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Geoscientific
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Discussions

Interactive comment on "The global chemistry transport model TM5: description and evaluation of the tropospheric chemistry version 3.0" *by* V. Huijnen et al.

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General comments

We thank the referee for his/her positive review of our manuscript. The reviewer questions whether the title should be changed to give a better reflection that not all chemical aspects of the model are evaluated. In our opinion the title is sufficiently concise. A model evaluation is limited by definition. In our case we choose to put the emphasis on tropospheric ozone and its key precursors, while for the model evaluation we use observational data that are frequently used in 3D global model studies. Therefore we think the title gives an acceptable summary of the contents of the evaluation paper.

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Actually, we include some evaluation of NMHC by comparing the simulated formaldehyde columns with satellite data, while the evaluation of nitrogen deposition depends on the simulation of ammonium and nitrate aerosols. As written in the manuscript, a more detailed evaluation of the aerosol chemistry in TM5 has recently been described in Aan den Brugh et al. (ACPD 2010).

Specific comments

P1014 l14-18 and P1014 l23-27 essentially repeat the same text.

We removed the second version of the paragraph.

P1016 l1: It is more usual to use 't' for time rather than 'T' (more normally temperature); I would suggest stick with convention, but possibly there is a good reason for this nomenclature.

We changed 'T' to 't'

P1019 l9: Clarify that the heterogeneous conversion of N2O5 on aqueous surfaces is included (forward reference to Section 3.3).

We changed the sentence to:

"Gaseous conversion of N2O5 to nitric acid (HNO3) via the reaction with water vapour is not included, as it has been found to exhibit a rather negligible effect in the presence of the heterogeneous conversion on aqueous surfaces (Williams et al., 2009b), which is parameterized as described in Sect. 3.3."

P1020 l4: Define AFGL.

The AFGL atmosphere refers to the standard atmosphere as defined once by the Air Force Geophysics Laboratory. We reformulate the sentence into:

. . .the standard AFGL (Air Force Geophysics Laboratory) atmosphere for the tropics

P1020 l21: The section also describes aqueous phase chemistry, so the section title

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should reflect this.

We changed the section header to:

"Heterogeneous and aqueous phase reactions"

P1020 l25: 'aquated', as far as I know, is not a word (although I know what you mean). Suggest change to 'hydrated' or similar. It also appears later in the text.

We changed 'aquated' to 'hydrated' throughout the manuscript.

P1021 l3: Isn't it the total surface area (not volume) that is the important quantity (at least for heterogeneous reactions)?

The reviewer is correct that the surface area is the relevant quantity. For clarity we changed "The total volume that" into "The SAD that".

P1021 l25: Strictly speaking, you are defining NOy, not the NOy budget (budget is a wider term that describes all the sources and sinks).

We indeed want to express that we assure the conservation of total mass of N per grid cell, as present in the NOy tracers. We reformulated into:

"A mass balance step is applied to the gas-phase components of NOy, where NOy is defined as the sum of NO, NO2, NO3, HNO3, HNO4, 2 $\times$ N2O5, PAN, ORGNTR, and NO_3^-)"

P1022 l2: Insert comma: 'Gas-aerosol partitioning,'.

OK

P1023 l25: What does 'assuming an interstitial fraction of 0.3' mean? Is it that 70% instantaneously partitions into the cloud drops? Clarify.

The interstitial fraction refers to the fraction of aerosol mass inside clouds which is not scavenged. We changed the formulation "Aerosol particles (...) are scavenged in clouds assuming an interstitial fraction of 0.3, and for gases, using the liquid water

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content" to:

"We assume that a constant fraction of 30

P1023 l26 (and next page): Strictly speaking, Henry's name is used with respect to Henry's Law, rather than for particular coefficients or solubilities.

We reformulated ". . .the effective Henry equilibrium coefficients are used" into

". . .the effective equilibrium coefficients based on Henry's law are used"

and "the respective Henry uptake coefficients" into:

"the respective Henry's law coefficients"

P1024 l1: Some aqueous phase reactions are pH dependent. It would be fairly straightforward to calculate pH in the model. Do you know if using a fixed pH has any influence on your results?

