Response to the review of the revised manuscript by an anonymous reviewer:

The authors have provided a comprehensive response to my first review. Although I still think lightning emissions should not be neglected in a global model run, I am OK to accept the authors' justification for this if it is made clear in the paper. I recommend stating this fact in the abstract.

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

Following the reviewer comment on lightning emissions, now the lack of this emission is stated in the abstract. Page 1 line 7-8: "We note that in this study, we omitted aerosol processes and some natural emissions (lightning and volcanoes emissions)."

Please indicate also in the response what you are going to change in the text. Without this information it is difficult for the reviewer to judge how the actual manuscript has been amended.

Response: An updated version of the "ResponsetoreviewersGMDD-141.pdf" document is provided to the reviewer explicitly indicating the changes introduced in the revised text at the end of each response. For clarity, a bold letter format is used.

No tracked changes were indicated in the reference section but I believe that at least some new references appeared. So please mark them in new response.

Response: It is true that the changes in the reference section were not tracked properly. The new references are there but they are not highlighted. In the updated version of the track changes document, this has been corrected, and now the new references are well identified.

I found that too few aspects of the response were actually included in the revised paper (or it was perhaps not marked as a track change). I would welcome if some of the discussion would appear also in the paper. So please briefly summarize the references on the impact of lightning NO, aerosol impact on photolysis rates, the performance of the COPCAT scheme, the meteorological performance of the model and the emission injection profiles.

Response: Now all these aspects are included in the revised manuscript. Particularly in the revised manuscript, the impact of lightning NO emissions are discussed in Page 12 line 23-27, Page 16 line 13-15, Page 19 line 25-26; the aerosol impact on photolysis rates are discussed in Page 5 line 31-33, Page 9 line 25-27, Page 12 line 27-28, Page 13 line 19-20; the performance of the COPCAT scheme is discussed in Page 7 line 28-29, Page 20 line 28-30; the meteorological performance is discussed in Page 3 line 27-29; and the discussion on the emission injection profiles used is in Page 9 line 21-25.
I asked for a better quantification for statements such as “good agreement”. I found a few examples where this has been done but there are still hardly any quantification in the conclusion. Please include a quantification next to the qualitative statements in the conclusion.

*Response:* Now, the qualitative statements in the conclusions appear with a clear quantification. Additionally, this has been revised through the manuscript, see Page 16 line 8, Page 17 line 15, Page 18 line 29, Page 20 line 16, Page 19 line 33, Page 20 line 25, Page 20 line 28, Page 20 line 30.

I suggested to use volume mixing ratios rather concentrations (microgram/m3) also for the evaluation with surface data. This was done not only for the reasons of consistency. Concentrations depend on density and therefore on the station height. Differences in the model orography and station height have therefore an impact on the evaluation results (beside the scientific problem of the vertical representativeness). This could be avoided by converting the concentrations in volume mixing ratios using the stations measurements of p and T. I recommend using ppb as unit for the comparison.

*Response:* Following the reviewer suggestion, now all the evaluation is done in volume mixing ratios. Figures 3,4,7,8,11 and 12 have been updated presenting the results in volume mixing ratios.

The wording and clarity of the paper has been improved a lot.

*Response:* An important revision of the English has been made.
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.

General remarks:

My two main concerns with the presented model run for 2004 is (i) that NOx 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-daymean 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.

Response: The authors wish to thank anonymous reviewer #2 for the 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”.

We agree with the reviewer that the model run presented in the manuscript has some shortcomings. However, we disagree that they undermine the scientific credibility of the paper. This contribution describes a first major step in the development of a new multiscale chemical weather prediction system and its thorough evaluation, all of which is within the scope of GMD. Improvements to the system are conducted on a regular basis in our group, and the inclusion of NOx emissions from lightning is planned for the next version of the model. The selection of the emissions was an intentional decision.
Not only the thorough evaluation with observations but also the comparison to other scientific studies was our priority. Considering the timeline of the Atmospheric Chemistry and Climate Model Intercomparison Project (ACCMIP) initiative, we made use of the emissions presented in Lamarque et al. (2010). Thus, we took advantage of this excellent opportunity to evaluate the NMMB/BSC-CTM model in a way that can be consistently compared to other modeling systems contributing to ACCMIP. In this sense, emissions representative of the 2000 decade were used both for anthropogenic and biomass burning sources. This decision implied some drawbacks in our work, such as the non specificity of the emissions for year 2004, or the lack of seasonal variability on anthropogenic emissions. We consider those limitations as minor compared to the benefits of comparing our simulations to those of the ACCMIP experiment. The selected year of simulation provides a reference to identify the skills and limitations of the methods applied, but it is not intended to reproduce exactly the same episodes observed during 2004. Our goal is to reproduce the annual trends and patterns of the main atmospheric chemistry components. In the manuscript, all the limitations of our simulations are identified and described. In the revised manuscript version, we have provided additional discussion on these issues along with their implications. For example, the lack of lightning emissions helps explaining the underestimation of OH in the middle to upper troposphere. The results of this evaluation and comparison have allowed us identifying the next steps that will be required to improve the modeling system.

The lack of seasonality in the anthropogenic emissions is considered a minor limitation. Currently, there are still important uncertainties on the estimation of anthropogenic emissions at global scale, and most of the available modeling studies of atmospheric chemistry at global scales run the experiments with constant profiles. This can be explained by the resolution of study, as models are configured at global horizontal resolutions of a few degrees. Under such conditions, the daily variability of the emissions may not be the most important issue to address. Regarding biomass burning emissions, Marlier et al. (2014) showed that going from monthly to daily fire emissions does not change the atmospheric composition very drastically, especially for gases. This supports our initial approach of not using specific daily emissions of biomass burning for 2004.

We believe that the constructive discussion of the model’s behavior in the absence of the aforementioned processes can provide useful insight into the importance of those processes.

This discussion is now introduced in the revised manuscript in Page 12 line 23-27, Page 16 line 13-15, and Page 19 line 25-26.


On the other hand, the presented model had two advanced properties, namely the online calculation of VOC emissions using the MEGAN model and the fact that the presented model is an on-line coupled chemistry – meteorological model (or ChemistryGCM). 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.

Response: This manuscript represents our first step towards the development of a fully coupled chemistry-meteorology model. We intend to present the current state of development of the model (version 1.0) with a comprehensive evaluation using a wide variety of observations. This will serve as a baseline for further studies where more complexity will be added to the system and where specific sensitivity studies may be conducted. Future work will analyze the impact of the online nature of the model, and to assess the sensitivity to the online calculation of biogenic emissions.

Concerning the name of the model (NMMB/BSC-CTM), we have considered the suggestion of the reviewer to not use the term CTM (Chemistry Transport Model) in the model name. CTM has been used traditionally for offline chemistry models, and we agree with the reviewer that using it may lead to some confusion. Considering the nature of our model, a pure online meteorology-chemistry system, we have decided to completely rename the model. The new name is NMMB-MONARCH, where MONARCH stands for "Multiscale Online Nonhydrostatic Atmosphere Chemistry model". We believe that with the new name, all the major characteristics of the system are clearly stated. Now in the revised manuscript all the references to NMMB/BSC-CTM have been replaced with NMMB-MONARCH. We only keep a reference to the old name in the abstract and introduction section to link the previous developments with the new naming adopted. Thus, the title of the manuscript is now "Description and evaluation of the Multiscale Online Nonhydrostatic Atmosphere Chemistry model (NMMB-MONARCH) version 1.0: gas-phase chemistry at global scale".

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.

Response: The results are now better quantified in the revised manuscript, and although expressions as “good agreement” are maintained, they come with a quantification of statistics to support them.

Some places where this has been addressed are Page 16 line 8, Page 17 line 15, Page 18 line 29, Page 20 line 16, Page 19 line 33, Page 20 line 25, Page 20 line 28, Page 20 line 30.

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.

Response: In our case, we attribute the CO overestimation mainly to emission sources. Even, if some other models using similar emissions don’t show this overestimation, we have detected significant sensitivity of the biogenic emissions from MEGAN (as discussed in the manuscript) to the temporal basis of the meteorological conditions. This could affect CO emission overestimation.

On the other hand, the O3 overestimation can be due to multiple factors, such as the lack of halogen and heterogeneous chemistry, an overestimation of the STE for some seasons, the lack of aerosols in the simulation, uncertainties in the solar radiation reproduced by the model, the lack of seasonal variation on anthropogenic emissions, and/or deviations in dry/wet deposition rates.

It is out of the scope of this work to develop specific sensitivity analysis to analyze the influence of each of those factors (as in Strode et al., 2015). Future works will be devoted specifically to these tasks.

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).

Response: The authors agree that the emphasis put during the discussion on the uncertainty of emissions and the lack of aerosols in the previous version of the paper could lead to the misconception that we believe those are the only causes for the disagreements found between model and observations. The revised manuscript has been thoroughly revised to point out other known causes for poor model performance, which have been referenced in all cases. See for example, Page 12 line 26, Page 13 line 21, Page 16 line 14, Page 19 line 1, Page 19 line 25, or Page 19 line 29.

For ozone, other factors contributing to the deviations could be the lack of halogen chemistry in the CB05 mechanism, an excess of STE towards the troposphere, or possible bias in the meteorology (solar radiation, temperature). Sherwen et al. (2016) show that the halogen chemistry reduces the global tropospheric ozone burden by 15%. On the other hand, Real and Sartelet (2011) studied the effect of aerosols on photolysis rates and gaseous species and found 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 are reduced over Europe by around 10 to 14% and their tropospheric burden by around 10%. The decrease in OH leads to an increase of the lifetime of several species such as VOC”. Additionally, 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. These last works, supports the idea introduced in our manuscript that the role of aerosols and photolysis is also important. In the revised manuscript, we have included these possible causes for the ozone bias discussion.

The aerosol impact on photolysis rates are discussed in Page 5 line 31-33, Page 9 line 25-27, Page 12 line 27-28, Page 13 line 19-20.


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.

Response: This manuscript aims to describe the tropospheric gas-phase chemistry of a new modeling system. Although, the comment of the reviewer asking for information about realism of the stratospheric ozone and clouds is relevant, we believe its discussion goes beyond the scope of the paper, and we have decided not to explicitly include it in the revised manuscript. However, we provide below information about the goodness of both simulated fields. In the revised manuscript we introduce the role of STE in the O3 biases.

The stratospheric ozone in the NMMB/BSC-CTM can be computed with linear models, on one hand the COPCAT scheme and on the other, as an alternative, the Cariolle scheme. In the PhD of Dr. Alba Badia (Badia, 2014), both systems were evaluated against satellite data. Relevant biases on the COPCAT stratospheric ozone are:

1) COPCAT underestimates the stratospheric ozone maximum between 50N-20S latitudes and the ozone above 10 hPa. This underestimation is consistent with the study by Monge-Sanz et al. (2011), which compares the annual average from the year 2000 ozone profiles between the COPCAT parameterization and the HALOE measurements in the tropics (4 N).

2) COPCAT also underestimate the ozone in the low stratosphere between 90N-50N during DJF and MAM.

3) Over high southern latitudes COPCAT tends to overestimate O3 concentrations during the months of DJF and MAM in the mid/low stratosphere below 20hPa.

In addition, Figure 1 shows the comparison of the ozone total column computed with Cariolle and COPCAT and with SCIAMACHY satellite retrievals. Additionally, the work of Monge-Sanz et al. (2011) further describes the biases associated to the COPCAT
linear model. It is important to note that COPCAT is used as the linear ozone scheme of the ECMWF global model system IFS.

In the revised manuscript, the reader is referred to Monge-Sanz et al. (2011) for further information on the scheme biases, see Page 7 line 28-29.

Figure 1. Comparison of the ozone total column monthly mean computed with NMMB/BSC-CTM using Cariolle scheme (CAR) and COPCAT scheme (COP), and compared with the SCIAMACHY satellite retrievals.

Regarding the clouds and possible biases. Clouds are simulated by the NMMB meteorological model. The model has been and is being extensively evaluated by NCEP in its both global and limited area configurations. Results of the skills of NMMB at global scale have widely been presented at several international conferences (e.g. AGU, EGU General Assembly). Comparisons of the NMMB initialized with GFS analysis show competitive results of the NMMB compared with GFS. For example, in Figure 2 the anomaly correlation at day 5-forecast shows excellent results with NMMB initialized with GFS or ECMWF analysis. Better results are obtained with ECMWF analysis. Such results wouldn’t be possible with a poor representation of the cloud cover, and provide clear information on the skill of the NMMB as a numerical weather forecast model at global scale.
Figure 2. Anomaly correlation day-5 forecast in the Northern and Southern Hemispheres obtained with the NMMB global model initialized with GFS and ECMWF analysis. Days selected when GFS forecast was poor. (Source: Z. Janjic personal communication).

However, some works have shown minor tropospheric effects of clouds on ozone (e.g., Voulgarakis et al., 2009; Liu et al., 2006). This points to that clouds are unlikely to have a large global effect on tropospheric gas-phase chemistry (only regionally).

In the revised manuscript it is mentioned that the verification of the meteorological model is conducted by NCEP on a regularly basis, Page 3 line 28. Thus, the authors consider that including information about the meteorological performance is beyond the scope of the present work.


Monge-Sanz, B. M., Chipperfield, M. P., Cariolle, D., and Feng, W.: Results from a new linear O3 scheme with embedded heterogeneous chemistry compared with the parent full-chemistry 3-D CTM, Atmos. Chem. Phys., 11, 1227-1242, doi:10.5194/acp-11-1227-2011, 2011.


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).

Response: The authors agree that the injection of emissions in global models is a critical point. However, there is no clear consensus within the modeling community in the approaches to be applied, as can be seen in different works where different approaches are applied from vertically distributing the emissions in height, injecting the emissions in the first model layers, or simply in the first model layer (i.e., Emmons et al., 2010, Huijnen et al., 2010). In our case, we consider that a global model with 1º of horizontal resolution and 64 vertical layers has limitations in the vertical diffusion of pollutants injected in the surface layer due to the lack of detail in the type of landuse or soil properties at specific locations. The coarse resolution strongly affects the surface concentration and we found beneficial to inject the emissions in the PBL for anthropogenic and biomass burning. In the case of anthropogenic emissions, we defined a constant injection height of 500 m that will distribute the emissions within the convective boundary layer during the day, and will trap the emissions in a stable boundary layer during nighttime. With higher resolutions this first approach might be detrimental to the model skills, and a more detailed injection of the emissions will be considered. Regarding biomass burning, the injection within the first 1300 m was considered a good approximation of injecting the emissions within the PBL and is within the range of emission heights recommended by Dentener (2006). Furthermore, some studies recommend an increase in the injection height in the tropics to 2 km based on the evidence from recent satellite observations (e.g. Labonne et al., 2007), which is a height much higher than that selected in our simulations.

The selection of the emission profiles has been indicated in the revised manuscript, see Page 9 line 24-25.


The paper would greatly benefit from proofreading for English language.

**Response:** The final version of the manuscript has been revised in detail.

Specific comments:

P1
Title: consider not calling the model a CTM as CTM’s are understood as “off-line”

**Response:** See the response to the general comment addressed above.

P2
L 27: a better reference for IFS-MOZART is Flemming et al. 2009, GMD

**Response:** Amended.

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

**Response:** Yes, the nonhydrostatic option was turned on in the run although it is not necessary for the global scales used in our study. The extra computational cost of the nonhydrostatic dynamics is on the order of 10% in global applications, or nonexistent if the nonhydrostatic extension is switched off at coarser resolutions. However, the relatively low cost of the nonhydrostatic dynamics allows its application even at transitional resolutions where the benefits due to the nonhydrostatic dynamics are small or uncertain. See Janjic and Gall (2012) for further clarifications.

