Review #2

General reply to reviewers

We thank the reviewer for helpful comments that make the paper clearer. Below we answer general and specific comments, but first we address some necessary manuscript changes due to correction of model/diagnostic errors identified during the course of responding to the reviews.

Important manuscript changes

1. During the review process, we located and corrected two errors in the CTM3 nitrate aerosol module. First, wet scavenging of these aerosols was too slow and when correcting this less HNO3 is removed through wet scavenging of nitrate aerosol. Second, nitrate aerosols were wrongly affecting stratospheric NOy/NOx, in effect reducing reactive NOx in the UTLS. Correcting this lead to about 10% higher tropospheric O3 burden in CTM3 and also affects the OH and hence CH4 lifetime. Stratosphere-to-troposphere exchange of O3 (STE) now does not change when including nitrate aerosols. When corrected, the effect of including nitrate aerosols in CTM3 is similar to that in CTM2. The discussions of OH, CH4 lifetime, O3 burden and STE are revised accordingly, and statements about nitrate being more effective in CTM3 have therefore been removed. These corrections do not change the conclusions of the work, but makes the results more consistent.

2. We have removed the lightning sensitivity section 4.2 and condensed the text into the lightning section 2.6. Figure 11 is therefore removed, along with the entry C3_PIC in Table 2. One additional simulation has been carried out with a fixed vertical scaling of L-NOx approximately similar to CTM2, revealing this to account for about 50% of the CTM2-CTM3 differences in OH, CH4 lifetime and O3 burden.
   The new lightning paragraph reads:
   "We evaluate the new L-NOx emission parameterization against a simulation that uses the previous vertical emission profiles. These old vertical profiles (Pickering et al., 1998) injected a large fraction of L-NOx near the surface and near the convective cloud top. In contrast, the new vertical profiles (Ott et al., 2010), which are based on more extensive in situ measurements, place most L-NOx below the convective cloud top with little near the surface. While the new L-NOx algorithm scales the new profiles to match the cloud-top heights hourly in each CTM column, the old CTM2 algorithm assumed a fixed convective top at 16 km, for purposes of calculating the vertical distribution of L-NOx. Thus CTM2 placed much more L-NOx in near the tropopause, or even in the stratosphere when the tropopause was below 16 km. The change in L-NOx between CTM3 and CTM2 is extensive, including location, flash-rate and scaling factors as well as vertical profiles of the injected NO.
   We have tested the change in profiles in CTM3, and cound that compared to the old profiles (Pickering et al., 1998), the new Ott et al. (2010) profiles cause modelled zonal mean NOx to increase by 10-15% (annually up to ~10%) in the middle troposphere (400-800 hPa) and decrease NOx by up to 15-25% (~15% annually) in the tropical upper troposphere (250-100 hPa). Accompanying O3 changes range from -2% to 2%. However, scaling the old profiles to fixed 16 km convective cloud tops has a larger effect, increasing NOx} by more than 100 % at ~ 200 hPa and O3 by ~35 % at 400 hPa. We come back to this in Sect. 3.3."

3. Because the lightning was removed from section 4.2, the transport sensitivity section 4.1 is now called section 4, "Transport sensitivity studies". It has been updated to take into account reviewers comments.

4. Simulation names are changed for clarity:
   C3_30MIN -> C3_1/2 (halving the operator split time step, i.e. 30min)
   C3_V2 -> C3_pole
   _NIT -> _ssn ('ssn' to reflect sea salt, sulfate and nitrate; less focus on nitrate)
   In addition we have included C3_1/4, where the original time step is quartered (i.e. 15min).

5. Table 2, showing age of air runs, has been removed, because the information is already in Table 1. Where needed, the text has been changed to reflect this.

6. We have shortened the CH4 lifetime section, to make it clearer. And we have changed the focus of O3 burden to the 150ppb O3 definition, to better be able to compare with Stevenson et al.

7. Wet scavenging section: The two paragraphs before the last, are modified to match the correction of nitrate aerosols:
"Our chemistry simulations C2 and C3 show similar results; lessHNO3 is scavenged in CTM3, giving the latter a higher tropospheric burden of HNO3. The very different pattern of scavenging from CTM2 to CTM3 also contributes to this, by changing the relative distribution of HNO3 throughout the troposphere. In the ssn-simulations, we see a reduction of tropospheric HNO3 by about 28% in CTM2 and 32 in CTM3. This agrees well with what we found for OH and lifetime ofCH4. The slightly higher effect on HNO3 in CTM3 is probably due to the less efficient wet scavenging in CTM3 also affecting sea salt and nitrate aerosols, allowing more HNO3 to be bound in nitrate particles. Also important are differences in other loss processes such as photodissociation and the loss to OH.

The higher HNO3 burden in CTM3 is not distributed evenly. In the tropics, below 400hPa, HNO3 is about 50% lower in CTM3, while in the extra-tropics CTM3 has up to twice as much HNO3, consistent with less scavenging. Inclusion of tropospheric aerosols reduces extra-tropical HNO3 by up to 100 % in both CTM2 and CTM3, slightly more pronounced in CTM3. Because of the less efficient scavenging of sea salt aerosols in CTM3, more HNO3 is taken up to form nitrate aerosols, explaining this difference. Above 200 hPa, in the stratosphere, CTM3 has about half of the HNO3 in CTM2, except in the upper stratosphere, where CTM3 again has somewhat higher HNO3."

General Comments

I recommend publication after the authors have addressed my comments, many of which are concerned with style and clarity rather than actual content. For instance some of the Figures are not useful in their current form either being too small or assuming the reader can calculate differences by eye (not so simple in some instances).

We appreciate helpful comments on the figures, and we have revised the figures to make them more readable. Figure changes are listed in the general reply at the top.

The many short paragraphs (sometimes only one line) should be condensed to remove sentences which are either repeating details or not providing relevant information. It is confusing for the discussion to flit about comparing many sensitivity tests often without any strong conclusions. I would advise that if the changes between two sensitivity studies are not important then do not mention them.

We have revised the document with respect to reducing the short paragraphs, making conclusions clearer.

I think that the naming of the models in the text should be sequential e.g. "when comparing CTM2 with CTM3" rather than being the other way around as it currently does not follow a logical style. This should be corrected throughout the text.

The focus is CTM3, and it is compared with CTM2, hence we keep our notation on several places.

