

## ***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 (RC1)

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 presented evaluation of TOMCAT is reasonably comprehensive but the juxtaposition of the two runs is of little scientific value. Such an inter-comparison experiment, as any scientifically sound experiment, should only differ in the specific aspects, which are

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under investigation. However, the two presented model runs differ in the year (2000 vs. 2008), in the biomass burning data set (GFED 3.1 vs. FINN), the anthropogenic emissions (ACCMIP vs Streets v1.2) and probably also in the prescribed CH<sub>4</sub>. Given the different years, not only the meteorology but also the biomass burning and VOC emissions will be different. It is not clear at all what reasonable conclusion can be drawn from the comparison of the two model runs. As the paper reports on the update of the chemical mechanism, one would expect that two model runs with and without this update, which are otherwise identical (i.e. w.r.t emissions, year, meteorology, CH<sub>4</sub>) are compared. One should choose a year for which there are many observations available from campaigns or satellite observations. The model description part is too short, in particular for the chemical mechanism. If this section – as the title suggest – is an important part of the paper, including only references or simply stating the fact of certain upgrades is not sufficient. It would be better to discuss the chemical mechanism in more detail and to give a motivation for the necessity and most importantly the impact of the upgrades."

The main purpose of this paper is to document the version of the TOMCAT chemical mechanism that is now being used for scientific studies. The current chemical mechanism has only been previously documented in a PhD thesis (Monks, 2011). This means that users of the TOMCAT model would benefit from the chemical mechanism being documented in a peer-reviewed journal which is easily accessible. The earlier version of the chemistry scheme described by Arnold et al., (2005) is now obsolete and we see little value in showing an evaluation of this old scheme. However, we have added in a short discussion to Section 2.1 (Section 2.1.1 Impacts of extended VOC chemistry), which describes the impact of changing the chemistry from Arnold et al., (2005) to the current scheme. These results have been taken from Monks (2011) and are used to show the change in CO, O<sub>3</sub> and OH. The description of the chemical mechanism has also been extended. The overall changes are an increase in CO and O<sub>3</sub> and a decrease in OH.

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On consideration of the reviewer comments, we agree that substantial confusion arises from the inclusion of both the RUN\_2000 and RUN\_2008 simulations, and so RUN\_2000 has been removed from the paper. Our original intention was to show the two separate RUN\_2000 and RUN\_2008 simulations in order to provide best representation of the year 2000 and 2008, respectively, and make use of different datasets that are more representative of early years (e.g. the aircraft climatology) and later years (satellite and surface data). We believed that this would be useful in identifying any errors that may exist in the model that are not systematic and may be confined to the set-up for the particular year. This was useful in showing species like PAN and NO<sub>x</sub> are very sensitive to how the model is set-up. However, since these simulations use very different emissions data and meteorology that differs according to year, as highlighted, it is difficult to know the exact cause of differences in the model (meteorology versus emissions). We have also removed comparisons to the aircraft climatology (data for 1992-2001) as this dataset was more comparable to RUN\_2000. We have kept RUN\_2008 and have performed some additional comparisons to the 2008 POLARCAT campaign aircraft data to provide some data for other VOCs and NO<sub>y</sub> species that are not regularly measured. The surface data is now also shown for the year 2008 only.

Specific remarks:

"P1 Abstract seems too long. It should be a single paragraph." The abstract has been shortened.

"P1 L18: The term "boundary conditions" is a bit confusing as you only change emissions. Better say "emissions" as deposition is not considered. The fixed CH<sub>4</sub> can be described as "effective emissions" Sentence no longer applies as RUN\_2000 has been removed.

"P2 L25: As a CTM does not simulate winds, it seems better to say the transport is "driven" rather than constrained." Changed to driven.