The meteorological model characteristics are described in Section 2.1 of the revised manuscript.


P3
L 26: better “in detail”

**Response:** Amended.

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.

**Response:** Table 1 presents the specific configuration of the NMMB used in this work. A comment regarding Table 1 is now introduced in the revised manuscript, Page 4 line 25.

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/

**Response:** In this first model version, we followed the approach to prescribe a constant methane concentration, which will be refined in future model versions. 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 the choice in our case was made to include the value the closest to the present time global background concentration (1.83 ppm, see WMO 2015, or Dlugokencky, 2016), because we aim to apply this first version of the NMMB/BSC-CTM in forecast mode. The global average for 2004 is reported to be 0.06 to 0.07 ppm lower, around 4%.

Moreover, the sensitivity of the model results to the background methane concentration is relatively low. We have performed a test using a value of CH4 of 1.786 ppm (reported level by WMO for 2004) and compared it to the simulation with CH4 at 1.85 ppm. Differences in daily mean O3 and CO concentration up to ±0.6 ppb and ±0.12 ppb, respectively, are found for randomly selected days of April and August (Figure 3 depicts the differences for the 5th of August 2004 at surface level). The largest differences for O3 are not at the surface, but at higher layers (plots depict only surface level, but the maximum differences reported consider the full atmospheric column). In relative terms, differences can be considered low.

(a)
Figure 3. Difference in daily mean surface level concentration of (a) CO (ppb), and (b) O3 (ppb) for the 5th of August 2004, between a simulation with background methane concentration set to 1.85 ppm and another with 1.786 ppm.

L 22: Add more information about the realism of the two input fields (overhead ozone and clouds)
Response: This has already been addressed in the reply of a general comment above.

P 6
L 10: Is this a monthly climatology?
Response: No. A specific value for season and landuse is provided.

L 13: “cloud processes” – does this also include wet-phase chemistry?
Response: No. The version 1.0 of the model does not consider aqueous phase chemistry. This is now clarified in the revised manuscript, Page 6 line 19-20.

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?
Response: The terms presented are common in the modeling context and are taken from Byun and Ching (1999) and Foley et al. (2010). The manuscript includes both references. The meaning of the terms used are:
- Grid-scale: it refers to those processes explicitly resolved by the model at the spatial scale of the model resolution.
- Subgrid-scale: it refers to those processes that are parameterized in the physics, i.e., convective clouds.
- Grid-scale Scavenging: it refers to the removal of gases by cloud droplets of clouds explicitly resolved by a numerical model, i.e., stratiform clouds.
- Grid-scale Wet deposition: it refers to the deposition by precipitation produced by explicitly resolved clouds.
- Subgrid-scale vertical mixing: it refers to the convective mixing that is produced within convective clouds.
- Subgrid-scale scavenging: it refers to the removal of gases by cloud droplets of clouds parameterized with a cumulus convection parameterization.
- Subgrid-scale wet deposition for precipitating: it refers to the deposition by precipitation produced by parameterized convective clouds.

The relevant non-precipitating cloud processes are scavenging and vertical mixing. Wet deposition is possible for precipitating clouds. In order to avoid a too long section with common terminology definition the reader is referred to the scientific literature from where this terminology comes from.


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

Response: The most relevant scavenging process affecting gases are the in-cloud scavenging. The American Meteorological Society glossary in its definition of scavenging by precipitation says “Rainout (or snowout), which is the in-cloud capture of particulates as condensation nuclei, is one form of scavenging. The other form is washout, the below-cloud capture of particulates and gaseous pollutants by falling raindrops. Large particles are most efficiently removed by washout. Small particles (especially those less than 1 µm in diameter) more easily follow the airstream flowing around raindrops and generally avoid capture by raindrops except in heavy rain events”. In the case of gases, neglecting below-cloud scavenging is not considered a strong limitation.

L 19: How is the cloud time scale derived?

Response: Following Byun and Ching (1999) and Foley et al. (2010) the time scale of a convective cloud is assumed to be 3600s, and for explicitly resolved clouds, the model times step is applied. See Page 7 line 7.


P7

L2: Do you refer to large-scale and convective precipitation here?
Response: Yes, grid-scale refers to large-scale processes (those resolved by the grid information solved by the model) and subgrid-scale to convective processes.

L7: “Convective mixing” do you mean transport by convective mass fluxes?
Response: Yes, convective mixing is referred to the processes that transport vertically within the convective cloud the air masses.

L 13: 100 hPa is a rather high tropopause for mid- and high latitudes.
Response: There is a common practice in modeling studies to implement stratospheric chemistry above 100 hPa (i.e., Lamarque et al., 2010; Voulgarakis et al., 2011). The chemistry in the tropopause is very slow and the transition within the tropospheric and stratospheric chemistry is wide. Considering the resolution of models in the upper troposphere and stratosphere makes the selection of 100 hPa a compromise solution.


L 13: Were these Mozart 4 fields evaluated for the stratosphere?
Response: Yes. MOZART-4 model is described in Emmons et al., (2010) : “Mixing ratios of several species (O3, NOx, HNO3, N2O5, CO, CH4) are constrained in the stratosphere since MOZART-4 does not have complete stratospheric chemistry. These mixing ratios have been updated to zonal means from a MOZART-3 simulation.” MOZART-3 is suitable for representing chemical/physical processes in stratosphere and for quantifying ozone fluxes from the stratosphere to the troposphere (Kinnison et al., 2007). The validity of STE processes in MOZART-3 has been evaluated in several studies (Park et al., 2004., Pan et al. 2007 and Liu et al., 2009). The model results in Park et al., 2004 showed good agreement for methane and water vapor, but underestimated the nitrogen oxide abundance.

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

**Response:** Following the reviewer’s comment, the description of the COPCAT linear scheme is removed from the manuscript and a reference, Monge-Sanz et al. (2011), is provided in Page 7 line 19.

Monge-Sanz, B. M., Chipperfield, M. P., Cariolle, D., and Feng, W.: Results from a new linear O3 scheme with embedded heterogeneous chemistry compared with the parent full-chemistry 3-D CTM, Atmos. Chem. Phys., 11, 1227-1242, doi:10.5194/acp-11-1227-2011, 2011.

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.

**Response:** As already discussed in the general comments, information on the stratospheric ozone simulated by COPCAT scheme can be found in the PhD of Dr. Alba Badia (Badia, 2014). From there, the most relevant biases on the stratospheric ozone identified are:

1) COPCAT underestimates the stratospheric ozone maximum between 50N-20S latitudes and the ozone above 10 hPa. This underestimation is consistent with the study by Monge-Sanz et al. (2011), which compares the annual average from the year 2000 ozone profiles between the COPCAT parameterization and the HALOE measurements in the tropics (4 N).

2) COPCAT also underestimate the ozone in the low stratosphere between 90N-50N during DJF and MAM.

3) Over high southern latitudes COPCAT tends to overestimate O3 concentrations during the months of DJF and MAM in the mid/low stratosphere below 20hPa.

In addition, Figure 1 shows the comparison of the ozone total column computed with Cariolle and COPCAT and with SCIAMACHY satellite retrievals. Additionally, the work of Monge-Sanz et al. (2011) further describes the biases associated to the COPCAT linear model. It is important to note that COPCAT is used as the linear ozone scheme of the ECMWF global model system.

In the revised manuscript the following sentence has been added “For further description of the approach and information on the biases of the stratosphere ozone simulated by the COPCAT scheme, the reader is referred to Monge-Sanz et al. (2011)”, in Page 7 line 28-29.


Monge-Sanz, B. M., Chipperfield, M. P., Cariolle, D., and Feng, W.: Results from a new linear O3 scheme with embedded heterogeneous chemistry compared with the parent full-chemistry 3-D CTM, Atmos. Chem. Phys., 11, 1227-1242, doi:10.5194/acp-11-1227-2011, 2011.

P 8

L 14: No lightning emissions is a severe shortcoming of the simulation and the paper (see my general comment).
Response: This has been highlighted in the revised manuscript. The following sentence has been included “The omission of lightning emissions is expected to have a significant impact in the oxidation of the middle and upper troposphere” in Section 2.2.6. See also mention in Page 12 line 23-27, Page 16 line 13-15, and Page 19 line 25-26.

L 15: Please explain in more detail how the MEGAN code was integrated in your model.
Response: The authors do not fully understand the comment of the reviewer regarding the coupling of MEGAN code with the NMGB/BSC-CTM. The MEGAN code has been prepared as a subroutine that is called by the chemistry driver of the model to compute an update on the biogenic emissions. The coupling is fully on-line integrated, so the meteorological variables (temperature and solar radiation) is passed as attribute to the subroutine. The authors consider that this explanation is not required in the manuscript.

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.
Response: The MEGAN model uses two different pieces of information regarding meteorological conditions. On one hand the model uses the actual temperature and solar radiation provided by the meteorological model at the required time-step of the simulation. On the other hand, the MEGAN model requires information of the previous day meteorological conditions. For that, several approaches are found in the literature, from implementations providing the average of the previous day temperature and solar radiation to approaches that work with monthly averages. MEGAN shows important sensitivity on these approaches as described in the manuscript: “[...]Marais et al. (2014) performed several sensitivity model runs to study the impact of different model input and settings on isoprene estimates that resulted in differences of up to 17% compared to a baseline”. In our study, weather inputs are based on previous day 24h averages and data for the hour of interest.


P 9
L3: better say “every 720 s”
Response: Amended.

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?
Response: The meteorological model NMGB is initialized with the Final Analysis (FNL) of NCEP. The fields initialized are temperature, winds, specific humidity, and cloud water mixing ratio. The work developed at NCEP shows that the Anomaly Correlation at 500 hPa at 24 h of forecast is nearly 1, see Figure 4. This provides clear evidence of the skill of the model in NWP. The skills are comparable to those of GFS.
Figure 4. Anomaly Correlation of the NMMB (green lines) and GFS (black lines) 9-day forecast of the 500 hPa Height.

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?

Response: The anthropogenic emissions and biomass burning emissions used in our work are those described in Lamarque et al. (2010), to allow an easy intercomparison. An error was introduced in the initial manuscript, because the emissions are not computed for the year 2004; we use the emissions derived by Lamarque et al. (2010) for the 2000. The details on how these emissions were derived can be read in Lamarque reference. Now, in the revised manuscript the sentence “Note that the 2004 emissions are derived from a linear interpolation between years 2000 and 2010” has been changed to “Note that this methodology involves assuming 2004 emissions equivalent to the best estimate reported by ACCMIP for year 2000” in Page 9 line 6. The biomass burning emissions provided by Lamarque et al. (2010) have a seasonal cycle, while anthropogenic emissions are constant throughout the year.


L 26: see my general comment, please justify the choices
Response: The injection height of the different emissions used in our model are derived from sensitivity runs. Please, see previous discussion in the general comments about the justification of the emission heights.

L 29: Please provide reference the strong impact of aerosol on the photolysis rates.
Response: Real and Sartelet (2011 ACP) studied the effect of aerosols in the photolysis rates and gaseous species. They state 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 are reduced over Europe by around 10 to 14% and their tropospheric burden by around 10%. The decrease in OH leads 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. We have included these references in the revised manuscript, Page 12 line 28.


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.
Response: If missing data are found in the observations, the corresponding time period is removed from the model data. Following this approach, model and observation derived averages are fully comparable (as only matching periods are used for the evaluation). This is clarified in the manuscript as follows: At the surface-level, daily O3 averages are computed from 3-hourly values of model and observations, applying a filter to model data whenever observations are missing, so as to consider timely/collocated values (See Page 11 line 21-23). Section 1 of the supplementary material presents the statistical measures calculated from the daily 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)
Response: The altitude 1000 m asl. has been shown as a clear transition from a boundary-layer to a free-tropospheric regime for ozone (Chevalier et al., 2007). We are interested in filtering mountain stations because they provide measurements that are normally representative of the local conditions, but not of the air parcel that would be included on a model grid cell, of 1.4°x1°. Applying this threshold for station filtering is a common practice for several works devoted to atmospheric chemistry-transport model evaluation (Solazzo et al., 2012; Solazzo and Galmarini 2016).


P 12

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

Response: We agree with the reviewer’s comment. However, one has to bear in mind that NOx concentrations in the tropopause region are very low, as the stratospheric NOx layer is located considerably higher in the stratosphere, and pollution NOx is mostly located closer to the surface. Therefore the tropospheric column does not significantly depend on the choice of tropopause level. In our work, we have considered the tropopause definition as the model level interface corresponding to approximately 100 hPa in the tropics and 250 hPa in the extratropics following Horowitz et al. (2003). In this sense, we don’t think that this inconsistency has a real impact in the results discussed.


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.

Response: The authors agree with the reviewer comment. In this sense, the sentence of L29 has been reformulated in the revised manuscript in the following way: “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), Page 12 line 26-28.

P 13

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

Response: CH4 is an hydrocarbon, so as stated in the manuscript, the photolysis of hydrocarbons are a source of CO. For clarification, “including methane” is now written in the revised manuscript, Page 12 line 32.
L 14 add reference for C-IFS
Response: Amended.

L 29: Could the high methane be a reason?
Response: The influence of CH$_4$ on modeled CO levels has been assessed through a sensitivity test, which resulted in a low impact (please, see our previous response). Changing the CH$_4$ prescribed value from 1.85 ppm to 1.78 ppm (2004 global average as reported by WMO) leads 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).


L 30: Figure 3. Please show separate plots for NH, SH mid-latitudes and tropics. The seasonality is obscured by averaging over all stations.
Response: We have only 14 stations available for CO, and only 2 of them are located in the SH. For that reason, we decided to average over all stations. The seasonality for the CO is represented in Figure 6.

P 14
L2: no seasonality of the anthropogenic emissions is an oversimplification.
Response: This limitation has been highlighted in the revised manuscript, Page 15 line 26-27.

L3: Figure 3 shows the relative bias (%), not MB as defined in the supplement.
Response: Figure 3 shows the MB, not the relative bias (%). This information is now corrected in the revised manuscript.

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.
Response: Time scale is described in the Model evaluation section: “For the evaluation of daily surface-level O3, we considered averages of temporally collocated 3-hourly values from the model and the observations. Section 1 of the supplementary material presents the statistical measures calculated from the daily data”, Page 11 line 21-23. We didn’t filter out seasonality.

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

P 15
L8: What do you mean by overestimated emissions above the PBL ?
Response: As we described in the document, all the land-based anthropogenic emissions are emitted in the first 500 m of the model and biomass burning emissions from forests in the first 1300 m. When the PBL is lower than this altitude, emissions
above the PBL can be overestimated and contribute to the positive bias identified in the CO.

L 8: Please discuss the role of convection
Response: The following sentence has been included in the revised manuscript: “Excessive vertical mixing by moist convection may explain the overestimation in the tropics”, Page 15 line 2-3.

L 32: What regime (rural, urban) was used?
Response: Only rural stations were used in this study. This information is now included in the revised manuscript.

P 16
L 8: Please discuss also PBL mixing during the night
Response: We have included the following sentence in the revised manuscript: “Also, excessive mixing within the PBL during the night could contribute to a decrease in ozone titration by NO and partially explain the bias”, Page 16 line 1-2.

P 17
L 8: see my general comment on the use of “good agreement”
Response: More quantitative statements have been included in the revised manuscript supporting the qualitative statements initially used.

