

RC1: 'Reviewer Comments', Anonymous Referee #1, 01 Aug 2016

The authors present a detailed description and evaluation of the tropospheric chemistry transport model NMMB/BSC-CTM. This model domain has been expanded from regional to global. The focus of their evaluation is gas-phase chemistry with emphasis on tropospheric ozone and its precursors. Several ground-based, aircraft and satellite data are used to show model strengths and weaknesses. The paper is well-written and is within the scope of the journal. I would recommend the publication of this paper after my minor comments below have been addressed:

***Response:** The authors wish to thank anonymous reviewer #1 for his/her valuable comments and suggestions.*

Note that we have decided to rename our model following a comment from reviewer #2 about avoiding the use of CTM for an online model. Thus, the new name is NMMB-MONARCH, where MONARCH stands for "Multiscale Online Nonhydrostatic Atmosphere Chemistry model". In the responses to the reviewer's comments we keep the NMMB/BSC-CTM name to keep consistency with the manuscript submitted to GMDD, but in the revised manuscript the new name, NMMB-MONARCH, is used.

Now, the revised manuscript is entitled "Description and evaluation of the Multiscale Online Nonhydrostatic Atmosphere Chemistry model (NMMB-MONARCH) version 1.0: gas-phase chemistry at global scale".

Page 2, line 15: replace "fed by emission inventories" with "emissions of chemical species".

***Response:** Amended.*

Page 2, Line 31: Define NMMB/BSC here.

***Response:** In the revised manuscript MONARCH is now defined there.*

Page 3, line 19: replace "main reactions occurring in the atmosphere by" with "atmospheric composition". It would be helpful to give a motivation for choosing year 2004 for evaluation.

***Response:** Amended. 2004 is a reference year for our modeling group that we already considered in previous studies (e.g., Pay et al. 2010; Baldasano et al. 2011). Therefore, our choice is based on the amount and variety of quality controlled and quality assured observations available in our group. We don't think this information is relevant for the manuscript.*

Pay, M. T., et al. "A full year evaluation of the CALIOPE-EU air quality modeling system over Europe for 2004." Atmospheric Environment 44.27 (2010): 3322-3342.

Baldasano, J. M., et al. "An annual assessment of air quality with the CALIOPE modeling system over Spain." Science of the Total Environment 409.11 (2011): 2163-2178.

Page 3, line 28: Insert "direct" before radiative effect.

***Response:** Amended.*

Page 5, line 10: A reference is needed here.

***Response:** The reference of Yarwood (2005) is now included.*

Page 5, line 17: 1.85 ppm is too high for this year of simulation. Can you please provide a justification for using this number? Also, is the CH₄ concentration constant throughout the troposphere in the model or only at the surface?

Response: *Considering a global background concentration of methane is a common practice in air quality modeling. Following this approach, current practices set the methane background level either as a default background concentration (i.e. 1.76 ppm, e.g. Shindell et al, 2006), or as the background level for the Northern Hemisphere (i.e. 1.85 ppm, used for instance within CMAQ). Including either of those concentrations would lead to differences with respect to reality, and we decided to select the latter, which on the other hand is the closest to the present time global background concentration (1.83 ppm, see WMO 2015, or Dlugokencky, 2016). The global average for 2004 is reported to be 0.06 to 0.07 ppm lower, around 4%. This small difference is not expected to cause any sizeable differences in the results shown here.*

Shindell et al. Multimodel simulations of carbon monoxide: Comparison with observations and projected near-future changes. Journal of Geophysical Research Atmospheres. VOL. 111, D19306, doi:10.1029/2006JD007100, 2006

WMO Greenhouse Gas Bulletin N° 11: November 2015

Ed Dlugokencky, NOAA/ESRL www.esrl.noaa.gov/gmd/ccgg/trends_ch4/, viewed on 30/09/2016

Page 8, section 2.2.6: Where does the MEGAN model implemented in this CTM derive the leaf-area index needed to calculate biogenic emissions?