For wet scavenging we indeed keep the pH constant, for simplicity. This is justified as we argue that the model uncertainty related to sub-grid scavenging and mixing processes are more relevant than the pH. However we note that the pH is explicitly calculated when the aqueous phase reactions involving oxidation of SO2 (by O3 and H2O2) are considered. We adapted the text in sec. 3.3:

For the loss of gaseous trace species via heterogeneous oxidation processes, the model explicitly accounts for the oxidation of SO2 in cloud and aerosol through aqueous phase reactions with H2O2 and O3, depending on the acidity of the solution.

P1024 l5-6: 'Using a maximum cloud fraction overlap scheme, rainfall rates are used to estimate the liquid water content, droplet radius and terminal fall velocity.' It is not obvious what this means, please clarify.

This concerns the description of below-cloud scavenging. We changed this sentence to:

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"For below-cloud scavenging the parameterization takes into account the estimated liquid water content, droplet radius and thus the terminal fall velocity."

P1024 l9: What does 'standard deviation 2.0' mean? 2.0%, 2.0μm?

The size distribution of aerosols is often described by a lognormal size distribution. The width of such a distribution is in log-space, and hence non-dimensional and always larger than 1. Formally, this is called the geometric standard deviations. We use now this more formal term but refrain from a further elaboration, since this is considered outside the scope of the paper.

"for aerosol the scavenging efficiency was calculated from a collection kernel assuming a lognormal aerosol distribution (dry particle geometric mean radius of 0.034 μm and geometric standard deviation 2.0)."

P1026 l2: 'This is the case when' – isn't this always the case in the model simulations described here? Or is it topography dependent? Clarify.

In the current model simulation we use 34 levels, which are constructed by merging various model levels from the ECMWF operational forecasts (91 for 2006). Concerning the first model level, this corresponds to the first two ECMWF levels, which actually exceeds a pressure difference at the surface of 3.5 hPa. This criterion is based on standard pressure at sea level. So in the current simulation we inject anthropogenic emissions in the bottom layer only. We added a sentence for clarification:

"In current model simulation with 34 levels the anthropogenic emissions are introduced in the surface layer only."

P1026 l13: 'fourth-order polynomial fit' – do you mean it is a fourth order polynomial function of cold cloud thickness? Clarify.

The reviewer is correct. The fraction of cloud-to-ground flashes depends on a 4-th order polynomial function, rather than a fit. This function was originally created by fitting the polynomial to observational data (Price and Rind, 1993). We adapted the

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text accordingly.

P1026 l17: 'scaled to 5 TgN/yr' – is total lightning NOx production scaled to this number every year (in multi-annual simulations), or just the first year (then allowed to vary inter-annually)? Your simulations here are for 2006, but with 2 years spin-up. Do you just repeat 2006 for 3 years, or do you run 2004-2006, and if so is the lightning total normalised to 5 TgN/yr each year?

The scaling parameter which leads to (approx) 5 TgN/yr is a fixed constant, i.e. it is not optimized for the year that is currently studied. This means that we allow for inter-annual variations, depending on the meteorology. We calculated the annual production for this simulation was calculated 6.6 Tg N/yr. We reformulated "The total annual production is scaled to 5 TgN/yr" to:

"The total annual production for the current model simulation is 6.6 Tg N /yr"

A two year spin-up period is adopted, using metrological data and emissions for the year 2006. This is made more explicit in the text, section 5.

"A spin-up of twice the year 2006 is applied"

P1029 l3: The CO burden is 354 Tg in the text, but 353 Tg in Table 9.

We have made this more consistent, by adapting the numbers in the text. The discrepancy is caused by different round-off methods.

P1029 l27: at -> in

OK

P1030 l6: Are the 'mean' CO values shown in Figure 5 area (or mass) weighted means?

Yes: mass weighted CO. We will adapt the figure caption.

P1030 l9-11: Clarify – is MOPITT V3 used in Shindell et al. (2006)?