"P3 L4: Mention the motivations of the update. Introduce the choice of the setup of

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the different model runs you want to compare." Changed to: "This paper summarises the latest tropospheric chemical mechanism scheme used in TOMCAT (Section 2). The scheme gives a more detailed representation of hydrocarbon chemistry than previously included in the model, with the inclusion of the oxidation of ethene, propene, butane, toluene and monoterpenes. Alkenes have the greatest potential for forming O<sub>3</sub> (Saunders et al., 2003), and previously, isoprene was the only alkene treated in the TOMCAT model. In addition, a more extensive VOC scheme makes it possible to couple the TOMCAT tropospheric chemistry to the formation of secondary organic aerosol in future versions of the GLOMAP aerosol model (Mann et al., 2010)."

"P3 L33: Aircraft emissions should be mentioned together with the rest of the emissions in section 2.2" This has been moved.

"P4 L12: Please provide more detail on the chemical scheme and motivate the several updates of the VOC chemistry. This would be of interest for the scientific community." This has been extended: "The previously documented TOMCAT tropospheric chemical mechanism included odd oxygen (O<sub>x</sub>), reactive nitrogen (NO<sub>y</sub>), carbon monoxide, methane, ethane, propane, acetaldehyde, acetone and formaldehyde chemistry (Arnold et al. 2005). TOMCAT also includes oxidation of isoprene based on the Mainz Isoprene Mechanism (MIM) scheme (Pöschl et al., 2000). The implementation of this scheme into TOMCAT is described by Young (2007). Isoprene is one of the largest single sources of VOCs to the atmosphere, accounting for around a third of total natural and anthropogenic VOC emissions (Guenther et al., 2006). After emission, isoprene is highly reactive and can influence O<sub>3</sub> concentrations both regionally (Chameides et al., 1988) and globally (Wang and Shallcross, 2000). O<sub>3</sub>, OH, CO and PAN from the condensed MIM scheme were found to agree within 10% of the concentrations calculated from a more explicit representation of isoprene chemistry in the Master Chemical Mechanism (Pöschl et al., 2000). However, significant uncertainties still exist in the representation of isoprene chemistry in models and chemical mechanisms will likely evolve in the future (Archibald et al., 2010; 2011; Squire et al., 2015). Most recently,

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the TOMCAT model chemistry has been expanded to include the emission and destruction of some C2-C7 unsaturated and aromatic hydrocarbons (ethene, propene, toluene and butane) based on the Extended Tropospheric Chemistry scheme (ExtTC), and monoterpenes based on the MOZART-3 chemical mechanism (Kinnison et al., 2007). Whilst this scheme has been used in the TOMCAT model for some scientific studies (e.g. Richards et al., 2013; Emmons et al., 2015), the expanded scheme was not fully documented or evaluated globally. This is the purpose of this study.

The updated model chemistry scheme has a total of 79 species, 16 of which are emitted (see Table 2), and approximately 200 chemical reactions. The biomolecular, termolecular and photolysis reactions are shown in Tables 3, 4 and 5, respectively. The chemical reactions are integrated in the model with a 15-minute chemical timestep using a software package, ASAD 5 (Carver et al., 1997), which allows the use of input files that contain the information listed in Tables 2-. The package allows fractional products and the use of families, which can be used for grouping very short-lived species together for transportation (see Table 2 for species treated as families). The bimolecular and termolecular kinetic rates are mostly taken from the 2000 – 2005 International Union of Pure and Applied Chemistry recommendations (<http://iupac.pole-ether.fr/>) and the Leeds Master Chemical Mechanism (MCM, 2004). Simplified ethene, propene and butane chemistry is based on von Kuhlmann et al., (2001), with reaction rates taken from IUPAC (Atkinson et al., 2006). Ethane chemistry adds 1 bimolecular reaction (R135-R136, Table 3) and 1 termolecular reaction (R17-19, Table 4) to TOMCAT. Oxidation of ethane by OH forms PrpeOO, a peroxy radical, which continues in the propene oxidation chain (von Kuhlmann, 2001). Propane oxidation adds 6 bimolecular reactions (R137-145, Table 3) and 1 termolecular reaction to TOMCAT (R17-19, Table 4) and butane adds 10 bimolecular reactions (R123-134, Table 3) and 5 photolysis reactions (R40a-46b, Table 5). Ethene, propene and butane emissions are emitted into the respective compounds, with no lumping of higher alkenes/alkanes. Toluene is emitted into a generic aromatic compound, AROM, which produces AROMO2 (peroxy radicals) and AROMOOH (hydroperoxides). Including AROM adds 11 additional