Response: *The leaf-area index is obtained from the MEGANv2.04 databases (<http://lar.wsu.edu/megan/guides.html>). The data is originally at 150 sec horizontal resolution and it is averaged to the NMMB/BSC-CTM model grid. It is described in Guenther et al. (2006).*

Guenther, A. et al. Estimates of global terrestrial isoprene emissions using MEGAN (Model of Emissions of Gases and Aerosols from Nature). Atmos. Chem. Phys 1–30 (2006).

Page 8, line 29: Need a period after “parameters”.

Response: *Amended.*

Page 9, section 3: What is the size of the bottom-most layer in the model? Also provide an estimate of the time it takes to run a year’s simulation.

Response: *The size of the bottom-most layer in the model is below 40 m. This information is now included in the revised manuscript. The time to run a yearly simulation is about 2 weeks using 132 cores in the Marenostrum supercomputer based on Intel SandyBridge-EP E5-2670/1600 20M 8-core at 2.6 GHz.*

Page 9, lines 12-13: Since emissions after year 2000 were not provided by Lamarque et al., which projection (RCP?) was used for 2010 emissions to perform linear interpolation?

Response: *Thanks for pointing this out. There was an error in the description of the methodology used to derive the 2004 anthropogenic and biomass burning emissions. We stated that these emissions were obtained by interpolation between years 2000 and 2010. In reality we considered the emissions for year 2000 from Lamarque et al. (2010). This issue has been clarified in the revised manuscript, as follows: “Note that this methodology involves*

assuming 2004 emissions equivalent to the best estimate reported for ACCMIP for year 2000”.

Lamarque, J.-F., Bond, T. C., Eyring, V., Granier, C., Heil, A., Klimont, Z., Lee, D., Liousse, C., Mieville, A., Owen, B., Schultz, M. G., Shindell, D., Smith, S. J., Stehfest, E., Van Aardenne, J., Cooper, O. R., Kainuma, M., Mahowald, N., McConnell, J. R., Naik, V., Riahi, K., and van Vuuren, D. P.: Historical (1850–2000) gridded anthropogenic and biomass burning emissions of reactive gases and aerosols: methodology and application, *Atmos. Chem. Phys.*, 10, 7017-7039, doi:10.5194/acp-10-7017-2010, 2010.

Page 10, line 25: Any particular reason why only two aircraft campaigns were used for the evaluation instead of several others available in Emmons et al. (2000).

Response: The model was evaluated against all the campaigns available in Emmons et al. (2000). However, for the paper we selected the two closest campaigns to year 2004. Figure 1 shows the comparison of the model with the PEM-Tropics and POLINAT-2 campaigns for HNO₃, NO_x and PAN. We have included Figure 1 in the supplementary material (see Figure S5) and additional text describing the results in the main manuscript (see also Table 5).

