Review of The high-resolution version of TM5-MP for optimised satellite retrievals: Description and Validation

August 25, 2016

Introduction

I think the work described in the manuscript is interesting. The manuscript clearly states the larger context and final aim of the work, and motivates reasonably well the changes made to the model. The work is reasonably focused, and in general rather well explained. The flow of the manuscript makes it motivating to read it.

However, there are also some major points of critic. The analysis of the results and explanation of the results is not thorough enough. Further, the writing of the manuscript is of poor quality. In addition, some useful information which would have been useful is forgotten.

General

The abstract is not attractive, as it is too much a listing of separate observations.

There is too much similarity between the abstract and the conclusions. The conclusion section should benefit from including an outlook paragraph (which, e.g., should not be in the abstract). Now that a study has been done on modifying the chemistry and the resolution, and improvements in the model performance are moderate, what could be the next points of focus for further development of TM5. What would be the way forward to further improve CTMs? Another aspect which could be discussed in the conclusion : as the differences between 1x1 and 2x3 are moderate, is it an option to still use the 3x2 version profiles as a priori for retrievals?

I have the impression that the manuscript is too strong in its argumentation that emissions are an important reason for discrepancies with observations. I would suggest that the authors make this claim more solid, e.g., by doing sensitivity experiments.

This manuscript is presenting a new version of TM5, and is mainly focusing on the new aspects. For most of the other information and comparisons, it refers to earlier publications. However, as the current manuscript is aimed to be a comprehensive description of the new TM5 version, it should also contain some relevant basic information. Essential information which is missing in such a model description is :

- What bout the number of levels in the model?
- What about the horizontal resolution towards the pole : is a reduced grid used?
- What about the number of species? Number of transported tracers? I assume that several new tracers have been added, which were not used in earlier versions.
- What about the turbulent diffusion in the model? What about the dry deposition scheme?
- Some lumped species which are mentioned in the text, should be shortly described : ORGNTR, ...
- What about the tracer transport scheme? Is it mass conservative?

The analysis of the differences between the two resolutions should be improved. What about the difference in turbulent vertical transport between both model simulations? What about dry deposition parameterisations affected by the different resolutions? What about lightning NO_x parameterisations? In the current manuscript, there is a lot of focus on convection, whereas other parameterizations might also play a role.

It is also not made clear how large the differences are in the meteorological fields seen by the different resolutions. E.g., is the total precipitation equal in $3x^2$ and $1x^1$? Are the convective mass fluxes, when globally averaged, equal in both versions? Is the cloud cover equal when globally averaged? And the albedo? If not, it would be informative to quantify that.

Similarly, are the ²²²Rn and other emission totals equal?

As one of the aims of the model is to use it for generating instantaneous a-priori profiles and columns, it is not sufficient in this study to look at biases only. One should also look at the high frequency behaviour and thus, e.g., at correlations. E.g., Table 9 gives seasonal biases, but I think it is necessary to also show correlations. In addition, as the satellite retrievals will be used globally, it is not sufficient to quantify the difference between the $3x^2$ and $1x^1$ versions only for the observation locations of the manuscript. As the (tropospheric) columns of NO₂, SO₂ and CH₂O from TM5 will be important for the retrieval, one could, e.g., estimate how well the $3x^2$ and $1x^1$ distributions are correlated spatially. This could also allow to better quantify whether using $3x^2$ in stead of $1x^1$ still makes sense.

The figures in the supplementary material are of poor quality.

It is not always clear from the text whether one is comparing resolutions, or models versus observations. Mentioning it always explicitly will make the text too heavy. However, the authors should be careful that their manuscript is not confusing.

The writing of the manuscript is of poor quality. This manifests itself in different ways :

- using abbreviations without or before they have been defined
- referring to wrong figures
- incorrect numbering of the sections
- using capital letters for words which do not need it (Forest, Tundra, ...)
- poor quality of the text in figure and table captions, the captions should also be much more homogeneous,
- starting to use a symbol for concentrations from page 12 onwards (e.g., [NO] and [NO₂])
- using expressions like : between $800\mathchar`embed{solution} between <math display="inline">800\mathchar`embed{solution} between the solution of the solution o$
- spelling of identical words in different ways.