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bimolecular reactions to the model (R146-R156, Table 3) and two photolysis reactions (R48a-R48b, Table 5), with reactions rates taken from Folberth et al. (2006). Including ethene, propene, butane and toluene will account for missing sources of carbon in the model and produces peroxy radicals that are important for O<sub>3</sub> production (von Kuhlmann, 2001). Alkenes are particularly efficient at producing O<sub>3</sub>, with photochemical ozone creation potentials of 100 and 105 for ethene and propene, respectively, compared to 8.8 and 18.3 for ethane and propane, respectively (Saunders et al., 2003). The lumped monoterpene compound (C<sub>10</sub>H<sub>16</sub>) is treated as alpha-pinene, with emissions made up of the sum of alpha-pinene, beta-pinene, limonene, myrcene, ocimene, sabinene, and delta-3-carene. Including monoterpene chemistry adds 6 bimolecular reactions (R116-R122, Table 3) and one photolysis reaction (R26-27, Table 5) to the TOMCAT extended scheme. Monoterpenes are the second largest biogenic source of VOCs, after isoprene, and play an important role in OH, NO<sub>3</sub>, O<sub>3</sub> and aerosol chemistry (Atkinson et al., 2003; Fuentes et al., 2000). Accounting for monoterpenes also allows the coupling of the atmospheric chemistry model, TOMCAT, to the aerosol model, GLOMAP.

Photolysis rates are calculated online at each chemical timestep based on the two-stream method of Hough (1988), which considers both direct and scattered radiation. The scheme has total of 203 wavelength intervals from 120nm to 850 nm, though only wavelengths above 175 nm are used for stratosphere-troposphere studies. These wavelength intervals are the same as those employed in the TOMCAT stratospheric chemistry scheme (e.g. Chipperfield et al., 2015; Sukhodolov et al., 2016). The top of the atmosphere solar flux spectrum is fixed in time and there is no account of, for example, the 11-yr solar cycle in the standard model. This photolysis scheme is coupled with the TOMCAT model by using the model temperature and ozone concentration profiles. The scheme is also supplied with surface albedo, aerosol concentrations and monthly mean climatological cloud fields. This scheme was first used in this manner by Arnold et al., (2005). Previously, an offline approach was used where photolysis rates were calculated offline and then read in to the model (e.g. Law et al., 1998). Where

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possible photochemical data is taken from Sander et al. (2011), for species which are also relevant for the stratosphere. Otherwise photochemical data is generally taken from IUPAC (Atkinson et al., 2004; 2006). The UV absorption cross-sections for methyl hydroperoxide (MeOOH), which are from JPL (Sander et al., 2006), are used for the hydroperoxides produced from the oxidation of butane (BtOOH), toluene (AROMOOH) and monoterpene (TERPOOH). For the photolysis of ONIT, which represents organic nitrates produced from higher alkanes (currently only butane), cross-sections for methyl nitrate are used based on IUPAC recommendations (Atkinson et al., 2006). ONIT can be an important reservoir of reactive nitrogen (von Kuhlmann, 2001). Stratospheric concentrations of O<sub>3</sub> and NO<sub>y</sub> calculated offline by the 2-D Cambridge model (Pyle and Law, 1993) are read in by TOMCAT in the absence of stratospheric chemistry. Hydrogen, carbon dioxide, oxygen and nitrogen are kept at fixed global mean volume mixing ratios in the model. Water vapour is calculated from the specific humidity field read in from the meteorological input data.”