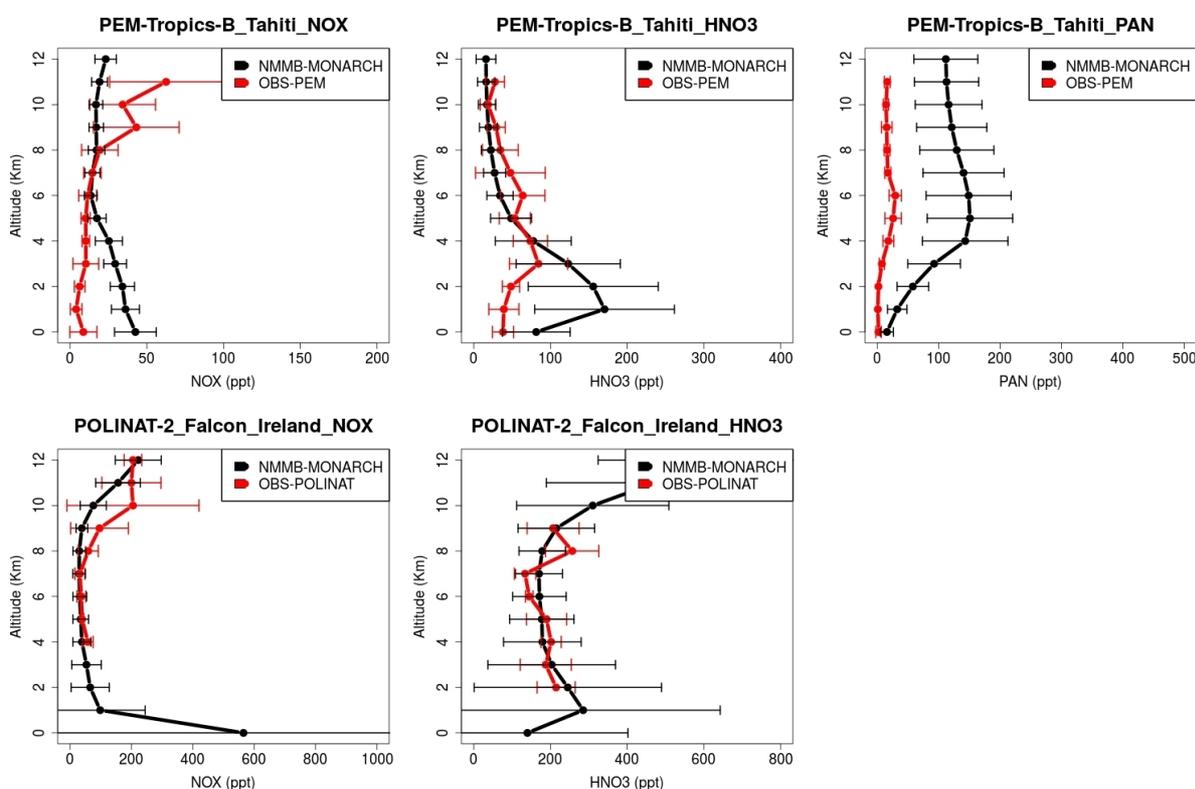


Figure 1. Comparison of modeled (black lines) and observed (red lines) vertical profiles of NO_x and HNO₃ and PAN for Tahiti and Ireland. Horizontal lines show the standard deviations.

Emmons, L. K., Hauglustaine, D. A., Müller, J.-F., Carroll, M. A., Brasseur, G. P., Brunner, D., Staehelin, J., Thouret, V., and Marenco, A.: Data composites of airborne observations of tropospheric ozone and its precursors, *Journal of Geophysical Research: Atmospheres*, 105, 20 497–20 538, doi:10.1029/2000JD900232, <http://dx.doi.org/10.1029/2000JD900232>, 2000.

Page 12, section 5.1: What is the simulated tropospheric lifetime of methane in the model and how does it compare with that from multi-model studies (e.g., Naik et al 2013a)?

Response: Methane lifetime was not explicitly calculated during model execution. While the burden can be calculated in post processing the estimation of the mean tropospheric methane – OH oxidation flux (needed to calculate lifetime) would require repeating the simulations. Therefore, we can neither include this information, nor discuss it at present in the manuscript.

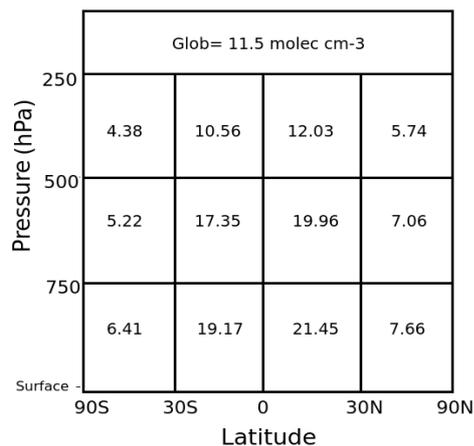
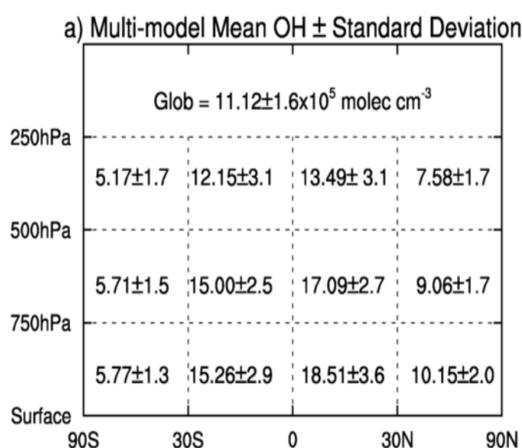
How does the simulated OH interhemispheric ratio compare with other studies (Naik et al., 2013a; Patra et al., 2014).

