

## ***Interactive comment on “The TOMCAT global chemical transport model: Description of chemical mechanism and model evaluation” by Sarah A. Monks et al.***

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Author Response to Reviewer Comments (RC2)

The authors would like to thank the reviewer for taking the time to read and comment on the submitted manuscript. The comments are repeated below (in quotation marks) followed by our responses. We have taken the comments on board and addressed them as described.

"The manuscript concerns the description and validation of the TOMCAT Chemistry Transport Model. One point which is not clear is what significant model improvements has been made, compared to recent versions used for intercomparison studies, that

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warrants a new benchmarking study at this point in time. There has been the addition of higher Volatile Organic Compounds and new Isoprene/Monoterpene chemistry, with no significant updates to the dynamics, microphysics or transport in the CTM. Currently the manuscript is written as such that the reader has no idea as to whether these chemistry updates improve the model performance, where a 'before' and 'after' simulation for a chosen year is not provided. Two arbitrary simulation years are presented (2000 and 2008), being run by different emissions and driven by different meteorology. Some of the validation is presented in a climatological sense, whereas some is presented for the simulation year in general, with some data being for different time periods. In general TOMCAT exhibits biases but the reasons given are not quantitative but rather speculative as the rigor of the analysis is not sufficient enough to provide concrete answers, with conclusions being used from previous studies presumably using an identical model version. One could exploit rarely used data such as from SAFARI, MINATROC and/or THESEO campaigns, which provides an unique snapshot across different chemical regimes if more data is necessary. In order to improve this manuscript I recommend a major revision following either (i) a focus on one year presumably the year 2000 using yearly specific measurements or (ii) do a sensitivity study with and without new chemistry for a chosen year to show effects and (hopefully) improvements."

The main purpose of this paper is to document the version of the TOMCAT chemical mechanism that is now being used for scientific studies. Whilst this scheme has been used in some recent scientific studies (Emmons et al., 2015 and Richards et al., (2013), the current chemical mechanism has only been previously documented in a PhD thesis (Monks, 2011). Emmons et al., (2016) and Richards et al., (2013) only compared the model to data in limited regions and over limited time periods. Therefore, a benchmarking paper is warranted, where evaluation of key components can be made globally throughout the year. Users of the TOMCAT model and GLOMAP-TOMCAT coupled model would also benefit from the chemical mechanism being documented as the only other published description of the TOMCAT chemistry scheme is described by Arnold

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et al., (2005), which is now obsolete. We see little value in showing an evaluation of this old scheme due to it not being used anymore, and therefore only evaluate the current scheme. However, a short discussion has been added to Section 2.1 (Section 2.1.1 Impacts of additional VOC chemistry) to describe the impact of changing the chemistry from Arnold et al., (2005) to the current scheme. These results have been taken from Monks (2011) and show the change in CO, O<sub>3</sub> and OH. The overall changes are an increase in the burdens of CO and O<sub>3</sub> and a decrease in OH. As with all models, TOMCAT is negatively biased in CO, so an increase in the CO burden reduces this bias for the TOMCAT model. Simulated global mean OH is also higher than estimates calculated from methyl chloroform, therefore a decrease in OH provides a better simulation of OH.

Following similar comments from Reviewer 1 on confusion caused by presenting the two simulations from two different years, we have chosen to remove the year 2000 simulation. The aircraft climatology has also been removed and we show surface and aircraft comparisons for the year 2008 only. Please see our response to Reviewer 1 for more details.

Main comments: "Are the 31 levels having a higher resolution in the troposphere? How many layers describe the UTLS and Stratosphere?" The levels have a higher resolution in the boundary layer/lower troposphere and near the UTLS. A figure has been added to the manuscript to show this.

"Is the meteorology smoothed between 6 hourly updates or are step edges applied to e.g. H<sub>2</sub>O fields?" The meteorological data is linearly interpolated between the 6-hour updates. This information has been added to the manuscript.