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"P4 L27: Please provide more detail, what uptake coefficients are used etc." We have added a table with the uptake coefficients.

"P5 L8: See my general comment. I think the two runs differ in too many and random aspect." RUN\_2000 has now been removed to avoid confusion.

"P5 L28: Explain how Streets differs from ACCMIP and what they have in common." The description of Streets has been extended.

"P5 L31: Are the biogenic emissions produced with exactly the same MEGAN model version and input." Changed to: "Monthly mean biogenic emissions are from the MACC (Monitoring Atmospheric Composition and Climate) project (MACCity), which provides simulated VOCs calculated offline by the Model of Emissions of Gases and Aerosols from Nature (MEGAN) v2.0 for a reference year 2000 (Guenther et al., 2006)."

"P5 L33: Explain the differences between FINN and GFED." No longer relevant.

"P5 L34: It is not clear if the same method to constrain CH4 is used in both runs." This no longer relevant as RUN\_2000 has been removed. A description of the two different ways of constraining methane is given earlier.

"P6 L8: Please add an explanation how you tackle the problem that different years are evaluated. Mention more clearly when use timely data and when climatological data" No longer relevant. RUN\_2000 has been removed and RUN\_2008 is compared to year

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2008 observations. Yearly specific observations are now used.

"P6 L39: Explain how you use the satellite for the different years. (GOME-2 was put in orbit only in 2006)" Satellite O3 data was only compared to RUN\_2008 as GOME-2 was only launched in 2006. RUN\_2000 has been removed so this should no longer be confusing.

"P7 L10: Should be "Surface data climatology"" No longer relevant. We use only 2008 observations.

"P7 L30: Should be "Ozone data climatology"" Changed.

"P8 L 37: Did you verify the importance for the suggest pathway. It should result in higher OH values. The surface ozone concentrations also depend strongly on deposition. Is any dry deposition active over the ocean? Please comment how H2O is represented in the model." No. This would be interesting to look at in the future. We have removed this explanation. Yes, O3 deposition does occur over the ocean and water vapour is now described in the model description section.

"P8 L 37: Please specify your choice of the tropopause." We have not used a tropopause here as we are simply discussing the general distribution of high/low ozone. We have changed the sentence to be clearer: "In the tropics, lower O3 concentrations are seen at 100-300 hPa due to a higher tropopause in this region and the uplift of air with low O3 within deep tropical convection. At around 20-40 S/N, evidence of the downward transport of stratospheric O3 by the Brewer-Dobson circulation (Butchart, 2014) can be seen.

"P9 L16-22: Consider putting the part on the observations in section 3." Moved.

"P10 L15: As you show the OH distribution of RUN\_2000 and RUN\_2008 please also discuss potential causes of the differences. Is the RUN\_2000, which seems to have larger biases also worse in other aspects? Are errors in OH related to errors in other species?" RUN\_2000 is no longer included.

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"P11 L14: Consider using the update of the MACCITY data suggest by Stein et al. (2014)" Whilst Stein et al., (2014) do show that increases in MACCity transportation emissions improve comparisons with some observations, they also show a similar performance when using the RETRO/REAS emissions. Streets emissions also uses REAS over Asia and other best estimates of emissions when regional inventories exist, with EDGAR to fill in the gaps. These emissions are a best guess for the year 2008 and were produced for the POLARCAT campaign. As we are using the POLARCAT data for comparison to RUN\_2008, we have chosen to retain the Streets anthropogenic emissions.

"P11 L14: Consider also the role of transport." Added sentence saying: "Transport errors in the model could also play a role, however, they are unlikely to cause such widespread biases of this magnitude."

"P 11 L16: According to your Figure 4 there is too much OH in Tomcat in NH and SH in the lower troposphere." We have changed the sentence to reflect this: "In addition to this, as mentioned in Section 4.2, OH in the TOMCAT model is most likely too high at the surface, particularly in the tropics, and the NH/SH OH ratio is higher than estimates based on observations. This is likely to influence the lifetime of simulated CO and will contribute to the NH and SH biases."