Response: The mean OH inter-hemispheric (N/S) ratio of the model is 1.18. This quantity is comparable with the present-day multi-model mean ratio (1.28 ± 0.1) shown in Naik et al., (2013). This information is now included in the revised manuscript.

Naik, V., Voulgarakis, A., Fiore, A. M., Horowitz, L. W., Lamarque, J.-F., Lin, M., Prather, M. J., Young, P. J., Bergmann, D., Cameron-Smith, P. J., Cionni, I., Collins, W. J., Dalsøren, S. B., Doherty, R., Eyring, V., Faluvegi, G., Folberth, G. A., Josse, B., Lee, Y. H., MacKenzie, I. A., Nagashima, T., van Noije, T. P. C., Plummer, D. A., Righi, M., Rumbold, S. T., Skeie, R., Shindell, D. T., Stevenson, D. S., Strode, S., Sudo, K., Szopa, S., and Zeng, G.: Preindustrial to present-day changes in tropospheric hydroxyl radical and methane lifetime from the Atmospheric Chemistry and Climate Model Intercomparison Project (ACCMIP), *Atmos. Chem. Phys.*, 13, 5277-5298, doi:10.5194/acp-13-5277-2013, 2013.

Lightning NO_x emissions have been shown to contribute significantly to tropospheric OH concentrations (Murray et al., 2013). Lightning NO_x emissions are not considered in the current model set-up. Please explain how the simulated OH concentrations match closely with those of other modeling studies that include lightning NO_x emissions.

Response: We calculated the regional mean air mass-weighted OH concentrations and they are close to the multi-model values in Naik et al. (2013a) (see Fig. 2). Over the tropics (30S-30N) our OH is slightly higher, and above 500 hPa, is lower than the multi-model mean. Labrador et al. (2004) studied the sensitivity of OH to NO_x from lightning. They showed that OH increases mostly in the middle to upper troposphere (500-200 hPa) when lightning emissions are considered. Accordingly, the lack of lightning emissions in our model could explain the lower OH values above 500 hPa reported here. This discussion is now included in the revised manuscript and Fig. 2 (right panel) is included in the supplementary material as Figure S2.



Naik et al. 2013

NMMB/BSC-CTM

Figure 2. Comparison between the model and Naik et al., (2013) regional mean airmass-weighted OH concentrations ($\times 10^5$ molecule cm⁻³).

Labrador, L. J., R. von Kuhlmann, and M. G. Lawrence (2004), Strong sensitivity of the global mean OH concentration and the tropospheric oxidizing efficiency to the source of NO_x from lightning, *Geophys. Res. Lett.*, 31, L06102, doi:10.1029/2003GL019229

Naik, V., Voulgarakis, A., Fiore, A. M., Horowitz, L. W., Lamarque, J.-F., Lin, M., Prather, M. J., Young, P. J., Bergmann, D., Cameron-Smith, P. J., Cionni, I., Collins, W. J., Dalsøren, S. B., Doherty, R., Eyring, V., Faluvegi, G., Folberth, G. A., Josse, B., Lee, Y. H., MacKenzie, I. A., Nagashima, T., van Noije, T. P. C., Plummer, D. A., Righi, M., Rumbold, S. T., Skeie, R., Shindell, D. T., Stevenson, D. S., Strode, S., Sudo, K., Szopa, S., and Zeng, G.: Preindustrial to present-day changes in tropospheric hydroxyl radical and methane lifetime from the Atmospheric Chemistry and Climate Model Intercomparison Project (ACCMIP), *Atmos. Chem. Phys.*, 13, 5277-5298, doi:10.5194/acp-13-5277-2013, 2013.

Page 12, line 28: Need a reference for aerosol influence on OH. Also, a larger oxidizing capacity would be simulated if lightning NO_x emissions were included in these simulations.