L 17: Please mention the value of the biases.
Response: The average wet deposition rates for the model and the observations are shown in Fig S4 (supporting material). Wet deposition MB for Europe, USA and Asia are -200.70 mg N/m², -36.87 mg N/m² and -163.27 mg N/m², respectively. These biases are now mentioned in the revised manuscript.

L 26: What is the seasonal cycle of the biomass emissions?
Response: The ACCMIP inventory has monthly variations for biomass burning. This is described in Lamarque et al. (2010).


P 18
L 1 “rural” (?) perhaps better remote
Response: Amended.

L 4: dominated by the tropics - perhaps simply because they are the largest region on earth (?)
Response: This point is now included in the revised manuscript.

L 7: TM5 has a similar chemical mechanism. Should it not be similar?
Response: NMMB/BSC-CTM and TM5 have a similar chemical mechanism. However, other processes that are represented differently in both models, such as the deposition, vertical mixing and emissions, also have an impact on the O3 burden. These processes can explain the differences in the O3 burdens between the models.

L 16: Please clarify how the STE is calculated in your model.  
Response: The stratosphere-troposphere ozone exchange flux is calculated as the annual balance of the ozone mass crossing the 100 hPa height. This approach is accurate on the global and long-term average (Hsu et al., 2005). However, there are considerable differences among the models for calculating or specifying the stratosphere-troposphere exchange of ozone. This is now indicated in the revised manuscript, Page 17 line 34.


L 27: "all day long"? Do you mean "throughout the year"  
Response: Yes. The expression has been changed to "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.  
Response: Amended.

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.  
Response: The authors agree with the reviewer’s comment. In the revised manuscript this possible explanation of the overestimation of O3 is included.

P 19  
L4 Please quantify biases, what do you mean by “error”.  
Response: In the revised manuscript the biases are now quantified and the sentence “error” has been substituted by “root mean square errors” to clarify the text. The quantification of the biases is provided in Figure 12.

L 15. This points to biases of the COPCAT ozone, which has consequences for the photolysis rates.  
Response: The authors agree with the reviewer on the possible effect of the COPCAT biases upon the tropospheric ozone. This has been introduced in the revised manuscript.

P 20  
L 8: The lack of aerosol modulation of photolysis is a 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.  
Response: The authors consider that the effect of aerosols on photolysis and radiation is not a minor aspect, as the results in Real and Sartelet (2011 ACP) indicate: “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 are reduced over Europe by around 10 to 14% and their tropospheric burden by around 10%. The decrease in OH leads to an increase of the lifetime of several species such as VOC. Furthermore, 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. We have included these references in the revised manuscript.

Following the reviewer’s suggestion now the main shortcomings of the simulation presented in the manuscript are clearly stated. The following sentence has been added in the conclusions section: “We note that in this contribution, we omitted aerosols and lightning emissions; anthropogenic emissions disregard seasonality; and biomass burning emissions are not specific to 2004”, Page 19 line 25.


L15: The paper provides no evidence for this claim - it can therefore not be a conclusion.
Response: The authors do not present as a conclusion the effect of aerosols on the concentrations of OH. We only suggest some possible reasons to explain why our results of OH at northern latitudes are slightly higher than the climatological mean of Spivakovsky et al. (2000). We agree with the reviewer that this cannot be a conclusion of the work, in this sense, we only highlight a possible explanation that deserves further research in the future studies with the model.

L 34 see above, no evidence in the paper
Response: Addressing the reviewer’s comment, the following statement has been removed from the revised manuscript: “CO production from VOCs biogenic emissions, calculated online and depending on meteorological variables such as radiation, might be overestimated too, due to the lack of aerosol attenuation of radiation”.

P 21
L 13 better “megacities”
Response: Amended.

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.
Response: We have only 14 stations available for CO, and only 2 of them are located in the SH. For that reason, we decided to average over all stations. The seasonality for
the CO is represented in Figure 6. Volume mixing ration has been used instead of concentration.

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.
Response: In the document the MB is calculated, not the relative bias (%). This information is now corrected in the revised manuscript. Surface observations are shown in ppb now.

Figure 7: better y-range, use ppb
Response: Surface observations are shown in ppb now.

Figure 8: as for Figure 4
Response: In the document the MB is calculated, not the relative bias (%). This information is now corrected in the revised manuscript.

Figure 11: use ppb
Response: Surface observations are shown in ppb now.

Figure 12: see Figure 4
Response: In the document the MB is calculated, not the relative bias (%). This information is now corrected in the revised manuscript.

Figure 13: choose x-range 0-100 for better clarity in the troposphere.
Response: The x-range 0-100 has been chosen to include both the tropopause and the lower stratosphere.

Figure 14: see Figure 4
Response: In the document, the MB is calculated, not the relative bias (%). This information is now corrected in the revised manuscript.
Gas-phase chemistry in the online multiscale NMMB/BSC Chemical Transport Model: Description and evaluation of the Multiscale Online Nonhydrostatic Atmosphere Chemistry model (NMMB-MONARCH) version 1.0: gas-phase chemistry at global scale

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Abstract.

This paper presents a comprehensive description and benchmark evaluation of the tropospheric gas-phase chemistry component of the Multiscale Online Nonhydrostatic Atmosphere Chemistry model (NMMB-MONARCH), formerly known as NMMB/BSC Chemical Transport Model (NMMB/BSC-CTM), an online chemical weather prediction system conceived for both the regional and the global scale. We can be run on both regional and global domains. Here, we provide an extensive evaluation of a global annual cycle simulation using a variety of background surface stations (EMEP, WDCGG and CASTNET), ozonesondes (WOUDC, CMD and SHADOZ), aircraft data (MOZAIC and several campaigns), and satellite observations (SCIAMACHY and MOPITT). We also include an extensive discussion of our results in comparison to other state-of-the-art models. We note that in this study, we omitted aerosol processes and some natural emissions (lightning and volcanoes emissions).

The model shows a realistic oxidative capacity across the globe. The seasonal cycle for CO is fairly well represented at different locations (correlations around 0.3-0.7 in surface concentrations), although concentrations are underestimated in spring and winter in the Northern Hemisphere, and are overestimated throughout the year at 800 and 500 hPa in the Southern Hemisphere.
Nitrogen species are well represented in almost all locations, particularly NO$_2$ in Europe (RMSE below 9 $\mu$g m$^{-3}$ 5 ppb). The modeled vertical distribution of NO$_x$ and HNO$_3$ are in excellent agreement with the observed values and the spatial and seasonal trends of tropospheric NO$_2$ columns correspond well to observations from SCIAMACHY, capturing the highly polluted areas and the biomass burning cycle throughout the year. Over Asia, the model underestimates NO$_x$ from March to August probably due to an underestimation of NO$_x$ emissions in the region. Overall, the comparison of the modeled CO and NO$_2$ with MOPITT and SCIAMACHY observations emphasizes the need for more accurate emission rates from anthropogenic and biomass burning sources (i.e., specification of temporal variability).

The resulting ozone (O$_3$) burden (348 Tg) lies within the range of other state-of-the-art global atmospheric chemistry models. The model generally captures the spatial and seasonal trends of background surface O$_3$ and its vertical distribution. However, the model tends to overestimate O$_3$ throughout the troposphere in several stations. This may be attributed to an overestimation of CO concentration over the Southern Hemisphere leading to an excessive production of O$_3$ or to the lack of specific chemistry (e.g., halogen chemistry, aerosol chemistry). Overall, O$_3$ correlations range between 0.6 to 0.8 for daily mean values. The overall performance of the NMMB/BSC-CTM-NMFB-MONARCH is comparable to that of other state-of-the-art global chemical transport models.

1 Introduction

Tropospheric ozone (O$_3$) is a radiatively active gas interacting with solar and terrestrial radiation that is mainly produced during the photochemical oxidation of methane (CH$_4$), carbon monoxide (CO) and non-methane volatile organic compounds (NMVOC) in the presence of nitrogen oxides (NO$_x$) (Crutzen, 1974; Derwent et al., 1996). Downward transport from the stratosphere, where O$_3$ is created by photolysis of oxygen (O$_2$) molecules, is also an important source of tropospheric O$_3$ (Stohl et al., 2003; Hsu and Prather, 2009). In urban areas, O$_3$ is a major component of ‘smog’, which can cause a number of respiratory health effects (WHO, 2014). Since the pre-industrial era, changes in emissions of O$_3$ precursors from anthropogenic and biomass burning sources have modified the distribution of tropospheric O$_3$ and other trace gases (Lamarque et al., 2013). Tropospheric O$_3$, with an average lifetime of the order of weeks, is highly variable in space and time, and Air Quality Models (AQM) are required to predict harmful levels of O$_3$ along with its precursors and other trace gases.

AQM are driven by meteorological fields and fed by emission inventories of chemical species. They include a chemical mechanism for representing gas-phase and aerosol atmospheric chemistry, a photolysis scheme describing the photo-dissociation reactions driven by sunlight, dry and wet deposition schemes to account for the removal of pollutants from the atmosphere, and the characterization of the downward transport of stratospheric O$_3$. The development of AQMs and meteorological models (MM) evolved as separate fields (offline approach) due to complexity and limitations in computer resources.

The offline approach requires lower computational capacity, but involves a loss of essential information on atmospheric processes whose time-scale is smaller than the output time rate of the meteorological model (Baklanov et al., 2014). Nowadays, owing to a general increase in computer capacity, online coupled meteorology-chemistry models are being increasingly devel-
oped and used by the scientific community, who recognizes the advantages of the online approach (Byun, 1990). Overviews of online AQM-MM models are available in the literature (Zhang, 2008; Baklanov et al., 2014).

Several global AQMs have been developed during the last decades: online, including the multiscale GEM-AQ (online, 1.5° x 1.5°) (Gong et al., 2012), offline-TM5-chem-v3.0 (offline, 3° x 2°) (Huijnen et al., 2010), online-LMDZ-INCA (offline, 3.8° x 2.5°) (Folberth et al., 2006), online-the GATOR-GCMM (online, 4° x 5°) (Jacobson, 2001), online-the IFS-MOZART used in the MACC project (80km x 80km), online, 80 km x 80 km (Flemming et al., 2009), C-IFS recently developed at ECMWF (80km x 80km, online, 80 km x 80 km) (Flemming et al., 2015), and offline-MOZART-4 (offline, 2.8° x 2.8°) (Emmons et al., 2010). Most of these models have been applied at coarse resolutions with simplified chemical schemes. Currently, the systems are being updated and prepared for higher resolution applications.

In this contribution, we describe and evaluate the gas-phase chemistry of the NMMB/BSC-Chemical Transport Model (NMMB/BSC-CTM), an online multi-scale non-hydrostatic Multiscale Online Nonhydrostatic Atmosphere Chemistry model (NMMB-MONARCH), a chemical weather prediction system formerly known as NMMB/BSC-CTM that can be run either globally or regionally (Pérez et al., 2011; Jorba et al., 2012). The NMMB-MONARCH, developed at the Barcelona Supercomputing Center, is based on the coupling of the meteorological Nonhydrostatic Multiscale Model on the B-grid (NMMB; Janjic and Gall, 2012) with a chemistry module. We provide a thorough evaluation of the gas-phase chemistry over a one-year period for the global domain using an horizontal resolution of 41.4° x 41.4°.

The NMMB/BSC-CTM model, NMMB-MONARCH configured as a limited area model, (regional) model has recently participated in the Air Quality Model Evaluation International Initiative Phase2 (AQMEII-Phase2) intercomparison exercise - A spatial, temporal and vertical (Im et al., 2014), Badia and Jorba (2014) also provided a detailed evaluation of the chemical model results - gas-phase chemistry for the year 2010 on a regional scale are presented in. Moreover, a comparison between other modeling systems currently applied in Europe and North America over Europe in the context of AQMEI phase 2 is presented in. Evaluations of previous version of the model include the dust implementation, presented in and, and the sea-salt aerosol module, described and evaluated at the global scale in and. The aerosol module for other relevant global aerosols (natural, anthropogenic and secondary) is currently under development within the NMMB/BSC-CTM-AQMEII-Phase2. The initial model developments focused on the implementation of the mineral dust aerosol component (NMMB/BSC-Dust; Pérez et al., 2011; Haustein et al., 2012) and the sea-salt aerosol component (Spada et al., 2013, 2015). The implementation and evaluation of other relevant aerosols will be soon described elsewhere (Spada et al., in prep). This initiative aims at developing a fully coupled chemical multiscale (global/regional) weather prediction system that resolves gas-aerosol-meteorology interactions and provides initial and boundary conditions for embedded high resolution nests in a unified dynamics-physics-chemistry environment.

The focus of this paper is to describe and evaluate the global atmospheric model NMMB/BSC-CTM in terms of the spatial distribution and seasonal variations of O3 and its precursors paper is organized as follows. In Sec. 2, we provide a description of the atmospheric driver, the gas-phase chemistry module, and the model configuration including the online biogenic emissions. Section 3 presents an overview of the model setup with a description of the chemical and meteorological initial conditions, and the anthropogenic and biomass burning emissions implemented for this experiment. We illustrate the capabil-
ity of the NMMB/BSC-CTM-NMMB-MONARCH to reproduce the main reactions occurring in the atmosphere atmospheric composition by evaluating the model with ground-based monitoring stations, ozonesondes, aircraft data, climatological vertical profiles and satellite retrievals, which are described in Sec. 4. The results of the model performance are discussed in Sec. 5 for year 2004. The last section summarizes the conclusions of this work.

5 2 Model description

The NMMB/BSC-CTM-NMMB-MONARCH is a fully online multiscale chemical transport model weather prediction system for regional and global-scale applications (Pérez et al., 2011; Jorba et al., 2012). The system is based on the meteorological Non-hydrostatic–Nonhydrostatic Multiscale Model on the B-grid (NMMB; Janjic and Gall, 2012), developed and widely verified at the National Centers for Environmental Prediction (NCEP). The model couples online the NMMB with the gas-phase and aerosol continuity equations to solve the atmospheric chemistry processes with detail. Due to its online-coupling approach, the model accounts for the feedback processes of in detail. The model is designed to account for the feedbacks among gases, aerosol particles and radiation meteorology. Currently, it can consider the direct radiative effect of aerosols while presently, while ignoring cloud-aerosol interactions. In the present this work, only the gas-phase chemistry is used, thus no interaction between gas-phase and aerosol-phase is applied. In this section we provide a concise description of the NMMB model and the gas-phase chemistry module of the BSC-CTM-NMMB-MONARCH.