Apart from the wet scavenging of trace gases is there any conversion of e.g. N2O5 on wet surfaces? Please add a statement.

We now have included a statement on this, in the chemistry section and OH section.

One weakness of the current paper is that there is no comparison of the distributions of either CO or CH4 against measurements which is provided. Considering the dominant effect of CO on oxidative capacity and CH4 lifetime it is necessary to provide at least one figure comparing some surface values from e.g. the GMD dataset. One dominant question related to CO is related to how good the seasonal cycle represented at the high northern latitudes and whether the mixing ratios in the Southern hemisphere are of the right order of magnitude and not over-estimated. One question related the CH4 is how the model captures the regional variability (polluted vs remote) which can then be used in the discussion related to burdens, etc.

We will answer this in the specific comments.
We find CO at the surface to match previous CTM2 results, which has been evaluated previously e.g. in the multi-model comparison of Shindell et al (2006, JGR, doi:1029/2006JD007100). Re-doing such a study is outside the scope of this paper, and should require inclusion of new CO inventories.
However, we have included comparison of free-tropospheric CO measured by CARIBIC. More on this is given in the specific comments.
A reference to the work describing the surface fixed fields of CH4 is added.
For the stratosphere, there is nothing new on CH4, better observations are available for N2O.

The LiNOx sensitivity study does not really add much to the paper as it is currently presented as it is not clear whether this update is important for improving e.g. NOx distribution of UT O3.

Have revised the section and moved it to the lightning section, as described in our general reply at the top. The change from Pickering to Ott is not important in terms of O3 change, which is because the profiles are scaled to convective top height. However, when scaling lightning to a fixed altitude, 16km, as in CTM2, we see a large impact. We have included this info in the new manuscript (see specific comments below).

Specific comments:
Pg 1563: lines 14-19: This paragraph should be removed as it is not providing any new information. It is almost a mini summary of the paper the authors are about the present.

Thanks for pointing this out. We have removed the sentence and put parts of it into the conclusions.

Pg 1563, lns 21-25. All these details regarding possible future aerosol versions of CTM3 should be shifted to the model description and not appear in the middle of the plan of the paper.

Good point, we have moved this sentence to section 2.

Pg. 1564, ln 1: More info on the sensitivity studies is needed. Why are they presented? What is the aim of each study? An introductory paragraph on some of the other contents related to the Polar transport issues and LiNOx update should be included.

We treat the sensitivity studies as effectively ‘perturbed physics” cases of the basic conceptual model CTM3, and we have added more info on the motivation for these. The lightning test has been removed from Table 2, and is discussed in Section 2.6 only. The new text:
“The sensitivity studies are all without sulfur and nitrate aerosols, included to assess the CTM3 transport, and are compared with C3. C3_30MIN is the C3 run with an operator split time step of 30 min, which is half of that in C3. It is included to assess transport errors. Similarly, we carry out a 12 month C3_15MIN, starting at 1 January 2005 from the C3_30MIN simulation, for testing the convergence of operator split time steps. C3_V2 is the C3 run with the optional more accurate polar cap treatment described in Sect. 2.”

Pg. 1564, ln 23: Where or what is section 2.5?

We have added info on section 2.5: emission inventories used.

Pg 1565, ln 18: Remove “in addition”.

Done

P 1567, ln 8: Please refrain from using the colloquial term “ blob”. Surely a more scientific notation is “local maxima”?.

Local maxima sounds good, but implies a peak value. We study a large relatively uniform region of elevated N2O. In literature and later in the paper we also refer to this as a Frozen-in Anti-Cyclone (FriAC), and we therefore use this term.
New sentence:
“First is the relatively large region of high-N2O air in the Arctic middle stratosphere that splits off from the tropics in March 2005 and survives into August (Manney et al., 2006), a so-called Frozen-in Anti-Cyclone (FriAC).”

P 1567, ln 13: The phrase “ . . . shredded the vortex” sounds like irreparable damage. I think you mean “ .. more diffusive therefore perturbing the isolation of the Arctic Vortex allowing air to leak out”.

In fact, the problem was more an irreparable damage; it was not a slow leak or “diffusive.” We have changed “shredded the vortex.” to: “… failed to correctly transport tracers across the pole, resulting in vortex air being transported out of the vortex and air from outside transported in.”

P 1568, ln 5-7: Merge paragraph with proceeding paragraph.

Done

P 1569, ln 4: Quantify ! Terms such as ‘slightly different’ are too subjective.

The new vegetation dataset has more categories and has a higher resolution. We see that the dry deposition velocities may change up to 100% in some grid boxes either due to vegetation differences or due to resolution differences. The overall patterns are similar. Due to computational limits, we could not test the effect of the dry deposition in a full CTM simulation. Also see comment on Pg 1585, ln 18-25.

We have changed the text to:
“Dry deposition rates are unchanged from CTM2 to CTM3 (Wesely, 1989). However, the CTM3 uses a more detailed land use dataset, hence weighting deposition rates for vegetation categories different than in CTM2. While CTM2 uses only 5 categories in T42 horizontal resolution, the CTM3 uses the 1°x1° 18-category ISLSCP2 MODIS dataset (9-category ISLSCP88 is also available). Differences in both resolution and vegetation fractions lead to changes up to 100% in some grid boxes, while the overall deposition rate pattern is maintained. We treat this change as part of the core update.”

P 1569, ln 8: solved -> dissolved

Thanks; corrected.

P 1569, ln 9: This implies that upon evaporation chemical constituents are dumped back into the gas phase but I am not so sure this is the case in the model. For HNO3 it is most likely to remain as NO3- on the CCN/particle.

In each model layer the amount of dissolved tracers are calculated and transported downwards by falling rain. If the dissolved tracer coming in from above is larger than what can be dissolved (based on cloud water and rain water from meteorological data), the difference is evaporated back to the original tracers. Dissolved HNO3 will then be put back into gas phase HNO3. (CTMs generally do not calculate rain or CCN/particles.)
This applies for both CTMs, but the treatments differ; CTM2 treatment is explained shortly in the next lines, while CTM3 is described by Neu and Prather (2012).
We have improved the section to make this clearer.

P 1569, ln 22: Is the HNO3 uptake reversible or irreversible ? Burial or Langmuir? Is this an important sink for NOx in the model?