"Emission inventories: In order to attain the most accurate simulations using a CTM requires time evolving emission input data which e.g. captures the development in the transport sector. In TOMCAT aircraft NO<sub>x</sub> emissions come from the QUANTIFY project for the fixed year of 2002 which will surely underestimate the contributions in

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aircraft NO<sub>x</sub> over the simulation period (year 2008). A valid reason of using these fixed emissions is not given where e.g. the MACCity inventory provides yearly specific aircraft NO<sub>x</sub> estimates." I was not able to find any yearly specific MACCity aircraft emissions to evaluate this discrepancy properly. As MACCity emissions are simply a linear interpolation between the data I am using and the RCP 8.5 2005 and 2010 estimates, I looked at the RCP 8.5 emission dataset to assess by how much emissions have changed. Annual aircraft emissions of NO<sub>x</sub> are estimated to have increased from 2.8 Tg/yr to 3.1 Tg/yr between 2000 and 2010 (10 –year period). Assuming a linear increase, this would be ~0.2 Tg (7%) change in aircraft emissions in the model over a 7-year period. We feel that the impact of using 2002 estimates instead of 2008 estimates will only cause a maximum difference of a few ppbv in O<sub>3</sub> in the UT. This is because simulations with and without aircraft emissions have shown a maximum of 7 ppbv difference during the summer (Gauss et al., 2006). For this reason, we feel that this does not warrant a change in the emissions at this time but will be worth changing them in the future when the emissions are being updated.

"Are any burning heights applied for the biomass burning emissions?" No.

"Heterogeneous conversion: What is happening to the conversion of N<sub>2</sub>O<sub>5</sub> into HNO<sub>3</sub> on cloud surfaces? How do you calculate the available surface area density? Why is there only one heterogeneous reaction when HO<sub>2</sub> conversion on aerosols is also now considered important?" There is no treatment of N<sub>2</sub>O<sub>5</sub> uptake on cloud surfaces currently due to the use of climatological clouds in the model. This is something that will be considered in future versions of the model. Surface area density is calculated from offline simulated aerosol radius and number density for the 5 different aerosol types that are described in the paper. HO<sub>2</sub> uptake onto aerosol is indeed important. The code exists for this to be included in the future but it has not been tested currently and is therefore not implemented in this version of the model. These points have been clarified in the manuscript.

"Photolysis rates: How old is the absorption spectral data and quantum yields em-

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ployed?. Many changes to the recommendations have been made since the 1980's but no details are given as to what improvements have been made. Photolysis drives tropospheric chemistry therefore an accurate description of diurnal variability is needed to capture e.g. good NO<sub>2</sub> lifetimes. How are cloud and aerosol treated in terms of optical density through the column? Sun-spectrum used with any modifications due to the sun-cycles? As this is a benchmark paper it is not attractive that the reader has to trawl through the literature to get such details." The photolysis scheme for the tropospheric chemistry scheme described here shares the 'library' of photochemical data with the stratospheric scheme (not discussed) and is regularly updated. Sun-cycles are not accounted for. More details of the model photolysis scheme have been added into the revised paper (see also response to Reviewer 1).

"Given that the chemical scheme employed is for the description of tropospheric processes (e.g. no CFC's), how is the stratosphere constrained in order to get correct seasonality in the overhead O<sub>3</sub> column and thus actinic flux profile?" There is no stratospheric chemistry in this version of the model. We constrain stratospheric O<sub>3</sub> with offline generated fields from the Cambridge 2-D model. A description of this has been added in Section 2.1 (also see RC1).

"Why use GOME-2 and not GOME O<sub>3</sub> data used for the year 2000 and 2008? Considering the current debate about the (potential) recovery of stratospheric O<sub>3</sub> it appears the wrong decade is being used for validation. Why use an O<sub>3</sub> sonde climatology from a completely different time period? This make little sense considering the choice of satellite data concerning O<sub>3</sub>. Both 2000 and 2008 annual means should be composed from the sondes and used to provide a better assessment of the vertical O<sub>3</sub> profiles as the reader is unable to assess the ability of TOMCAT to capture inter-annual variability in the distributions. Were O<sub>3</sub> measurements extracted on identical days for a valid comparison?" We have removed RUN\_2000 so using GOME data is no longer required. The GOME Ozone record experienced a problem where the tape recorder failed on the ERS-2 satellite on 22nd June 2003. Therefore, data could only be

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download while it was within direct line of sight of a ground station resulting in reduced spatial. GOME-2 has global coverage for 2008 with improvements in the resolution and quality of the data. That is why we chose to retain RUN\_2008.