"P 11 L23: Do you discuss here RUN\_2008 (?). In this run the FINN data and not GFAS is used. Please be more specific. Is the difference between FINN and GFED (table 1) due to the different years used or is there a systematic bias between FINN and GFAS?" Yes, this is RUN\_2008 with FINN data. RUN\_2000 has been removed so there should be no more confusion in terms of which emissions are being used.

"P11 L33: Please specify what years do you use in the comparison. Note there is a CO trend of about 1%/yr (Worden et al. 2013)." MOPITT 2008 data is used to compare to RUN\_2008. It was previously noted in the Figure 4 caption, however, this has been made more clear in the text.

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"P13 L34: It would be nice to also show the OH observations at Hohenpeissenberg. As far as I know OH is measured there as well." We have been unable to locate any OH measurements for the year 2008. There were a few campaigns where measurements were made intensively and there was also some data that existed over a five-year period 1999-2003. We chose not to pursue this any further as we are not sure what this would add to the paper. It would be difficult to use them to evaluate the model due to OH being very short-lived and the model grid box being so large.

"P13 L42: Some comments on the assumed temporal profiles of the anthropogenic and biogenic VOC emissions would be interesting here." This is described in Section 2.2 so the reader could find out the emission temporal resolution here. As the measurements are made continuously and we are using monthly means to compare to the model we are not sure why this should be added to the discussion. If we were using observation made at specific time of the day, then yes this would be worth discussing.

"P14 L8: The relation between emissions and concentrations seems a bit trivial if you do not specify in more detail, why the VOC emissions are different." We are unable to find P14 L8. We assume that the comment refers to a difference in concentrations between RUN\_2000 and RUN\_2008. RUN\_2000 is no longer included so we believe this comment no longer applies.

"P14 L25: Please specify how r was calculated, i.e. using hourly, daily or monthly averages?" The correlations are calculated between the observations and the model data shown in Figure 15. This is monthly mean data. We have added this information into the figure caption.

"P14 L33: Is the overestimation over Europe also related to emissions?" Yes, it is most likely linked to the emissions. The largest biases occur over the Baltic and North Seas where ship emissions are located. This could indicate a positive bias in these emissions.

"P 14 L38 Which biomass data set and year you are referring to? (see above)." We

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are referring to FINN fire emissions as this is the RUN\_2008 comparison. We have clarified this. Also, now RUN\_2000 has been removed there will be less confusion.

"P15 L1 -5: It would be great if the paper could show in any way the benefit of this upgrade. This is unfortunately not the case with the presented two model runs." This is now discussed with the addition of a section showing the difference between two simulations with and without the extended chemistry. The new chemistry slightly increases CO in the model and therefore reduces the model negative bias. Global mean OH is reduced slightly to lower the positive bias compared with estimates based on methyl chloroform. Ozone is also higher due to the extra VOC emissions in the extended chemistry version of the model.

"P16 L 19: Is there an explanation for the OH differences? Has the model upgrade improved the OH bias? Is the 2000 run generally worse because of the larger biases against Spivakovsky?" No, it is still not fully understood why models and observations show this difference in the NH/SH OH ratio. We have expanded on this in the text. The model global mean OH is slightly lower with the extended chemistry, so yes this does improve the bias. RUN\_2000 has been removed.

"Figure 2 Use different colour range for O3 cross section to show more structures in the troposphere." Done.

"Figure 4. Rotate latitude vs CO graphs." While we understand it seems to make more sense that the y-axis is CO in terms of it being the dependent variable, we have chosen to keep the figure as it is so you can compare c to a/b and f to d/e by eye.

"Figure 6 Which colour for which run?" RUN\_2000 has been removed. Also, the label is already shown.

"Figure 9. Mark a-d) on panels. Consider including graph titles." Done.

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