HNO3 uptake is reversible in that virga releases all gases trapped in the ice/water according to the algorithm. For temperatures below 258K, Kärcher and Voigt (2006) treatment used here is burial (HNO3 uniform throughout the ice), while for warmer temperatures a modified Henry's law is used. For the former, evaporation of the precipitation releases proportional amounts of HNO3, but for the latter, the Henry's law is updated for precipitation (adding or removing HNO3) at each level.

Neu and Prather (2012, ACP) found that ice uptake, at least at a moderate rate, is a very important (critical) component in the upper tropospheric NOx budget, and hence also for the tropospheric O3. These results also apply to CTM3, which has the same algorithm and fields for clouds and precipitation.

New paragraph:
CTM3 also uses Henry's Law to calculate the solution of species in rainfall, but has a~more complex cloud model that accounts for overlapping clouds and rain (Neu and Prather, 2012). This scheme also covers HNO3 removal on ice for temperatures 258 K < T < 273 K, for which the uptake is calculated by Henry's law modified by a retention coefficient. For temperatures below 258 K, HNO3 uptake on ice follows Kärcher and Voigt (2006) (burial where tracer is uniform through the ice). Further details, e.g. on evaporation, can be found in Neu and Prather (2012).

Pg 1570, ln 22: 'individually tuned'. This opens a whole realm of possibilities concerning the treatment of the absorption characteristics for use in fast-JX. Is this to reduce errors compared to a full solution of the J value on a higher resolution grid. Have the J-values been compared against measurements at all? If not how do you know this aspect of the model upgrade results in an improvement in terms of performance.

This section can be misread – the tuned Stern-Vollmer quantum yields are tuned/parameterized individually by authors in IUPAC. See revised/clarified sections below. The photolysis code was one of the core in PhotoComp2008 (now referenced) in which J's were tested and included models (TUV) that were part of the IPMMI studies.

Revised text:
"An important update connected to fast-JX <u>(Photocomp2008, 2010)</u> is the new cloud overlap treatment, described by Neu et al. (2007). The only difference …"

And:
"Photolysis of volatile organic compounds (VOC) is taken solely from the latter, and uses <u>the provided, individually tuned,</u> Stern-Vollmer pressure dependencies for the quantum yields.

New ref
PhotoComp 2008 (2010), Chapter 6 – Stratospheric Chemistry in SPARC Report N°5 (2010) Chemistry-Climate Model Validation (V. Erying, T. Shepherd and D. Waugh, eds.), World Climate Research Programme Report No. 30, World Meteorological Organization TD No. 40, Geneva, http://www.sparc-climate.org/publications/sparc-reports/sparc-report-no5/

Pg 1572, ln 3-5: Is sulphate aerosol included in the scattering component? If not then why only account for the BC absorption as the scattering component above the boundary layer will introduce an additional redistribution of the UV flux in the vertical (more important on clear days).

Only a climatology of BC is included, similar to the treatment in CTM2. When all aerosol modules are included and tested we will include these in J-value calculations. We could have included a climatology of sulfate aerosols, but this has not been done previously in CTM2 and spending time on this is not a priority. It is important to do so later, but not critical for C2-C3 comparison because both have the same treatment.

Pg 1572, ln 8: Energy and Industrial production will NOT have a pronounced diurnal cycle (there is never a shut down for industrial processes as this costs too much money and time). Is this diurnal cycle going to zero during the night?

The diurnal cycle for industrial emissions is not pronounced. In the RETRO diurnal cycles, which assumes the production rate to be daily average, production for industrial combustion goes down to 75% at night and up to 130% in daytime.
Energy emissions go down to 71% at night and up to 122% during day.
E.g. transport, solvent use agricultural emissions have larger diurnal variations.

Pg 1572, ln 9: Motivation for the 96%/4% split? Is this based on measurements.

The motivation is that there will always be fast small scale processes that cannot be resolved in the model grid resolution. Some of the emitted NO will react to produce NO2.
For historical reason we used Petry et al (1998, JGR, doi:10.1029/97JD03749) for all surface NOx emissions, but this has very little impact since NOx resets quickly and the implied O3 emissions from the NO2 are also small. We have added a note on this in the text.

Pg 1572, ln 15: Again motivation for the 90%/10% split.

We have added reference for this: Andreae, M. O. and P. Merlet., Emissions of trace gases and aerosols from biomass burning, Global Biogeochemical cycles, 15, 955-966, 2001, saying that "The most abundant N species in the emissions is typically NO, and it has been frequently assumed that all NOx is released in this form. Where NO2 has been determined selectively, however, It has been found that it represents some 10% of NOx already directly above the fire (Griffith et al. 1991; Yokelson et al. 1996)."

Pg 1572, ln 19: What effects did the plume parameterization have on the results ? Summarise in one sentence. It could ultimately be tailored to the new model set-up.

Kraabøl et al. (2002) found the NILU plume to cause a ~20% reduction in the effect on O3. This was also found in a report by Gauss et al. (2003) using the Oslo CTM2 with trop+strat chemistry. A sentence about this has been added.
The model code used for creating the parameterization is no longer available, making an update to the new resolution too large work. Including other parameterizations could be possible in the future.

Pg 1572, ln 21: Please give the total emission N yr-1 from lightning.

Done

Pg 1572: Please add a sentence explaining what is used to constrain the CH4 in the model (climatology based on measurements or is it a fixed mixing ratio). Is there a seasonal cycle and is there a latitudinal gradient as observed?

The surface CH4 is set as fixed mixing ratios each month, based on a model simulation which used CH4 emissions and surface deposition. We have included this in the text, and also in Section 3.3.2.
HYMN Report added to reference list.

Pg 1573, ln 8: remove 'with'.

Done

Pg 1573, ln 9: add '('

The reference is corrected.

Pg 1573, ln 16-20: Are these profiles in the convective column also used in other parameterizations in the model or are they not from the meteorology used to drive the model?

We have updated the text to make this clear. The sentence now says:
Lightning is diagnosed in the Oslo CTM3 only when convection out of the boundary layer exceeds a grid-averaged updraft velocity of 0.01 m/s at ~850 hPa, as specified by the input meteorology.

Pg 1575, ln 6-9: Condense into proceeding paragraph.

Done

Pg 1575, ln 16: is -> 'has been'.

Corrected

Pg 1575 Ln 19: 'transport-chemistry' -> 'chemistry-transport'.