"Why lump NO and NO<sub>2</sub> together?. Seasonality in NO<sub>2</sub> mixing ratios is a proxy for the performance of the photolysis routine which is masked by lumping both oxides together. Even if there is a persistent bias the seasonality should be captured in some way." This has been changed so we now show NO<sub>2</sub> comparisons.

"Have the satellite comparisons for e.g. NO<sub>2</sub> been done at the local overpass time for the days when measurements are available or in more ad-hoc way?" TOMCAT composition data have been co-located in time and space to both the GOME-2 Ozone and OMI NO<sub>2</sub> products. Here the closest model grid box to the satellite pixel is sampled, within 3 hours of the satellite daytime overpass (e.g. 13.30 LT for OMI) as the model output is every 6 hours.

Specific comments: "Pg 8, In 17: Providing seasonal means would be more informative for the reader, where significant changes should occur in the hemispheric zonal means. " We have chosen to show annual means so they can be compared to the papers that are referenced in the discussion (Young et al., 2013; Voulgarakis et al., 2013), which show annual mean concentrations of O<sub>3</sub> and OH from the mean of multiple models. Seasonal O<sub>3</sub> and CO are shown in comparisons to observational data.

"Pg 8, In 45: Please introduce a table with the O<sub>3</sub> diagnostics (Burden, Lifetime, Strat-Trop exchange and deposition) and then place in context using the new multi-model means. Also for the other gases such as CO and CH<sub>4</sub>, as information is scattered throughout the text." A table has been added for the O<sub>3</sub> burden, global mean OH concentration and CH<sub>4</sub> lifetime. Some O<sub>3</sub> diagnostics (strat-trop exchange and deposition) have not been calculated as diagnostics from these experiments unfortunately.

"Pg 9, In 40: Indeed, it would be even more illuminating by providing a Table of global mean OH from various studies." A table has been added.

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"Pg 13, In 32: Not surprising considering the mitigation practices over the last decade or two. This bias is potentially exaggerated by using non-yearly specific measurements. Many studies have focused on VOC emissions so can the authors state whether this is an artifact of CTM's in general?" These comparisons are no longer included. ARCTAS 2008 data and 2008 surface data show a similar discrepancy, therefore, it is not due to a mismatch between emissions in the model and the years that the observations were made. This appears to be an artifact in CTMs in general. The paper already referenced (Emmons et al., 2015) in the discussion shows that this is widespread across different models with different OH concentrations so it likely due to underestimated emissions.

"Pg 15, In 15: Looking at Figure 17 I can see discrepancies of >100 percent for some of the points. Better to discuss seasonality to identify which season has the largest bias given that PAN is temperature sensitive. " The aircraft climatology has been removed so this figure is no longer included.

"Pg 15, In 15: " : : : may be too high : : : ". If a comparison has been made then surely it either is or isn't too high for this period. " The 'may be too high' phrasing was due to the mismatch in the aircraft data climatology in terms of years of observations and the year which the model simulation represents. As mentioned the aircraft climatology has been replaced with yearly specific aircraft observations.

"Pg 15, In 19: What is the relevance of GEOS-chem to this paper? What about the multitude of other CTM's? " This section has been removed as I no longer use the aircraft data climatology. GEOS-Chem was discussed because they used the aircraft data climatology to evaluate PAN in a similar way and there are limited papers that evaluate simulated PAN in such a manner.

"Figure 14 and 15: What is the motivation for lumping the measurements but not the model results? There are trends in surface observations related to emission trends. Is the bias larger for 2008 than 2000 i.e. does the deviance increase with time due to incorrect emission estimates?" No longer relevant, 2008 observations are now used

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instead of a climatology.

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Interactive comment on Geosci. Model Dev. Discuss., doi:10.5194/gmd-2016-212, 2016.

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