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Yes. We adapted the text:

concentrations for MOPITT V4 are 5–15 ppbv larger than those in MOPITT V3, as presented in Shindell et al. (2006).

P1032 l2: Delete ',CH2O' in the units for clarity.

OK

P1033 l2: Suggest change 'also given' to 'which may also be related to'?

OK

P1034: First two paragraphs are essentially the same.

We removed the second version of this paragraph.

P1034 l14-15: I'm not sure the model-data comparison in Figure 11 tells us much about the model ability of the model to simulate the boundary layer. Explain why or delete.

The reviewer is correct in that the statement on the model ability of TM5 to simulate the boundary layer thickness is inaccurate. In fact, the daily development of the boundary layer height is a parameter that is taken from the meteorological driver. The model ability to describe mixing in the boundary layer has been shown in separate studies (e.g. Peters et al. JGR 2004). We removed the statement here.

P1034 l18: NOy was already defined on p1021.

We removed the explanation here.

P1036 l18-24: The rather homogenous vertical O3 profiles in the model possibly suggest

too vigorous convection?

The reasons for the mismatch in tropical O3 concentrations in the free troposphere can be numerous. These include (amongst others) uncertainties in the lightning NOx

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parameterization (e.g. Barret et al. ACP 2010), the effect of the stratospheric boundary conditions for HNO3, and consequently the downwelling NOx from the stratosphere, the convective transport of biomass burning emissions in Africa (Williams et al., ACPD 2010), missing long range transport from SH biomass burning (Peters et al., Tellus 2004) or missing ozone production in the FT due to O2 photolysis (Prather, GRL 2010). Because of the speculative nature of any of these explanations we would prefer keep the formulation "this requires further investigation"

P1037 l5: 'a slope of 0.8 with an offset of 21 ppbv' – slightly more information is needed for this to be immediately useful.

We chose to remove this sentence, as it is not essential for the analysis.

P1039 l17: Clarify that 'these' refers to the fields from the 2006 simulation described in the main text.

We changed "To validate the OH field from the TM5 tropospheric chemistry model, these monthly mean OH fields" to:

"To validate OH in the TM5 tropospheric chemistry model, the monthly mean OH fields from this study for the year 2006"

P1040 l5: The formula for the tropopause pressure in the text initially confused me – I suggest clarify this.

We think that the details on the definition of the tropopause pressure is not essential for the understanding. Therefore we modified this sentence to:

"In these simulations OH in the stratosphere was obtained from. . ."

P1063, Table 8: Clarify what's in brackets (presumably SH/tropics/NH).

We now clarified this.

P1064, Table 9: Do you mean 'levels' or grid-boxes? (I think you mean grid-boxes).

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The trend should have units of Tg/yr, and I would say it was negative.

The reviewer is correct. We indeed mean "grid-boxes". We now write "tendency" instead of "trend" and have adapted the unit.

P1069, Figure 2: The vertical scale cannot strictly be pressure; if it were then there should be no results over the Antarctic at pressures above 800 hPa. Presumably it is some sort of hybrid level, please clarify.

The reviewer is correct. We modified the figures, to include the effect of the orography.

P1072, Figure 5: Clarify if these are area (mass) weighted means.

These are mass weighted. We adapted the caption

P1074, Figure 7: The description of this figure in the text (p1032 l4-7) suggests what is plotted is more complicated than as described in the figure caption. Clarify.

We extended the figure caption

P1075, Figure 8: Clarify that the model results shown in this figure are now sampled as per SCHIAMACHY (which is different to Figure 7).

This is not the case: We perform similar area-averaging for TM5 as SCIAMACHY data in Figure 8, but we again do not apply sampling to SCIAMACHY observations. So the data in Figure 8 is fully consistent with the results presented in Figure 7. To clarify this aspect we changed in the text "The seasonal cycle of CH2O. . ." to

"The corresponding seasonal cycle of CH2O. . ."

Interactive comment on Geosci. Model Dev. Discuss., 3, 1009, 2010.