Corrected

Pg 1577, ln 26-27: Figure 4 needs to be assessed for clarity. Showing the absolute differences between CTM2 and CTM3 would provide more information related to the changes introduced between both models.

Figure 4 is included for informative reasons, showing that the models capture the seasonal changes. Instead of showing the difference plot, we introduce a new figure showing only 2005 results. In this figure we show the flexibility of the CTM3 by showing how the operator split time step affect the polar vortex, and that by cutting the time step we increase the accuracy.
These show the differences of C3 vs C2, C3_1/2 vs C2 and C3_1/4 vs C2. It should be noted that some of the differences near the South Pole is due to the improvements in polar cap transport in CTM3 compared to CTM2.

Additional text:
"In Figure 5 we show the CTM3 vs CTM2 difference for year 2005, for different operator-split time steps in CTM3. Clearly, CTM3 converges towards CTM2 for shorter time steps, with the 15min time step close to the ~10 min [U]-flux time step in CTM2. We note, however, that CTM2 does not necessarily show the truth and also has its errors, especially near the pole where CTM3 resolves transport better."

Pg 1578, ln 27: 'produce' -> produces

Done

Pg 1578, ln 29: Fig. 6 is not currently legible being much too small !!. Split into Fig 6a/b using the different months as the division criteria. This should really have been amended in the technical review before publication in GMDD.

We have revised the figure. Before GMDD it was found to give reasonable figures in A4-printing, but not in GMDD format. We have reduced the space between the figures and made the figures larger, and we have split the figure into two.
If the editor wishes us to keep it in one figure, we will be happy to comply.

Pg 1579, ln 5: The statement regarding the stratospheric differences are insignificant between C3 and C3NIT should be moved forward to directly after where the simulations are introduced to avoid repeating this multiple times further through the paper.

We agree, and have moved the statement to the beginning of Section 3, and removed several re-statements.

Pg 1579, ln 11: under-estimates "it". It being . . . ?

MLS, corrected to "underestimate at altitudes…"

Pg 1579, ln 22: "similar distributions in N2O".

Corrected: "similar distributions of"

Pg 1580, ln 10: " although the amplitude " . . . amplitude of what ? Seasonal cycle ?

We refer to the amplitude of N2O in the intrusion. We have revised the text to make this clearer:
"The Oslo~CTM3 reproduces transport of the high-N2O intrusion well, although the mixing ratio amplitude is underestimated."
We have also added a note on the color scale range, which differs from the Allen et al. paper, and we have added a dashed-dotted line 75ppb, and a solid line 100ppb, to be better comparable with Allen et al.

Pg 1582, ln 3-5: This previous multi-model inter-comparison study allows the authors to place the performance of CTM3 in the context of other models which are used in the community. Some comment should be made regarding what deficiencies CTM2 exhibited and whether these have been improved upon.

Compared to Hoyle et al., we use a different set of meteorological data. They used 40-layer meteorology from cycle 29 (model top at 2hPa), while we use 60-layer from cycle 36 (model top ~0.1hPa). This results in different convection and hence model result profiles, and it is difficult to conclude whether the CTM3 is better than their CTM2 version. But from previous studies, we have seen that CTM2 generally overestimate short lived components at higher altitudes (e.g. Skeie et al., 2011), and from this test we see that CTM3 will produce lower abundances.

If we assume that the effect of entrainment/detrainment data would reduce CTM2 upper-tropospheric mixing ratios, as for our CTM3 results. In Hoyle et al., this would shift T6h for CTM2 towards the FRSGC/UCI results below 100hPa, while T20d would be shifted closer to the other models.

Added paragraph:
"Hoyle et al. (2011) used a different set of meteorological data, so a direct comparison is not possible. However, from our results we can assume that inclusion of downdrafts and detrainments would shift their Oslo CTM2 towards the FRSGC/UCI results for T6h below 100hPa, while T20d would be shifted closer to the other models."

Pg 1583, ln 4-9: The scale used for Fig 9 means that changes in tropospheric profiles are not discernible. It appears as if CTM2 has a better performance around 250hPa across all profiles. This figure should be replaced by something more useful such as either the CO or CH4 surface comparisons.

We thank the reviewer for pointing this out, and have instead of these four sondes included a supplementary document where all the sondes in the average profiles are plotted, with both CTM2 and CTM3, similar to Fig. 9.

We agree that CO is interesting, and have included a discussion on surface CO, along with two figures in the supplementary material. CTM3 is in general higher than CTM2 over land, but lower over ocean, consistent with the reduced upward transport found in the convection section.
However, after the vertical profiles section, we have included a comparison of the models with CO from CARIBIC measurements. The figure shows all measurements done in 2005, and shows excellent agreement between models and CARIBIC. It also shows that CTM3 catches large spikes in CO due to better vertical treatment of transport.
Additional text in a separate section has been added to the manuscript:

New text:
"3.2.5 Modelling CARIBIC CO measurements
The atmospheric abundance of CO is important for OH. On a global average, we find surface CO in CTM3 to match CTM2. On a monthly basis, CTM3 has a more pronounced seasonal variation, being up to 5 % lower in NH spring and 5 % higher in NH autumn. As found in Sect. 3.2.2, CTM3 transports less out of the boundary layer, and for CO we see that CTM3 has up to 50 % higher abundances over land, while having up to 10 % less over ocean. A comparison modeled and measured CO at station Alert (shown in supplementary material) reveals that while CTM3 is 5--15% lower than CTM2, both models catch the seasonal variation, but underestimates at NH winter, as was found in the multi-model CO-comparison of Shindell et al. (2006). E.g. at Hohenpeissenberg, CTM3 is on average about 15 % higher than CTM2 (also in supplementary material). Unfortunately, it has not been possible to compare our results directly with CTM2 results from the comparison of

Shindell et al. (2006), a study which used different emissions, with slightly higher total amount of CO emissions, 1077 Tg/yr compared to our 1012 Tg/yr from RETRO, POET and GFEDv3 datasets. It can be noted that the anthropogenic emissions in RETRO do have a month-to-month variation, in contrast to the emissions in their study. Shindell et al. (2006) suggest NH emissions are too low, and recently other studies also suggest that CO emissions are low in general (Lamarque et al., 2010; Pison et al., 2009; Kopacz et al., 2010).