Finally, it is not clear what selection criterium is used for putting some figures and tables in the main document and others in the supplementary material? If a reader decides not to read the supplementary material, he should at least have an idea of what he will miss.

Comments

- page 2, line 55 The text mentions current resolutions of 2–4° in latitude and 2–6° in longitude. There are however currently models with higher resolutions, see, e.g., Yu et al. [2013.]
- page 3, line 113 which replaces the parameterization of Tiedtke (1989). Be clearer about what sub-grid scale parameterizations are still calculated in TM5. E.g., are turbulent diffusion coefficients calculated, or have they been archived too?
- page 3, line 117-119 Concerning large scale transport, one mentions the CFL criterium. In addition, maybe it is interesting for the reader to mention which transport scheme is used.

- page 6, line 231 The Schery et al. (2004) reference for the ²²²Rn emissions is difficult to find, as it is part of a book. It would therefore be useful to describe shortly some aspects of the ²²²Rn emission map: is it only from continents, is there a latitudinal gradient, are there emissions at high latitudes, is it very patchy or rather homogeneous? Is it dependent on soil moisture or precipitation?
- page 8, line 290-293 Differences are attributed to the resolution of the emission data set, and the convection. It is not clear why the temporal resolution of the emissions should play a large role. Isn't it mainly the horizontal resolution which plays a role in explaining the difference between 3x2 and 1x1? In addition is written earlier in the text (page 8, line 283) that ²²²Rn is emitted at a steady rate.
- page 10, line 368-369 Is this relevant as only 90° S- 30° S, 30° S- 30° N, and 30° N- 90° N are shown?
- page 10, line 372-373 The abbreviations O3S (tagged O_3 tracer which undergoes only ...) and BO3S (stratospheric burden of O_3) are confusing. If O3S is a tracer as defined above, it would be logical that BO3S would be just the total burden of that tracer, whereas here that is not the case.
- page 11, line 414-417 Differences are attributed to only 4 causes. Might there not be an impact of the resolution on the dry deposition, the turbulent mixing, the large scale transport, or the mass conservation of the transport?
- page 12, line 428 shows that differences are small : across resolutions?
- page 11-12, line 436-438 this does not seem to hold on a regional scale.
- page 13, 1 501 What is ORGNTR?
- page 15, line 581 splitting the atmosphere in 3 regions (NH extra-tropics, tropics, SH extra-tropics) is much rougher than "zonally integrated"
- page 16, line 603 Fig. S13 : is this the correct figure to be referred to?
- page 16, line 616-617 Isn't it the increase in spatial resolution which helps?
- page 25, line 744-745 Those with differences <5% are considered to exhibit no discernible change in the bias. An interpretation should not be written in the caption of a figure.
- page 31-32, Figs. 4-5 Are the observations also just the 13:00 values?
- page 35, line 823 during September 2006 : but October 2006 is apparently also shown.
- **SM** Add page numbers in the supplementary material.

Questions

- page 1, line 15-17 differences ... differences
- page 1, line 15-17 increases/decreases : it is not clear which resolution is the reference.
- page 1, line 18 "strength" of convective activity is rather vague. Is CAPE meant, updraft velocity, updraft mass flux?
- page 1, line 19 NH is not yet defined. What is meant by "NH (tropics)"?
- page 1, line 20-21 from simulations at 1x1 horizontal resolution. Isn't it also done for 3x2?
- page 1, line 31-32 not clear whether for both resolutions.
- page 1, line 34 shouldn't 20 and 35 sum up to 100? At this stage, the reader is not yet aware of the fact that changes of less then 5% are not accounted for.
- page 1, line 35 in TM5-PP : only the high resolution version.
- page 8, line 301-303 are the globally averaged ²²²Rn emissions equal in 3x2 and 1x1? Are the globally average convective mass fluxes equal in 3x2 and 1x1?

