

P4

L15-25: please clarify which of the options is actually used in the presented run. The other options don't need to be mentioned. They could be referenced.

P 5

L 17: 1850 ppb of methane seems too high for 2004. The value should be 1775 ppb
http://www.esrl.noaa.gov/gmd/ccgg/trends_ch4/

L 22: Add more information about the realism of the two input fields (overhead ozone and clouds)

P 6

L 10: Is this a monthly climatology ?

L 13: “cloud processes” – does this also include wet-phase chemistry ?

L 14: The presented terms are not clear. Please clarify what you mean by all the mentioned processes. For example, what is wet deposition for non-precipitating cloud?

L16: Why only in-cloud scavenging and not all the other processes ?

L 19: How is the cloud time scale derived?

P7

L2: Do you refer to large-scale and convective precipitation here?

L7: “Convective mixing” do you mean transport by convective mass fluxes ?

L 13: 100 hPa is a rather high tropopause for mid- and high latitudes.

L 13: Were these Mozart 4 fields evaluated for the stratosphere ?

L 18ff: There is no need to present the COPCAT scheme here, a reference to the paper is enough.

L 18: Please provide information on the biases of your stratosphere ozone simulated by the COPCAT scheme because they have an impact on the photolysis rates.

P 8

L 14: No lightning emissions is a severe shortcoming of the simulation and the paper (see my general comment).

L 15: Please explain in more detail how the MEGAN code was integrated in your model.

L34: I don't understand the 24h averages here. I thought (L23) the actual hourly meteorological data were used for the calculation of the Megan emissions.

P 9

L3: better say “every 720 s”

L 5: Which fields are initialised (also clouds or only T, v,w,q). What is known about the biases of the 24 h forecasts?

L 12: not using 2004 fire emissions is a severe omission (see my general comment). Please clarify what fire emissions have been used. Was it an average for the period? It is not clear what “interpolated” means. Do the fire emissions have a seasonal cycle ?

L 26: see my general comment, please justify the choices

L 29: Please provide reference the strong impact of aerosol on the photolysis rates.

P 11

L27: It is not clear how missing data in the surface observations were considered. If you compare only averages without timely match give numbers of the amount of missing data.

L 30: 1000m asl.? This could be a mountain stations near to the coast or a station on a flat plateau inland. It would be better to include the model orography in the choice of the mountain stations. (say 500 m above orography)

P 12

L1: This choice of the tropopause is not consistent with the choice of the tropopause for the chemical boundary conditions (P7L13).

L 29: see my general comment. The aerosol effect may not be the most important one. There are many other possible explanations: high CH₄, water vapour, clouds and photolysis, excessive mixing of emissions etc.

P 13

L1: CH₄ is also a CO source. Please also reformulate the sentence.

L 14 add reference for C-IFS

L 29: Could the high methane be a reason ?

L 30: Figure 3. Please show separate plots for NH, SH mid-latitudes and tropics. The seasonality is obscured by averaging over all stations.

P 14

L2: no seasonality of the anthropogenic emissions is an oversimplification.

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L3: Figure 3 shows the relative bias (%), not MB as defined in the supplement.

L3 Please clarify correlation of which time scale is shown, i.e. of the hourly, daily monthly values? Did you filter out seasonality? How important is the diurnal cycle to the correlation.

L 28 Stein et al. and many other authors find a general underestimation in winter and spring NH.

P 15

L8: What do you mean by overestimated emissions above the PBL ?

L 8: Please discuss the role of convection

L 32: What regime (rural, urban) was used ?

P 16

L 8: Please discuss also PBL mixing during the night

P 17

L 8: see my general comment on the use of “good agreement”

L 17: Please mention the value of the biases.

L 26: What is the seasonal cycle of the biomass emissions ?

P 18

L 1 “rural” (?) perhaps better remote

L 4: dominated by the tropics - perhaps simply because they are the largest region on earth (?)

L 7: TM5 has a similar chemical mechanism. Should it not be similar ?

L 16: Please clarify how the STE is calculated in your model.

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L 27: “all day long “ ? Do you mean “throughout the year”

L 27: For global models the values are commonly given in volume mixing ratios (ppb). Try to avoid mg/m³ throughout the paper.

L 28: The emission injection (500m) leads to a dilution of NO and therefore a reduction of the ozone titration. This could also explain the overestimation.

P 19

L4 Please quantify biases, what do you mean by “error”.

L 15. This points to biases of the COPCAT ozone, which has consequences for the photolysis rates.

P 20

L 8: The lack of aerosol modulation of photolysis is probably a minor aspect. Lack of heterogeneous chemistry (N₂O₅) might be more important. Please also mention the main shortcomings of this simulation: (1) no lightning, (2) no 2004 biomass burning emissions and (3) no seasonal cycle for anthropogenic emissions.

L15: The paper provides no evidence for this claim - it can therefore not be a conclusion.

L 34 see above, no evidence in the paper

P 21

L 13 better “megacities”

Figure 3: better to have plots for different regions (NH, SH, Tropics) to better see the seasonal cycle. Choose a smaller y-range for more clarity. Show plot in ppb (as for the profiles) rather than microgramm/m³.

Figure 4: MB (see supplement) is defined without scaling (i.e. not relative in %). Please show the MB as defined in the supplement. Use ppb as unit.

Figure 7: better y-range, use ppb

Figure 8: as for Figure 4

Figure 11: use ppb

Figure 12: see Figure 4

Figure 13: choose x-range 0-100 for better clarity in the troposphere.

Figure 14: see Figure 4

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