With our focus on transport differences between CTM2 and CTM3, we look more specifically at the capability of CTM3 to properly incorporate emissions originating from biomass burning, compared to that of CTM2. For this we compare our modelled CO in the troposphere with measurements carried out in 2005 by CARIBIC (Civil Aircraft for the Regular Investigation of the atmosphere Based on an Instrument Container, http://www.caribic-atmospheric.com, Brenninkmeijer et al., 2007). In the year 2005 most CARIBIC flights were operated between Europe and South America, and model results are interpolated on-line to the spatial and temporal locations of all measurements. Figure 12 shows all CO measurements for all flights in 2005 (black), along with corresponding model output (red for CTM3 and green for CTM2). Measurements located in the CTM stratosphere (2.5 PVU) are shown in blue. Both CTM2 and CTM3 produce CO remarkably close to measured values, also for the spikes originating from biomass burning events in August and October (Ebinghaus et al., 2007), which is due to the combination of realistic meteorological data and a proper estimation of CO from biomass burning events in the monthly GFEDv3 emissions. CTM3 produces larger spikes than CTM2 does, due to the improved vertical transport in CTM3. It is expected that with a higher temporal resolution on GFEDv3 emissions, these spikes would be even higher."

Pg 1583, ln 11-14: Remove short paragraph.

We agree, it is removed.

Pg 1583, ln 22-26: By showing a correlation plot for the troposphere, upper troposphere a more quantitative discussion could be included. I would present a seasonal comparison for the 3 latitude bands used in the paper. You could add the various comparisons in an appendix if you feel that they are necessary. By using the correlation co-efficient a much stronger case can be made regarding improvements (or degradations).
If there is any degradation in performance then one dominant term for O3 is photolytic destruction. Maybe this has degraded (become too efficient) in the upper troposphere ?

We have changed the sonde comparisons to be more helpful, where all single sondes used in the monthly means are included as a supplementary document. We do not understand what would come from the correlations since the identical meteorology means that the synoptic overlap would give $R^2 = 1$.

Pg 1584, ln 27: More photochemically active because of the new solar spectrum or . . .? Is there a sun cycle imposed on the TOA flux ?

In fast-JX the annual sun-earth distance is accounted for, but there is no 11-year solar cycle. The solar flux is derived from the SUSIM data set as an average over active and inactive solar conditions (Hsu and Prather, 2009, JGR, doi:10.1029/2008JD010942). The section is revised to:

"CTM3 has a larger OH than CTM2, partly due to updates in solar fluxes and O(1D) quantum yields (Hsu and Prather, 2009), but also due…"

Pg 1585, ln 15: Conversion of N2O5 to HNO3 is another important process that is not mentioned yet.

The conversion of N2O5 to HNO3 is the same in CTM2 and CTM3. The treatment is a simple parameterization which will be revised in the future. A note on this is included.

Pg 1585, ln 18-25: There seems to be a wide range of opinion regarding the dry deposition flux of CO for use in global models. What is the total deposition flux per year in CTM2 and CTM3 ? Although high variable with respect to land type, neglecting dry deposition in arid and ocean regions could be another way of improving the CO distributions.

The CO dry deposition velocity in CTM2 and CTM3 is 0.03cm/s in forest and on grass, and zero elsewhere. The rate is reduced in stable air to account for slow mixing close to the surface. In general, this is a small sink for CO (in CTM3 it is <0.1% of the total atmospheric CO burden over 24 hours), and some models even skip it (e.g. GEOS-Chem, http://wiki.seas.harvard.edu/geos-chem/index.php/Dry_deposition).
To change CO significantly, there would have to be a large change in this rate, and studying it is out of scope of this work. We have, however, performed a test where anthropogenic emissions are increased by 20%, which only changed OH by <1%. This has been included into the OH-section.

Pg 1586: A figure comparing surface monthly mean mixing ratios of CH4 would give more confidence towards any improvements in the model performance and the subsequent discussion related to CH4 lifetime.

The surface CH4 is fixed, and is the same in CTM2 and CTM3: This is not the reason for differences in CH4 lifetime. We have included a statement about this in the lifetime section, including a reference to the fixesd CH4 values (EU project HYMN).

Pg 1585, ln 22: Rather than discussing the performance of other models (not relevant to this study) comparing the global CO distribution in CTM3 against measurements would significantly enhance the quality of the discussion regarding lifetimes considering it is a well-mixed gas which scavenges OH effectively.

The CO distribution of CTM2 was published earlier (Shindell et al., 2006, JGR, doi:10.1029/2006JD007100), and CTM3 produce similar distributions. Re-doing such a study is out of scope for this work, even if it could change the OH. Our new lightning test indicates that differences in vertical distribution of L-NOx in CTM2 and CTM3 is one of the main reasons for higher OH in CTM3.

However, as noted above, we have included a comparison with CARIBIC measurements of CO for 2005, flying between Europe—South America, to study transport changes from CTM2 to CTM3.
See comment P1583, ln 4-9.

P 1587, ln 3: If you use a different set of anthropogenic and biomass burning NOx emissions then I am not so sure how, when comparing values, you can determine the ‘correct’ value for CH4 lifetime. Maybe there are other studies that have used the RETRO inventory (without aerosols) which could be used for comparison which maybe more relevant.

We can evaluate the “correct” atmospheric CH4 lifetime from observations. We believe that that value is the one we should be comparing with. The disagreement in modeled lifetime is large, factor of two, even when models purport to be using the same conceptual model and emissions. The reviewer’s suggestion is interesting but not practical.

P 1587, ln 6: There have been other studies looking at this effect which need to be referenced such that the 8% effect exhibits an uncertainty range (e.g. 5-15% OH decrease). E.g. Martin et al, JGR, 108, doi:10.1029/2002JD002622.

We thank the reviewer for pointing this out. Martin et al. found a 9% reduction in global OH, and that the reduction is largest in the Northern Hemisphere (13% vs 4% in SH). Also Bian et al (2003) found regional differences, however, we do not focus on that here. We have changed the text in section 3.3.1 to:
"Bian et al. (2003) found the effect of aerosols on photolysis to reduce global OH by ~8 %, similar to Martin et al. (2003) who also found a larger reduction in the Northern Hemisphere than in the Southern Hemisphere."

