

Response to the review from Dr. Nick Savage

We would like to thank Nick Savage for his review of our paper and respond in the following way. The reviewer's comments are put in italics. Suggested changes to the text are given in quotation marks.

Title Please include the version of the IFS used in the title of the paper as required by GMD (CY40r1)

We would prefer not to include the cycle number of the IFS in the title of the paper because the IFS cycles (of which there are about 2 - 3 per year) reflect the development of the NWP code but are not yet linked to the development of the chemistry modules. The current chemistry scheme has been used for several IFS cycles. It is the plan to introduce a proper version naming convention for the chemistry modules in the Copernicus Atmosphere Monitoring Service.

Section 2.2 How large an impact does the correction of negative MMRs have on the budget of transported species?

Negative interim MMR can occur because of the chosen operator splitting and lack of implicitness in the parameterisations and numerical solvers. They occur more often when a large time step is chosen. Using the Quasi-monotonic option for the Semi-Lagrangian Advection scheme avoids negative concentrations after the advection therefore quasi-monotonic restricting of the interpolation result is equivalent to a negative fix. To answer the questions of the magnitude of the fix to chemical zero, we compared the source because of the chemical zero fixer to the largest of the four global sink and source terms (emission, dry and wet deposition, chemistry). For the majority of the species the contribution was below 0.1% . The negative fixer source was in the order of 1% for nitrogen species such as NO, N₂O₅ as well as up to 3% for highly soluble species such HNO₃, HO₂, NO₃_A. We suspect that areas of large gradients such as for the NO_x chemistry at the terminator in the stratosphere as well as intensive wet deposition are the reasons for the need to apply the negative fixer under these circumstances.

Section 2.3 Are the biogenic emissions calculated from MEGAN offline and read from a file or is MEGAN used online?

The MEGAN biogenic emissions were calculated off-line and accumulated in monthly means. We plan to use on-line calculated values in the future. We will change the text as follows:

“The biogenic emissions were simulated off-line by the MEGAN2.1 model (Guenther et al., 2006). The anthropogenic and natural emissions were used as monthly means. Daily Biomass ...”

Section 2.4.3

Please note that Price and Rind (1994) derived a correction factor for cloud top height as the resolution of the model is decreased. They found that as resolution decreases the global lightning frequencies decrease exponentially and corrected for this with a calibration factor. Was this calibration factor used in this implementation?

We are aware of the paper (as the factor is used in the MOZART CTM) but we did not use it. We suspect that the correction factor also depends strongly on the parameterisation of convection in the model. Instead we scaled the emission to be 4.9 Tg yr⁻¹ at a T159 (110 km) resolution. Running the model at T255 (80km) resolution - as done for the paper - led to an increase (without changing coefficients in the parameterisation) to 5.7 Tg yr⁻¹. Please note that the parameterisation of Meijer et al. 2001 based on convective precipitation was used.

Section 2.5.1 Please give more details of the heterogeneous chemistry of SO₂ - what reactions are included, how is the pH calculated and how is the relationship of pH and reaction rate calculated? For the heterogeneous conversion of N₂O₅ into nitric acid how is the surface area of aerosols calculated - does it account for hygroscopic growth? Which aerosols are used - the prognostic scheme in the IFS or a climatology? Is the surface area of water droplets based on a calculated size distribution or is there an assumed size distribution used in the cloud scheme?

Then heterogeneous chemistry is treated in a simple way and it follows the implementation of the TM5 model as describe in Huijnen et al. 2010. We will rephrase the corresponding text as follows:

“For the loss of trace gases by heterogeneous oxidation processes, the model explicitly accounts for the oxidation of SO₂ in cloud through aqueous phase reactions with H₂O₂ and O₃, depending on the acidity of the solution. The pH is computed from the SO₄, MSA, HNO₃, NO₃_A, NH₃ and NH₄ concentrations, as well as from a climatological CO₂ value. The pH, in combination with the Henry coefficient, defines the factor of sulphate residing in the aqueous phase, compared to the gas phase concentration (Dentener and Crutzen, 1993). The heterogeneous conversion of N₂O₅ into HNO₃ on cloud droplets and aerosol particles is applied with a reaction probability (γ) set to 0.02 (Evans and Jacob, 2005). The surface area density is computed based on a climatological aerosol size distribution function, applied to the SO₄, MSA and NO₃_A aerosol, as well as to clouds assuming a droplet size of 8 μ m.”

Dentener, F. J. and Crutzen, P. J.: Reaction of N₂O₅ on tropospheric aerosols: Impact on the global distributions of NO_x, O₃ and OH, J. Geophys. Res., 98(D4), 7149–7163, 1993.

Section 2.5.4 Please explain in more detail what data is used to prescribe surface methane concentrations - are they based on observations or a model (and give a reference). Spatially is a single number used everywhere, is a zonal mean or a 2D fields? Temporally is there a seasonal cycle or is it the same all through the year? (I note that it is mentioned later that monthly zonal mean concentrations are prescribed, but still no reference, and it would be better to say that here)

CH₄ is nudged towards zonal-mean monthly varying surface concentrations derived from a latitudinal interpolation of the monthly-mean observations at the stations South Pole, Cape Grim, Mauna Loa, Mace Head, Barrow, and Alert. These are the same CH₄ surface concentrations as used in the work of Bândă et al. (2015)

We will add the following in section 2.6

“The CH₄ surface concentrations were derived from a latitudinal interpolation of observations from the stations South Pole, Cape Grim, Mauna Loa, Mace Head, Barrow and Alert, as discussed in Banda et al. (2014)”

Banda, N., M. Krol, T. van Noije, M. van Weele, J. E. Williams, P. Le Sager, U. Niemeier, L. Thomason, and T. Röckmann (2014), The effect of stratospheric sulfur from Mount Pinatubo on tropospheric oxidizing capacity and methane, J. Geophys. Res. Atmos., 119, doi:10.1002/2014JD022137.