2.1 The Non-hydrostatic–Nonhydrostatic Multiscale Model on the B-grid

The Non-hydrostatic–Nonhydrostatic Multiscale Model on the B-grid (NMMB; Janjic and Black, 2005; Janjic and Gall, 2012) was conceived for short- and medium-range forecasting over a wide range of spatial and temporal scales, from large eddy simulations (LES) to global simulations. Its unified non-hydrostatic nonhydrostatic dynamical core allows regional and for running either regional or global simulations, both including embedded regional nests. The NMMB has been developed within the Earth System Modeling Framework (ESMF) at NCEP, following the general modeling philosophy of the NCEP regional Weather Research and Forecasting (WRF) Non-hydrostatic–Nonhydrostatic Mesoscale Model (NMM; Janjic et al., 2001; Janjic, 2003). The regional NMMB has been the operational regional North American Mesoscale (NAM) model at NCEP since October 2011. The numerical schemes used in the model were designed following the principles presented in Janjic (1977, 1979, 1984, 2003). Isotropic horizontal finite volume differencing is employed so a variety of basic and derived dynamical and quadratic quantities are conserved. Among these, the conservation of energy and entrophy enstrophy (Arakawa, 1966) improves the accuracy of the nonlinear dynamics. The hybrid pressure-sigma coordinate is used in the vertical direction and the Arakawa B-grid is applied in the horizontal direction. The global model on the latitude-longitude grid with polar filtering was developed as the reference version, and other geometries such as the cubed-sphere are currently being tested. The regional model is formulated on a rotated longitude-latitude grid, with the Equator of the rotated system running through the middle of the integration domain resulting in more uniform grid distances. The non-hydrostatic nonhydrostatic component of the model dynamics is introduced through an add-on module that can be turned on or off, depending on the resolution. The
operational physical package includes: (1) the Mellor-Yamada-Janjic (MYJ) level 2.5 turbulence closure for the treatment of turbulence in the planetary boundary layer (PBL) and in the free atmosphere (Janjic et al., 2001), (2) the surface layer scheme based on the Monin-Obukhov similarity theory (Monin and Obukhov, 1954) with introduced viscous sublayer over land and water (Zilitinkevich, 1965; Janjic, 1994), (3) the NCEP NOAH (Ek et al., 2003) or the LISS land surface model (Vukovic et al., 2010) for the computation of the heat and moisture surface fluxes, (4) the GFDL or RRTMG long-wave and shortwave radiation package (Fels and Schwarzkopf, 1975; Mlawer et al., 1997), (5) the Ferrier gridscale clouds and microphysics (Ferrier et al., 2002), and (6) the Betts-Miller-Janjic convective parametrization (Betts, 1986; Betts and Miller, 1986; Janjic, 1994, 2000). Vertical diffusion is handled by the surface layer scheme and by the PBL scheme. Lateral diffusion is formulated following the Smagorinsky non-linear approach (Janjic, 1990). Table 1 describes the main configuration of the meteorological model used in this work.

2.2 Gas-phase chemistry module

The tropospheric gas-phase chemistry module is coupled online within the NMMB code. Different chemical processes were implemented following a modular operator splitting approach to solve the advection, diffusion, chemistry, dry and wet deposition, and emission processes. Meteorological information is available at each time step to solve the chemical processes properly. In order to maintain consistency with the meteorological solver, the chemical species are advected and mixed at the corresponding time step of the meteorological tracers using the same numerical schemes implemented in the NMMB. The advection scheme is Eulerian, positive definite and monotone, maintaining a consistent mass-conservation of the chemical species within the domain of study (Janjic et al., 2009; Tang et al., 2009; Janjic and Gall, 2012).

2.2.1 Chemical-phase reaction mechanism

Several chemical mechanisms can be implemented within the NMMB/BSC-CTMNMMB-MONARCH. A modular coupling with the Kinetic PreProcessor (KPP) package (Damian et al., 2002; Sandu and Sander, 2006) allows the model to maintain wide flexibility. Additionally, an Eulerian-Backward-Iterative solver (Hertel et al., 1993) was implemented as a complementary option to the KPP solvers to allow the model to run with a fast ordinary differential equation solver at global scales. For the present study, we use a Carbon-Bond family mechanism, the Carbon Bond 2005 (CB05; Yarwood et al., 2005), an updated version of the Carbon-Bond IV (CB4) lumped-structure-type mechanism (Gery et al., 1989). CB4 was formulated focusing on limited domain extent, urban and regional environments and for planetary boundary layer chemistry. CB05 extends its applicability from urban to remote tropospheric conditions and is suitable for global applications. CB05 was evaluated against smog chamber data from the University of California, Riverside and University of North Carolina (Yarwood et al., 2005). It includes 51 chemical species and solves 156 reactions (see Tables S1 and S2 in the supplementary information). Both the organic chemistry of methane and ethane, and the chemistry of methylperoxy radical, methyl hydroperoxide and formic acid are treated explicitly. The higher organic peroxides, organic acids, and peracids are treated as lumped species. Following its main design, CB05 defines proxy single and double carbon bond species, paraffin and an olefin bond respectively, and it introduces the internal olefin species. The rate constants were updated based on evaluations from Atkinson et al. (2004) and Sander et al.
Organic compounds not explicitly treated are apportioned to the carbon-bond species based on the molecular structure and following Yarwood et al. (2005) assignments from VOC species to CB05 model species. The concentration of methane is considered constant (1.85 ppm) in this study.

### 2.2.2 Photolysis scheme

One of the most important processes determining tropospheric composition is the photo-dissociation of trace gases. Table S2 in the supplementary information lists the photolysis reactions considered. To compute the photolysis rates, we implemented the Fast-J (Wild et al., 2000) online photolysis scheme. Fast-J has been coupled with the physics of each model layer (e.g., clouds and absorbers such as O$_3$). The optical depths of grid-scale clouds from the atmospheric driver are considered by using the fractional cloudiness based on relative humidity (Fast et al., 2006). The main advantages of Fast-J are the optimization of the phase function expansion into Legendre polynomials and the optimization of the integration over wavelength (Wild et al., 2000). The Fast-J scheme has been upgraded with CB05 photolytic reactions. The quantum yields and cross section for the CB05 photolysis reactions have been revised and updated following the recommendations of Atkinson et al. (2004) and Sander et al. (2006). The Fast-J scheme uses seven different wavelength bins appropriate for the troposphere to calculate the actinic flux covering from 289 to 850 nm (see Table VIII from Wild et al. (2000)). In this work, aerosols are not considered in the photolysis rate calculation. This might produce an atmosphere excessively oxidized in regions where aerosols are significant (e.g., Bian et al., 2003; Real and Sartelet, 2011).

### 2.2.3 Dry-deposition scheme

The dry-deposition scheme is responsible for computing the flux of trace gases from the atmosphere to the surface. It is calculated by multiplying the concentration in the lowest model layer by the spatially and temporally varying deposition velocity:

$$\frac{\partial C_i}{\partial t}_{\text{dry-dep}} = -C_i v_d$$

where $t$ is the time, $i$ the gas-phase species, $C_i$ is the concentration of the gas in the lowest model layer, and $v_d$ is the dry-deposition velocity. At each time step, $v_d$ is calculated according to:

$$|v_d| = \frac{1}{(R_a + R_b + R_c)}$$

where $R_a$ is the aerodynamic resistance (depends only on atmospheric conditions), $R_b$ is the quasilaminar sublayer resistance (depends on friction velocity and molecular characteristics of gases), and $R_c$ is the canopy or surface resistance (depends on surface properties and the reactivity of the gas). $R_a$ and $R_b$ are computed following their common definition (Seinfeld and Pandis, 1998), while $R_c$ is simulated following Wesely (1989), where the surface resistance is derived from the resistances of the surfaces of the soil and the plants. The properties of the plants are determined using land-use data (from the meteorological
driver USGS land-use) and depend on the season. The surface resistance also depends on the diffusion coefficient, the reactivity, and water solubility of the reactive trace gases.

### 2.2.4 Wet-deposition scheme

We use the scheme of Byun and Ching (1999) and Foley et al. (2010) to resolve the cloud processes affecting the concentration of 36 gases from the CB05 chemical mechanism. The processes included are grid-scale scavenging and wet-deposition, subgrid-scale vertical mixing, and scavenging and wet-deposition for precipitating and non-precipitating clouds. **Aqueous chemistry is neglected in version 1.0 of the model.** At the moment, we consider only in-cloud scavenging, which is computed using the Henry’s Law equilibrium equation. The rate of change for in-cloud pollutant concentration is given by:

$$\frac{\partial C_{icld}}{\partial t} = C_{icld} \frac{e^{-\alpha_i \tau_{cld}} - 1}{\tau_{cld}}$$  \hspace{1cm} (3)

where $C_{icld}$ is the gas concentration within the cloud [ppm], $\tau_{cld}$ is the cloud timescale [s], and $\alpha_i$ is the scavenging coefficient for the gas species that is calculated as:

$$\alpha_i = \frac{1}{\tau_{washout}(1 + \frac{TWF}{H_i})},$$  \hspace{1cm} (4)

where $H_i$ is the Henry’s Law coefficient for the gas species [M/atm], $TWF = \rho_{H_2O}/(W_T RT)$ is the total water fraction (where $\rho_{H_2O}$ is the density of water [kg/m$^3$], $W_T$ is the total mean water content [kg/m$^3$], $R$ is the Universal gas constant, and $T$ is the in-cloud air temperature [K]), and $\tau_{washout}$ is the washout time [s], i.e., the amount of time required to remove all of the water from the cloud volume at a specified precipitation rate [m/s], which is given by:

$$\tau_{washout} = \frac{W_T \Delta Z_{cld}}{\rho_{H_2O} P_r}$$  \hspace{1cm} (5)

where $\Delta Z_{cld}$ is the cloud thickness [m] and $P_r$ is the precipitation rate [m/s]. Both grid-scale and subgrid-scale scavenging are computed with equation 3, where $\tau_{cld}$ is 1 hour for subgrid-scale clouds, and the chemistry timestep for grid-scale clouds.

Wet deposition is computed following the algorithm of Chang et al. (1987), which depends upon $P_r$ and the gas concentration within the cloud $C_{icld}$. Thus, the wet deposition is given by:

$$wdep_i = \int_0^{\tau_{cld}} C_{icld} P_r \, dt$$  \hspace{1cm} (6)

The sub-grid cloud scheme implemented solves convective mixing, scavenging and wet deposition of a representative cloud within the grid cell following the CMAQ and RADMv2.6 model schemes (Byun and Ching, 1999; Chang et al., 1987). Precipitating and non-precipitating sub-grid clouds are considered. The latter are categorized as pure fair weather clouds and non-precipitating clouds and may coexist with precipitating clouds (Byun and Ching, 1999; Foley et al., 2010).
2.2.5 Upper boundary conditions

Because the model focuses on the troposphere, stratospheric chemistry is taken into account using a simplified approach. Above 100 hPa, mixing ratios of several species (NO, NO\(_2\), N\(_2\)O\(_5\), HNO\(_3\) and CO) are initialized each day from a global chemical model MOZART-4 (Emmons et al., 2010). For O\(_3\), an important reactive gas requiring a more refined representation in the stratosphere, we use a linear O\(_3\) stratospheric scheme, COPCAT (Monge-Sanz et al., 2011). COPCAT is based on the approach of Cariolle and Déqué (1986), which represented the first effort to include a linearized O\(_3\) scheme (named Cariolle v1.0) in a three-dimensional model.

Following the COPCAT approach, the change in O\(_3\) with time due to local chemistry is given by:

\[
\frac{\partial C_{O_3}}{\partial t} = \frac{d\chi}{dt} = (P - L)\chi, T, \Phi
\]

where \((P - L)\) represents the O\(_3\) tendency as a linear function depending on \(\chi\), the O\(_3\) mass mixing ratio \((kgkg^{-1})\), \(T\), the temperature \((K)\), and \(\Phi\), the column O\(_3\) above the point under consideration \((kgm^{-2})\).

Equation (??) is expanded to first order in a Taylor Series as follows:

\[
\frac{\partial C_{O_3}}{\partial t} = \frac{d\chi}{dt} = (P - L)_0 + a + \frac{b + c}{d}
\]

The second term in the expansion represents variations in the local O\(_3\) amount \((a)\), the third represents temperature effects \((b)\) and the last term, called radiation term, accounts for the influence of non-local O\(_3\) on the amount of solar radiation reaching the level under consideration \((c)\). Specific terms in this equation (represented with the subscript 0) are coefficients applicable at the equilibrium state. In COPCAT these coefficients are obtained at equilibrium from the TOMCAT/SLIMCAT box model (Chipperfield, 2006). These terms are presented as functions of 24 latitudes, 24 model vertical levels and 12 months.

Heterogeneous processes describing the polar stratospheric chemistry are non-linear and depend on the three-dimensional structure of the atmosphere. COPCAT includes complete heterogeneous processes in their coefficients, considering heterogeneous and gas-phase chemistry to be consistent when applied in this linear O\(_3\) parameterization. This kind of parameterization is in better agreement with the current state of knowledge of stratospheric heterogeneous chemistry than previous schemes (Monge-Sanz et al., 2011). For further description of the approach and information on the biases of the stratosphere ozone simulated by the COPCAT scheme, the reader is referred to Monge-Sanz et al. (2011).
2.2.6 Online natural emissions

Natural emissions of gaseous pollutants include biogenic emissions, soil emissions, emissions from lightning, and emissions from oceans and volcanoes. Currently, soil and oceanic emissions in the model are prescribed as described in Sect. 3.1 and emission from lightning and volcanoes are not considered. The omission of lightning emissions may have a significant impact in the oxidation of the middle and upper troposphere. Only biogenic emissions, which strongly depend on meteorological fields and vegetation cover, are calculated online. They are computed using the Model of Emissions of Gases and Aerosols from Nature version 2.04 (MEGANv2; Guenther et al., 2006). MEGAN is able to estimate the net emission rate of gases and aerosols from terrestrial ecosystems into the above-canopy atmosphere. MEGAN canopy-scale emission factors differ from most other biogenic emission models, which use leaf-scale emission factors, and cover more than 130 Non-Methane Volatile Organic Compounds (NMVOCs). All the MEGAN NMVOCs are speciated following the CB05 chemical mechanism; thus, emissions for isoprene, lumped terpenes, methanol, nitrogen monoxide, acetaldehyde, ethanol, formaldehyde, higher aldehydes, toluene, carbon monoxide, ethane, ethene, paraffin carbon bond, and olefin carbon bond are considered within the model. Biogenic emissions are computed every hour to account for evolving meteorological changes in solar radiation and surface temperature. Thus, weather- driving variables considered are temperature at 2 m and incoming short wave radiation at the surface.

Figure S1 in the supplementary information shows the modeled emission for isoprene and terpenes for January and July 2004, and Table 2 lists the global annual emissions for isoprene, monoterpenes and other important NMVOCs. Biogenic isoprene emissions used in this study amount 683.16 Tg/year. While other global models have lower estimates (Huijnen et al., 2010; Horowitz et al., 2003; Emmons et al., 2010), MEGAN isoprene emissions typically range from about 500 to 750 Tg/year (Guenther et al., 2006). These estimates largely depend on the assumed land cover, emission factors, and meteorological parameters. Therefore, the emission uncertainties and their impacts upon surface \(O_3\) are associated with uncertainties in these inputs. Ashworth et al. (2010) obtained emission reductions of 3% and 7% when using daily and monthly meteorological data, respectively, instead of hourly data, with reductions reaching up to 55% in some locations. Marais et al. (2014) performed several sensitivity model runs to study the impact of different model input and settings on isoprene estimates that resulted in differences of up to ± 17% compared to a baseline. In our study, weather inputs are based on previous day 24h averages and data of the hour of interest.

3 Model setup

The model is set up as global with a horizontal grid spacing of \(4.4^\circ \times \frac{1}{2} \times 1.4^\circ \times 1^\circ\) and 64 vertical layers up to 1 hPa. The depth of the bottom layer is below 40 m. The dynamics fundamental time step is set to 180s and the chemistry processes are solved every 4 fundamental time steps 720s. The radiation, photolysis scheme and biogenic emissions are computed every hour. We use NCEP/Final Analyses (FNL) as initial conditions for the meteorological driver, and we reinitialize the meteorology every 24 h to reproduce the observed transport. We performed a spin-up of 1-year using initial chemistry conditions from the global atmospheric model MOZART-4 (Emmons et al., 2010) prior to the 2004 annual cycle simulation that is evaluated in the
present study. Table 1 describes the main configuration of the model. The feedback between chemistry and meteorology is not considered in this study.