P1587, ln 25-27. Another factor could be errors in the emission estimates which build up over time when there are no constraints applied towards climatologies in the stratosphere. This should also be mentioned in the text.

We have made this clearer. Both CTM2 and CTM3 use climatologies in the uppermost model layer, and the same surface emissions for species. We do not model N2O sources, but set a fixed surface concentration of N2O (due to its long lifetime). If too much is transported to the stratosphere, the calculated loss may be too large, while if too little is transported, the loss would be too small.
We do, however, not see how any of the emission errors or climatology boundary conditions could affect the lifetime of N2O.

Transport differences between CTM2 and CTM3 are minor in the tropical stratosphere. The most important difference between CTM2 and CTM3 is J-values as stated.

P1588, ln 14: I do not agree with the statement regarding the definition of the tropospheric domain in Stevenson et al., 2006 for calculating O3 burdens. It uses the 150 ppb O3 contour taken from Prather et al., 2001 and states this. Whether this is the "correct" definition is another matter. Without a clear definition of what is considered the troposphere the comparison of the model values would not be valid. Please remove this sentence.

We stand corrected on this, thanks, and have removed the sentence. We have also included a comment that the 150ppb O3 surface is used, and that CTM2 and CTM3 with nitrate aerosols produce burdens close to their mean values. New text is:
"Based on several model studies, Stevenson et al. (2006) presented a tropospheric burden range of 344±39 Tg, calculated from all air masses with O3 less than 150 ppb. The C3_ssn and C2_ssn runs lie well within this range, with values of 346 and 363 Tg, respectively. When excluding nitrate aerosols our models lie in or just above the upper end of the range."

P1588, ln 17-19: Surely it is the non-linearity in the O3 production calculated by the chemical solver introduced when decreasing the time step which also plays a role?

The chemical time step is not changed, only the overall operator split time step. The standard is to do multiple chemistry steps for a total of one hour, then transport. The chemistry sub-step is 15 minutes in the troposphere, and for an operator split time step of one hour is therefore done 4 times.

Also, we see that a bit more information on the time steps may be wise, because it makes the talk about operator split time step less confusing. We have therefore included a text on the time step in the beginning of the model section:
"All modelled processes, or operations, are carried out sequentially and asynchronously with the only requirement that all process sub-cycles must synch at the end of the operator-split time step, which is typically 60 min but can be shorter or longer. As will be explained in Sect. 2.2.1, the large-scale transport selects a maximum global time step from the Lifshitz criterion, and then sets the number of sub-cycle steps, e.g. within 60 min, that meet that criterion. For emissions, boundary layer mixing, chemistry and dry deposition (called the EBCD-sequence below), CTM3 retains the CTM2 method, with an internal cycling of maximum 15 min. If a

60min sub-step is set for the EBCD-sequence in an operator-split time step of 60 min, the EBCD-sequence is carried out 4 times with 15 min time step. Note that the time step used in chemical integrations may be shorter than this (see Sect. 2.1), and does not change unless the operator split time step is shorter than 15 min. As an example, suppose the Lifshitz maximum time step is 42 min, then two 30 min sub-cycle advection steps will be calculated. If e.g. the EBCD-sequence is calculated every 10-min step and transport and wet scavenging request a~15-min step, then the sub-cycle picks 1/12 of 60 min, or 5 min, as its basic cycle. At 5 min, there are no ops; at 10 min, EBCD is calculated; at 15 min, transport and wet scavenging are done; at 20 min, EBCD again, at 25 min, there are again no ops; at 30 min all three processes are calculated, and so on up to 60 min"

We have also included a separate text on the chemical time step at the end of Sect 2.1:
"As in CTM2, the chemical integrator is the quasi steady-state approximation (QSSA, Hesstvedt et al., 1978). Tropospheric chemistry is integrated with a maximum time step of 15 min (shorter if the operator split time step is shorter), except for the OH-chemistry which uses a 1/3 of this time step (Berntsen and Isaksen, 1997). Stratospheric chemistry is carried out with a maximum of 5 min."

P 1589, ln 1: Associated uncertainty on mean model values for STE?

We have added the range from Stevenson et al. 552±168Tg

P 1589, ln 18-24: As you have used a different definition to define the STE it is no longer directly comparable to the previous multi-model comparison values you quote on line 1. What is the CTM2 and CTM3 values using the Stevenson definition of STE=L + D –P?

Our definition of STE **<u>is</u>** equivalent to the Stevenson definition. Instead of separating the P and L terms for tropospheric O3 (which is somewhat arbitrary and often based on a concept of odd-oxygen) we diagnose the sum P-L (= dM_chem) and the deposition (D = S in our equation). Thus our equation gives STE = D + (L-P) when there is no tropospheric trend (which implies that dM_tot + F_t→t = 0 in our equation).
We have made this clear in the text.

P 1590, ln 3-4: More realistic compared to the previous model values or the measurements?

The higher flux is closer to what is inferred from theory and observed trace-gas correlations in the lower stratosphere and what is and modeled by others, so it suggests that the meteorology of the older meteorological data is more realistic than for the new data, since it does not seem to have partly stagnant air in the tropical lower stratosphere.
We have changed the text to make this clearer:
"While this higher flux indicates that the old cycle may provide a more realistic circulation, it should be noted…"

P 1590, ln 8: You weaken the entire section by stating that the chosen isopleths may not be the correct threshold for use in your calculation of STE.

This is a problem because the fraction of the mass of the atmosphere without an Antarctic ozone hole that has O3 < 120 ppb is very close to that having O3 < 150 ppb, and is similar to that estimated from the tropopause height (80%) see Prather et al. 2011 JGR – but with an ozone hole, a large fraction of the lower Antarctic stratosphere appears to be tropospheric under the O3 isopleth condition.

The text is fixed to note that the problem is related to ascribing the ozone hole to be tropospheric air:
"We have found the 150 ppb isopleth of O3 used in S2006 to confuse stratospheric air with tropospheric air for O3-hole conditions, hence we have lowered this isopleth to 120 ppb to minimise this problem."

We also have updated the last paragraph to avoid weakening the section:
"Due to the difficulties with a calculated O3 isopleth, such as stratospheric air being counted as tropospheric air for low O3, and tropospheric O3-rich air in polluted areas being counted as stratospheric air, we have also tested calculating STE through an alternative surface, namely the troposphere defined by the artificial e90-tracer (Prather et al, 2011). With this we find the annual STE of 296 Tg(O3)/yr, close to the value from using the 120\,ppb isopleth. Also the reduction when halving the operator-split time step is similar to what was found using the 120 ppb isopleth."