- page 8, line 304-306 I would expect that, if archived mass fluxes are used, the global total mass flux is equal, independent of the resolution (1x1 or 3x2). Secondly, if mass fluxes were stronger in 1x1, I would expect for 1x1 (compared to 3x2) lower ²²²Rn concentrations at the surface and higher concentrations between 900 and 700 hPa. But for DJF in Paris and London one sees the inverse.
- page 9, line 341 and around Iceland for JJA. This difference is hard to distinguish.
- page 9, line 344-346 Here no averaging is performed towards an identical hor ... : does it mean that the value of a 1x1 grid box is compared with the value of a 3x2 grid box? Is there a spatial interpolation between 3x2 ... grid points, and 1x1 grid points?
- page 10, line 390-391 "At 1x1 the largest increase in STE occurs in the SH during JJA." : This cannot be seen from the numbers in the tables. Isn't the aim explaining the 7% reduction?
- page 10, line 391-393 "Comparing the 0.2 contour ...". I have the impression, looking at the upper panels in Fig. 2, that the change in the SH is as large as in the NH. One sees a lowered contour line between 50°N-90°N and 50°S-90°S, but a lifted contour line between 30°N-50°N and 30°S-50°S, leading to a more tilted contour line representing stronger horizontal gradients (as if horizontal gradients can be conserved better in the 1x1 simulation).
- page 13, line 482-483 In the lower troposphere (<900 hPa) : as it is written here, one interpretes it as if (<900 hPa) is the definition of the lower troposphere. It is however meant to be an extra condition.
- page 13, line 490-493 And what about October?
- page 13, line 504 to quantify the effect on higher spatial resolution.
- page 13, line 515 decrease marginally by 2-3% : looking at Table S1, shouldn't it be 1-3%?
- page 16, line 619 aggregated on a weekly basis does not matter if one looks at seasonal biases; it would have played a role if one also shows correlations.
- **page 24, Table 4** Whereas most terms in this table are in units of $Tg O_3 yr^{-1}$, it is unclear in what units BO3 and Strat. BO3 are. If these are burdens, one would expect Strat. BO3 to be a larger fraction of BO3, than the values shown here (e.g., on the global scale 80 and 378).
- page 24, Table 4 It is not clear where one can find "The fraction of the tropospheric burden originating from the stratosphere is also given." Does one mean Strat BO3? Are these absolute values Tg, or is it %?
- page 24, Table 4 Why no % for the NH/SH/tropical STE changes? As these values are not given, the sentence on page 10, line 379 is rather unclear : "The increase in STE in the SH, with an associated decrease in the NH ..."

Inconsistencies

Below a list can be found of inconsistent use of abbreviations, capital letters, etc. Please make the manuscript more self-consistent.

- Sect. and Sect
- Correct the numbering of the sections and subsections.
- Fig. versus Figure. One should use Figure at the beginning of a sentence, but Fig. within a sentence.
- Free Troposphere : should just be written "free troposphere"
- Marine Boundary Layer should be just written "marine boundary layer".
- Chemistry Transport Models (CTM) versus vertical column densities (VCD).
- The naming of the campaigns should be coherent throughout the manuscript: INTEX-B versus INTEX B, Texas-AQS versus TexAQS II.
- earth-orbiting versus earth orbiting.

- TROPOMI versus tropOMI.
- supplementary material versus Supplementary Material.
- TM5-chem-v3.0 versus TM5 v3.0.
- grid cell versus grid-cell.
- gas phase versus gas-phase.
- BL : definition of boundary layer given much later than first three appearances.
- LT : used but never defined.
- SH, NH : used but never defined.
- BO3 : used but not defined.
- Actinic Fluxes versus actinic fluxes.
- um versus μ m.
- [SO₂] (page 27, line 759) versus NO surface concentrations (page 31, line 793).
- 10 Tg S/year (page 6, line 211) versus 49 Tg N yr⁻¹.
- The Netherlands versus the Netherlands.
- 500 hPa versus 500hPa.
- J values versus J_{O3}, J_{NO2}.
- Tg O₃ versus TgO₃.
- Strat. nudging versus Strat BO3 versus Trop.Chem.Prod (Table 4).
- (i.e.) versus i.e.
- TES versus MLS : for TES the full name is given, whereas not for MLS.
- methodology outline versus methodology outlined.
- Stratosphere versus stratosphere.
- monthly-mean versus monthly mean.
- recycling versus re-cycling
- TR : used but never defined.
- UT : used but not defined.
- i.e. is sometimes used where e.g. should be used.
- overestimate versus over estimate.
- underestimate versus under-estimate.
- high bias versus high-bias.
- NOy versus NO_y .
- Strat. Nudge versus Strat. Nudging.
- N reservoir versus N-reservoir.
- $1^{\circ} \ge 1^{\circ}$ versus $1^{\circ} \ge 1^{\circ}$.
- cloud Surface Area Data : no capital letters needed.