Response: Real and Sartelet (2011) studied the effect of aerosols in the photolysis rates and gaseous species, showing that differences in photolysis rates lead to changes in gas concentrations, with the largest impact simulated on OH and NO concentrations. At the ground, monthly mean concentrations of both species were reduced over Europe by around 10 to 14% and their tropospheric burden by around 10%. The decrease in OH led to an increase of the lifetime of several species such as VOC. On the other hand, Bian et al. (2003) evaluated the effect of aerosols on the global budgets of O₃, OH and CH₄ through their alteration of photolysis rates. The impact identified was to increase tropospheric O₃ by 0.63 Dobson units and increase tropospheric CH₄ by 130 ppb (via tropospheric OH decreases of 8%). Although the CH₄ increases were global, the changes in tropospheric OH and O₃ were mainly regional, with the largest impacts in northwest Africa for January and in India and southern Africa for July.

As we have described in a previous comment, a larger oxidizing capacity would be simulated, especially above 500 hPa, if lightning NO_x emissions were included in our model run (Labrador et al., 2004).

Both aspects, and corresponding references, are now discussed in the revised version of the manuscript with the following paragraph: “Therefore, the lack of lightning emissions in our model run could at least partly explain the lower OH values above 500 hPa reported here. Another potential explanation is the lack of aerosols in our simulation, which may overestimate photolysis rates in polluted regions (e.g., Bian et al., 2003; Real and Sartelet, 2011).”

Real, E., and K. Sartelet. "Modeling of photolysis rates over Europe: impact on chemical gaseous species and aerosols." *Atmospheric Chemistry and Physics* 11.4 (2011): 1711-1727.

Bian, H., M. J. Prather, and T. Takemura, Tropospheric aerosol impacts on trace gas budgets through photolysis, *J. Geophys. Res.*, 108(D8), 4242, doi:10.1029/2002JD002743, 2003.

Labrador, L. J., R. von Kuhlmann, and M. G. Lawrence (2004), Strong sensitivity of the global mean OH concentration and the tropospheric oxidizing efficiency to the source of NO_x from lightning, *Geophys. Res. Lett.*, 31, L06102, doi:10.1029/2003GL019229

Page 13, line 9-10: Please give the reason why there were low CO concentrations in 2004 despite large Alaskan and Canadian wildfires.

Response: Elguindi et al., 2010 presented a global analysis of observed CO seasonal averages and interannual variability for the years 2002-2007. They analyzed the CO concentrations during this period: “In JJA 2003, the anomalously high concentrations of CO due to the intense heat wave experienced in Europe, especially in August (Tressol et al., 2008; Ordoñez et al., 2010), are well represented in the data. Likewise, the high concentrations seen in SON 2002 are due to exceptional circumstances, namely the intense boreal forest fires which occurred over western Russia (Edwards et al., 2004; Yurganov et al., 2005; Kasischke et al., 2005).”

In summary, there were also important fires during the period 2002-2007 and meteorological conditions that could have an impact to the CO concentrations, like the intense heat wave or the photochemical conditions. This is why Elguindi et al., 2010 concluded that “despite the intense boreal forest fires that occurred during the summer in Alaska and Canada, the year 2004 had comparably lower tropospheric CO concentrations”.

Elguindi, N., Clark, H., Ordóñez, C., Thouret, V., Flemming, J., Stein, O., Huijnen, V., Moinat, P., Inness, A., Peuch, V.-H., Stohl, A., Turquety, S., Athier, G., Cammas, J.-P., and Schultz, M.: Current status of the ability of the GEMS/MACC models to reproduce the tropospheric CO vertical distribution as measured by MOZIC, *Geoscientific Model Development*, 3, 501–518, doi:10.5194/gmd-3-501-2010, <http://www.geoscientific-model-dev.net/3/501/2010/>, 2010.

Page 13, 11-15: Other modeling studies suggest even lower CO burden (e.g., Naik et al., 2013b). Could higher CH₄ concentration prescribed in the model play a role in the simulated high CO burden?