Section 3.1 One model resolution is given here a spectral truncation and the other in lat-lon spacing. It would be useful here if the approximate resolution of both is given in km to make it easier to compare the resolution of the two models.

We will add the approximate resolution in km in brackets. It was 80x80 km for C-IFS CB05 and about 120 x 120 km for MOZ.

Section 3.2.1 How is the model sampled for comparisons with MOZAIC data?

The model column is obtained over the airport at the middle time between start and end of the profile observation. The columns are interpolated in time between two subsequent output time steps. This procedure does not take into account the horizontal movement of the plane, which could be up to 200 km during the ascent and descent.

“The model column for the comparison with the profile was obtained at the middle between start and end time of the profile observation. The model columns were interpolated in time between two subsequent output time steps.”

"Only the rural Airbase O3 observations have been selected for the evaluation of the diurnal cycle". Please clarify - two plots for surface ozone over Europe are shown - a seasonal and a diurnal cycle. I assume that both of these analyses were carried out using all EMEP data and the Airbase observations at rural sites. If this is correct, this would be better phrased as "For evaluation over Europe, the EMEP observations and the rural Airbase O3 observations were used."

The assumption is correct. We will change the text following your suggestions.

Section 3.3 Does the lower bias in C-IFS imply that the Cariolle scheme is performing better in the lower stratosphere than the MOZART chemistry? If so, please comment on this and implications for future choices of upper boundary conditions for ozone - if the Cariolle scheme is cheaper and better, why are you planning to add a detailed stratospheric chemistry scheme to CB05?

Please note that the stratospheric ozone was nudged to the MACC re-analysis above the tropopause (see section 2.5.4). As shown in Flemming et al., (2011a), both the MOZART stratospheric chemistry and the Cariolle parameterisation have specific issues and strengths. We are planning to implement stratospheric chemistry schemes not only to provide boundary conditions for the troposphere and to assimilated total columns ozone observations but also to provide more specific information about stratospheric composition in CAMS.

Section 3.4 I would say that the model reproduces well only the location of the global maxima (the manuscript correctly identifies the commonly observed underestimation of CO in the NH later on).

We will correct the text as follows:

“ reproduced well the location of the observed ...”

Section 3.5 The low bias in the outflow regions may also be related to insufficient production of NOx reservoir species such as PAN and alkyl nitrates.

We agree that limitations of the chemical mechanism CB05 and not only the emissions are the reason for the underestimation. For example, Alkyl nitrates are not considered. We will add the following line:

“Further, the insufficient simulation of NO_x reservoir species such as PAN and the lack of alkyl nitrates in CB05 might be a reason for the underestimation.”

The overestimation of NO₂ in the biomass burning region coupled with the underestimation at this time suggest that the emissions modelling may be the issue here rather than the fire count.

As pointed out by the reviewer, emission factor for NO may need to be re-considered. We will add:

“The overestimation during biomass burning events could be related to the assumed NO emission factor.”

Section 3.6 Is the underestimation of winter HCHO in Eastern US possibly linked to the ozone bias here as well? Is there some important winter time chemistry missing in both CB05 and MOZAIK?

Without further investigation we find it difficult to give a good explanation of the HCHO underestimation over the eastern US in January and February. Emissions of VOC as well as limitations of the chemical scheme are a possible explanation. The good match in the rest of the years is however encouraging. Uncertainties in the satellite retrievals could also play a role here. The observed values in December are much lower (and the model shows better agreement) than in January, when the model underestimates.

Section 3.8 I found the following sentence confusing. "The additional resources allocated to the IFS are however mostly latent as the coupled MOZART model and the coupler software could not be made faster by using more resources." I think what is implied here is that in order to get enough memory, the coupled MOZART model needed to be run using a large number of CPUs. However, this is inefficient because there is insufficient parallelism in the coupled model to exploit this large CPU count. Please clarify.

The assumption is correct. We will re-phrase the text as follows.

“However, there is insufficient parallelism in MOZART to exploit a larger number of CPUs for speeding up the simulation of the coupled system.”

The more complex chemistry schemes presumably require more resources to run because of both the costs of tracer transport and the extra chemistry. It would be helpful to indicate how the additional costs are spread between these two aspects.

Most of the increase in cost between C-IFS CB05, C-IFS MOZART and C-IFS MOCAGE are because of the demands of the chemistry. The overhead because of the advection of more tracers (CB05 56 tracers, MOZART 108, MOCAGE 112) is in the range of 10%. The semi-Lagrangian advection is very efficient to simulate a larger number of tracers as the departure point is the same for all tracers.

We will add:

“The overhead because of the doubled number of advected species in C-IFS RACMOBUS and MOZART is small because of the efficiency of the semi-Lagrangian advection scheme. “

Section 4 If a consistent chemistry scheme were used in the C-IFS to that of the coupled framework, this would help resolve whether the improvements in SO₂ for example

are due to difference in the diffusion schemes as suggested in the paper.

We agree that a scientifically sound comparison of the MOZART and CB05 chemistry is only possible if the two schemes are implemented in C-IFS. This is now the case and work on this has started.