- UTLS : not defined.
- $ug m^{-3}$ versus $\mu g m^{-3}$.
- monthly mean versus monthly-mean.
- down-welling versus down welling.
- underestimation versus under-estimation.
- both * and x in mathematical expressions.
- style for labeling panels in figures : sometimes is the position of the panel (left/right/top/bottom) given before mentioning the element it describes (e.g., in Figs. S5 and S7a), and sometimes it is given after the element it describes (most other figures).
- tropical cities versus Tropical cities.
- (-) versus ().
- (1x1 3x2)/3x2 (Fig. 1) versus 1x1/3x2 (most other figures).
- chose vs choose.
- Marine Boundary Layer does not need capital letters.
- Di-Methyl Sulphide : no capital letters needed.

Inappropriate or unclear language

Below are examples of the poor language of the manuscript. Sometimes suggestions are given for improvement, otherwise it is up to the authors to find a better expression.

page 1, line 24 by between 5-10%.

- page 1, line 39-40 vertical column densities : is column-integrated values meant?
- page 2, line 50-53 from earth-orbiting satellites including ... : The use of "including" gives the impression that a list of satellites will follow, whereas actually a list of instruments follows.
- page 2, line 56 of hundreds of kilometers in area.
- page 2, line 57 sampling is not the same as resolution.
- page 2. line 59 constraints \rightarrow limitations.
- page 2, line 60 NO is not yet defined.
- page 2, line 67 the information ... are.
- page 2, line 67 the coarsening procedure.
- page 2, line 71 VCD is never used later. So I would not define it.
- **page 2, line 79-80** is for placing \rightarrow is to place, or is placing.
- page 2, line 80 constraints in \rightarrow constraints on.
- page 3, line 83 yields.
- page 3, line 83-84 whose spatial location is also smeared via the coarsening procedure : poor language.
- page 3, line 85 deriving biases \rightarrow finding/estimating biases.
- page 3, line 89 add i.e. when you start the summing of all the modifications.
- page 3, line 93 Radon : without capital letter. Or use ²²²Rn.

- page 3, line 118 criteria : is this meant to be plural?
- page 4, line 121 place emphasis of \rightarrow on.
- page 5, line 169-170 between 0.25-0.27µm.
- page 5, line 197 between 2003/2004 and 2001-2009.
- page 5, line 195 mean ratios between CO/O_3 .
- page 6, line 208 Non-Methane Hydrocarbons (NMVOC). The abbreviation does not seem to correspond with the full expression.
- page 6, line 237 at both 3x2 and 1x1 : add resolution(s).
- page 7, line 238 diverse ... diverse.
- page 7, line 252 insures \rightarrow ensures.
- page 7, line 253 O_3 , $\rightarrow O_3$,
- page 7, line 259 around Alaska : all those flights start at the southern tip of Alaska and continue southwards, so around is not the appropriate wording.
- page 7, line 260 and bin \rightarrow and we bin.
- page 7, line 265 ... we supplement the INTEX-B comparisons with those ...
- page 7, line 266 as part of the \rightarrow as part of, or as part of the ... campaign.
- page 8, line 285 deposition \rightarrow dry deposition.
- page 8, line 288 averaged between 800-900 hPa.
- page 8, line 288-289 highlighting the spatial variability in convective upwelling near the top of the convective boundary layer : As the boundary layer depth is different for different locations, just sampling the 800-900 hPa altitude does not guarantee that you sample everywhere the top of the convective boundary layer. This sentence should be improved.
- page 8, 1 294 with increases/decreases : not clear whether 1x1 is compared to 3x2, or the inverse.
- page 8, l 295 coasted regions \rightarrow coastal regions.
- page 8, l 295 Madagaskar \rightarrow Madagascar.
- page 8, 1 306-307 range between 2-10%.
- page 9, line 318-319 potential differences [=differences] ... can be considerable compared to those [=absolute profile] ...
- page 9, line 322-323 For this comparison no averaging is employed [is this absolute?], where the selected grid cells are near the centre of each urban conurbation [or is this a condition?].
- page 9, line 323-324 residual(s) \rightarrow ratio.
- page 9, line 329 changes ... has \rightarrow have.
- page 10, line 372-373 zonal STE.
- page 10, line 375 the multi-model STE mean.
- page 10 ,line 376 This 7% reduction ... : the use of "this" is strange because the reduction has not been mentioned before.
- page 10, line 380 the stratospheric BO3.
- page 10, line 383 "recent" study for 2011.