### 3.1 Emissions

The global emissions used in this study are based on the Atmospheric Chemistry and Climate Model Intercomparison Project (ACCMIP; Lamarque et al., 2013), which includes emissions from anthropogenic and biomass burning sources at 0.5° x 0.5° horizontal resolution (Lamarque et al., 2010). Note that the methodology involves assuming that 2004 emissions are derived from a linear interpolation between years 2000 and 2010—equivalent to the best estimate reported for ACCMIP for year 2000. Therefore specific events occurred during 2004 (e.g., large summer wildfires in Alaska and Canada) are not described. The ACCMIP inventory is a combination of several existing regional and global inventories. The surface anthropogenic emissions are based on two historical emission inventories, namely RETRO (1960-2000; Schultz and Rast (2007)) and EDGAR-HYDE (1890-1990; Van Aardenne et al. (2005)) and monthly variations for biomass burning, ship and aircraft emissions are provided. One limitation is that land-based anthropogenic emissions have constant values for the entire year. Lamarque et al. (2010) presents a comparison of the annual total CO anthropogenic and biomass burning emissions (Tg(CO)/year) for different regional and global emission inventories for year 2000 (see Table 5 of this paper). Note that ACCMIP global CO anthropogenic emissions are significantly higher (610.5 Tg CO/year) than other emissions inventories (e.g. RETRO with 476 Tg CO/year, EDGAR-HYDE with 548 Tg CO/year, and GAINS with 542 Tg CO/year).

Ocean and soil natural emissions are based on the POET (Granier et al., 2005) global inventory. Lightning and volcano emissions are not considered in this simulation. Biogenic emissions are computed using MEGANv2.04 model as described in Sec. 2.2.6. NO emissions for January and July 2004 are shown in Figure S1 in the supplementary information and yearly totals for anthropogenic and biomass burning, biomass burning, biogenic, soil, and ocean emissions are summarized in Table 2.

To account for the sub-grid scale vertical diffusion within the planetary boundary layer (PBL) all the land-based anthropogenic emissions are emitted in the first 500 m of the model, biomass burning emissions from forests in the first 1300 m, biomass burning emissions from grass in the first 200 m, ocean emissions on the first 30 m and shipping emissions on the first 500 m. This vertical distribution of emissions has been derived after some sensitivity runs and it may not be appropriate for higher resolution runs. The model does not include the attenuation of radiation due to aerosols in the photolysis scheme. Therefore, regions with strong biomass burning emissions may significantly overestimate chemical photolysis production (e.g., Bian et al., 2003; Real and Sartelet, 2011).

### 4 Observational data

#### 4.1 Surface concentration and wet deposition

For the evaluation of ground-level gas concentrations, we selected background stations having hourly data (Fig. 1 left panel) from the World Data Center for Greenhouse Gases (WDCGG; http://gaw.kishou.go.jp/wdcgg/), the European Monitoring and
Evaluation Programme (EMEP; http://www.emep.int/), the Clean Air Status and Trends Network in USA (CASTNET; http://java.epa.gov/castnet/) and the Acid Deposition Monitoring Network in East Asia (EANET; http://www.eanet.asia/). For O\textsubscript{3}, we used data from 41 WDCGG, 52 EMEP, 64 CASTNET and 11 EANET stations, covering Europe, United States (US), and a few locations in East Asia. We also selected 21 EMEP stations for NO\textsubscript{2}, 10 EANET stations for NO\textsubscript{x} and 14 WDCGG stations for CO.

The simulated wet deposition of HNO\textsubscript{3} is also compared against observed nitrate (HNO\textsubscript{3} and aerosol nitrate) wet deposition, including 260 measurements from the National Atmospheric Deposition Program (NADP; http://nadp.sws.uiuc.edu/) network in North America, 51 from the EMEP network in Europe and 28 from EANET in East Asia.

4.2 Vertical structure: ozonesondes and MOZAIC measurement campaigns

The surface evaluation is complemented with an assessment of the vertical structure of O\textsubscript{3} using ozonesondes from the World Ozone and Ultraviolet Radiation Data Center (WOUDC; http://www.woudc.org/), the Global Monitoring Division (GMD; ftp://ftp.cmdl.noaa.gov/ozone/) and the Southern Hemisphere ADditional OZonesondes (SHADOZ; http://croc.gsfc.nasa.gov/shadoz/; Thompson et al., 2003a, b). Most stations provide between 4 and 12 profiles per month each year with a precision of \(\pm 3-8\%\) in the troposphere (Tilmes et al., 2012). We followed the methodology of Tilmes et al. (2012) for the selection and treatment of the measurements. Table 3 lists the locations and the number of available measurements per season of the 39 ozonesonde stations used (also displayed in Fig. 1), as well as the regions where stations with similar O\textsubscript{3} profiles were aggregated.

Additional observations considered in this study are CO vertical profiles from Measurement of Ozone, Water Vapor, Carbon Monoxide, Nitrogen Oxide by Airbus In-Service Aircraft (MOZAIC; http://http://www.iagos.fr). Based on the availability of data, we selected 14 airports (displayed in Fig. 1 right panel) covering different regions of the world during 2004. The number of vertical profiles available per season are provided in Table 4.

Nitric oxide (NO\textsubscript{x}), peroxyacetyl nitrate (PAN) and acid nitric (HNO\textsubscript{3}) vertical profiles are used from two-four different measurement campaigns: TOPSE (Atlas et al., 2003; Emmons et al., 2003) and TRACE-P (Jacob et al., 2003), PEM-Tropics-B (Raper et al., 2001) and POLINAT-2 (Schumann et al., 2000). Tropospheric data from these previous campaigns were gridded onto global maps with resolution 5\(\degree\) \(\times\) 5\(\degree\) \(\times\) 1km \(\times\) 1 km, forming data composites of important chemical species in order to provide a picture of the global distributions (Emmons et al., 2000).

When running an AQM model, it is preferable to compare the model output with an observational database from the same year as the model simulation. Nevertheless, in our case, there are insufficient global observations to achieve this goal for any full year. Hence, in this model evaluation, all the observations are for considered are within the simulated year (2004, except for the vertical profiles obtained from measurement campaigns. Hence, in this study, model output from selected regions are compared with this campaign from the same regions regardless of the year of the measurements. In addition, it is valuable to compare the same regions for different species which allows identification of systematic differences between the model results and observations. Details of these campaigns describing their geographical
Details on the geographical regions and periods for these campaigns are described in Table 5, and the location locations are displayed in Fig. 1 (right panel).

4.3 Satellite data

Modelled Modeled tropospheric NO$_2$ columns are compared with SCanning Imaging Absorption spectroMeter for Atmospheric CHartographY (SCIAMACHY, http://www.sciamachy.org/) satellite data. SCIAMACHY (on board of ENVISAT, which was operational from March 2002 to April 2012) is a passive remote sensing spectrometer measuring backscattered, reflected, transmitted or emitted radiation from the atmosphere and Earth’s surface with a wavelength range between 240-2380 nm. The SCIAMACHY instrument has a spatial resolution of typically 60 km x 30 km$^2$, and has three different viewing geometries: nadir, limb, and sun/moon occultation. Alternating nadir and limb views, global coverage is achieved in six days.

NO$_2$ daily data was obtained from the Institute of Environmental Physics, the University of Bremen (http://www.iup.uni-bremen.de/doas/scia_no2_data_tropos.htm), based on Version 3.0 data product (Hilboll et al., 2013). This dataset is an improved extension of the data presented in Richter et al. (2005). Validation of the data product was performed in several studies (e.g., Petritoli et al., 2004; Heue et al., 2005). We used daily satellite overpasses of cloud-free (<20% cloud fraction) tropospheric vertical column densities (VCDtrop NO$_2$) from SCIAMACHY measurements using the limb/nadir matching approach, whose total uncertainty is estimated to vary between 35 and 60% in heavily polluted cases and >100% in clean scenarios (Boersma et al., 2004).

Additionally, CO mixing ratios at 800 and 500 hPa were evaluated with the Measurement of Pollution in the Troposphere (MOPITT, http://www2.acd.ucar.edu/mopitt) instrument retrievals. The MOPITT, aboard MOPITT, on board of the NASA EOS-Terra satellite, is a gas filter radiometer and measures thermal infrared (near 4.7 $\mu$m) and near-infrared (near 2.3 $\mu$m) radiation, only during clear-sky conditions, with a ground footprint of about 22 km x 22 km. We used the MOPITT Version 5 (V5) Level 2 data product, which provides daily CO mixing ratios. MOPITT CO mixing ratios have been validated with in situ CO profiles measured from numerous NOAA/ESRL aircraft profiles in Deeter et al. (2013), and they were found to be positively biased by about 1% and highly correlated (r = 0.98) at the surface levels.

5 Model evaluation

This section presents the evaluation model evaluation with observations of relevant trace gases from the NMMB/BSC-CTM using the observations described in the previous section. It also, and compares the results with other modeling studies available in the literature. For the evaluation of daily surface-level comparison, three hourly averages from O$_3$, we considered averages of temporally collocated 3-hourly values from the model and the observations and model are used to compute daily O$_3$ averages and calculate the statistical measures defined in section observations. Section 1 in the supplemental material of the supplementary material presents the statistical measures calculated from the daily data. Ground-monitoring stations were selected with a maximum altitude of 1000 meters. In the case of ozonesondes and MOZAIC the comparison is made only when vertical profile observations
are available, i.e., the data from the model and the observations are collocated/simultaneous. Similar criteria is used in the case of Temporal collocation was also considered when comparing to ozonesondes, MOZAIC, MOPITT and SCIAMACHY for NO2. Moreover, For CO, averaging kernels for CO are accounted were considered to represent the observational sensitivity at different pressure levels. When computing the modelled tropospheric columns of NO2 the tropopause was assumed to be fixed at 100 hPa in the tropics and 250 hPa in the extratropics.

Similarly, when comparing model data with data composites from aircraft campaigns the same period of the year at the same location is selected and mapped into the same grid resolution, the evaluation with aircraft campaigns was performed after remapping the model output to the resolution of the observed data composites (5° x 5° x 1 km, before the comparison is made 1 km). For some species, the model evaluation is given per seasons: DJF for December-January-February, MAM for (March-April-May), JJA for (June-July-August) and SON for (September-October-November).

### 5.1 Hydroxyl Radical (OH)

One of the means for characterizing the general properties of an AQM is through its ability to simulate OH oxidation. OH is the main oxidant in the troposphere and is responsible for the removal of many compounds, thereby controlling their atmospheric abundance and lifetime. OH is mostly found in the tropical lower and mid troposphere with a strong dependence and strongly depends on the levels of ultraviolet radiation and water vapor. Tropospheric OH formation is mainly due to O3 photolysis, dominated by the tropics. Also, OH is directly connected to the chemistry of O3 production since the initial reactions of O3 formation (VOC+OH and CO+OH) are driven by OH. Hence, O3 production rates depend on the sources and sinks of odd hydrogen radicals. Primary OH formation also includes the photolysis of HCHO and secondary VOC.

The tropospheric mean (air mass weighted) OH derived by the model is 11.5 molec x 10^5 molec cm^{-3}, assuming a tropospheric domain ranging from 200 hPa to the surface. Note that previous studies suggest that the estimation of the mean OH does not depend on the definition of the tropopause (Voulgarakis et al., 2013). This value is in good agreement with other studies, e.g., Voulgarakis et al. (2013) where the mean OH concentration from 14 models for 2000 was estimated to be 11.1 ± 1.8 x 10^5 molec cm^{-3}; Spivakovsky et al. (2000) with 11.6 x 10^5 molec cm^{-3}, and Prinn et al. (2001) with 9.4 ± 0.13 x 10^5 molec cm^{-3}.

The zonal mean OH concentrations for January, April, July and October 2004 are shown in Fig 2. Seasonal differences reflect the impact of water vapor concentration and stratospheric O3 column upon incident ultraviolet (UV) radiation (Spivakovsky et al., 2000; Lelieveld et al., 2002). The highest OH concentrations arise in the tropics throughout the year. In northern midlatitudes, the highest OH concentrations are found during summer in the lower to middle troposphere. The latitudinal and seasonal variations are similar to the climatological mean in Spivakovsky et al. (2000), particularly the lower values in the extratropics. Peak concentrations are slightly larger compared to this climatology and other studies (e.g., Horowitz et al., 2003; Huijnen et al., 2010). During January and October the peaks appear in the southern tropics between 700-1000 hPa and 800-1000 hPa, respectively. The peak in April and July is found in the northern tropics between 800-1000 hPa and 700-1000 hPa, respectively.
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. (2013b). In addition, the model regional annual mean air mass-weighted OH concentrations have been calculated and are in general agreement with the multi-model values (Naik et al., 2013b) (see Figure S2 in the supplementary information). However, concentrations over the tropics (30S-30N) are slightly higher than the multi-model mean and concentrations above 500 hPa are lower than the multi-model mean. Labrador et al. (2004) studied the sensitivity of OH to NOx from lightning, showing that OH concentrations increase mostly in the middle to upper troposphere (500-200 hPa) if lightning emissions are considered. 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).

5.2 Carbon monoxide (CO)

CO is one of the most important trace gases in the troposphere exerting a significant influence upon the concentration of oxidants such as OH and O3 (Wotawa et al., 2001). Main sources of CO in the troposphere are the photochemical production from the oxidation of hydrocarbons (including methane) and direct emissions, mainly fossil fuel combustion, biomass burning and biogenic emissions. CO main loss is by reaction with OH, which occurs primarily in the tropics, but also in the extratropics.

In the northern extratropics, the elevated CO concentrations are dominated by anthropogenic emissions and precursor hydrocarbons, which leads to a net CO export to the tropics (Shindell et al., 2006; Bergamaschi et al., 2000). Although most of the biomass burning occurs in the tropics, gases and aerosols emitted from large wildfires can be transported to the southern extratropics, where emissions and chemical production are lower. Moreover, due to the strong convection, enhanced by forest fire activity, emissions can reach the upper troposphere and the lower stratosphere (Jost et al., 2004; Cammas et al., 2009). CO has a chemical lifetime of a few months (~1-3), and therefore it is a useful tracer for evaluating transport processes in the model. It is important to keep in mind that despite large Alaskan and Canadian wildfires occurred during the summer, globally 2004 had lower CO concentrations than other years during the decade (Elguindi et al., 2010).

An analysis of the CO burden in different regions is presented in Table 6. The global and annual mean burden of CO for 2004 is 399.03 Tg, with higher abundances in the tropics (229.43 Tg CO), followed by the Northern Extratropics (101.71 Tg CO), and the Southern Extratropics (67.88 Tg CO). Other model estimates of the CO burden (Horowitz et al., 2003; Huijnen et al., 2010; Flemming et al., 2015) are also shown in Table 6. Our estimates are higher by ~ 46-48 Tg CO in comparison with these studies. We performed tests comparing the annual mean burden of tropospheric CO with and without biomass burning emissions in the model. Neglecting biomass burning emissions only reduced 7% of the tropospheric CO annual mean burden. Therefore, other factors should explain our higher CO burden. On the one side, biogenic emissions are computed online every hour in
order to account for evolving meteorological changes, such as solar radiation and surface temperature (see section 2.2.6). Also this simulation neglects the attenuation of radiation due to aerosols, which may produce an overestimation of VOCs biogenic emissions and the derived CO.

