

Reply to Referee #2

P1163 L7. What other options were considered for washout rates? What AOD observations were used, and how much did the agreement improve with this choice? Has there been any evaluation with observations over land?

MODIS AOD observations were used. The last sentence of Section 2.2 has been revised to:

“Washout of all aerosols, except hydrophobic black carbon and organic carbon, is set to 20% of the washout rate of HNO₃ (Horowitz, 2006, Tie et al., 2005). Comparison of calculated aerosol optical depth (AOD) over ocean to AOD retrievals from the MODIS satellite instrument indicate this is a reasonable washout rate.”

We have made comparisons to AERONET measurements over land, however it is difficult to use those results to evaluate the washout.

Sections 2.1 and 2.2. If there are known areas where the mechanism and aerosol chemistry is most uncertain, this could be acknowledged. For example, isoprene oxidation chemistry is rapidly evolving with new field and lab experiments suggesting that current schemes are incorrect, with implications for both gas-phase chemistry and secondary organic aerosols.

Global modeling is inherently uncertain and a discussion of the uncertainties of aerosol and chemical schemes could be a paper in itself. The point of this paper is to document what is currently included in MOZART-4, and I do not think it is necessary to include discussion of possible new schemes.

Section 2.3. How different are the results for some key photolysis rates (e.g., J O1D) when FTUV is used rather than the lookup table? It might be clearer to state at the beginning that a major improvement here is the inclusion of aerosols in the photolysis calculations, in addition to the on-line (vs. lookup table) approach.

The first couple of sentences of Section 2.3 have been re-written to emphasize the improvement of including aerosol impacts. The differences between FTUV and TUV are given in Tie et al., 2003. It is difficult to generalize the differences in MOZART-4 between FTUV and the lookup table, as they depend on the presence of aerosols and clouds.

Section 2.5. A more explicit description of “strongly dependent on the soil moisture” (L14) and “vegetation-dependent” (L16) would be helpful, as would a rough idea of how variable the deposition velocities are when calculated online as compared to the climatology.

The soil moisture dependence is taken directly from Sanderson’s paper:

v = dry deposition velocity (10–4 m s⁻¹), x = soil moisture content (as volume ratio, i.e., volume of water per unit volume of soil), z = log_e (soil moisture content). Savannah: $v = az^2$

+ $bz + c$, $a = 0.270$, $b = -0.472$, $c = 1.235$. Agricultural land: $v = bx + c$, $b = -41.39$, $c = 16.85$. Forest: $v = bx + c$, $b = -41.9$, $c = 19.7$. Grasslands: $v = bx + c$, $b = -41.39$, $c = 17.7$.

We have not included these equations in the revised text, but do include a more specific reference and explanation. The vegetation dependence for PAN deposition is taken from (Sparks, J. P., J. M. Roberts, and R. K. Monson, GRL, 2003) so this reference has been added.

Section 2.7. It seems appropriate to briefly comment on the major differences in switching isoprene emission factor maps and the vegetation maps (range of emissions from Pfister et al 2008 seems to be a factor of 2 from their Table 2), and why the combination used here is preferred.

We have used the vegetation maps that have been adopted for use by the NCAR CCSM, so as to provide consistency with them. Also, these maps resulted in emissions totals that fall between the other 2 maps considered in (Pfister et al., 2008) and closest to currently accepted global isoprene totals. We use the most recent (ver. 2.1) emission factor maps; the differences from ver 2.0 are not large. Text added clarifying this.

Section 2.8. For comparison with implementations in other models, consider giving the dependence on the soil dryness dependence more specifically. What is the contribution from fertilizer use to total global soil NO emissions? Please describe how the vertical distribution of lightning NO has been modified from Pickering et al [1998].

The soil dryness dependence is directly from Yienger and Levy (1995).

NO from fertilizer contributes about half of the NO soil emissions from agricultural lands (also from Yienger and Levy).

We already state that the lightning vertical distribution has lower emissions near the surface (so is no longer a “c-shape”). The result is similar to that used by DeCaria et al. (2005), so that reference is added.