- page 10, line 382 in the horizontal and the vertical.
- page 11, line 425 vertical gradients \rightarrow vertical profile.
- page 12, line 446 13:00 local time close to \rightarrow which is close to.
- page 12, line 441-442 improving the bias \rightarrow reducing the bias.
- page 12, line 456 implying \rightarrow concluding.
- page 12, line 459 accuracy \rightarrow inaccuracy.
- page 12, line 463 For a 20% of sites ...
- page 12, line 463-465 should be improved.
- page 13, line 482 their ratio. These are $\dots \rightarrow$ their ratio. It is \dots
- page 13, line 486-487 during ... during.
- page 13, line 488 1-sigma variability. One should explain better what is meant.
- page 13, line 501-503 Considering ... means ...
- page 14, line 521 advective mixing terms.
- page 14, line 524 this intermediate become \rightarrow becomes.
- page 15, line 571 that is it \rightarrow that it is.
- page 15, line 590 3-5 Tg less $CH_2O \text{ yr}^{-1} \rightarrow 3-5 \text{ Tg } CH_2O \text{ yr}^{-1}$ less.
- page 16, line 607 at number of EMEP sites.
- page 16, line 607-608 Forest, Rural, $\dots \rightarrow$ forest, rural, \dots
- page 16, line 609-610 Energy Sector \rightarrow energy sector.
- page 16, line 609-611 strange sentence.
- page 16, line 610-611 varying from ... and $\dots \rightarrow 1$) varying between ... and ..., or 2) varying from ... to ...
- page 16, line 618 overview of the changes : that would be correct if the difference was shown in Table 8. However, now just the values are shown in that table.
- page 16, line 631 being described better as for that shown for NO_2 in Fig.6.
- page 17, line 651-652 we show that differences exist at higher resolution.
- page 17, line 653 location orography.
- page 17, line 673-674 in only a of the order of few percent.
- page 18, line 680 For SO₂ comparison with surface observations in Europe show \rightarrow shows.
- page 18, line 681 at 20% of sites.
- page 18, line 683 associated with either precursor or direct emission terms : shouldn't there be also the word underestimation in the last part of the sentence?
- page 19, line 697 ... applied for NO_x radical-radical reactions and nitrogen reservoirs.
- page 19, line 699 is taken \rightarrow are taken.
- page 21, Table 21 [E] is not defined.
- page 21, Table 21 $IC_3H_7O_2 \rightarrow C_3H_7O_2$ or i-C₃H₇O₂.
- page 21, line 705 and 707 Branching ratio and Rate have a capital letter, whereas assumed has not.