The CO anthropogenic emissions used in this study (610.5 Tg/year) are also higher than those in other inventories (see 3.1). The dry deposition of CO is significantly weaker in the NMMB/BSC-CTM-NMNB-MONARCH (24 Tg CO) than the global model TM5 (184 Tg CO) and the study of Bergamaschi et al. (2000) (292-308 Tg CO). By contrast, other global models such as MOZART-2 have significantly lower dry deposition (2 Tg CO) and the study of Wesely and Hicks (2000) suggests that CO and other relatively inert substances are deposited very slowly. Clearly, there are major uncertainties in the sources and sinks of CO that could be responsible for modeled CO differences.

Fig. 3 shows the time series of CO daily mean concentration over 14 ground-monitoring stations from the WDCGG database (primarily in the northern mid-latitudes, but with a few of them in the tropics and southern mid-latitudes). The solid red line and the solid black line represent, respectively, the average of observations and the model simulation. Bars show the 25th-75th quartile interval of all observations (orange) and the model simulation (grey). The model is in good agreement with the CO field in the surface layer (daily correlations between 0.3-0.7). However, the model is not able to fully capture the seasonal CO variability, with a slight underestimation during cold months (-10.65 ppb) and overestimation during warm months (28.67 ppb). Such a model limitation could be explained by the fact that most of the stations are closer to anthropogenic polluted areas, where its concentration is primarily determined by local emissions, and the CO land-based anthropogenic emissions inventory does not have any seasonal variation in this study (see Sec.3.1).

Fig. 4 shows the CO mean bias (MB), correlation and root mean square error for all rural WDCGG stations. The model has a negative MB over stations in Europe and Japan and a positive bias in stations in Canada and Africa, where the correlations are low. The negative bias for several of the northern mid-latitude stations indicates that the higher CO burden found in our model compared to other models in these areas is a feature mainly driven by free tropospheric abundances. Higher correlations are found in northern regions of Europe, southern Africa and eastern Asian countries. Correlation: The daily correlation in Canadian stations is between 0.3-0.5. In most of the stations, the RMSE is found to be less than 60-40 \( \mu g \text{m}^{-3} \text{ppb} \), with only 4 stations having a RMSE higher than 60 \( \mu g \text{m}^{-3} \text{ppb} \).

Additionally, the model was compared with the seasonally averaged vertical profiles of temporally collocated CO from MOZAIC aircraft observations (therefore not available; see section 2.2.6). The comparison is made only when observations are available; i.e., the same data from the model and the observations are used. Measurements are represented by the solid red line and the model simulation by the solid black line. To understand the variability of the data, standard deviation is plotted in each vertical layer for both model and observations. It is important to Dhabe and Niamey (shown in Figs. 5 and S3 in the supplementary information). Observations and model results (both mean and standard deviation) are represented in red and black, respectively. We note that the number of flights is significantly different between the different airports; therefore, they differ among airports (therefore not all comparisons are statistically robust. In addition, note that the scale), and the CO range represented for Beijing is different (0-1000 ppb) from the than for others stations (0-400 ppb).
The model captures reasonably well the vertical profiles during the first part of the year, with higher and shows larger biases during the warm months. Generally, it overestimates CO from the middle to the upper troposphere in most of the stations throughout the year. Over Frankfurt, the model is in good agreement with the observation observations during the entire year, despite slight underestimation during MAM and overestimation during SON-slight underestimations during MAM (∼ 31 ppb) and overestimations during SON (∼ 12 ppb) in the middle troposphere.

For Beijing, one of the most polluted cities in the world, the model shows a clear tendency to underestimate CO in the lower atmosphere (below 600 hPa). This is very probably due to an underestimation in the of CO anthropogenic emissions. Most of the AQMs seem to be AQMs are unable to capture the extreme growth of anthropogenic emissions in China (Akimoto, 2003; Turquety et al., 2008). Over Atlanta, the model performs much better during the better in winter and spring along the troposphere but positive biases (throughout the troposphere than in summer and autumn, when positive biases reach ∼ 20-25 ppb) are seen during the summer and autumn. Regions.. In regions with biomass burning and biogenic influence, such as Abu Dhabi and Niamey, show a significant overestimation the model significantly overestimates CO during warm months throughout the tropospheric column. During winter and spring, Stein et al. (2014) also obtain an underestimation of CO vertical profiles in airports located in the Northern Hemisphere (NH).

To complete this CO evaluation, seasonal averages are compared with data from the MOPITTT instrument at 800 hPa and 500 hPa in (Figs. 6 and S3-S4 in the supplementary information, respectively). At 800 hPa, the largest differences are seen detected during boreal winter and spring, when the model clearly overestimates in the tropics and underestimates in the north extratropics and north of northern extratropics and North Africa. The negative bias during winter wintertime negative bias (∼ -10-35 ppb) in the NH could further may be explained by either the lack of a seasonal cycle anthropogenic emissions

However, the underestimation during NH winter, which appears most state-of-the-art AQMs could also be originated from seasonally varying anthropogenic emissions in our simulation, an underestimation of CO emissions (Stein et al., 2014) - or a combination thereof. There are significant positive biases over west-central Africa and also over western South America, Indonesia and the surrounding Pacific and Indian oceans during the dry season. Sources of CO over west-central Africa are mainly from biomass burning and biogenic emissions. Uncertainties in the emission inventories have probably contributed probably contribute to the CO overestimation for in these regions. Due to the long-range transport of CO, higher CO concentrations are seen throughout all concentrations are also seen throughout the year over the tropics and are extended over some parts of the extratropics from June to November. Hence, during JIA and SON the model overestimates CO concentrations in most places including south and central EU and USA Europe and US (∼ 10-25 ppb).

At 500 hPa, the model presents similar results, with a clear underestimation in the north extratropics and overestimation clear underestimations in the northern extratropics and overestimations in the tropics and southern latitudes. Excessive vertical mixing by moist convection may explain the overestimation in the tropics. Overestimated emissions in Africa or Asia above the PBL can could also lead to this positive bias in the middle of the troposphere.

Naik et al. (2013b) presents an annual average bias of compared the multi-model (annual mean from 17 global models) mean CO for models for year 2000 against average 2000-2006 MOPITTT CO with the average CO from MOPITTT at 500 hPa. These between 2000 and 2006. The 17 models used the same anthropogenic and biomass burning emissions as our study model,
and a priori and averaging kernels are taken into account for each model before computing the biases. The biases in the tropics and extra tropics are similar to those presented here. Hence, these biases might be related to discrepancies in, suggesting systematic model errors due to inaccurate anthropogenic and biomass burning emission inventories, where the magnitude, and perhaps location of emission is not completely understood or correctly modelled. We note that MOPITT V4 CO retrievals are affected by biases of about -6% at 400 hPa when evaluated with in-situ measurements (Deeter et al., 2010), which are low compared to current model discrepancies. Naik et al. (2013b) discussed a too high OH concentration possibly leading also discussed how an overestimated OH concentration may lead to the northern mid-latitude underestimates of CO, which is also a possibility in our case, given the high OH concentrations that the model shows underestimation of CO. This may partly explain our results given the higher OH concentration simulated by our model compared to other models. Numerous studies show that the variability in simulated significant differences in CO among AQMs is large, and uncertainties are diverse including emissions, which may emerge from a diversity of uncertainties including those in emission inventories and injection height heights (Elguindi et al., 2010; Shindell et al., 2006; Prather et al., 2001). A detailed evaluation of MOPITT V4 CO retrievals between 2002–2007 with in-situ measurements shows a bias of about -6% at 400 hPa. However, this bias is not able to explain the model biases that vary in sign and magnitude between different global regions. For example, Stein et al. (2014) suggest 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.

5.3 Nitrogen compounds

The NO\textsubscript{x} (= NO\textsubscript{2} + NO) family is one of the key players in the formation of O\textsubscript{3} in the troposphere, and during pollution episodes it causes causing photochemical smog and contributes contributing to acid rain during pollution episodes. Because it has a relatively short lifetime, consequently a few hours within the PBL and up to a few days in the upper troposphere; Tie et al. (2001, 2002), it is generally restricted to emission sources, both natural and anthropogenic (mainly fossil fuel combustion). The seasonal cycle of NO\textsubscript{x} near the surface is controlled by the seasonality of anthropogenic emissions (especially in the northern hemisphere NH) and biomass burning emissions (especially in the tropics and the southern hemisphere SH). As a result, NO\textsubscript{x} concentration is more sensitive to errors in emissions than other pollutants, and errors in NO\textsubscript{x} emissions can change NO\textsubscript{x} concentrations even more drastically (Miyazaki et al., 2012).

Figure 7 shows the time series of NO\textsubscript{2} and NO\textsubscript{x} daily mean surface concentrations over 21 EMEP and 10 EANET rural ground-monitoring stations from the EMEP and EANET networks, respectively. In both cases, the model is able to successfully reproduce the seasonal cycle of NO\textsubscript{2} and NO\textsubscript{x}. However, a positive bias (< 3 ppb) is found during the summertime for NO\textsubscript{2} in Europe (Fig. 7 top panel). Such a result could be explained by the limitation on the anthropogenic emissions that are constant during the entire year. Because of that, the model cannot reproduce the decrease in anthropogenic emissions during the summertime, which leads to overestimated concentrations.

Daily profiles show that the, which may result from the lack of seasonality in the anthropogenic emissions. The modeled NO\textsubscript{2} tends to be too high concentration is excessive during nighttime (not shown). This result may be due to may result from the lack of heterogeneous formation of HNO\textsubscript{3} through N\textsubscript{2}O\textsubscript{5} hydrolysis, an important sink of NO\textsubscript{2} at night. In addition, the
model does not consider secondary aerosol formation for in the present study, which might result in an atmosphere that is too oxidising resulting in an excessively oxidising atmosphere (overestimation of OH radicals). In combination with the nocturnal chemistry this that in turn may lead to an accumulation of NO$_2$ in the surfacelayers. However, near the surface, Between 9 and 18 UTC there is a slight underestimation is observed between 9 – 18 UTC. Looking at the annual time series of NO$_x$ in the Asian network of NO$_2$. In Asia (Fig. 7 bottom), it is observed that the model does not reproduce the observed NO$_x$ values, with a sizeable showing a large negative bias during the summer. This underestimation could be attributed to an underestimation in the emission inventories, which do not capture the extreme increase of anthropogenic emissions over Asia during the last decade probably due to underestimated emissions (Akimoto, 2003; Richter et al., 2005), as was the case for CO in the case of CO. Also, an excessive mixing within the PBL during the night could contribute to a decrease in ozone titration by NO and explain the bias.

Concerning the spatial statistics (see Fig. 8), the model’s prediction capabilities are lower in some. The model correlation is lower in regions such as the Iberian Peninsula and most of the stations in Japan, showing poor correlations. (Fig. 8). The best performance is seen in central EU occurs in central Europe and stations in Japan that are not in the main island. In general there is a negative bias in most of the stations for these two regions.

The comparison of modelled and observed vertical profiles of NO$_x$. Fig. 9 displays the comparison of NO$_x$. HNO$_3$ and PAN are presented in Fig. 9. Vertical profiles for several regions over-in the US, China, Hawaii and Japan (see Table 5). The comparison over Tahiti and Ireland is shown in Fig. S5 of the supplementary information. As explained in Sec. 4.2, the observed vertical profiles do not correspond to the simulated year (see Table 5 for more details), but the qualitative patterns can provide insights on the model capability to reproduce the chemistry involved. Fig. 9 (first column) shows that vertical profiles of NO$_x$ are in very good agreement with the observed values, really well captured by the model. The model has a tendency to overestimate NO$_x$ concentrations near the surface ($\sim 400 \text{ ppt in Japan and } \sim 300 \text{ ppt in China}$); it is likely that NO$_x$ emissions used in this study are higher than the real-actual emissions during the campaign periods. Another reason for these higher values over island locations (Japan and Hawaii) could be that emissions are spread throughout the entire low resolution-over the coarse model grid box while the measurements were taken in the cleaner marine boundary layer. In the middle and upper troposphere, the model produces the concentrations well, with a slight underestimation in most of the locations. Note that NO$_x$ lightning emissions are not included in this simulation, which may explain part of this underestimation, particularly in the upper troposphere.

PAN is the main tropospheric reservoir species for NO$_x$ with important implications for the tropospheric O$_3$ production and the main atmospheric oxidant, OH (Singh and Hanst, 1981). PAN is mainly formed in the boundary layer by oxidation of NMVOCs in the presence of NO$_x$. NMVOCs and NO$_x$ have both natural and anthropogenic sources. Rapid convection can transport PAN to the middle and upper troposphere and enables the long-range transport of NO$_x$ away from the urban and polluted areas, where it can produce O$_3$ and OH remotely.

Some features of the vertical profiles are well-captured by the model, although it significantly overestimates PAN concentrations (see Fig. 9, second-third column). We find overestimations from the surface to the middle atmosphere in Japan, China, Boulder and Churchill, which are possibly explained by an overestimation of biogenic and anthropogenic NO$_x$ surface emiss-
sions in this area at surface level. Another possibility for this overestimation is a too long lifetime for PAN. At an excessive lifetime of PAN, in most sites, PAN model concentrations tend the modeled PAN concentration tends to increase with altitude, reaching maximum mixing ratios at about 6 km, from where they progressively decrease. This behavior it progressively decreases. This behavior explains the long thermal decomposition time of PAN (lifetime of approximately a month) and the slow loss by photolysis in the cold middle-upper troposphere. Fischer et al. (2014) analyze the sensitivity of PAN to different emission types, showing that most of the northern hemisphere NH and Japan are more sensitive to anthropogenic emissions, while the southern hemisphere SH and the west coast of the USA-US are more sensitive to biogenic emissions, both contributing to 70-90% of the PAN concentrations.

HNO₃ is mainly produced by the reactions of NO₂ with OH and by the heterogeneous hydrolysis of N₂O₅ on aerosols (we do not account for this reaction-the latter in this simulation), and then it is removed by wet and dry deposition. HNO₃ is the main sink of NO₂ chemistry. In general, the modeled and observed nitric acid concentrations are in good agreement of the same magnitude throughout the troposphere, although the model reveals a tendency to overestimate HNO₃ concentrations that is even more pronounced, particularly in US regions. In the regions of Hawaii, Japan and China the model overestimates HNO₃ in the lower-middle troposphere (up to 5 km) and underestimates it in the upper troposphere (above 6 km). Overestimation of HNO₃ in the troposphere is a common problem in global models (Hauglustaine et al., 1998; Bey et al., 2001; Park et al., 2004; Folberth et al., 2006). HNO₃ concentrations are highly sensitive to the parameterization of wet deposition. One possible reason for this overestimation is that the scavenging from convective precipitation is underestimated.