P1169 L24-26. It'd be helpful to make a stronger recommendation as to when to use the SYNOZ tracer, and how to judge whether it is needed (P1170 L13-15 should explain how that conclusion was reached)

The primary determination was that ozone was too high in the upper troposphere compared to ozonesondes. This statement has been added to Sec 2.10.

Section 3. As suggested by the first reviewer, this section could point out where there are changes adopted (or not) from these prior studies or updates reflecting newer information since those studies, such as in the chemical mechanism (e.g. rate differs from Pfister et al 2008a Table 1 for ISOP₂+NO and several differences from Horowitz et al 2007).

The ISOP₂+NO reaction rate has been increased based on some recent measurements. No other significant changes have been made to the chemical mechanism or other model processes since previous published studies.

Section 5.1. The methane lifetime is heavily weighted towards the OH in the tropical

lower troposphere, so it'd be useful to also provide the methyl chloroform lifetime and compare both to those derived from observations [Prinn et al., 2005, Geophys Res Lett, 32, L07809]. Further, as suggested by the first reviewer, the relative distribution of the OH in MOZART-4 vs. the climatology could be discussed, with suggestions for what contributes to the differences.

We do not have the methyl chloroform lifetime for this simulation. Some discussion of OH vs the climatology has been added, also in response to reviewer 1.

P1175 L1-2. Some elaboration on the variation of the bias with season would be appropriate. For example, over the U.S., the model is too low in winter, but seems too high in summer (the summer overestimate is consistent with the Hudman et al [2008, Geophys Res Lett, 35, L04801] findings from the ICARTT field campaign).

As the comparison with MOPITT is primarily evaluating the model performance in the free troposphere it is difficult to distinguish emission errors from model transport (boundary layer and convection) errors. These plots show the general ability of the model to reproduce observations, and comparison with field campaigns, for example, for case studies must be used to further identify the causes of regional differences. Some discussion has been added to Section 5.2.

P1177 L3-5. This broad statement doesn't seem consistent with some of the problems highlighted in earlier sections.

We don't feel this is inconsistent and feel it unnecessary to restate all of the problems.

Figure 3. How typical is this profile? Some context should be given in the text.

The point of Fig. 3 is simply to show the relationship between SYNOZ, O3 and O3RAD. That relationship between these variables is always the same.

Figure 5. Any suggestions for what is causing the model underestimate in 2003 at Tenerife and Mauna Loa, where it otherwise seems to capture the yearly variability?

The underestimate of CO at Tenerife and Mauna Loa during Spring 2003 is probably due to the strong biomass burning in Siberia during that time. Sentence added to Section 5.2.

Technical corrections:

P1169 L18-20. Is some stratospheric chemistry being done online, or is there a parameterization to aerosol surface area density being used?

The chemical mechanism, along with the heterogeneous reactions, including hydrolysis of N2O5, operates throughout the model domain, including the stratosphere. Sentence added to Sec.2.10.

P1174 L19-20. Is the MOPITT averaging kernel sensitive near the surface? If not, then would releasing the emissions at the surface lead to an apparent underestimate?

No, the MOPITT averaging kernel is not sensitive to the surface. An error in the modeled vertical distribution may look like an error in the total column amount because

of the non-uniform vertical sensitivity of the MOPITT retrievals.

Section 4.1 Do the anthropogenic emissions include hourly, daily or seasonal variability? Which years are included in the REAS inventory? When only one year is given (e.g. Bond et al 2004 inventory; aviation and military traffic), are those emissions applied as annually invariant, or is some scaling assumed? It'd be useful to have a sentence stating which emissions vary on what time scales.

The anthropogenic emissions that have been used are only available on a yearly basis, without seasonal variation.

The REAS inventory is used for each year. No scaling is applied to the other anthropogenic emissions. Sentence added.

Table 2. Is there a reason why some species are given comments?

Comments are included where explanation of lumped species is warranted, and for common names.

Tables 9 and 10. The 2006 totals disagree for ISOP and C10H16, possibly an issue with Tg C vs. Tg species?

Table 9 has been corrected.

Figure 7. Please provide the latitude and longitude ranges for the regions considered.

Will be added.