- page 24, line 722-723 The definition ... and the ... are defined.
- page 24, line 724-725 nudging (=constraining) to constraints : one nudges to values.
- page 24, line 725-726 The contribution ... are provided.
- page 24, line 732 The RO2 term is $\dots \rightarrow$ The NO + RO2 term \dots
- page 25, line 742-743 The seasonal mean absolute biases of weekly [NO] (μ g m⁻³) composed from daily measurements at 13:00 for DJF and JJA. This sentence is hard to grasp, as there are four different references to time.
- page 25, line 743 (measurements-model).
- page 26, line 750 except.
- page 27, line 759-760 taking the difference between measurements-model values.
- page 28, line 766 between 800-900 hPa.
- page 28, line 766-768 (right) is mentioned, but not (left).
- page 33, line 814-817 (left) and (middle) are mentioned, but not (right).
- **page 37**, line 846 as part of the INTEX $B \rightarrow 1$) as part of INTEX B, or 2) as part of the INTEX B campaign.
- page 38, line 857-859 Is "weekly" missing?
- page 39, line 866 as part of the Texas-AQS II.
- page 39, line 867 for each of the days?
- page 39, line 867-878 details ... details.
- SM, Fig. S1 residual \rightarrow ratio.
- SM, Fig. S1 differences in ratio \rightarrow ratios between.
- SM, Fig. S1 between 1x1/3x2.
- SM, Fig. S1 for January and July during $2006 \rightarrow$ in 2006.
- SM, Fig. S1 red-line \rightarrow red line.
- SM, Fig. S3 1x1/1x1 (Tiedtke) (poor description).
- SM, Fig. S4 Monthly mean comparisons of $J_{O3} \dots \rightarrow$ Comparison of monthly mean $J_{O3} \dots$
- SM, Fig. S4 type of scenario : does this refer to "High Arctic", "Tundra", "Industrial", ...? I think scenario is not the correct word.
- SM, Fig. S6a Residual \rightarrow Ratio.
- SM, Figs. S7a, S7b, and S8 Why expressing "Comparisons are shown for volume mixing ratios." in Figs. S7a and S7b, and not in Fig. S8? Why not just mentioning the units in the first sentence after O₃?
- SM, Figs. S8 and S9 Is there a difference in interpretation between "The dotted line represents the 1-sigma variability associated with the measurements." and "The 1-sigma deviation from the measurements is shown as the dotted line for each of the days." If not, I suggest to homogenize the captions.
- SM, Fig. S11 except for.
- SM, Fig. S12 The units for the panels in the left column should be mentioned.
- SM, Fig. S14 details ... details.
- SM, Fig. S15 also October is shown.

Additional corrections

Below can be found a list of additional errors which should be corrected.

- page 2, line 47 (CTM) \rightarrow (CTMs).
- page 2, line 49 earth-orbiting \rightarrow Earth-orbiting.
- page 2, line 53 (Valks et al., 2011)) \rightarrow (Valks et al., 2011).
- page 2, line 50 (TES, Worden et al., 2007) \rightarrow (TES, Worden et al., 2007),
- page 2, line 68-69 an associated uncertainty of $2 \rightarrow$ an associated uncertainty of a factor of 2.
- page 3, line 103 regional \rightarrow region.
- page 3, line 105-106 massivelyntersta .
- page 4, line 123 and 125 is \rightarrow are.
- page 4, line 145 Details \rightarrow For details
- page 4, line 149 land and ocean ... 40 and 900, respectively. I presume 40 and 900 should be inversed.
- page 6, line 221 6Tg/N yr⁻¹ \rightarrow 6Tg N yr⁻¹.
- page 6, line 225 SO_2 (117 Tg Syr⁻¹) : this seems a lot. Could it be meant SO_2 (117 Tg SO_2 yr⁻¹)?
- page 6, line 234-235 (VOC) \rightarrow (VOCs).
- page 9, line 323 that the significant differences exist.
- page 9, line 342 Figure S3 refers to ²²²Rn, whereas here J-values are discussed
- page 10, line 362 the also the efficiency.
- page 10, line 396 from the Stratospheric.
- page 11, line 379 (Verstraeten et al. (2015)) \rightarrow (Verstraeten et al., 2015).
- page 12, line 444 Figures 4 and 5 shows \rightarrow show.
- page 12, line 453 due \rightarrow due to.
- page 12, line 475 are the order of.
- page 13, line 487-488 in NO and NO₂, mixing ratios.
- page 15, line 564 Goncalves et al., $2012 \rightarrow$ Goncalves et al., 2012.
- page 15, line 596-597 or that the chemical production term is too.
- page 16, line 626 significantly \rightarrow significant
- page 16, line 628 Figure S14 \rightarrow Figure S16.
- page 13, line 637 Figure $13 \rightarrow$ Figure 12.
- page 16, line 637 September 2006 is mentioned in the text, whereas also October is shown in the figures.
- page 21, line 705 $1/(1+498.*\exp(-1160/T) \rightarrow 1/(1+498.*\exp(-1160/T))$.
- page 34, line 823 and page 13, line 487 October is also shown.
- page 34, line 822-825 (left), (middle) \rightarrow (top), (middle), and (bottom).
- SM, Table S1 Tg/N.
- SM, Tables S1 and S2 chemical troposphere \rightarrow chemical tropopause.