Figure S4-S6 in the supplementary information presents the wet deposition fluxes of HNO₃ in comparison with nitrate observations for three different networks located in Europe, USA-US, and Asia. Satisfactory agreement is found in the HNO₃ wet deposition fluxes with correlations of 0.63 in Europe, 0.80 in USA-US, and 0.52 in Asia. There is a tendency to underestimate in most of the stations, principally in Asia and Europe. Part of this underestimation is because the comparison is between nitric acid (gas) and nitrate (nitric acid + particulate nitrate) wet depositions. However, this tendency to underestimate mainly in Asia (MB = -163.27 mg N/m²) and Europe (MB = -200.70 mg N/m²). We note that these observations include particulate nitrate in addition to HNO₃, and our model omitted nitrate in this study. Therefore, while the underestimation may be partly due to this omission, it is consistent with the higher values of HNO₃ observed at in the lower and middle troposphere. Seasonal averages of vertical tropospheric columns-VTTC (VTC) of NO₂ are compared with SCIAMACHY satellite data in Fig.10. The model is in good agreement with the observations, capturing the high NO₂ values over the most polluted regions, such as Europe, USA and Eastern-US and eastern Asia. The phase in the seasonal cycle of the NO₂ columns is performed well-captured satisfactorily by the model. During the entire year, the model tends to underestimate NO₂ VTCs in big cities megacities, especially during the colder months, and overestimate them in rural regions. The largest discrepancies are seen in eastern China, which suggests an underestimation of emissions regionally. The biomass burning cycle is captured remarkably well, with higher NO₂ VTC in central Africa during DJF and in South America in JJA. Over the sea, the model is in good agreement with SCIAMACHY, showing only small differences. The model does really well over the ocean, where only small biases are detected (± 0.5 ±10⁻¹⁵ molec/cm² cm⁻²).
5.4 Ozone (O\textsubscript{3})

Ozone is one of the central species that drive tropospheric chemistry, and for that reason it is essential that a model reproduces the spatial and temporal concentrations of Tropospheric O\textsubscript{3} well, both at the surface and across the troposphere and stratosphere. O\textsubscript{3} found in the troposphere is originated from in situ photochemical production and from intrusions of O\textsubscript{3} from the stratosphere. O\textsubscript{3} photochemical production in the troposphere involves stratospheric intrusions. Its photochemical production involves the oxidation of CO and hydrocarbons in the presence of NO\textsubscript{x} and sunlight. In rural remote areas, CO and CH\textsubscript{4} are the most important species being oxidized in the oxidized during O\textsubscript{3} formation. However, in polluted areas, short-lived NMVOCs (e.g. HCHO) are present in high concentrations and are the most important species.

The simulated global burden of tropospheric O\textsubscript{3} is shown in Table 7. In the troposphere, O\textsubscript{3} chemical sources and sinks are dominated by the tropics, where high concentrations are found (171.60 Tg O\textsubscript{3}). Low concentrations lower values are predicted in the northern extratropics (101.56 Tg O\textsubscript{3}) and especially the Southern Extratropics in the southern extratropics (75.41 Tg O\textsubscript{3}), where precursors are not present in high amounts the presence of precursors is limited. Similar results are found from in other global models, such as MOZART-2 (Horowitz et al., 2003) and TM5 (Huijnen et al., 2010). In general, with MOZART-2 having a higher and TM5 a lower annual mean burden of O\textsubscript{3} than the NMMB/BSC-CTM. The annual mean O\textsubscript{3} burden predicted by our model is higher than the two models in the southern extratropics (by 10-14 Tg O\textsubscript{3}) than the other two models. Higher CO concentrations in the southern hemisphere SH (see Table 6) might lead to excessive production of O\textsubscript{3} in this area. In addition, the region. Our global tropospheric O\textsubscript{3} burden in our model (348 Tg O\textsubscript{3}) is also in good agreement with the C-IFS global model (Flemming et al., 2015) and the two multimodel ensemble means of 25 and 15 state-of-the-art atmospheric chemistry-global models, the GFDL AM3 chemistry-climate model (Naik et al., 2013a) and multi-model means (Stevenson et al., 2006; Young et al., 2013).

According to our calculations, 1200-1201 Tg O\textsubscript{3} are removed from the troposphere by dry deposition at the surface. This quantity is higher in comparison with the global models, a value well above TM5 (829 Tg O\textsubscript{3}) and MOZART-2 (857 Tg O\textsubscript{3}). In this sense, the model is in good agreement with the global model estimates, but in agreement with LMDz-INCA (1261 Tg O\textsubscript{3}) and with the multimodel GFDL AM3 (1205 ± 20) and the multi-model ensemble study by Stevenson et al. (2006) (1003 ± 200 Tg O\textsubscript{3}). The net stratospheric input, Stratosphere-Troposphere Exchange (STE), annual rate of the model (384 Tg O\textsubscript{3}) is also shown in Table 7. STE exchange flux is calculated as the annual balance of the ozone mass crossing the 100 hPa height. The model’s STE is in good agreement with other modelling studies, especially with the multimodel ensemble in Stevenson et al. (2006) (552 ± 168 Tg O\textsubscript{3}).

Fig.11 shows the time series of O\textsubscript{3} daily mean concentration averaged over all available monitoring sites (from top to bottom, WDCGG, CASTNET, EMEP and EANET) over the entire simulation period. The solid red line and solid black line represent the average of observations and the model, respectively. Bars show the 25th-75th quartile interval of all observations (orange) and model simulation (grey). As illustrated in Fig.11, there is is an overall good performance although there are significant positive bias from May to October in the US and Japan. The seasonal cycle of O\textsubscript{3} from the model agrees well with the observations, showing the highest concentrations during July-August and the lowest concentrations...
during Nov-Dec over all stations. Although, the model captures the seasonal $O_3$ variability along this period, there is a tendency to overestimate concentrations during the warmer months, i.e. May-September. This positive bias is significantly higher in the US, where the overestimation occurs all day long ($10-20 \mu g m^{-3}$ through the year (5-15 ppb). Over Europe, the overestimation of $O_3$ levels during summer is lower than in other regions. Over East Asia the model captures reasonably well the peaks in April and May, although a positive bias is seen during the cold months in contrast to Europe where the model concentrations agree with the observations. Overall the observational networks show a reduction of $O_3$ concentrations from May-June, but the model has a tendency to simulate an annual cycle with higher concentrations until July. Moreover, a recent study by explains the importance of the dry deposition in controlling surface $O_3$. Possible reasons for the overestimation could be the reduction of the ozone titration due to an excessive emission injection height prescribed in the model, or the dry deposition processes included in our model. Val Martin et al. (2014) shows that accurate dry deposition processes can reduce the summertime surface $O_3$ bias from 30 ppb to 14 ppb and from 13 ppb to 5 ppb over eastern US and Europe, respectively. Thus, part of this positive bias could be related to the dry deposition processes included in our model. Further investigation is required to understand model behavior during this period.

Fig.12 displays the spatial statistics for $O_3$ over all in-situ monitoring sites using daily mean data. Areas without emissions, such as the south pole and isolated islands in the tropics, have low small mean biases and errors root mean square errors, and good correlations ($>0.80$). In polluted areas, a good performance is observed in the US midlands, and parts of central and southern Europe ($0.60 < r < 0.80$ and RMSE < $20 \mu g m^{-3}$, 12 ppb). Large errors are seen in northwestern and southern US and Northern Europe. Although, large errors are seen in all the stations over Japan, the two stations farthest from the main island show high correlation ($r > 0.7$).

In order to assess the vertical distribution of $O_3$, the model results are compared with available ozonesondes. The seasonal vertical profiles of $O_3$ for both the model and observations are compared in Figs.13 and S5 of the supplementary information for the period of study during the study period (see Table 3 and Fig. 1 for more details). The comparison is made only when ozonesonde observations are available. Fig.13 and S5 from the supplementary information figures show (from top to bottom) four panels: DJF, MAM, JJA and SON for each region. Measurements are represented by the solid red line and the model results by the solid black line. The variability of the data is shown in the form of standard deviation for both the model and observations. The magnitude and vertical profile of $O_3$ are in good agreement with the observations.

However, the model shows a positive bias of ~ 5-20 ppb along the troposphere in most of the regions during the entire year. As shown in Sec. 5.2 there is a significant overestimation of CO, especially in the free troposphere for some regions, which may account for the positive $O_3$ biases, although the CO overestimation mostly occurs in the tropics where $O_3$ biases are not so large. Another reason for this result could be that anthropogenic aerosols and secondary aerosol formation are neglected in this simulation, leading to a higher $O_3$ formation in regions with more precursors. However, this should have
more localised effects and therefore it cannot fully explain the biases throughout the troposphere. Possible biases in the stratospheric O\(_3\) or the lack of other specific chemistry (e.g., halogen chemistry) could also contribute to this positive bias.

The vertical profile is in good agreement with the observations, with O\(_3\) increasing from lower to higher tropospheric layers. In the lower-middle troposphere the model overestimates O\(_3\) in regions with high emissions (Japan, Canada, USA and W. Europe), a feature that is more significant in DJF stands out in DJF (< 20 ppb). In Western Europe and the US, this bias is reduced at the surface level. In tropical areas (Equator, NH tropical and W. Pacific) the model captures well the observed concentration and vertical structure of O\(_3\) in the lower to middle troposphere. However, the model tends to overestimate the O\(_3\) in the vicinity of the tropopause layer in these regions (10-20 ppb). At polar regions (NH and SH Polar) the model also presents a tendency to overestimate the vertical structure of O\(_3\). O\(_3\) in the tropopause layer is underestimated in the NH Polar case, and overestimated in SH Polar case.

Finally, statistics were computed to identify those areas where the errors are more important. Fig. 14 shows the mean O\(_3\) bias (left), correlation (middle) and RMSE (right) of the model with respect to ozonesondes (data is averaged between 400 and 1000 hPa over the year 2004). As we have shown, the mean bias is positive for most stations (MB > 10 \(\mu g m^{-3}\)). Large RMSE are seen in northern high latitudes (< 50 \(\mu g m^{-3}\)) and in two stations from the US, Europe and Japan present an RMSE around 30 \(\mu g m^{-3}\) and the tropics and subtropics are regions with lower errors, i.e. RMSE below 30 \(\mu g m^{-3}\). The highest correlations are seen in polar regions.

6 Conclusions

A new global chemical transport model, NMMB/BSC-CTM, has been presented. A comprehensive description has been provided for the different components. The model consists of a comprehensive description and evaluation of the gas-phase chemical module coupled online within the NMMB atmospheric driver. This model, which includes chemistry component of the NMMB-MONARCH model version 1.0 at global scale. The model considers 51 chemical species and solves 156 reactions, and simulates the global distributions of ozone and its precursors, including CO, NO\(_x\), and VOCs. The model simulation presented here was configured with a horizontal resolution of 1.14° x 1.14°, with 64 vertical layers and a top of the atmosphere at 1 hPa. Emissions from Modeled tropospheric ozone and related tracers were evaluated for year 2004 using data from surface-monitoring stations, ozonesondes, satellite and aircraft campaigns. We used emissions from ACCMIP (Lamarque et al., 2010) are considered and include for fossil fuel combustion, biofuel, biomass burning, soil and oceanic emissions. Biogenic emissions are calculated online with the MEGANv2.04 model (Guenther et al., 2006). In this simulation, aerosols are neglected, thus, no interaction between gas-phase and aerosol-phase is considered. We note that in this contribution, we omitted aerosols and lightning emissions; anthropogenic emissions disregard seasonality; and biomass burning emissions are not specific to 2004.

Modelled tropospheric ozone and related tracers have been evaluated for the year 2004 and compared with surface monitoring stations, ozonesondes, satellite and aircraft campaigns.
The evaluation of OH concentrations shows a good agreement with previous studies (Spivakovsky et al., 2000; Voulgarakis et al., 2013). The peak concentrations of OH seen in April and July at northern latitudes are slightly higher than the climatological mean calculated in Spivakovsky et al. (2000). This may be possibly explained by the fact that neglecting anthropogenic aerosols and secondary aerosol formation are neglected in this simulation; hence, a higher oxidized atmosphere is obtained may be leading to a more oxidized atmosphere due to higher photolysis when aerosols are not present. However, overall, the widespread positive ozone biases identified seem to be responsible for the higher OH concentrations.

The global annual mean burden of CO (399 Tg) is higher than in other studies, with larger concentrations located in the tropics (229.43 Tg CO). The model is in relatively good agreement with CO observations at the surface (daily correlations between 0.3-0.7), and shows negative biases at stations over Europe and Japan, and positive biases in Canada and Africa. The largest correlations are found in northern Europe, southern Africa and eastern Asia.

Concerning the vertical structure of CO, the model presents a good performance during the DJF and MAM, while positive biases are seen during JJA for most of the stations. In general, the model overestimates CO from the middle to the upper troposphere in most of the stations throughout the year. Significant underestimation of CO is seen in Beijing below 600 hPa. This result is similar to other evaluation studies, which indicates that emission inventories are not able to capture the extreme growth, a common result in other studies which strongly suggests an underestimation of anthropogenic emissions in China. The phase and amplitude of the seasonal cycles of CO at 800 and 500 hPa in NMMB/BSC-CTM, NMMB-MONARCH and MOPITT are quite similar.

Overestimations of CO are mainly located over west-central Africa, western South America, Indonesia and the surrounding Pacific and Indian oceans during the dry season. At 800 hPa, a significant negative bias is observed over the northern latitudes during winter. These results are most likely related to errors in anthropogenic and biomass burning emission inventories, where the magnitude and the location of emission are not correctly represented. In addition, CO production from VOCs biogenic emissions, calculated online and depending on meteorological variables such as radiation, might be overestimated too, due to the lack of aerosol attenuation of radiation.

Nitrogen oxide abundances are well simulated in almost all locations. Looking at the annual time series of NO$_2$ in Europe, the model captures the higher peaks during winter, although a positive bias is observed during summer. Nitrogen compounds are more sensitive to errors in emissions than other pollutants. We note that the emission inventory neglects seasonal variations for land-based anthropogenic emissions, and therefore we do not account for the potential reduction of NO$_x$ emissions during summer. Over Asia, there is a negative bias of NO$_x$ from March to August, probably due to underestimated emissions in this area. Vertical profiles of NO$_x$ are in good agreement with the observed values are really well captured by the model, although there is some underestimation in the upper troposphere, possibly due to the lack of lightning NO$_x$ emissions. Vertical profiles of PAN and HNO$_3$ were also compared with observations. Some agreement is seen in these vertical profiles, although the model has a tendency to overestimate, HNO$_3$ wet deposition fluxes tend to be underestimated, and are better captured in the US compared to Europe and Asia.
The comparison with observed NO₂ VTC from SCIAMACHY shows that the model reproduces the seasonality and the spatial variability reasonably well, capturing higher NO₂ over the most polluted regions. However, the results show a tendency to underestimate NO₂ VTC in big cities, especially during DJF and SON, possibly due to a low bias in the NO-background emissions. The biomass burning cycle is well captured by the model with higher NO₂ VTC in central Africa during DJF and in South America during JJA.

The ozone burden (348 Tg O₃) is in good agreement with other estimates from state-of-the-art global atmospheric chemistry models. The ozone burden in the southern extratropics is higher in our model, suggesting that higher CO concentrations in the southern hemisphere SH could lead to excessive production of ozone in this area. It seems unlikely that the positive ozone biases are caused by too much STE. STE is in good agreement (384 Tg O₃) is consistent with other evaluation studies. In addition, STE has stronger effects in the upper troposphere, hence, the related. Therefore, biases should increase with height, which is not the case in our simulations.

The surface O₃ results show a reasonable agreement with the observations, with significant positive biases (15 ppb) from May to October in the regions of the US and Japan. Surface O₃ concentrations are very sensitive to the emissions; consequently, the variability of ozone concentrations can be enhanced by improving the spatio-temporal distribution of the ozone precursor emissions.

The model captures the spatial and seasonal variation of observed background tropospheric O₃ profiles with a positive bias of 5-20 ppb throughout the troposphere in most of the regions during the whole year. The positive bias may be due to the significant overestimation of CO, especially in the free troposphere. The reason for this positive ozone bias, potential biases in stratospheric O₃ or the lack of halogen and aerosol chemistry.

In summary, NMMB/BSC-CTM the NMMB-MONARCH provides a good overall simulation of the main species involved in tropospheric chemistry, although with some caveats that we have highlighted here. Future versions of the model will address problems identified in this study and will include the effect of aerosols in the system.

7 Code Availability

Copies of the code are readily available upon request from the corresponding authors.