Response: The influence of CH₄ on CO has been assessed through a short sensitivity test. Changing the CH₄ prescribed value from 1.85 ppm (NH background average) to 1.78 ppm (global average for 2004) lead to changes in daily average CO concentration up to ±0.12 ppb, which leads us to believe that other factors have a larger impact on CO burden (see for instance Shindell et al., 2006).

Shindell, D. T., et al. (2006), Multimodel simulations of carbon monoxide: Comparison with observations and projected near-future changes, *J. Geophys. Res.*, 111, D19306, doi:10.1029/2006JD007100

Page 13, last paragraph: the role of seasonal CO emissions in explaining the low northern hemisphere wintertime bias has been highlighted by Stein et al., (2014), which should be noted here.

Response: Stein et al., (2014) is already discussed in the manuscript on page 14 and 15. On page 14 of the manuscript “During winter and spring, Stein et al. (2014) also obtain an underestimation of CO vertical profiles in airports located in the Northern Hemisphere (NH)” and “The wintertime negative bias (~ - 10-35 ppb) in the NH may be explained by either the lack of seasonally varying anthropogenic emissions in our simulation, an underestimation of CO emissions (Stein et al, 2014), or a combination thereof”. On page 15 “Stein et al. (2014) suggests that the persistent negative bias in northern mid-latitude CO in models is most likely due to a combination of too low road traffic emissions and dry deposition errors.”

Stein et al., (2014) On the wintertime low bias of Northern Hemisphere carbon monoxide found in global model simulations, *Atmos. Chem. Phys.*, 14, 9295-9316, doi:10.5194/acp-14-9295-2014.

Page 15, line 25: Give the lifetime of NO_x.

Response: The lifetime of NO_x varies considerably with altitude, being only a few hours near the PBL and up to a few days in the upper troposphere (Tie et al., 2001 and 2002). This information is now included in the revised manuscript with the following sentence: “It has a relatively short lifetime (a few hours near the PBL and up to a few days in the upper troposphere; Tie et al., 2001 and 2002)”.

Tie, X., R. Zhang, G. Brasseur, L. Emmons, and W. Lei (2001), Effects of lightning on reactive nitrogen and nitrogen reservoir species in the troposphere, *J. Geophys. Res.*, 106(D3), 3167–3178, doi:10.1029/2000JD900565.

Tie, X., Zhang, R., Brasseur, G. et al. *Journal of Atmospheric Chemistry* (2002) 43: 61. doi:10.1023/A:1016145719608

Page 18, line 13-18: How do the calculated dry deposition estimates compare with those from more recent chemistry-climate model simulations (e.g., Naik et al., 2013b).

Response: The calculated dry deposition (1209 Tg O₃) is higher than in TM5 (829 Tg O₃) and MOZART-2 (857 Tg O₃) and similar to LMDz-INCA (1261 Tg O₃) and the multimodel ensemble in Stevenson et al. (2006) (1003 +- 200 Tg O₃). In addition, the model shows similar results to the GFDL AM3 chemistry-climate model (Naik et al., 2013b). The reference to GFDL AM3 model is now added in the revised manuscript.

Naik et al., 2013b, Impact of preindustrial to present day changes in short-lived pollutant emissions on atmospheric composition and climate forcing, *J. Geophys. Res.*, doi: 10.1002/jgrd.50608.

Page 19: What fraction of model O₃ biases could be related to biases in the simulated meteorological fields (e.g., temperature)?

Response: This is not an easy question to answer. Ozone is sensitive to temperature, solar radiation and vertical mixing. It is clear that biases in the meteorology will have a significant impact on the ozone biases. Another study would be required to provide a thorough quantification of the biases and this is beyond the scope of the present work. In any case, the NMMB meteorological skills are under constant improvement at NCEP and the authors consider the computed meteorology to lie within the skills of current state-of-the-art meteorological models.

Map figures: Please remove the grey background on the maps as this makes it difficult to read the colours.

Response: *We think that the color scale is readable, according to the figure's purpose, and if we remove the grey background the white dots in the figure would not be visible. Therefore, we kept the figures with the grey background.*

Figure 6, 10: Colour bar text is too small to read.

Response: *Amended.*