We have revised the STE section with updated numbers, after correcting the error in the nitrate treatment (noted in our general reply at the top). STE now does not change when including nitrate aerosols.

P 1591, ln 8-9: Remove text after "1 January 2005". Details are provided in the next paragraph.

The spatial distribution of scavenging was not repeated; the fact that scavenging is reduced in the upper tropospheric while increased in the tropical lower troposphere, is what was also found by Neu and Prather (2012).
We have moved the sentence to the end of the next paragraph.

P 1591, ln 12: typo: NHO3.

Corrected.

Pg 1591, ln 18-20 and 29: What is the lifetime of HNO3 (H2O2) given in the literature? Is CTM3 improving compared to what is commonly accepted regarding atmospheric lifetimes?

The definition of atmospheric "lifetime" for either HNO3 or H2O2 is ambiguous. The local scavenging frequency depends on altitude and location (wet or dry season in tropics) thus the lifetime depends on where you inject the gas, see Figure 1 of Neu and Prather (2102, ACP). For example, for a given surface emission pattern of HNO3 one could define a lifetime based on the total atmospheric burden, but that lifetime will change if the emission pattern is changed. There are no useful observational lifetimes or accepted values.

Pg 1592, ln 15: Why the focus on HO2NO2? There are no recommendations for the scavenging parameters for this species so it is speculation regarding the potential importance of trapping on ice. In fact you could argue that a motivation for excluding it in CTM3 is that the evidence of trapping is weak. Other species such as HCHO and N2O5 are scavenged on ice and do have recommendations but are not mentioned in the text. I would like a clear statement about N2O5 here as most global models do include this heterogeneous surface process (on aquated aerosol and cloud droplets) whereas the HNO4 loss can be considered to be much more exotic and less well measured. This will have implications for the O3 burden, etc.

Neu and Prather (2012) highlighted the potential importance of scavenging HO2NO2 because of its rapid exchange with NOx (unlike HNO3). We decided to test that out in our model, but agree that it adds not a lot new to the original work and will drop this section. In terms of why we did not try ice scavenging of other species, see answer below.

N2O5 conversion to HNO3 on aerosols is included as a chemical parameterization, which is similar in CTM2 and CTM3. Updating this parameterization has unfortunately been outside the scope of this paper.

Pg 1593, ln 8: Values of H2O2 trapping on ice are available so I am not so sure excluding it in the model update is a good thing to be honest. Ideally it should have been implemented along with HNO3 (although the competition for active surface sites on the ice surface has to be ignored).

This is a useful recommendation, but in this first, major update from CTM2 to CTM3, it was essential to adopt process models that were already developed and published where possible. This allows us to refer to these publications for the basic characteristics of the process model as implemented in CTMs or compared with other models and measurements. The Neu and Prather (2012, ACP) scavenging scheme was one of these; and in its assessment and testing, the H2O2-ice scavenging was deemed not as critical, nor established as the HNO3. We agree, however, that with the next version of CTM3, ice scavenging of other species should be implemented. (It is trivial to implement in the current code, but much more difficult to test and evaluate the impact of each change.)

Pg 1595, ln 25-28: In that the time-step can be iterative and have different values a different latitudes raises the question of the usefulness of the 30 min time-step sensitivity test when constraining at global scale. If the reason is the poles why not limit the time-step at latitudes above 60 degrees? Also why not reduce the time-step to 15 minutes to prove that 30 minutes is sufficient for capturing the polar vortexes well? In an operational mode what is the typical time-step for the tropics and mid-latitudes?

The 30 min operator-split time step test is a global time step, so unfortunately it is not possible to use a shorter operator split time step only at high latitudes. We agree that the text on the internal time step is confusing and have rephrased it, and also added more info on time steps in the model section. One of the large benefits for CTM3 is that the internal time step and asynchronization makes it possible to increase the operator-split time step and not force all processes to match the shorter requirements of any one.

To meet the question of 15min run, we have also carried out a 12-month simulation with operator split time step of 15 minutes (C3_1/4), starting from the 30min instant field of 1 Jan 2005. Due to the sharp gradients in the Southern Hemisphere polar vortex, the percent change can be large: For the modeled O3 total column in the ozone hole in the beginning of October 2005, the 30min simulation reduces vortex O3 by up to 20% compared to 60min, while extra-vortex O3 is increased by 5%. Comparing 15-min to 60-min, the values are 25% and 9%, respectively.
"Total O3 hole column values at the polar side of the SH vortex edge are reduced by up to 20 % while on the mid-latitude side the column values are increased by up to 5 %. These relatively large changes are only found across the sharp gradients. Using a 15 min operator split time step increases these numbers to 25 % and 9 %, respectively, showing rapid convergence to CTM2, as was shown in Sect. 3.1.2. A similar effect can also be seen in the Northern Hemisphere, however to a smaller extent."

The same features are seen in the monthly zonal means of O3, where we see that the changes in O3 column arise around the vortex: Equatorward of the vortex edge O3 is increased by up to ~25% for 30min, which

increases to 40% in 15min. Within the vortex, O3 is reduced by up to ~25% in 30min and by up to 30% in 15min. In general the max/min changes are enhanced by up 10-15% when going from 30min to 15min operator-split time step.
For the March NH polar vortex, we see a weaker change in O3, and the max/min changes between 30min and 60min are enhanced by 5-7% when using 15min step.
We have rewritten the next paragraph and joined it with the previous one:
"The largest differences occur at mid to high latitudes between 300 hPa and 20 hPa, in O3-hole conditions with strong asymmetric vorticies. Here, as we have discussed, the jets may overshoot. When halving the operator-split time step, the maximum increase outside the vortex and the maximum decrease inside the vortex are of the same order: In SH, this maximum change from C3 to C3_1/2 is ~25%, enhanced further by 10--15% for C3_1/4. In NH these numbers are smaller (<1/2)"

We have rewritten the next-to-last paragraph to make it clearer.
"Using the more accurate polar transport (C3_pole) increases the computing time by about 50 %. Hence, not all model time steps require shorter transport time steps than for C3. The changes from C3 are small, and generally confined to the polar cap. However, the shorter transport time steps produce a somewhat sharper gradient across the vortex edge than for C3. C3_pole may impose a shorter global transport time step, but keeps operator-split time step unchanged. Thus transport will essentially be similar to C3_1/2 when the transport time step is reduced, but still carried out for 60min before chemistry is calculated. However, if the global transport time step does not need to change, it may be that the internal transport step (1-D pipe flow) requires a shorter time step, but this will not produce large differences between C3 and C3_pole. Again, this was also seen for the age of air studies."