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Science Research School (ESSReS), an initiative of the Helmholtz Association of German research centres (HGF) at the Alfred Wegener Institute for Polar and Marine Research. All the numerical simulations were performed with the MareNostrum Superccomputer hosted by the Carlos Pérez García-Pando acknowledges the AXA Research Fund for sustained support. The authors thankfully acknowledge the computer resources at MareNostrum and the technical support provided by Barcelona Supercomputing Center (RES-AECT-2015-1-0007). Comments from two anonymous reviewers are gratefully acknowledge. We also thank Beatriz Monge-Sanz for providing the COPCAT coefficients.
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Figure 1. Stations used for the evaluation of the NMMB/BSC-CTM-NMMB-MONARCH model. On the left, surface-monitoring rural stations of $O_3$ (blue triangle), CO (red circle), $NO_2$ (green square cross) and $NO_x$ (black diamond) are shown. Moreover, wet deposition of HNO$_3$ measurement locations (yellow cross) are presented. On the right, locations of the different ozonesondes used (O$_3$ vertical profiles) are shown. Ozonesonde measurement locations are grouped by the following regions: NH Polar (brown circle), Canada (cyan circle), W. Europe (purple circle), USA-US (pink circle), Japan (orange circle), SH Midlat (blue circle), SH Polar (green circle), NH Subtropics (black circle), W. Pacific (red circle), Equator (yellow circle) and Others (grey circle). In addition, CO vertical profiles from the aircraft campaign MOZAIC (pink square) are presented. Finally, large rectangles show areas for the climatology analysis (NO$_x$, PAN and HNO$_3$) for Boulder (blue), Churchill (red), China (orange), Hawaii (black) and Japan (purple).
Figure 2. Zonally monthly mean OH concentrations ($10^5$ molecules/molec cm$^{-3}$) for January, April, July and October by the NMMB/BSC-CTM-NM-B-MONARCH model.
Figure 3. Time series of CO daily mean concentration in \( \mu g m^{-3} \) averaged over all the rural WDCGG stations used. Observations are depicted with a solid red line and model data in with a solid black line. Bars show the 25th-75th quartile interval for observations (orange bars) and for the model simulation (grey bars).

Figure 4. CO spatial distribution of mean bias (MB, ppb) (left panel), correlation (r) (middle panel) and root mean square error (RMSE, \( \mu g m^{-3} \) ppb) (right panel) at all the rural WDCGG stations used.
Figure 5. CO vertical profile seasonal averages over Frankfurt, Beijing and Atlanta (from left to right) for the whole year 2004–2004 from MOZAIC. Observations are depicted with a solid red line and model data in with a solid black line. Horizontal lines show the standard deviations. The number of observations flights is given on the top of each plot.
Figure 6. Comparison of modeled CO mixing ratio at 800 hPa against satellite data (MOPITT) for (from top to bottom: DJF for December-January-February, MAM for March-April-May, JJA for June-July-August and SON for September-October-November) for the whole year 2004. NMMB-MONARCH data is displayed in the left panel, MOPITT data in the middle panel and the bias in the right panel.
Figure 7. Time series of NO$_2$ (top) and NO$_x$ (bottom) daily mean concentration averaged over all the rural EMEP and EANET stations, respectively, used in $\mu$g m$^{-3}$ ppb. Observations are depicted with a solid red line and model data with a solid black line. Bars show the 25th-75th quartile interval for observations (orange bars) and for model simulation (grey bars).
Figure 8. NO$_2$ (top) NO$_x$ (bottom) and spatial distribution of mean bias (MB, ppb) (left panel), correlation ($r$) (middle panel) and root mean square error (RMSE, µg m$^{-3}$ ppb) (right panel) at all rural EMEP and EANET stations, respectively. Stations used.
Figure 9. Comparison of modeled (black lines) and observed (red lines) vertical profiles of NO\textsubscript{X} (first column), HNO\textsubscript{3} (second column) and PAN (third column) for several regions over the US, China, Hawaii and Japan. Horizontal lines show the standard deviations.
Figure 10. Comparison of modeled NMMB/BSC-CTM NO₂ vertical tropospheric columns against satellite data (SCIAMACHY) for (from top to bottom) DJF, MAM, JJA, and SON for the entire year 2004. NMMB/BSC-CTM 2004, NMMB-MONARCH data is displayed in the left panel, SCIAMACHY data in the middle panel and the bias in the right panel.
Figure 11. Time series of O₃ daily mean concentration averaged over all the rural WDCGG, CASTNET, EMEP and EANET stations (from top to bottom) used in ppb. Observations are depicted with a solid red line and model data with a solid black line. Bars indicate the 25th-75th quartile interval for observations (orange bars) and for model simulation (grey bars).
Figure 12. Spatial distribution of the O₃ mean bias (MB, ppb) (left panel), correlation (r) (middle panel) and root mean square error (RMSE, µg m⁻³ ppb) (right panel) at all rural WDCGG, CASTNET, EMEP and EANET stations (from top to bottom) stations used.
Figure 13. Comparison of ozonesonde measurements (red lines) and simulated (black lines) seasonal vertical profiles of O₃ (ppb) and standard deviations (horizontal lines). The region name and the number of stations, using brackets, are given above each plot between brackets.
Figure 14. Mean tropospheric ozone bias spatial distribution of NMMB/BSC-CTM minus ozonesondes (MB, ppb) (left panel), correlation (middle panel) and root mean square error (RMSE, µg m⁻³ ppb) (middle panel) and correlation (right panel) for the whole year 2004, averaged between 400-1000 hPa. The diameter of the circles indicates the number of profiles over the respective stations. The bias is calculated as model minus observation.
Table 1. Model characteristics and experiment configuration

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| Physics                       | Ferrier microphysics (Ferrier et al., 2002)   |
| BMJ cumulus scheme            | BMJ cumulus scheme (Betts and Miller, 1986)   |
| MYJ PBL scheme                | MYJ PBL scheme (Janjic et al., 2001)          |
| LISS land surface model       | LISS land surface model (Vukovic et al., 2010) |
| RRTMG radiation               | RRTMG radiation (Mlawer et al., 1997)         |

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Table 2. Emissions totals by category for 2004 in Tg(species)/year -- for 2004. Anthropogenic and biomass burning emissions applied in this study are based on Lamarque et al. (2013). Ocean and soil natural emissions are based on the POET global inventory (Granier et al., 2005). Biogenic emissions are computed online from the MEGAN (Guenther et al., 2006).

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Table 3. Ozone sondes main information about the ozonesondes used in this model evaluation study, including the year 2004. Location of these ozonesondes is displayed in the third location and fourth columns. Columns 6-9 display the number of available measurements for each season (DJF for December-January-February, MAM for March-April-May, JJA for June-July-August and SON for September-October-November).

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<td>89.6W</td>
<td>Equador</td>
<td>7</td>
<td>4</td>
<td>10</td>
<td>13</td>
</tr>
<tr>
<td>Boulder</td>
<td>USA</td>
<td>40.0N</td>
<td>105.26W</td>
<td>USA</td>
<td>12</td>
<td>11</td>
<td>17</td>
<td>16</td>
</tr>
<tr>
<td>Trinidad Head</td>
<td>USA</td>
<td>40.8N</td>
<td>124.2W</td>
<td>USA</td>
<td>4</td>
<td>7</td>
<td>5</td>
<td>8</td>
</tr>
<tr>
<td>Suva</td>
<td>Fiji</td>
<td>18.13E</td>
<td>178.4E</td>
<td>W Pacific</td>
<td>13</td>
<td>12</td>
<td>48</td>
<td>11</td>
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</tbody>
</table>
Table 4. MOZAIC aircraft information used in this model evaluation for including the year 2001. Location of the MOZAIC measurements is displayed in the third, and fourth columns. Columns 5-8 display the number of available measurements for each season (DJF for December-January-February, MAM for March-April-May, JJA for June-July-August and SON for September-October-November).

<table>
<thead>
<tr>
<th>Station</th>
<th>Country</th>
<th>Latitude</th>
<th>Longitude</th>
<th>DJF</th>
<th>MAM</th>
<th>JJA</th>
<th>SON</th>
</tr>
</thead>
<tbody>
<tr>
<td>Abu Dhabi</td>
<td>United Arab Emirates</td>
<td>24.44N</td>
<td>54.65E</td>
<td>11</td>
<td>17</td>
<td>58</td>
<td>20</td>
</tr>
<tr>
<td>Atlanta</td>
<td>&amp;</td>
<td>United States</td>
<td>33.63N</td>
<td>84.44W</td>
<td>24</td>
<td>130</td>
<td>168</td>
</tr>
<tr>
<td>Beijing</td>
<td>China</td>
<td>40.09N</td>
<td>116.6E</td>
<td>5</td>
<td>12</td>
<td>23</td>
<td>17</td>
</tr>
<tr>
<td>Cairo</td>
<td>Egypt</td>
<td>30.11N</td>
<td>31.41E</td>
<td>19</td>
<td>16</td>
<td>2</td>
<td>8</td>
</tr>
<tr>
<td>Caracas</td>
<td>Venezuela</td>
<td>10.6N</td>
<td>67W</td>
<td>21</td>
<td>9</td>
<td>9</td>
<td>21</td>
</tr>
<tr>
<td>Dallas</td>
<td>&amp;</td>
<td>United States</td>
<td>32.9N</td>
<td>97.03W</td>
<td>8</td>
<td>24</td>
<td>24</td>
</tr>
<tr>
<td>Douala</td>
<td>Cameroon</td>
<td>4.01N</td>
<td>9.72E</td>
<td>7</td>
<td>0</td>
<td>10</td>
<td>6</td>
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<tr>
<td>Frankfurt</td>
<td>Germany</td>
<td>50.02N</td>
<td>8.53E</td>
<td>169</td>
<td>295</td>
<td>286</td>
<td>192</td>
</tr>
<tr>
<td>New Delhi</td>
<td>India</td>
<td>28.56N</td>
<td>77.1E</td>
<td>30</td>
<td>24</td>
<td>72</td>
<td>38</td>
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<tr>
<td>New York</td>
<td>&amp;</td>
<td>United States</td>
<td>40.7N</td>
<td>74.16W</td>
<td>79</td>
<td>23</td>
<td>41</td>
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<tr>
<td>Niamey</td>
<td>Niger</td>
<td>13.48N</td>
<td>2.18E</td>
<td>4</td>
<td>0</td>
<td>12</td>
<td>12</td>
</tr>
<tr>
<td>Portland</td>
<td>&amp;</td>
<td>United States</td>
<td>45.59N</td>
<td>122.6W</td>
<td>5</td>
<td>8</td>
<td>5</td>
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<tr>
<td>Tehran</td>
<td>Iran</td>
<td>35.69N</td>
<td>51.32E</td>
<td>8</td>
<td>11</td>
<td>31</td>
<td>18</td>
</tr>
<tr>
<td>Tokyo</td>
<td>Japan</td>
<td>35.76N</td>
<td>140.38E</td>
<td>38</td>
<td>50</td>
<td>56</td>
<td>34</td>
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</table>

Table 5. Description of additional aircraft campaign data. Location of the measurements campaigns is displayed in the third, including location and fourth columns. The fifth column lists the date of these campaigns.

<table>
<thead>
<tr>
<th>Region Name</th>
<th>Expedition</th>
<th>Latitude</th>
<th>Longitude</th>
<th>Date</th>
</tr>
</thead>
<tbody>
<tr>
<td>Boulder</td>
<td>TOPSE</td>
<td>37.47N</td>
<td>110-90W</td>
<td>5 February to 23 May 2000</td>
</tr>
<tr>
<td>Churchill</td>
<td>TOPSE</td>
<td>47.65N</td>
<td>110-80W</td>
<td>5 February to 23 May 2000</td>
</tr>
<tr>
<td>China</td>
<td>TRACE-P</td>
<td>10.30N</td>
<td>110-130E</td>
<td>24 February to 10 April 2001</td>
</tr>
<tr>
<td>Hawaii</td>
<td>TRACE-P</td>
<td>10.30N</td>
<td>170-150W</td>
<td>24 February to 10 April 2001</td>
</tr>
<tr>
<td>Japan</td>
<td>TRACE-P</td>
<td>20.40N</td>
<td>130-150E</td>
<td>24 February to 10 April 2001</td>
</tr>
<tr>
<td>Tahiti</td>
<td>PEM-Tropics-B</td>
<td>20S-0</td>
<td>160W-130W</td>
<td>6 March to 18 April 1999</td>
</tr>
<tr>
<td>Ireland</td>
<td>POLINAT-2</td>
<td>50-60N</td>
<td>15-5W</td>
<td>19 September to 25 October 1997</td>
</tr>
</tbody>
</table>

Table 6. Annual mean burden of tropospheric CO (Tg CO) in NMMB/BSC-CTM the NMMB-MONARCH, MOZART-2 and TM5 and C-IFS global models.

<table>
<thead>
<tr>
<th>Model</th>
<th>Global</th>
<th>NH</th>
<th>SH</th>
<th>Trop.</th>
<th>N. Extratrop.</th>
<th>S. Extratrop.</th>
<th>Dry depo.</th>
<th>Reference</th>
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</thead>
<tbody>
<tr>
<td>NMMB/BSC-CTM</td>
<td>399</td>
<td>221</td>
<td>177</td>
<td>229</td>
<td>101</td>
<td>67</td>
<td>24</td>
<td>This study</td>
</tr>
<tr>
<td>NMMB-MONARCH</td>
<td>351</td>
<td>210</td>
<td>142</td>
<td>199</td>
<td>102</td>
<td>59</td>
<td>2</td>
<td>Horowitz et al. (2003)</td>
</tr>
<tr>
<td>MOZART-2</td>
<td>353</td>
<td>-</td>
<td>-</td>
<td>188</td>
<td>106</td>
<td>59</td>
<td>184</td>
<td>Huijgen et al. (2010)</td>
</tr>
<tr>
<td>TM5</td>
<td>-</td>
<td>-</td>
<td>-</td>
<td>-</td>
<td>-</td>
<td>-</td>
<td>-</td>
<td>Fleming et al. (2015)</td>
</tr>
<tr>
<td>C-IFS</td>
<td>361</td>
<td>-</td>
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<td>-</td>
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</table>
Table 7. Annual mean burden, dry deposition of tropospheric O$_3$ and stratospheric inflow (Tg O$_3$) for the NMMB/BSC-CTM, NMMB-MONARCH, MOZART-2, TM5, LMDz-INCA, GFDL AM3, C-IFS global models, and two different Multimodel multi-model ensembles (including 25 and 15 global models each).

<table>
<thead>
<tr>
<th>Model</th>
<th>Burden</th>
<th>Dry deposition</th>
<th>Stratospheric inflow</th>
<th>Reference</th>
</tr>
</thead>
<tbody>
<tr>
<td></td>
<td>Global</td>
<td>NH</td>
<td>SH</td>
<td>Trop.</td>
</tr>
<tr>
<td>NMMB/BSC-CTM, NMMB-MONARCH</td>
<td>348</td>
<td>189</td>
<td>158</td>
<td>171</td>
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<tr>
<td>MOZART-2</td>
<td>362</td>
<td>203</td>
<td>159</td>
<td>203</td>
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<tr>
<td>TM5</td>
<td>312</td>
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<td>165</td>
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<td>LMDz-INCA</td>
<td>303</td>
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<td>125</td>
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<td>GFDL AM3</td>
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<td>C-IFS</td>
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<tr>
<td>Multimodel Multi-model</td>
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<td>±39</td>
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<td>Multimodel Multi-model</td>
<td>337</td>
<td>±23</td>
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