We have also added a paragraph about 15 min time step to age of air:
"By halving the operator-split time step as in C3_1/2, the maximum difference relative to C2 at this location is reduced to 0.29 yr (Fig.3b). A time step of 15min reduces the difference, C3_1/4 minus C2, further to 0.12 yr (Fig. 3c). Thus the error induced by a finite time step can be readily evaluated from the sequence C3, C3_1/2, C3_1/4 and it is not clear whether the differences in Fig 3c are from errors in C3_1/4 or C2."

Pg 1596, ln 1-5: Suggests that the sensitivity study is far from complete and the final limitations/settings needed regarding capturing transport around the poles are yet to be determined in CTM3.

Thanks for pointing this out.
The choice of polar transport settings is important. C3_pole utilizes full grid resolution at the poles but at a significantly greater cost, especially if one moves the CTM3 to higher resolution such as T159. C3_pole is thus more accurate, as would be a higher resolution, and such accuracy may only be important for the simulation when there are large gradients across the polar cap.

C3 is still far better than CTM2. For example the basic fast version C3 will enable more science to be done at T159 (1.1°) resolution than will C3_pole, but will have better effective polar resolution (2.2°) and accuracy than C3_pole at T42 (2.8°).
We have included some more text to clarify this.

We have changed the text to:
"Studies involving stratospheric polar cap transport, e.g. Arctic and Antarctic O3 loss studies, will clearly benefit from using the C3_pole version of CTM3 combined with 30 min operator-split time step in C3_1/2, depending on the required accuracy at the vortex. When reduced accuracy at the vortex is acceptable, e.g. when computational costs would otherwise prevent a calculation, the standard set-up C3 should be used. However, at higher horizontal resolutions, e.g. T159, the need of C3_pole may be reduced because C3 will have better effective polar resolution (2.2°) and accuracy than C3_pole at T42."

Pg 1596: Sect 4.2: The discussion related to the influence of changing the L-NOx profile is so short as to be uninformative, especially as it is a CTM3 vs CTM3 comparison. Without comparing model distributions against satellite NOx distributions and in-flight O3 measurements it is not possible to determine if any improvement occurs directly from changing the Li-NOx profile.

After localizing a bug in the input data, the effect of shifting from Pickering to Ott vertical profiles is in fact about half of what we showed in Sect 4.2. Therefore we have condensed section 4.2 to fit into the lightning section, and removed Figure 11. Due to this, the Section 4 is now called "Transport sensitivity studies" The C3_PIC has been removed from Table 2.

Text added to section 2.6 is listed at the top of our reply.

If only satellite NO2 (not NOx) and aircraft O3 measurements would help us quantify L-NOx and its impact on O3, we would have done that a decade ago. Yes, they provide critical constraints, but there is still no agreement on L-NOx – alas. The study noted in this section was intended as a sensitivity study to determine IF details of the L-NOx profile (not the absolute amount or location) is important for CTM3. As already mentioned, we have shortened section 4.2 and moved it to section 2.6, noting the effect of shifting from Pickering to Ott vertical profiles.

Pg 1597, ln 4-5: Well there is an improvement regarding model speed-up for a start. The improvement regarding the distribution of trace species is a rather difficult to determine from the paper. This sentence gives doubt as to whether it was worth including extra complexity to the model (excluding the improvements to parallelization).

We thank the reviewer for pointing this out, and have changed the text accordingly. We still keep the last sentence, because it refers to the evaluation, not to the speed. The new text:
"We have presented the chemical transport model Oslo CTM3 and documented the changes from the earlier version Oslo CTM2. The Oslo CTM3 provides a large improvement over CTM2, due to updated and more physically based parameterizations, cleaner code, and increased computational efficiency due to the new structure. The latter makes the Oslo CTM3 better suited for parallelization and up to 40 % faster, enabling much higher spatial resolution or multi-year ensemble/sensitivity simulations. CTM3 also has improved the representation of sub-grid processes such as convection, scavenging, and fractional cloud cover. Our evaluation of the Oslo CTM3 against the Oslo CTM2 and against measurements shows that the new version is as good as, if not better than the old model, and is much more capable, enabling ready advancement to T159 resolution (1.1°)."

Pg 1597, ln 17: Too subjective.

Changed to 20-50% less.

Pg 1598, ln 27-29: Move this paragraph up to the beginning of the conclusions. There is nothing in the conclusions about the age of air which should appear before the discussion on scavenging. Each section of a paper introducing results should have a sequential conclusion synthesized in this final section.

We agree, and have moved the paragraph.
We have included a paragraph on age of air in the conclusions:
"The oldest stratospheric age of air in CTM3 is about 5.8yr, slightly lower than in CTM2 (5.9yr). The spatial distributions are similar, except close to the southern hemisphere polar vortex edge. We have shown that reducing the transport time step causes CTM3 to converge to CTM2 results at equivalent time steps. "

Table 1: The title states “improvements” but point f doesn’t sound like an improvement.
I would use the word “model updates”. Many of these improvements are also based on previous work so remove the “New” (use “ Updated ..”) and provide the reference for e.g. the wet scavenging in a separate column next to each point.

Good idea. The references are given as text in each point. We have removed point f, which was not an update.

Table 2: The last column doesn’t provide much information and could be included in the Table legend (All simulations use an operator time-step of 60 mins apart from C3_30MIN).

As we have included some text on the operator split time step versus the U-flux time step, we keep this column and add the U-flux time step also. The U-flux time step is the global time step for the horizontal transport operations, and is the step mainly controlling the overshooting of the jets. This inclusion is to point out the flexibility of CTM3, where the user has larger control over the accuracy and model speed. A discussion is included in Section 2 and also in Section 4.

As noted by the other referee the figures need to be improved in many instances to be used as evidence of an improvement due to the model updates.

We have improved the figures.

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